Very Dilute Aqueous Solutions — Structural and Electromagnetic Phenomena

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Abstract

Very dilute aqueous solutions have properties which are of importance for wastewater treatment, toxicology and pharmacology. Such solutions typically have concentrations below about 10^{-6} - 10^{-10} mol/liter. According to the customary theories, such solutions and distilled water should have similar structural, physicochemical and biological properties. However, previous experimental studies revealed that this is not always the case. When these solutions are prepared by serial dilutions and vigorous shaking after each dilution step, their properties may considerably differ from those of distilled water. Their properties may also significantly differ from those of serially diluted solutions with the same chemical composition, which were not vigorously shaken after each dilution step. In the current study, the following phenomena of serially diluted, vigorously shaken aqueous solutions are analyzed: (a) their self-similar topology, (b) their emission of ultra low frequency (ULF) radiation; (c) 7.85 Hz alternating magnetic fields affecting their structure, and (d) their structure after dilution below 10^{-24} mol/liter. Since ambient electromagnetic fields affect these phenomena, the analyses are carried out within the context of quantum electro-dynamics (QED). The analyses show that the QED model for serially diluted, vigorously shaken aqueous solutions, as developed by Yinnon and Yinnon [*Int J Mod Phys B* 25:3707-3743 (2011)], provides consistent explanations for phenomena (a)-(d).

Introduction

During the last two decades, research on serially diluted, vigorously shaken aqueous solutions intensified, as reviewed by Konovalov and Ryzhkina (2014) and Elia *et al.* (2015). These solutions are prepared from stock solutions with concentration (C) in the ~4 - 10^{-3} mol/liter (M) range. The stock solutions are repetitively (serially) decimally or centesimally diluted, at ambient conditions. After each dilution step, the liquids are vigorously shaken with a lab dancer shaker, by vertical vortexing or other methods.

The study of serially diluted, vigorously shaken aqueous solutions was stimulated by their now amply demonstrated significance for water reclamation, toxicology and medicine. The bioactivity of solutions of many (but not all) inorganic-, organic- or bio-molecules non-monotonically changes upon serial diluting to low (picomolar) or ultralow (femtomolar) concentrations or beyond (Burlakova *et al.*, 1986, 2004,

2005; Davenas *et al*, 1988; Palmina *et al.*, 1994; Pynzar *et al.* 1995; Lobyshev *et al.*, 2005; Ryzhkina *et al.*, 2010, 2011a; Belov *et al.*, 2011; Konovalov, 2013; Mishina *et al.*, 2015; Voeikov and Yablonskaya, 2015). The effects of serially diluted, vigorously shaken aqueous solutions on bio-systems may follow an upside-down U-shaped concentration dependent response curve. Such a curve is observed for many but not for all solutes. The slope reversal appears at a solute type dependent threshold concentration (C_{thr}). Typically, C_{thr} is of the order of about 10^{-6} - 10^{-10} M.

Compounds released by glass containers are present in serially diluted, vigorously shaken aqueous solutions (Witt *et al.*, 2006). Such compounds and other contaminants affect the physicochemical variables of these liquids (Elia and Niccoli, 2004a&b). However, in depth analyses of such liquids prepared in glass or plastic vessels show that such compounds and contaminants alone cannot account for their typical properties (Elia and Niccoli, 2004a&b; Ciavatta *et al.*, 2008; Montagnier *et al.*, 2009; Elia and Napoli, 2010; Upadhyay and Nayak, 2011; Demangeat, 2009, 2010; Pershin *et al.* 2015; Yinnon and Liu, 2015a,b).

Many aspects of serially diluted, vigorously shaken aqueous solutions have been measured by independent research groups. The main aspects are cited in the next section. The most pronounced aspect is that for some (but not for all) solutes, at C below C_{thr}, their physicochemical, structural and bioactive properties differ from those of serially diluted solutions with equivalent chemical composition, which were *not* vigorously shaken after each dilution step (Elia and Niccoli, 1999, 2000, 2004a; Elia *et al.*, 2004b). Solute attributes underlying the differences are not yet clarified (Konovalov, 2013; Ryzhkina *et al.*, 2015a; Yinnon and Liu, 2015c). In this paper, the denotation SDVSAS (serially diluted, vigorously shaken aqueous solutions) will be used for referring to the following liquids: Solutions which were prepared by serial dilutions and vigorous shaking after each dilutions step, for which at $C<C_{thr}$, their measured physicochemical properties significantly differ from those of serially diluted solutions with equivalent chemical composition which were not vigorously shaken after each dilution step.

Modeling SDVSAS requires a quantum electro-dynamic (QED) approach. Electromagnetic (EM) radiation majorly affects the structure and physicochemical properties of these liquids (Ryzhkina *et al.* 2011b, 2012a-c; Konovalov, 2013). For example, for $C<C_{thr}$, ambient EM radiation underlies formation of their 10^{-7} m sized nano-associates. Such large molecular associates are manifestations of QED effects (Preparata, 1995; Yinnon and Liu, 2015a; Fiorini, 2016). The QED model of SDVSAS, proposed by Yinnon and Yinnon (2011), has provided consistent explanations for many of their phenomena (Yinnon and Elia, 2013; Yinnon and Liu, 2015b,c).

The goal of this paper is to employ the QED model for clarifying some hitherto unexplained characteristics of SDVSAS reported in the literature. In-depth explanations for ultra violet (UV) and infra red (IR) spectral features of SDVSAS will be presented in a separate publication. Some reported characteristics will not be analyzed. Seemingly not all researchers are familiar with the hysteretic and far-from-equilibrium dissipative system properties of SDVSAS. These properties have implications for assuring SDVSAS phenomena are significant. For example, the following parameters affect the physicochemical properties of SDVSAS samples: volume, storing conditions, the passage of time after preparation of the samples, shaking procedure, ambient EM radiation, temperature, chemical composition of the stock solutions from which SDVSAS are prepared and the purity of the water used for its dilutions (Elia *et al.*, 2004a,

2008a&b, 2012; Lobyshev *et al.*, 2005; Yinnon and Elia, 2013; Rhyzkina *et al.*, 2010, 2011a-c, 2012a-c, 2013, 2015c; Konovalov and Ryzhkina, 2014; Mishina *et al.*, 2015). The group of Miranda *et al.* (2011) demonstrated that the electrical impedance of SDVSAS of LiCl samples varies when prepared with distilled water procured from different laboratories. The group carefully controlled the SDVSAS preparation procedures, their storing conditions and their impedance measurements. Another implication of the hysteretic and far-from-equilibrium dissipative system properties of SDVSAS is that the controls (blanks) should be carefully designed. Perturbing pure water with serial dilutions and vigorously shaking after each dilution step also alters their structure and physicochemical properties (Elia *et al.*, 2000, 2004a; Lobyshev *et al.*, 2005; Rhyzkina *et al.*, 2015b).

The outline of the paper is as follows: It starts with a concise overview of the main measured properties of SDVSAS. The overview is important for appreciating the issues presented in the subsequent sections. Next, in the "Theory Section", the SDVSAS model is concisely summarized.[a] In the subsequent section, *i.e.*, the "Analyses of Reported Data Section," explanations are presented for the following phenomena: the self-similar topology of SDVSAS, the emission of ultra low frequency (ULF) radiation by SDVSAS, 7.85 Hz alternating magnetic fields affecting the structure of SDVSAS, and ordering of H_2O in SDVSAS diluted below the Avogadro limit. After that, in the "Discussion Section," the strengths and the shortfalls of the current version of the SDVSAS model are discussed.

In this section, also hypotheses concerning the bioactive principle in SDVSAS are addressed. No new experimental results are presented in this paper. All the experimental results, which will be analyzed, were obtained in previous studies and reported in the literature. A list with abbreviations is presented at the end of this paper.

Measured Properties of SDVSAS

During the last decades, many properties of SDVSAS have been measured. For reviews, see Konovalov and Ryzhkina (2014) and Elia *et al.* (2015). These properties have been consistently explained with the QED model (Yinnon and Yinnon, 2011; Yinnon and Elia, 2013; Yinnon and Liu (2015a-c). The main measured properties of SDVSAS, pertaining to the goals of this paper, are:

1) At $C<C_{thr}$, molecular associates are present in SDVSAS (Elia and Niccoli, 1999, 2000, 2004a; Elia *et al.*, 2004b). The associates are mainly composed of solvent molecules (Elia and Niccoli, 1999, 2000, 2004a; Elia *et al.*, 2004b; Ryzhkina *et al.*, 2011a-c, 2012a-c, 2013,2015a&b; Konovalov 2013; Konovalov and Ryzhkina, 2014). Conversely, in a serially diluted solution made of the same solute but which was not vigorously shaken, at $C<C_{thr}$, all their molecules are randomly distributed and participate in the Brownian movement.

2) Hysteretic properties (Samal and Geckeler, 2001; Elia *et al.*, 2004a, 2005, 2008 a&b; Lobyshev *et al.*, 2005; Belon *et al.*, 2008; Konovalov and Ryzhkina, 2014; Miranda *et al.*, 2011).

3) For $C<C_{thr}$, the electrical conductivity (χ) and the heat of mixing with acids or bases (Q) are linearly correlated (Elia *et al.*, 2004b; Belon *et al.*, 2008; Elia *et al.*, 2015). At this C range, the pH and log χ are also linearly correlated (Elia *et al.*,

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[a] Elaborate discussions on the various aspects of the model are published in Yinnon and Yinnon (2011), Yinnon and Elia (2013) and Yinnon and Liu (2015b&c). Since QED of aqueous solutions has hitherto mainly been employed for explaining special phenomena, many readers may be unfamiliar with it. Its aspects relevant to this paper's analyses are concisely summarized in Yinnon and Liu (2015a).

2015). The correlations indicate that a single cause underlies the specific properties of SDVSAS (Elia *et al.*, 2015, 2016).

4) The quasi-periodic variations with macroscopic times (months and years) of the χ, Q and pH values of SDVSAS indicate that these liquids are far-from-equilibrium dissipative systems (Elia *et al.*, 2000, 2008a&b; Belon *et al.*, 2008; Elia *et al.*, 2015). Hence, SDVSAS properties are not quantitatively reproducible but are qualitatively repeatable. It should be stressed that SDVSAS properties depend on the time of their storage, the volume of the samples and ambient EM fields.

5) For $C<C_{thr}$ but above 10^{-24} M (*i.e.*, above the so-called Avogadro limit), EM radiation crucially affects the structure, physicochemical properties and bioactivity of SDVSAS (Ryzhkina *et al.* 2011b, 2012a-c; Konovalov, 2013). These SDVSAS lose their characteristic properties when stored for many hours (24 h or more) in a Permalloy container, *i.e.*, under hypo electro-magnetic conditions (Konovalov and Ryzhkina, 2014). Within Permalloy containers the residual field is about 10 - 50 nano Tesla. In other words, on shielding these SDVSAS from some ambient EM radiation, their properties become the same as those of solutions with equivalent chemical composition but which were not prepared by serial dilutions and vigorous shaking after each dilution step.

6) The dependence of the bioactivity of SDVSAS on the number of dilution steps (NDS) is interrelated with their physi-cochemical and structural properties (Lobyshev *et al.*, 2005; for review see Konovalov and Ryzhkina, 2014).

7) The sizes of the associates in SDVSAS at $C<C_{thr}$ are in the range of 10^{-7} - 10^{-4} m (Ryzhkina *et al.*, 2010, 2011a-c, 2012ac-c, 2013; Konovalov and Ryzhkina, 2014; Elia *et al.*, 2015). The associates underlie the peculiar properties of these SDVSAS. Second order phase transitions typify their formation (Pershin *et al.*, 2016). Since SDVSAS are far-from-equilibrium dissipative systems, the associates constantly reform. Perturbations (*e.g.*, dilutions and shaking) reorganize their molecules. The reorganization is reflected foremost in the non-monotonic dependence of the dielectric permittivity of SDVSAS on C (*i.e.*, this variable often is significantly affected by each dilution step and by the shaking of the liquid (Ryzhkina *et al.*, 2011c, 2012b, 2013; Lunev *et al.*, 2014; Yinnon and Liu, 2015c)). The reorganization also underlies the non-monotonic dependence of other physicochemical variables of SDVSAS on NDS [*e.g.*, the surface tension, χ, the size and electrokinetic potential (ζ-potential) of the associates (Konovalov and Ryzhkina, 2014; Yinnon and Liu, 2015c)]. In addition, the reorganization underlies the non-monotonic dependence of the bioactivity of SDVSAS on NDS. The aforementioned non-monotonic dependencies are interrelated (Konovalov and Ryzhkina, 2014). The various non-monotonic dependencies have been observed by several independent research groups[b] and were shown not to be attributable to contaminants.

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[b] Miranda *et al.* (2011) showed that the electrical impedance of SDVSAS of lithium chloride (LiCl) depends non-monotonically on these liquids' NDS. Duval *et al.* (2012) showed that the Landau-Placzek ratio, *i.e.*, the ratio of the Rayleigh scattering intensity over the total Brillouin intensity, for SDVSAS of LiCl and for SDVSAS of sodium chloride (NaCl) depends non-monotonically on these liquids' NDS. Pershin *et al.* (2015, 2016) demonstrated that for SDVSAS of potassium phenosan, the following variables were all found to depend non-monotonically on this liquid's NDS: the frequency of the center of the Raman scattering OH vibration band of its H_2O, the Rayleigh line width in its four-wave mixing spectra, its integrated elastic scattering profile of 527-nm laser pulses, its fluctuation of the elastic-scattering coefficient and its optical activity. Kozumi and Kitagawa (2016) demonstrated that the aquaporin permeability of serially diluted, vigorously shaken water kept for 24 h in contact with ceramics non-monotonically changes with the NDS. This liquid was 15 times centesimally diluted. After each dilution step, the aquaporin permeability changed non-monotonically.

Theory

The customary models of water and their solutions predict that serial dilutions, combined with vigorous shaking and exposure to ambient EM radiation, do not affect their characteristics (Horne, 1972; Robinson and Stokes, 2002). In addition, these models predict that H_2O molecules move randomly, with the exception of solvation shells' H_2O. Moreover, these models predict that solvated solutes distribute homogenously, move independently and randomly. These customary models explicitly describe electro-static forces and *assume that electro-dynamic ones can be treated perturbatively* or often even may be ignored. However, QED models *explicitly* (non-perturbatively) describing electro-dynamic forces show that these forces may lead to association of H_2O and solute molecules (Del Giudice *et al.*, 1988, 1998, 2000, 2002, 2006; Preparata, 1995 chapters 2, 5, 10; Arani *et al.*, 1995; Bono *et al.*, 2012; Yinnon and Yinnon, 2012). Interactions between EM radiation and electrolytic solutes, polar solute molecules, the dipole moments of H_2O or the electrons of H_2O may lead to formation of various domain types. Here such domains are denoted QED domains. Formation of these domains occurs only in specific concentration ranges, which depend on solute type. These domains may agglomerate into supra-domains. These supra-domains are not ensembles of molecules but agglomerates of domains, like domains in liquid crystals.

A concise review of the QED theory of water and their solutions has been presented by Yinnon and Liu (2015a). The various hitherto identified QED domains present in these liquids, their schematic pictures, their properties and the physics underlying the dependence of the domains' formation on concentration are all summarized in Yinnon and Liu (2015a). In their review, Yinnon and Liu (2015a) emphasized that the QED theory of water is based on an ex-

plicit non-perturbative description of the van der Waals' dispersion forces. The imperative for adequately describing van der Waals' dispersion interactions and other electro-dynamic interactions in various condensed matter systems has also recently been pointed out by Ferri *et al.* (2015), Fiorini (2016) and Ambrosetti *et al.* (2016).
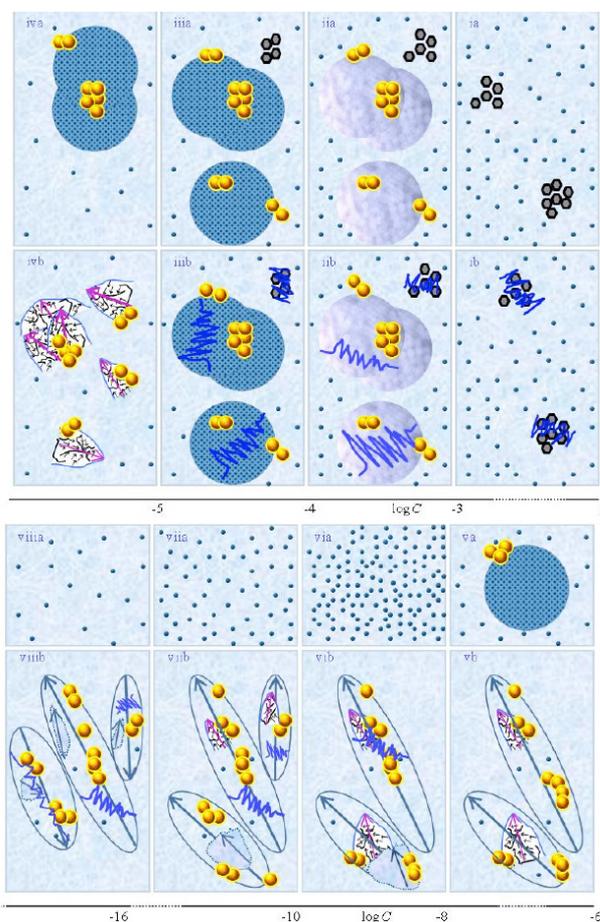

Figure 1. *This figure presents a schematic picture of serially diluted solutions of weak electrolytes or non-electrolytic compounds. The top row (a) and bottom row (b) series pertain to solutions which, respectively, were not vigorously shaken and those which were vigorously shaken after each dilutions step.* Figure ia *illustrates that for C larger than a transition concentration $C_{trans}^{CDplasma}$, all solvated solutes move randomly, i.e., do not organize in a QED domain. The tiny blue balls represent randomly moving ~10^{-9} - 10^{-8} m solvated solutes. The irregularly shaped bunches of black hexagons represent aggregates of non-solvated solutes.* Figure iia *illustrates that on dilution below $C_{trans}^{CDplasma}$, solvated solutes organize in a QED domain type denoted CD_{plasma} (symbolized with purple-blue colored balls). The yellow-brown balls*

and their agglomerates represent, respectively, $\sim 10^{-7}$m sized QED domains denoted $CD_{elec}^{H_2O}$ and supra-$CD_{elec}^{H_2O}$, which both are stabilized by CD_{plasma}. Figures iia *and iiia illustrate the transformation of* CD_{plasma} *into another type of QED domain denoted* IPD_{plasma}. *The transformation occurs at the transition concentration* $C_{trans}^{IPDplasma}$. *Figures ia-iiia illustrate that on dilution the non-solvated solutes diminish, i.e., solvate. Figures iiia-va illustrate that on dilution the diameter of* IPD_{plasma} *does not change, but the number of* IPD_{plasma} *diminishes. Figures via-viiia illustrate that below a certain concentration there are insufficient solutes to form* IPD_{plasma}. *The concentration below which no* IPD_{plasma} *form has yet not been theoretically derived. Figures via-viiia illustrate that whenever there are too few solutes to form* IPD_{plasma} *the solution has the characteristics predicted by the customary models, i.e., all solvated solutes move randomly and their number diminishes on dilution.*

In the Figure 1b *series, the blue zigzag curves symbolize that shaking excites or cracks domains and aggregates. Figure iib illustrates that excitations or cracking does not significantly alter the internal structure of* CD_{plasma}, *which just as in* Figure 1iia *are represented with purple-blue colored balls. Figures iib and iiib illustrate the transition from* CD_{plasma} *to* IPD_{plasma}, *with the latter pictured as blue-crystalline balls (just as in* Figure iiia*). Figures iiib and ivb illustrate that shaking excites or breaks up* IPD_{plasma}. *The excited or broken* IPD_{plasma} *pieces, which in the text are denoted electric dipole aggregates (*$EDA^{IPDplasma}$*), are pictured as irregularly shaped aggregates in* Figure ivb. *Their aligned black arrows orderings symbolize* $EDA^{IPDplasma}$*'s distorted ferroelectric* H_2O *orderings. The purple arrow in the* $EDA^{IPDplasma}$ *symbolizes these domains' dipole moments. Figures ivb and vb illustrate that on diluting below a solute type dependent critical concentration (* C_{crit}^{CDrot} *) a QED domain denoted* CD_{rot} *becomes stabilized by* $EDA^{IPDplasma}$, *i.e., the irregular shaped* $EDA^{IPDplasma}$ *are located within the elongated ovals representing* CD_{rot}. *The dark blue arrows symbolize the dipole moment of* CD_{rot}. *Figure vib shows that vigorous shaking excites or breaks up* CD_{rot}, *thus creating entities denoted* EDA^{CDrot}. *The lump outlined with an irregular shaped broken curve and located at the bottom of one of the left* CD_{rot}, *represents the* EDA^{CDrot}. *Figures vib-viib show that at certain concentrations both* $EDA^{IPDplasma}$ *and* EDA^{CDrot} *are present within* CD_{rot}, *though the sizes of* $EDA^{IPDplasma}$ *diminish with concentration. Figure viiib shows that on diluting further, no* $EDA^{IPDplasma}$ *persist, i.e., there*

are too few solute particles to sustain $EDA^{IPDplasma}$. *At these concentrations, vigorous shaking just breaks up* CD_{rot} *and creates new* EDA^{CDrot}. *These in turn stabilize new* CD_{rot}, *as pictured in* Figure viiib. *Figures vb-viib illustrate that* CD_{rot} *may align with their dipole moments parallel. Figure viiib illustrates that at certain concentrations their dipoles may be aligned anti-parallel. (Note that the sizes of the various domains, their broken pieces and the sizes of the solvated solutes with their hydration shells are not presented according to their realistic scale ratios.)*

SDVSAS MODEL

Fig.1 presents a schematic picture of the SDVSAS model proposed by Yinnon and Yinnon (2011) (*i.e.,* the structure of SDV-SAS for different *C* ranges). The figure is a reprint of *Fig.1* in Yinnon and Liu (2015c). The figure pertains to SDVSAS of weak- or non-electrolytic compounds. The schematic picture for the model of SDVSAS of strong electrolytes is very similar. It appeared in *Fig. 1* of Yinnon and Liu (2015b). The differences between the models of weak-, non- and strong electrolytes are minor ones, as discussed below in paragraphs I and II. The various aspects of the models are discussed in the following paragraphs I-VI and i-vii. Experimental results, obtained in independent research groups, agree with these aspects, as shown by Yinnon and Yinnon (2011), Yinnon and Elia (2013) and Yinnon and Liu (2015b&c). The few aspects which still have to be definitely confirmed by measurements will be explicitly pointed out below.

I. In solutions of weak- or non-electrolytes, for *C* above a transition concentration $C_{trans}^{CDplasma}$, only few solute molecules solvate. The solvated solutes move randomly (see *Fig.1i*). For $C_{trans}^{IPDplasma} < C < C_{trans}^{CDplasma}$, part of the solvated weak electrolytes, or solvated non-electrolytes with a sufficiently large electric dipole moment, aggregate in a QED domain type denoted CD_{plasma} (see *Fig.1 ii*). Within these domains, the solute molecules together with

their solvation shell H_2O perform coherent plasma oscillations. The diameter of a CD_{plasma} is of the order of 10^{-6} m. CD_{plasma} may agglomerate into supra-CD_{plasma}. A schematic picture of CD_{plasma} is presented in *Fig. 3a* in Yinnon and Liu (2015a). QED theory predicts that the interactions between the solute molecules and Tera Hertz (Hz) to Mega Hz EM radiation underlie formation of CD_{plasma}, but this has not yet been confirmed by experimental data. The transition concentrations $C_{trans}^{CDplasma}$ and $C_{trans}^{IPDplasma}$ depend on solute type. Typically, $\sim 10^{-4}$ M $< C_{trans}^{CDplasma}$ $<$1M and $\sim 10^{-6}$ M $< C_{trans}^{IPDplasma}$ $< \sim 10^4$M.

In solutions of strong electrolytes, at any C larger than $C_{trans}^{IPDplasma}$, part of the solvated solutes together with numerous H_2O organize in CD_{plasma}. Thus, the difference between solutions of weak- or non-electrolytes versus those of strong electrolytes is that in the latter there exist no concentration above which CD_{plasma} do not form, *i.e.*, there is no $C_{trans}^{CDplasma}$.

According to QED theory, CD_{plasma} and supra-CD_{plasma} may stabilize a QED domain type denoted CD_{elec}^{H2O}. These domains may aggregate into supra-CD_{elec}^{H2O}. A CD_{elec}^{H2O} is composed of H_2O, which coherently oscillate between their electronic ground state and an excited electronic state. Interactions between H_2O and UV radiation underlie formation of CD_{elec}^{H2O}. The diameter of a CD_{elec}^{H2O} is about 10^{-7} m. These domains cannot contain solutes. A schematic picture of CD_{elec}^{H2O} is presented in *Fig. 1* in Yinnon and Liu (2015a).

The abovementioned characteristics for $C>C_{trans}^{CDplasma}$ hold independent of the solutions' preparation procedure. In other words, these do not just hold for SDVSAS, but also for solutions prepared without serial dilutions or vigorous shaking. Serial dilutions or vigorous shaking affect CD_{plasma} and CD_{elec}^{H2O}, mainly causing their breakup. However, after perturbations are over these domains reform, as illustrated in *Figs.1 iia&b*.

The energetics of processes induced by vigorous shaking of SDVSAS have been evaluated by Yinnon and Liu (2015b) on pages 59-60 (footnote g). The evaluation indicates that vigorous shaking provides about 10^{10} - 10^{15} eV per aggregate (domain). The energy required for desorption of a molecule from a CD_{elec}^{H2O} is of the order of 10^{-1} eV, while a few eV are required for desorption of a molecule from a CD_{plasma}. Thus, the energy applied to SDVSAS by vigorously shaking is sufficient to breakup CD_{elec}^{H2O} and CD_{plasma}.

II. In solutions of *strong* electrolytes, at $C \leq C_{trans}^{IPDplasma}$, CD_{plasma} transform into another type of QED domain, *i.e.*, IPD_{plasma} (see *Figs.1ii-iii*). QED theory predicts that the interactions between the solute molecules and Tera Hz to Mega Hz EM radiation underlie formation of IPD_{plasma}, but this has not yet been confirmed by experimental data. IPD_{plasma}, just as CD_{plasma}, are composed of few solvated solutes and numerous H_2O. The solvated solutes in IPD_{plasma} are crystalline ordered. The solvated solutes and H_2O constituting IPD_{plasma} perform in-phase plasma oscillations. A schematic picture of IPD_{plasma} is presented in *Fig. 3b* in Yinnon and Liu (2015a). The diameter of an IPD_{plasma} is about 10^{-6} m. IPD_{plasma} may aggregate in supra-IPD_{plasma}. Dilution below $C_{trans}^{IPDplasma}$ diminishes the number of randomly moving solvated solutes as well as the number of solvated solutes incorporated in IPD_{plasma}. At very low concentrations, the number of solutes is too low for formation of IPD_{plasma}. As shown in *Fig. 1iiib*, IPD_{plasma} may stabilize CD_{elec}^{H2O} and supra-CD_{elec}^{H2O}.

The abovementioned characteristics for $C<C_{trans}^{IPDplasma}$ hold independent of the solutions' preparation procedure, *i.e.*, not just for SDVSAS, but also for solutions prepared without serial dilutions or vigorous shaking.

For solutions of weak- or non-electrolytes, for $C \leq C_{\text{trans}}^{\text{IPDplasma}}$, QED theory also predicts that CD_{plasma} transform into IPD_{plasma}. The differences between the transition concentrations $C_{\text{trans}}^{\text{CDplasma}}$ and $C_{\text{trans}}^{\text{IPDplasma}}$ may be tiny. However, definite experimental evidence for these aspects of such solutions has yet to be provided, as discussed by Yinnon and Liu (2015c).

III. For solutions containing IPD_{plasma}, their agitation by vigorous shaking affects their properties. Vigorous shaking transforms IPD_{plasma} into aggregates with an electric dipole moment (see *Fig.1iv*). These aggregates are denoted electric dipole aggregates $EDA^{\text{IPDplasma}}$. The sizes of $EDA^{\text{IPDplasma}}$ may reach 10^{-6} m, but may also be much smaller.

The formation of $EDA^{\text{IPDplasma}}$ results from agitations exciting or breaking up IPD_{plasma}. A few eV are required for desorption of a molecule from an IPD_{plasma}. Since vigorous shaking provides about 10^{10} - 10^{15} eV per (domain) (Yinnon and Liu, 2015b), vigorous shaking of SDV-SAS can break up IPD_{plasma}.

The excitations or break up of IPD_{plasma} partly destroy the spherically symmetric alignments of the dipole moments of the numerous H_2O in the large solvation shells surrounding its crystalline ordered solvated solutes. The partial destruction of the spherically symmetric alignments endows the disturbed IPD_{plasma} with an electric dipole moment.

Vigorous shaking of SDVSAS containing CD_{plasma}, *i.e.*, at $C > C_{\text{trans}}^{\text{IPDplasma}}$, does not create electric dipole aggregates. Solutes are randomly distributed in a CD_{plasma}, *i.e.*, are not crystalline ordered. Moreover, only few H_2O constitute its solvation shells, *i.e.*, only few H_2O are spherically symmetric aligned. Therefore, excitation or break up of CD_{plasma} does not create aggregates with a sizable electric dipole moment.

IV. $EDA^{\text{IPDplasma}}$ may stabilize $CD_{\text{elec}}^{\text{H2O}}$ and supra-$CD_{\text{elec}}^{\text{H2O}}$ (see *Fig.1 iv*).

V. For C less than the critical concentration $C_{\text{crit}}^{\text{CDrot}}$, yet another type of QED domain may form, *i.e.*, CD_{rot}. This domain is composed of ferroelectric ordered H_2O. A CD_{rot} has an electric dipole moment due to the ferroelectric ordering of its H_2O. The H_2O constituting CD_{rot} coherently oscillate between two of their rotational states. Interactions between the H_2Os' electric dipole moments and IR EM radiation underlie formation of CD_{rot}. For a schematic picture of CD_{rot} see *Fig. 2* in Yinnon and Liu (2015a). In bulk water at ambient conditions, CD_{rot} do not auto-organize. The energy gained by a H_2O joining a CD_{rot} is of the same order as the energy of the thermal fluctuations. Therefore, thermal aggression prevents auto-formation of CD_{rot} at ambient conditions. However, objects with sizable asymmetric charge distributions (*e.g.*, macromolecules or hydrophylic membranes) may induce formation of CD_{rot} (*i.e.*, stabilize CD_{rot}). Their stabilization causes a permanent time-dependent polarization of the liquid. The sizes of CD_{rot} may increase with the passage of time. At ambient conditions, their stabilization, such as by hydrophilic membranes, may require many hours or days. Their sizes may reach 10^{-4} m.

Solutes are pulled into CD_{rot}. Few solute particles can locate in CD_{rot} and not wreck their host. Many solute molecules destroy CD_{rot}. The critical concentration below which CD_{rot} persist ($C_{\text{crit}}^{\text{CDrot}}$) depends on the solute, typically ~10^{-10} M < $C_{\text{crit}}^{\text{CDrot}}$ < ~10^{-6} M. Analyses of experimental data of many aqueous solutions have shown that $C_{\text{crit}}^{\text{CDrot}}$ equals C_{thr}.

For $C < C_{\text{crit}}^{\text{CDrot}}$, due to the interactions between the electric dipoles of $EDA^{\text{IPDplasma}}$ and the electric dipoles of H_2O, $EDA^{\text{IPDplasma}}$ stabilize CD_{rot} (see *Fig.1vb*).

In other words, EDA$^{\text{IPDplasma}}$, due to their significant asymmetric charge distributions, stabilize CD$_{\text{rot}}$. Stabilization of CD$_{\text{rot}}$ also may be induced by solutes with sufficiently large permanent or induced electric dipoles. Even when no IPD$_{\text{plasma}}$ and hence EDA$^{\text{IPDplasma}}$ form in SDVSAS, solvated solutes with a sufficiently large asymmetric charge distribution still might induce CD$_{\text{rot}}$ stabilization for $C <$ $C_{\text{crit}}^{\text{CDrot}}$.

Vigorous shaking excites or breaks up CD$_{\text{rot}}$ (see *Fig.1v*). The energy required for desorption of a molecule from a CD$_{\text{rot}}$ is of the order of 10^{-2} eV. Since vigorous shaking provides about 10^{10} - 10^{15} eV per (domain) (Yinnon and Liu, 2015b), vigorously shaking of SDVSAS can break up CD$_{\text{rot}}$. Due to the ferroelectric ordering of the H$_2$O constituting CD$_{\text{rot}}$, excited or broken CD$_{\text{rot}}$ also are electric dipole aggregates, which are denoted as EDA$^{\text{CDrot}}$ (see *Fig.1vi*). Due to interactions between the electric dipoles of EDA$^{\text{CDrot}}$ and the electric dipoles of H$_2$O, EDA$^{\text{CDrot}}$ also stabilize CD$_{\text{rot}}$. Therefore, serial dilutions with vigorous shaking at each dilution step diminish solvated solutes and EDA$^{\text{IPDplasma}}$, but EDA$^{\text{CDrot}}$ persist. These EDA$^{\text{CDrot}}$ stabilize CD$_{\text{rot}}$ and supra-CD$_{\text{rot}}$ too. As a result, CD$_{\text{rot}}$ persists up to ultra-low concentrations and beyond (see *Figs.1vib-viiib*).

CD$_{\text{rot}}$ and supra-CD$_{\text{rot}}$ may stabilize CD$_{\text{elec}}^{\text{H2O}}$ and supra-CD$_{\text{elec}}^{\text{H2O}}$. Such agglomerates are denoted [supra-CD$_{\text{rot}}$ <supra-CD$_{\text{elec}}^{\text{H2O}}$>].

CD$_{\text{rot}}$ may agglomerate into supra-CD$_{\text{rot}}$ with their dipole moments more or less parallel or anti-parallel oriented. The vigorous shaking applied to SDVSAS after each dilution step restructures the orientations of the dipole moments of its supra-CD$_{\text{rot}}$. The restructuring strongly affects the properties of SDVSAS. For instance, it underlies the non-monotonic dependence of its physicochemical variables on C. Moreover, it affects the stabilization of CD$_{\text{elec}}^{\text{H2O}}$ and the formation of supra-CD$_{\text{elec}}^{\text{H2O}}$.

According to the QED model of SDVSAS, when these liquids are slightly or extremely diluted below $C_{\text{crit}}^{\text{CDrot}}$, the entities underlying their extraordinary physicochemical properties are CD$_{\text{rot}}$, EDA$^{\text{CDrot}}$ and CD$_{\text{elec}}^{\text{H2O}}$. In addition, it has been hypothesized that for $C < C_{\text{crit}}^{\text{CDrot}}$ $= C_{\text{thr}}$, CD$_{\text{elec}}^{\text{H2O}}$ underlies the bioactivity of SDVSAS. Therefore, on screening SDVSAS from the ambient EM radiation that mediates formation of their CD$_{\text{rot}}$ and CD$_{\text{elec}}^{\text{H2O}}$, these domains disintegrate and the SDVSAS lose their typical characteristics. Placing SDVSAS in Permalloy containers screens the EM radiation which mediates formation of their CD$_{\text{rot}}$ and CD$_{\text{elec}}^{\text{H2O}}$.

Explaining some measured properties of SDVSAS requires knowledge of several characteristics of CD$_{\text{elec}}^{\text{H2O}}$. Therefore, in the following paragraphs, these characteristics will be concisely summarized from the papers published by Preparata (1995), Arani *et al.* (1995), Del Giudice and Preparata (1998), Del Giudice *et al.* (2002, 2010a&b), Del Giudice (2007), Bono *et al.* (2012) and Montagnier *et al.* (2011, 2015).

i. Within a CD$_{\text{elec}}^{\text{H2O}}$, the photons underlying the electronic transitions of its H$_2$O are condensed (Bose-Einstein condensation). These photons mediate the attractions between its H$_2$O. These condensed photons are trapped and cannot be irradiated outward. Or more accurately, only when a CD$_{\text{elec}}^{\text{H2O}}$ disintegrates, for example due to heating, the trapped photons "evaporate."

ii. The H$_2$O constituting a CD$_{\text{elec}}^{\text{H2O}}$ coherently oscillate between their ground electronic state and an excited electronic state |b>. The energy H$_2$O gain by joining a CD$_{\text{elec}}^{\text{H2O}}$ [*i.e.*, the energy (E_{coh}) a H$_2$O gains by participating in the coherent oscillations]

depends on temperature. At ambient conditions, E_{coh} is of the order of 10^{-1} eV.

iii. For bulk water at ambient conditions, the energy of its thermal fluctuations is about an order of magnitude smaller than E_{coh}. Therefore, its CD_{elec}^{H2O} are meta-stable. The competition between QED interactions and collisions (thermal noise) causes a continuing adsorption and desorption of H_2O from its CD_{elec}^{H2O}. In other words, there are everlasting transitions of H_2O between a coherent and a non-coherent phase. The transitions are like those in superfluidic Helium. The transitions produce a continuously changing space distribution of the coherent and non-coherent fractions of H_2O. For bulk water at ambient conditions, only about 20 percent of its H_2O belong to the former; the time scale of the transitions is below 10^{-14} seconds (s). Therefore, observation of a CD_{elec}^{H2O} requires very fast probes. Indirect experimental evidence for presence of CD_{elec}^{H2O} in bulk water has been provided by De Ninno and Congiu Castellano (2011). Experimental data indicating stabilization of CD_{elec}^{H2O} by membranes or other QED domain types has recently been forwarded (Elia *et al.*, 2015, 2017; Yinnon *et al.*, 2016; Yinnon and Liu, 2015b,c).

iv. Whenever external forces affect a CD_{elec}^{H2O} and the energy transferred to the domain is less than E_{coh}, due to the coherence, the whole domain excites. In other words, these forces do not just perturb one of its molecules but excite the whole domain.

v. A CD_{elec}^{H2O} contains many coherent quasi-free electrons (QFE), because its H_2O molecules oscillate between the ground electronic and the nearly free |b> electronic states.[c] Exciting a CD_{elec}^{H2O} with

energies less than E_{coh}, *e.g.*, 0.17 eV at ambient conditions (4×10^{13} Hz), leads to coherent vortices in its pool of QFE. Due to their coherence, the vortices have no internal friction. The vortices cannot decay thermally because the coherence prevents random (thermal) fluctuations. In other words, the vortices are cold. Therefore, their lifetimes are very long and depend on those of the CD_{elec}^{H2O}, *e.g.*, the vortices may follow the dissipative dynamics of the CD_{elec}^{H2O}.

The energy spectrum of the vortices of the QFE of CD_{elec}^{H2O} is typified by a lowest lying excited state with a rotational frequency of the order of 1000 Hz. The energy spacing of the spectrum is of the same order, *i.e.*, in the ultra low frequency (ULF) range. The magnetic dipoles of these vortices may become aligned by external magnetic fields, *e.g.* the terrestrial magnetic field.

vi. The phases of the coherent oscillations of the H_2O of different CD_{elec}^{H2O} may become correlated by electric or magnetic fields, *e.g.*, the field due to the electric dipole moment of a CD_{rot} or external fields. Such correlation leads to a coherence among CD_{elec}^{H2O}, which enhances E_{coh}. Therefore, the presence of such fields may stabilize CD_{elec}^{H2O}, *e.g.*, on placing bulk water in external fields may enhance the prevalence of its CD_{elec}^{H2O}.

vii. Ions close to a CD_{elec}^{H2O} get attracted by the fields of the photons condensed in the domain. When the motion of these ions does not get altered by collisions with other molecules, they orbit around the CD_{elec}^{H2O}. The motion of ions incorporated in CD_{rot}, CD_{plasma} or IPD_{plasma} does not get altered by collisions. The energy such ions gain by their participation in the coherent dynamics of these domains protects them from the thermal

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[c] Fingerprints of these quasi-free electrons endowing CD_{elec}^{H2O} with a negative charge were revealed by electrophoresis carried out by Konovalov and Ryzhkina (2014), as discussed by Yinnon and Liu (2015c).

aggression of neighboring molecules. Hence, when a CD_{elec}^{H2O} is stabilized by a CD_{rot}, CD_{plasma} or IPD_{plasma}, part of the ions present in these domains orbit around CD_{elec}^{H2O}.

The circular speed of ions orbiting around a CD_{elec}^{H2O} is proportional to the ion cyclotron frequency (v_c). For v_c holds $v_c=(qB)/2\pi m$, where q and m are the electric charge and the mass of the ion, respectively, and B is the strength of the static magnetic field (*e.g.* the terrestrial magnetic field).

On applying an alternating magnetic field with a frequency matching v_c, ions are extracted from their orbits around CD_{elec}^{H2O}. The extraction, due to the conservation of angular momentum, causes rotational excitation of the QFE of the CD_{elec}^{H2O}, that is, formation of coherent vortices. When the ion concentration is about uniform in a mesoscopic region and an external field also spans over mesoscopic distances, the excitation energy of the vortices of many CD_{elec}^{H2O} may be uniform. A uniform excitation of CD_{elec}^{H2O} leads to coherence among them, which in turn enhances their E_{coh} and their stabilization. Recent experimental findings by D'Emilia *et al.* (2015, 2016) seemingly support this characteristic. The findings suggest that on exposing pure water to sufficiently strong magnetic fields with a frequency equaling that of one of the ion cyclotron frequencies of H_3O^+, *e.g.*, $v_c=7.83$ Hz, enhances stabilization of CD_{elec}^{H2O}.

Analyses of Published Experimental Data

SELF SIMILAR TOPOLOGY OF SDVSAS

Investigations of the dielectric permittivity and electrical conductivity of SDVSAS of membranotropic amphiphilic calix[4] resorcinarene with tris (hydroxymethyl)

methylamide groups (MAC4RWTHMG) at concentrations in the range of 10^{-11}-10^{-3} M have shown that its molecular associates have fractal structures (Lunev *et al.*, 2014). Analyses by Yinnon and Liu (2015c) have shown that this SDVSAS has the characteristics specified in paragraphs I-V — the analyses pertain to the experimental data on the physicochemical properties of these liquids and on the sizes and electrokinetic potential of their molecular associates measured by Ryzhkina *et al.* (2012b).

Self similarity is a property of the QED domains (Vitiello, 2012, 2014), *e.g.*, of supra-CD_{rot}, supra-CD_{elec}^{H2O}, supra-CD_{plasma} and supra-IPD_{plasma}. According to the above presented QED model, for $C < C_{trans}^{CDplasma}$, SDVSAS contain one or more of these domain types. Therefore, the molecular associates in SDVSAS, which are mediated by EM radiation, indeed should have fractal orderings. To the best of my knowledge, the analyses of the experimental data of the SDVSAS of MAC4RWTHMG, carried out by Lunev et al. (2014), provide the first confirmation of the self similarity aspect of the SDVSAS model. It should be noted, however, that the self similarity of the H_2O orderings in water perturbed with a Nafion membrane (iterative Nafionized water) or in water perturbed by iterative filtrations was recently proven by Capolupo *et al.* (2014). These researchers showed that the linear correlations between the logarithm of various physicochemical properties (*e.g.*, χ, Q or density) and the pH of these perturbed waters [measured by the group of Elia *et al.* (2013a,b, 2014a,b, 2015)] reflect the self similarity of their H_2O orderings. These orderings have been identified as CD_{elec}^{H2O} and CD_{rot} by Yinnon and Elia (2013), Yinnon *et al.* (2016) and Elia *et al.* (2017). Linear correlations between the logarithm of χ, the logarithm of Q and pH have also been observed for SDVSAS (Elia *et al.*, 2004b; Belon *et al.*, 2008; Elia *et al.*, 2015). Thus, from the analyses of Capolupo *et al.* (2014),

Elia *et al.* (2004b), Belon *et al.*(2008) and Elia *et al.* (2015), one could infer that the H_2O orderings observed in SDVSAS should have a fractal structure. The study by Lunev *et al.* (2014) confirmed this inference.

EMISSION OF ULF RADIATION BY SDVSAS

Data on SDVSAS emitting ULF radiation reported in the literature — Montagnier *et al.* (2009, 2011, 2015) demonstrated that SDVSAS of some types of deoxyribonucleic acid (DNA) emit ULF (500-3000 Hz) EM radiation.[d] The SDVSAS were prepared in plastic 1.5 mL Eppendorf tubes. Specifically, they demonstrated the following:

- The number of dilution steps required for the SDVSAS to emit the radiation depends on the type of the DNA (*e.g.*, DNA extracted from bacteria or viruses). Typically, at least six decimal dilutions of a few nano gram (ng) of DNA are required for the emission of ULF radiation to be detectable. After applying more than about 10 to 17 decimal dilutions, the emission is no longer observable. Expressed differently, the emission of ULF radiation is typically observed for SDVSAS of DNA with concentrations in the 10^{-5} M to 10^{-18} M range. The intensity of the emitted radiation non-monotonically depends on the concentration. The fact that no emission was detectable for SDVSAS of DNA with $C<10^{-18}$ M might be due to limitations of the experimental set up. The emission might be too weak to be observable with the simple apparatus employed by Montagnier *et al.* (2009, 2011, 2015).[d] No emission of ULF radiation was detectable for serially diluted DNA solutions which were not vigorously shaken after each dilution step.

- The SDVSAS of DNA only emit ULF radiation when these are stimulated by background extremely low frequency (ELF) ra-

diation. The sources of the ELF radiation may be the natural Schumann resonances of the geomagnetic field or artificial ones. The frequency of the Schumann resonance, which has the lowest frequency, is 7.83 Hz (Nickolaenko and Hayakawa, 2002). Storing the SDVSAS in Permalloy containers, which screens the background EM radiation, affects their emission of the ULF radiation. For some samples, after several hours of such storage, no emission was detectable anymore. For other samples, after having been stored for 48 h under equivalent conditions, emission of ULF was still detectable.

- The radiation emitted by the SDVSAS of DNA is absorbable by neighboring pure water samples. The absorbed radiation alters the physicochemical properties of the neighboring pure water samples. These samples acquire properties reminiscent of those of the SDVSAS of DNA.

Elia *et al.* (2012) demonstrated that also SDVSAS of other solutes (*e.g.*, fullerene), which were 5, 7, 9, 12 or 30 times centesimally diluted, emit radiation which alter the physicochemical properties of neighboring water samples.

Explanations for the emission of ULF radiation by SDVSAS — Montagnier *et al.* (2011, 2015) provided explanations for some of their experimental results. Their explanations, which are based on the processes detailed in paragraphs (v) and (vii), are as follows.

a) DNA are poly-electrolytes surrounded by a cloud of positive counter-ions. These ions may orbit around CD_{elec}^{H2O} (Del Giudice *et al.*, 2002). The circular speed of the orbiting ions is proportional to the ion cyclotron frequency (v_c). Typically, v_c falls in the 1-100 Hz range. Ions or-

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[d] Montagnier *et al.* (2009, 2011, 2015) detected the emitted radiation with an EM coil (bobbin of copper wire), which they connected to an amplifier (a sound blaster card). They linked the card to a computer.

biting around CD_{elec}^{H2O} get extracted from their loops by background alternating magnetic fields (Schumann resonances or artificial fields). Extraction of the ions from their orbits (because of conservation of angular momentum) causes a rotation of the plasma of the quasi-free electrons (QFE) present in the CD_{elec}^{H2O} (*i.e.*, the extraction excites the domain and induces vortices). Since the mass of electrons is much smaller than that of the ions, the rotating plasma has a frequency v_p that is much higher than v_c. Typically, the lowest v_p value is of the order of 1000 Hz and the spacing between the rotating plasma frequencies is of the same order, *e.g.*, in the ULF range.

b) The rotation of the QFE produces the magnetic component of the ULF radiation.

c) The frequency and intensity of the emitted ULF radiation depend on the concentration of the counter-ions, *i.e.*, the degree of dilution of the SDVSAS of DNA. The following causes underlie this dependency: Firstly, v_p depends on the number of counter-ions participating in the orbiting and extraction processes. This number is a function of their concentration. v_p is also determined by the quantization of angular momentum. Secondly, the co-resonating fields appearing in the surroundings of the rotating CD_{elec}^{H2O} depend on the concentration of the counter-ions. These fields may produce coherence among the individual CD_{elec}^{H2O} and thus affect the frequency and intensity of the emitted ULF radiation.

Montagnier *et al.* (2011, 2015) emphasized that their conjecture — that the emission of ULF radiation is due to magnetic activity — is supported by the absence of such emission after the SDVSAS samples were stored in Permalloy containers.[e]

As to the solute reminiscent alterations of pure water samples neighboring on SDV-SAS of DNA, Montagnier *et al.* (2011, 2015) conjectured that it is due to the SDVSAS samples emitting ULF radiation which triggers coherence among the CD_{elec}^{H2O} of the pure water samples. They noted that the physics underlying the copying of properties of SDVSAS to neighboring pure water samples is similar to that of: "the proximity effect observed in two superconducting samples or in the arrays of Josephson junctions, by which the samples or the junctions fall into a phase-locking regime."

Unexplained phenomena pertaining to emission of ULF radiation by SD-VSAS — To the best of my knowledge, no explanations have yet been forwarded for several of the observations made by Montagnier *et al.*'s (2009) and Elia *et al.* (2012). In the following, I attempt to clarify them with the QED model of SDVSAS:

♦ **Why was emission of ULF radiation not detectable for serially diluted aqueous solutions of DNA, when these solutions were not vigorously shaken after each dilution step?**

According to the QED model's aspects summarized in paragraph I: For $C_{trans}^{IPDplasma} < C < C_{trans}^{CDplasma}$, part of the solvated solutes aggregates in CD_{plasma}. Organization of DNA molecules (or their counterions) in domains with characteristics of CD_{plasma} has been observed by Samal and Geckeler (2001), as discussed by Yinnon and Yinnon (2009).

According to paragraph II: for $C < C_{trans}^{IPDplasma}$, CD_{plasma} transform into IPD_{plasma}. Typically 10^{-6} M $< C_{trans}^{IPDplasma} < 10^{-4}$ M. According to paragraph III, vigorous shaking of solutions containing IPD_{plasma}

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[e] EM radiation at frequencies of kHz - Tera Hz can also be attributed to vibrations of electrically polar structures (Cifra, 2015), *e.g.*, proteins, EDA^{CDrot} or $EDA^{IPDplasma}$.

excites or break up these domains and creates $EDA^{IPDplasma}$.

According to paragraph V: Firstly, for $C < C_{crit}^{CDrot}$, $EDA^{IPDplasma}$ stabilize CD_{rot}. Secondly, vigorous shaking of the liquid excites or breaks up the CD_{rot} and thus creates EDA^{CDrot}. Thirdly, the CD_{rot} and EDA^{CDrot} may agglomerate into supra-CD_{rot} with their electric dipole moments ordered more or less parallel or anti-parallel. The orientation of the electric dipole moments of the CD_{rot} affects the physicochemical properties of the SDVSAS.

As noted in paragraphs I-V, CD_{elec}^{H2O} may get stabilized by all the domains mentioned in the last three paragraphs, *i.e.*, by CD_{plasma}, IPD_{plasma}, $EDA^{IPDplasma}$, EDA^{CDrot} or CD_{rot}. For C above the Avogadro limit, all these domains contain coherently oscillating ions, which can orbit around CD_{elec}^{H2O}. Vigorous shaking may excite the CD_{elec}^{H2O}, *i.e.*, its plasma of QFE.

In regards to the information presented in the last four paragraphs and in paragraphs (a)-(c), the observation that only serially diluted solutions of DNA which were vigorously shaken after each dilution step emitted ULF radiation, alludes to the following: Only the micron sized CD_{rot}, EDA^{CDrot}, $EDA^{IPDplasma}$ or their agglomerates (creation of which requires serial dilutions and vigorous shaking after each dilutions step, and which all contain ferroelectric ordered H_2O) can induce coherence among many of the 10^{-7} m sized CD_{elec}^{H2O}. The coherence assures that the intensity of the ULF emitted by the excited plasma of QFE of the CD_{elec}^{H2O} is strong enough to be detected with a solenoid.

The abovementioned shows that the observed emission of ULF radiation by SDVSAS provides additional support for the main aspect of its model. In other words, CD_{rot}, EDA^{CDrot}, CD_{elec}^{H2O} and $EDA^{IPDplasma}$ are the entities underlying these liquids' extraordinary properties. It also substantiates the conjecture of Montagnier *et al.* (2015) that the emission of ULF radiation by SDVSAS of DNA is the outcome of symmetry breaking. As they formulate it: "the symmetry which gets broken is the rotational symmetry of the electrical dipoles of the H_2O and correlation modes are the ones associated to the dipole waves (similar to spin waves in ferromagnets)."

♦ **Which ions orbit around CD_{elec}^{H2O} in SDVSAS diluted below the Avogadro limit?**

Elia *et al.* (2012) demonstrated that SDVSAS diluted below the Avogadro limit emit EM radiation. Their experiments, however, did not reveal the wavelengths of the radiation. In case the radiation includes ULF radiation, the question arises: Which ions orbit around CD_{elec}^{H2O} in these extremely diluted liquids? Elia *et al.* (2012) prepared these SDVSAS in glass vessels. Therefore, these liquids contained dissolved components of the glassware containers at impurity level concentrations. However, in the absence of such impurities, such as when SDVSAS are prepared in plastic containers, H_3O^+ or OH^- ions are present. The pH of SDVSAS is affected by their molecular associates (Elia *et al.*, 2004a; Ryzhkina *et al.*, 2010, 2012b, 2013). [The effect of the associates on the pH has been explained in paragraph vi of the discussion section in Yinnon and Liu (2015c).] Hence, H_3O^+, OH^- or impurity ions may all orbit around CD_{elec}^{H2O} in SDVSAS.

♦ **Why is emission of ULF radiation no longer detectable for some samples, after their storage for a few hours in Permalloy containers, while other samples stored for 48 hours under equivalent conditions still emitted ULF radiation?**

For clarification of these phenomena, it should be emphasized that experiments have shown that the dynamics of SDVSAS are that of far-out-of equilibrium dissipative systems (Elia *et al.*, 2008a,b, 2010). The dissipative dynamics of their molecular associates occur over macroscopic time scales, *i.e.*, the dynamics may continue for months and even many years when the SDVSAS are kept at room temperatures and pressures.

The dissipative dynamics of SDVSAS, according to the QED model of SDVSAS, are due to a constant reorganization of the CD_{rot} and EDA^{CDrot} constituting the supra-CD_{rot} (Yinnon and Elia, 2013). It is also due to the stabilization and disintegration of CD_{elec}^{H2O} and reorganization of supra-CD_{elec}^{H2O}. Agitating SDVSAS by vigorously shaking accelerates reorganization of its domains. It also enhances excitation of the QFE of its CD_{elec}^{H2O}. The dynamics of CD_{rot} (which is related to the rotational excitation of H_2O) is much slower than that of CD_{elec}^{H2O} (which is related to the electronic excitation of H_2O).

The continuous reorganization of the domains in SDVSAS affects the radiation emitted and absorbed by these liquids. On disintegration of CD_{elec}^{H2O}, the EM radiation condensed within these domains is released (see paragraph i). The released radiation may enhance stabilization of new CD_{elec}^{H2O}, it may escape to the environment or it may transform to heat. The same holds for the EM radiation condensed within CD_{rot}. Reorganization of the domains in SDVSAS also affects the vortices of the QFE of CD_{elec}^{H2O}, as it affects the emission of ULF radiation. Part of the emitted ULF radiation may get absorbed by QFE of neighboring CD_{elec}^{H2O}.

Placing SDVSAS in Permalloy containers screens its CD_{rot} and CD_{elec}^{H2O} from ambient IR and UV radiation. These are the sources of the radiation mediating the interaction between the molecules constituting these domains (see paragraphs I and V). However, the EM radiation released by the reorganization and disintegration of CD_{rot} and CD_{elec}^{H2O} cannot escape through the walls of the Permalloy container to the environment. Thus, on placing SDVSAS samples in Permalloy containers, the non-linear dissipative processes taking place in these liquids imply that in some samples the CD_{rot} and CD_{elec}^{H2O} persist for a long time and continue to emit ULF radiation. Conversely, in other samples most of these domains quickly disintegrate.

♦ **Why does the intensity of the ULF radiation emitted by the SDVSAS of DNA depend non-monotonically on their concentration?**

The non-monotonic dependence on concentration of the intensity of the ULF radiation emitted by SDVSAS [as with similar dependencies of other physicochemical properties of SDVSAS, noted in paragraph (7)] result from the reorganization of their associates. More specifically, according to the model of SDVSAS, every perturbation like a dilution or shaking alters the ordering of the electric dipole moments of the supra-CD_{rot}. These alterations affect the prevalence of the CD_{elec}^{H2O} and supra-CD_{elec}^{H2O}. As such, the alterations also affect the ULF radiation emitted by the pool of quasi free electrons of CD_{elec}^{H2O}.

Hitherto, to the best of my knowledge, emission of ULF radiation has been reported only for SDVSAS of some types of DNA. This calls for research aimed at searching for SDVSAS of other solutes which emit ULF radiation. Such research might verify the various explanations presented in this section.

IMPACT OF EM RADIATION ON THE FORMATION OF ASSOCIATES IN SDVSAS

This section shows that recent experimental data, obtained by A.I. Konovalov's group (D.A. Konovalov *et al.*, 2015), support some aspects of the explanations of Montagnier *et al.* (2011, 2015) discussed in the previous section. The data supports the hypothesis that the CD_{elec}^{H2O} present in an aqueous SDVSAS with concentration less than C_{thr} but above the Avogadro limit are affected by 7.85 Hz EM fields.

A.I. Konovalov's group was the first to demonstrate that only when SDVSAS with *C* less than C_{thr} but above the Avogadro limit were stored under ambient EM fields, the presence of molecular associates in these liquids were observable with dynamic light scattering (DLS) (Ryzhkina *et al.*, 2011a; Konovalov and Ryzhkina, 2014). After storage of these SDVSAS for 24 hours in Permalloy containers, DLS could not distinguish associates in these liquids. Moreover, after such storage, these liquids' characteristic physicochemical and bioactive properties disappeared. Analyses of the associates and their impact on the physicochemical properties of SDVSAS indicate that these associates include CD_{elec}^{H2O}, CD_{rot} and EDA^{CDrot} (Yinnon and Liu, 2015b&c).

Recently, A.I. Konovalov's group built a hardware complex, which enabled controlling the amplitude and frequency of the magnetic field in a Permalloy container (D.A. Konovalov *et al.*, 2015). The group prepared samples of SDVSAS of biologically active *p*-sulfonato-calix[6]arene with $C=1\times10^{-9}$ M. The C_{thr} of this SDVSAS is about 10^{-7} M, significantly larger than *C*. One series of the samples was kept for 24 hours on the laboratory bench. In this paper, these samples are denoted as SDVSAS$_{LB}$. A second series was kept for 24 hours under hypo electromagnetic conditions, in the Permalloy container. Here these samples are denoted as SDVSAS$_P$. A third series was placed for 24 hours in the hardware com-

plex, with the frequency of its magnetic field tuned to 7.85 Hz. These samples are denoted as SDVSAS$_{MF-7.85}$. This third series was subdivided into four sub-series, each of which was subjected to magnetic fields with different amplitudes, respectively, 48, 24, 12 and 6.4 A/m. These samples are denoted as SDVSAS$_{48}$, SDVSAS$_{24}$, SDVSAS$_{12}$ and SDVSASL$_{6.4}$, respectively.

D.A. Konovalov *et al.* (2015) showed that molecular associates were detectable with DLS in their SDVSAS$_{LB}$, but were not detectable in their SDVSAS$_P$. The average hydrodynamic diameter of the associates present in the SDVSAS$_{LB}$ samples was 230 nm. These results conform to those of many previously studied SDVSAS (Konovalov and Ryzhkina, 2014). The 230nm average hydrodynamic diameter is typical for the supra-CD_{elec}^{H2O} present in SDVSAS for $C<C_{thr}$ (Yinnon and Liu, 2015c). DLS also evidenced associates in SDVSAS$_{48}$, SDVSAS$_{24}$ and SDVSAS$_{12}$, but did not uncover any associates in SDVSAS$_{6.4}$. The size distributions of the associates in SDVSAS$_{48}$ and SDVSAS$_{24}$ were mono-modal. These distributions were the same as those of SDVSAS$_{LB}$. However, decreasing the magnetic field's amplitude from 24 to 12A/m led to poorly reproducible results, as the SDVSAS$_{12}$ samples had poorly reproducible bimodal associate size distributions.

In the following, two complementary possible explanations for the findings of D.A. Konovalov *et al.* (2015) are offered:

♣ As noted in paragraphs I and V, interactions between UV EM fields and H_2O underlie formation of CD_{elec}^{H2O}, while interactions between IR EM fields and H_2O underlie formation of CD_{rot}. Despite Permalloy screening ambient UV and IR EM fields, the analyses of the experimental data obtained by Montagnier *et al.* (2009) have indicated that placing SDVSAS samples in Permalloy containers does not lead to immediate disinte-

gration of all its molecular associates. In some samples, the associates seem to persist for at least 48 hours. Thus, D.A. Konovalov *et al.* (2015)'s finding that DLS measurements did not reveal associates in $SDVSAS_P$, likely does not mean that no associates were any longer present in samples after these were screened from ambient EM fields for 24 hours. It is conceivable that the screening considerably diminished the number of associates, so that their prevalence was below the detection limit of DLS. According to QED, on disintegration of part of the supra-CD_{rot} and supra-CD_{elec}^{H2O}, the remaining domains are less stable. For CD_{elec}^{H2O}, their destabilization implies that the continuing adsorption and desorption of their H_2O (described in paragraph iii) take place on shorter timescales, so that it becomes more difficult to observe these domains.

The difference between $SDVSAS_P$ and $SDVSAS_{MF-7.85}$ is that the latter were exposed to 7.85 Hz EM fields. Since DLS showed that the size distribution of the domains in $SDVSAS_{48}$ and $SDVSAS_{24}$ was similar to that of $SDVSAS_{LB}$, *i.e.*, these liquids contained nano-associates with a 230 nm average hydrodynamic diameter, the presence of the 7.85 EM field seemingly enhanced stabilization of CD_{elec}^{H2O}. The stabilization is likely attributable to processes involving the vortices of the plasma of the QFE of CD_{elec}^{H2O}. These vortices are created by the 7.85 Hz EM field (see paragraph vii). For example, one possible stabilization process is that of the vortices of different CD_{elec}^{H2O} interacting with each other. Another process is that these vortices interact with the terrestrial magnetic field. This explanation implies that the intensity of the alternating 7.85 Hz EM field must be sufficiently strong to extract ions cycling around CD_{elec}^{H2O} from their orbit, because such extraction creates vortices (see paragraph vii). In other words, on lower-

ing of the intensities of the applied 7.85 Hz field, fewer or no vortices are created in the plasma of QFE of CD_{elec}^{H2O}. It is to be expected that few vortices cannot considerably stabilize CD_{elec}^{H2O}, and that these cannot significantly reduce the rate of the continuous adsorption and desorption of the H_2O of these domains. This explanation is compatible with the difficulties D.A. Konovalov *et al.* (2015) faced in observing molecular associates in $SDVSAS_{12}$. It is also compatible with them not being able to discern associates in $SDVSAS_{6.4}$.

In paragraph V it was emphasized that CD_{elec}^{H2O} may get stabilized by CD_{rot}. In other words, that the CD_{elec}^{H2O}, which are meta-stable in bulk water at ambient conditions (see paragraph iii), get stabilized by CD_{rot}. The stabilization of CD_{elec}^{H2O} in SDVSAS occurs when these liquids are diluted below C_{crit}^{CDrot} and CD_{rot} domains are stabilized by $EDA^{IPDplasma}$ or polar macromolecules. *The findings of D.A. Konovalov et al. (2015) imply that SDVSAS stabilization of* CD_{elec}^{H2O} *requires the presence of* CD_{rot} *as well as the presence of 7.85 Hz magnetic fields*. Indeed, the recent experiments by D'Emilia *et al.* (2015, 2016) hint that in bulk water, such fields reduce the continuously changing space distribution of the H_2O constituting its meta-stable CD_{elec}^{H2O} and its non-coherent H_2O. Their findings suggest that exposing pure water to sufficiently strong magnetic fields with a frequency equaling that of one of the ion cyclotron frequencies of H_3O^+, *e.g.*, v_c=7.83 Hz, reduces the flickering of its CD_{elec}^{H2O} landscape. In other words, it reduces the frequency of the adsorption and desorption of these domains' H_2O.

♣ Non-linear optical processes, induced by 7.85 Hz magnetic fields, are also possible sources for stabilizing the CD_{elec}^{H2O} in $SDVSAS_{48}$ and $SDVSAS_{24}$. In other words,

the 7.85 Hz EM field, the ULF radiation emitted from the QFE of CD_{elec}^{H2O}, the UV radiation emitted from disintegrating CD_{elec}^{H2O}, and the IR radiation emitted from disintegrating CD_{rot} all may non-linearly interact with the molecules constituting these domains. These non-linear interactions may re-stabilize disintegrating CD_{elec}^{H2O} and CD_{rot}. Since the H_2O organized in the supra-CD_{elec}^{H2O} oscillate coherently in phase with a coherently condensed EM field, and the same holds for the H_2O constituting the supra-CD_{rot}, non-linear optical processes may play significant roles. Coherence of photons is known to enhance non-linear optical effects (Zheltikov *et al.*, 2007).

As to the likelihood of non-linear optical processes playing a role in stabilization of CD_{rot} and CD_{elec}^{H2O}, phenomena pertaining to the exclusion zone (EZ) water forming in water adjacent to a hydrophilic membrane should be mentioned. The analyses by Del Giudice *et al.* (2013) and Yinnon *et al.* (2016) have indicated that EZ water is composed of CD_{elec}^{H2O} and CD_{rot}. The width of the EZ zone has been observed to expand on its irradiation with UV-visible and IR radiation, such as 270 - 700 nm and 1750 - 4200 nm radiation (Chai *et al.*, 2009). On irradiation, up to 300 percent widening of the EZ has been observed. The degree of widening increased with wavelength, except for a few local maxima. These maxima appeared at wavelengths for which the absorption of radiation by the EZ is maximal, *e.g.*, at 270 nm and 3.1 μm. The widening of the EZ was observed to be a function also of the irradiation time and intensity. These phenomena allude that non-linear optical processes affect the expansion of the EZ (*i.e.*, affect the stabilization of CD_{elec}^{H2O} and CD_{rot}).

To investigate the role of non-linear optical processes in stabilization of CD_{elec}^{H2O} and CD_{rot} in SDVSAS, research is called

for that focuses on excitation of these liquids not just by 7.85 Hz radiation but also with EM radiation at a large variety of frequencies.

STRUCTURAL ASPECTS OF SDVSAS DILUTED BELOW THE AVOGADRO LIMIT

Recently, the group of A.I. Konovalov extended its DLS measurements to SDVSAS diluted below the Avogadro limit (Ryzhkina *et al.*, 2015b). The group measured the DLS of SDVSAS of polyclonal antibodies to the protein interferon gamma. The SDVSAS were prepared with freshly double distilled water. Glass vials (Wiegand 120011543) were used. The stock solution was centesimally diluted. The concentration of the stock solution was 0.1 mg/ml. The SDVSAS samples were labeled as C2-C15, C30 and C50. The C indicates that the SDVSAS were centesimally diluted. The number denoted the number of dilutions steps (NDS). For the C7 samples, the molarity was calculated. It was of the order of 10^{-18} M. The properties of all the samples were measured after they were kept for 18 - 20 h on the laboratory bench or in Permalloy containers. The former are denoted as $SDVSAS_{LB}^{a-b}$ (*i.e.*, SDVSAS of anti-bodies kept at the laboratory bench). The latter are denoted as $SDVSAS_p^{a-b}$ (*i.e.*, SDVSAS of anti-bodies kept in Permalloy containers).

Ryzhkina *et al.*(2015b) also measured the electrical conductivity (χ), the surface tension (σ) and the ΔpH of $SDVSAS_{LB}^{a-b}$ and $SDVSAS_p^{a-b}$. ΔpH is the difference between the pH of the SDVSAS and the pH of the double distilled water with which the SDVSAS were prepared, (the pH of the distilled water on the day the SDVSAS were prepared).

For controls, Ryzhkina *et al.*(2015b) prepared samples of serially diluted, vigorously shaken water (SDVSW), (double distilled water which was serially diluted with double distilled water and vigorously shaken

after each dilution step).

A concise summary of the findings of Ryzhkina *et al.* (2015b) is presented in the next three paragraphs. Following the summary, these findings are analyzed within the context of the QED model of SDVSAS.

For $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ with concentrations above the Avogadro limit, just as Konovalov and Ryzhkina (2014) had observed for SDV-SAS of many other solutes, Ryzhkina *et al.* (2015b) found that their χ, σ and ΔpH values vary non-monotonically with NDS. For $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ diluted below the Avogadro limit, they observed that these values also vary non-monotonically with NDS. Moreover, for SDVSW, they observed that the values of these variables vary non-monotonically with NDS and differ from those of $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$. In addition, they measured significant differences between the values of these variables for $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ or SD-VSW versus those of double distilled water. [Elia and Niccoli (1999, 2000, 2004a) and Elia *et al.* (2005, 2015) did also report non-monotonic dependencies of χ, pH and other physicochemical properties of SDVSAS and SDVSW on NDS. They observed such non-monotonicity for SDVSAS of many different solutes. These SDVSAS had concentrations above the Avogadro limit or were diluted below this limit.]

For $\text{SDVSAS}_{\text{p}}^{\text{a-b}}$, Ryzhkina *et al.* (2015b) found that their χ and ΔpH variables only significantly vary non-monotonically with NDS when these liquids were less than 13 times centesimally diluted. When $\text{SDVSAS}_{\text{p}}^{\text{a-b}}$ and $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ were 13 times or more centesimally diluted, the χ and ΔpH of the former are higher or equal to those of the latter. As to σ, Ryzhkina *et al.* (2015b) observed an analogous behavior when $\text{SDVSAS}_{\text{p}}^{\text{a-b}}$ and $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ were more than 12 times centesimally diluted.

As to the DLS measurements of $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ and $\text{SDVSAS}_{\text{p}}^{\text{a-b}}$, the main results of Ryzhkina *et al.* (2015b) are:

A. For $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ with C2-C6, C8, C10-C12, C14 and C15, DLS does not indicate presence of associates.

B. For $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ with C7 or C9, DLS revealed presence of nano-associates with a monomodal size distribution. The average diameters of the associates are respectively, ~170 nm and ~140 nm.

C. The associates in C7 $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ are absent in C7 $\text{SDVSAS}_{\text{p}}^{\text{a-b}}$.

D. For C9 $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ and C9 $\text{SDVSAS}_{\text{p}}^{\text{a-b}}$, their associates have nearly the same dimensions. The monomodal size distribution of $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ is a bit narrower than that of $\text{SDVSAS}_{\text{p}}^{\text{a-b}}$.

E. In C13, C30 or C50 $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$, as well as in C13, C30 or C50 $\text{SDVSAS}_{\text{p}}^{\text{a-b}}$, DLS revealed particles with a trimodal size distribution. For such a distribution, the DLS method is inadequate for accurately measuring the average sizes of the particles. Yet the DLS measurements hint that particles with sizes of the order of 10^{-9} m, 10^{-8} m and 10^{-7} - 10^{-6} m are present in these liquids.

For analyzing the above cited findings of Ryzhkina *et al.* (2015b), foremost, the following should be emphasized: although DLS does not indicate presence of associates in C2 -C7, C8, C10-C12, C14 and C15 of $\text{SDVSAS}_{\text{LB}}^{\text{a-b}}$ (see paragraph A), these liquids' physicochemical variables and the non-monotonic variance of these variables with NDS reflect that these liquids' properties differ from those of double distilled water. Analyses of experimental data reported in earlier publications, pertaining to measured physicochemical variables of SDVSAS of other solutes with concentrations above the Avogadro limit or which were diluted below this limit (Elia and Niccoli, 1999, 2000, 2004a; Elia *et al.*, 2005, 2015, 2008a&b; Belon *et al.*2008), as well as those pertaining to SDVSW, suggest that for $C < C_{\text{crit}}^{\text{CDrot}} = C_{\text{thr}}$, in these liquids, CD_{rot} underlie the non-monotonic variance of

their physicochemical variables (Yinnon and Yinnon, 2011; Yinnon and Elia, 2013; Yinnon and Liu, 2015b&c). The analyses indicate that the impact of CD_{rot} on the physicochemical properties of SDVSAS or SDVSW slowly increases with the age (months, years) of these liquids. Since the data show that the physicochemical variables greatly vary with the age of those SD-VSAS which were only a few times diluted, shortly shaken and stored for less than a year, it suggests the following: the sizes of the CD_{rot} in such "young" SDVSAS are still small and the ordering of these domains in supra-CD_{rot} vary greatly with time. Based on these analyses, the findings presented in paragraph A hint that DLS is insensitive to the presence of young CD_{rot}. This insensitivity will be discussed further on.

As to the findings presented in paragraphs B and C, monomodal distributions of molecular associates with average diameters of the order of 10^{-7} m, which disappeared on screening of the liquid by Permalloy, (like the one observed for C7 SDVSAS$_{LB}^{a-b}$) have been detected in SDVSAS of many solutes (Konovalov and Ryzhkina, 2014). The associates underlying such distributions were shown to have the typical characteristics of CD_{elec}^{H2O} and supra- CD_{elec}^{H2O} (Yinnon and Liu, 2015b&c). As noted in previous sections, for $C < C_{crit}^{CDrot} = C_{thr}$, these domains are stabilized by CD_{rot} and low frequency magnetic fields. It is thus most likely that the 170 nm sized associates observed in C7 SDVSAS$_{LB}^{a-b}$ are supra-CD_{elec}^{H2O}. Measurements of their electrokinetic potential are called for. Such measurements could provide additional evidence for this conclusion.

A question arising from the above presented data is: "Why did not DLS detect a monomodal distribution of associates with sizes of the order of 10^{-7} m in SDVSAS$_{LB}^{a-b}$ which were less than 7 times centesimally diluted (see paragraph A)?" The concentrations of the antibodies in C2 - C7 SDVSAS$_{LB}^{a-b}$ are in the range of 10^{-8} - 10^{-18} M. A value as low

as about 10^{-18} M has not been reported for $C_{crit}^{CDrot} = C_{thr}$ of SDVSAS of other solutes. Typically, ~10^{-10} M$< C_{crit}^{CDrot} = C_{thr} <$ ~10^{-6} M.

A possible answer is that in SDVSAS$_{LB}^{a-b}$ which were less than 7 times centesimally diluted, the alignment of CD_{rot} in supra-CD_{rot} and the incorporation of the solutes in these domains are detrimental to stabilization of CD_{elec}^{H2O} in numbers sufficient for these domains to be observable with DLS. Such an answer might also be applicable to C8, C10-C12, C14 and C15 SDVSAS$_{LB}^{a-b}$, in which as noted in paragraph A, DLS did not detect associates with diameters of the order of 10^{-7} m. A similar explanation has been given for the relation between the prevalence of molecular associates with sizes of the order of 10^{-7} m observed in SDVSAS of many solutes and the dielectric permittivity of these liquids (Konovalov and Ryzhkina, 2014; Yinnon and Liu, 2015c). (The dielectric permittivity reflects the alignment of the electric dipoles of CD_{rot} in supra-CD_{rot}.)

The above analyses raise the issue: "How is information on CD_{rot} and supra-CD_{rot} reflected in DLS data?" At least theoretically, these domains may grow to sizes of 10^{-5} - 10^{-4} m. The issue has not been clarified in the earlier published analyses of SDVSAS. Some clues are provided by atomic force measurements (AFM) of the residues left over after evaporating drops of SDVSAS. AFM data revealed that in addition to more or less spherical 10^{-7} m sized chunks, micron sized elongated molecular associates were present in these residues [see Fig. 3f in Ryzhkina *et al.* (2012b); Fig. 1 in Ho (2014); Fig. 5 in Elia *et al.* (2015)]. The widths of these elongated associates were of the order of 10^{-9} - 10^{-7} m. Their lengths were of the order of 10^{-6} - 10^{-5} m. These elongated associates were more or less linear or curled up. The diameters of the ball-shaped curled up ones were of the order of 10^{-7} - 10^{-6} m. Earlier published analyses of the various physicochemical properties of these SD-VSAS evoked that the elongated associ-

ates are CD_{rot}, or supra-CD_{rot} organized in chains with their electric dipole moments parallel oriented (Yinnon and Liu, 2015c). The analyses also indicated that the chunks are supra-CD_{elec}^{H2O}. Therefore, in relation to the abovementioned issue, it is my conjecture that DLS cannot clearly distinguish between the presence of supra-CD_{elec}^{H2O} and the presence of the supra-CD_{rot} when these elongated associates are curled up in balls. Moreover, it is likely that DLS is insensitive to very thin linear molecular associates, such as the more or less linearly micron sized CD_{rot} or supra-CD_{rot} with width of the order of 10^{-9} m.

In regard of this conjecture, the data reported in paragraphs B and D possibly can be understood as follows: The monomodal distribution of associates with sizes of the order of 10^{-7} m which were present in C9 $SDVSAS_{LB}^{a-b}$ mainly reflect the presence of curled up supra-CD_{rot} and few CD_{elec}^{H2O}. On screening of this $SDVSAS_{LB}^{a-b}$ by Permalloy for 18 - 20 h, most of these few CD_{elec}^{H2O} disintegrated. However, as noted above, CD_{rot} have much slower dynamics than CD_{elec}^{H2O}. Therefore, CD_{rot} disintegrate slower than CD_{elec}^{H2O}. So many supra-CD_{rot} could persist in the samples after their storage for 18 - 20 hours in Permalloy containers. Moreover, since CD_{rot} formation is mediated by far IR radiation, while CD_{elec}^{H2O} formation is mediated by UV radiation, it is possible that screening by Permalloy affects the former less than the latter. These explanations potentially clarify why the difference between the observed DLS results for C9 $SDVSAS_{LB}^{a-b}$ and C9 $SDVSAS_{p}^{a-b}$ is small.

As to the findings presented in paragraph E, my conjecture also enables explanation of the characteristics of the associates in C13, C30 or C50 of $SDVSAS_{LB}^{a-b}$ and $SDVSAS_{p}^{a-b}$. The associates with diameters of the order of 10^{-7} - 10^{-6} m, which were revealed in these $SDVSAS_{LB}^{a-b}$ by DLS, likely mainly are curled up CD_{rot} and supra-CD_{rot}. These CD_{rot} may have stabilized few 10^{-7} m sized CD_{elec}^{H2O}. On

screening these $SDVSAS_{LB}^{a-b}$ with Permalloy for 18 - 20 hours, many of the CD_{rot} prevailed, while the CD_{elec}^{H2O} mostly disintegrated. The persistence of CD_{rot} explains that the χ, σ and ΔpH values of these $SDVSAS_{p}^{a-b}$ significantly differed from those of double distilled water.

As to the 10^{-9} m and 10^{-8} m sized particles which were observed in C13, C30 or C50 of $SDVSAS_{LB}^{a-b}$ and in $SDVSAS_{p}^{a-b}$, these may include broken CD_{rot} pieces (*i.e.*, EDA^{CDrot}) created by the vigorous shaking of these liquids. The slow dynamics of CD_{rot} implies that their size distribution is wide. The 10^{-9} m sized particles also may be (hydrated) compounds released by the containers or other impurities. Such particles were not discerned with DLS in $SDVSAS_{LB}^{a-b}$ or in $SDVSAS_{p}^{a-b}$ diluted less than 13 times. Likely, in these solutions, their concentrations were very low, *i.e.*, too low to be detectable with DLS. The concentration of compounds released by the glass container slowly increases with the number of dilution steps, because after each dilution step the liquid is vigorously shaken and a percentage of the liquid is transferred to a new vial containing double distilled water.

The upshot of the above analyses is that in the $SDVSAS_{LB}^{a-b}$, which were diluted below the Avogadro limit, CD_{rot} and supra-CD_{rot} are apparently present. Also analyses reported in earlier publications pertaining to SDVSAS of other solutes which were diluted below this limit, as well as those pertaining to SDVSW, suggested presence of such domains in these liquids (Yinnon and Elia, 2013). However, the analyses presented in the previous paragraphs hint that the capability of DLS to detect these domains depends on their shape. Since experimental data hint that on aging of SDVSAS, CD_{rot} grow and organize in supra-CD_{rot} (Yinnon and Elia, 2013), DLS measurements of aged SDVSAS are called for. In addition to DLS, it seems desirable to employ other techniques for revealing typifying fingerprints of CD_{rot}

and supra-CD_{rot} in SDVSAS diluted below the Avogadro limit (*e.g.*, their ferroelectric orderings and the electric dipole rotations of their H_2O.) Their ferroelectric orderings may be revealed by dielcometric titrations, as the experimental data obtained by Ryzhkina *et al*. (2012b, 2013) and their analyses by Yinnon and Liu (2015c) have indicated. Moreover, as the recent results obtained by Mahata (2013) suggest, such orderings and rotations of the electric dipoles of their molecules may possibly be revealed by dielectric dispersion measurements.

Dielectric dispersion measurements have provided evidence for the presence of about 3×10^{-5} m sized molecular structures in water samples to which a few drops of one of the following liquids were added: 6 or 30 times serially diluted solutions of Graphites or Copper, vigorously shaken after each dilution step (Mahata, 2013). The solvent of these serially diluted solutions was ethanol. After addition of the drops to the water samples, the samples contained about 3 percent ethanol. The 3×10^{-5} m sized structures affected the polarization of the samples. The effect became distinguishable about 24 - 48 hours after the drops were added to the water samples. The detailed experimental results indicate that formation of the structures involves electric dipole rotations of the liquids' molecules. More accurately, it involves rotations of the solvent molecules, but does not involve rotations of Copper or Graphite molecules. The QED model of serially diluted, vigorously shaken polar liquids indicates that such liquids should contain CD_{rot} (Yinnon and Yinnon, 2011). Analyses of experimental data pertaining to such liquids revealed fingerprints of such domains (Yinnon and Liu, 2015c). So the drops which Mahata (2013) added to the water samples probably contained CD_{rot} composed of the polar ethanol molecules. Since such CD_{rot} are aggregates with an electric dipole moment,

QED predicts that these may stabilize CD_{rot} composed of H_2O in the water samples (Del Giudice *et al*., 1988). So it is likely that the 3×10^{-5} m sized molecular structures are CD_{rot}. This is likely because according to QED, electric dipole rotations underlie the CD_{rot} composed of ethanol or water. However, it should be emphasized that Mahata (2013) observed the structures in water containing about 3 percent ethanol. In such aqueous solutions, as when neither the water nor the ethanol are serially diluted and vigorously shaken, domains are present (Sedlak, 2006; Sedlak and Rak, 2013). These domains have characteristics of QED coherent domains (Yinnon and Yinnon, 2009). Recent analyses indicated that in water containing a few percentage of ethanol, fractal structured molecular associates are present (Ghosh and Bagchi, 2016). As noted above, fractality is a property of supra-QED coherent domains[f]. So repeating the experiments of Mahata (2013) for controls is called for. For example, the controls may be water samples to which drops of SDVSAS of Copper or Graphite were added — according to the above and previous published analyses, these drops should contain CD_{rot}. Also, water samples to which drops of non-serially diluted non-vigorously shaken ethanol solutions of Copper or Graphite are added may serve as controls — these liquids should not contain CD_{rot}. Yet, even in the absence of data for controls, the findings of Mahata (2013) are significant. They suggest that dielectric dispersion measurements for SDVSAS diluted below the Avogadro limit seem to be capable of providing more direct evidence of CD_{rot} in these liquids.

Discussion

The above analyses show that the QED model of SDVSAS explains the recently observed self-similarity topology of these

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[f] See the discussions of Vitiello (2012, 2014), respectively, on *"Fractals, Dissipation and Coherent States"* and on *"The isomorphism between dissipative systems, fractal self-similarity and electro-dynamics."*

liquids. This self-similarity, together with the dissipative properties of SDVSAS identified in previous studies (Elia *et al.*, 2000, 2008a&b; Belon *et al.*, 2008; Elia *et al.*, 2015), in combination with the analyses by Vitiello (2012, 2014), corroborates that QED coherence plays a significant role in these liquids.[f] In other words, the above analyses substantiate the model's assumption that QED coherence domains (*e.g.*, CD_{rot}, CD_{elec}^{H2O} and IPD_{plasma}), underlie the structural properties of SDVSAS.

Furthermore, the above analyses show that the QED model of SDVSAS clarifies the EM phenomena of these liquids. The model elucidates some missing links in the explanations of Montagnier *et al.* (2011, 2015) pertaining to the emission of ULF radiation by SDVSAS of DNA.

The above analyses also complement previous ones, which showed that the QED model of SDVSAS consistently explains many structural and physicochemical properties of these liquids (Yinnon and Yinnon, 2011; Yinnon and Elia, 2013; Yinnon and Liu, 2015a-c). The above analyses of the recent DLS, electrical conductivity, pH and surface tension measurements carried out by Ryzhkina *et al.* (2015b) support the model. These recent measurements, which pertain to SDVSAS diluted beyond the Avogadro limit, complement the many earlier measurements for SDVSAS diluted up to this limit. The analyses indicate that the nano-associates observed in SDVSAS diluted below the Avogadro limit are also QED coherence domains, *i.e.*, CD_{rot} and CD_{elec}^{H2O}.

The analyses presented above, together with those presented in earlier publications and schematically portrayed in the lecture by Konovalov *et al.* (2014)[g] and the poster by Yinnon and Konovalov (2014)[h], accentuate that an appropriate description for the molecular associates in SDVSAS is the term recently coined by Konovalov and Ryzhkina (2016): *"nano-sized self-organized substrate-induced molecular ensembles."* The term "substrate" here refers to the solute with which the SDVSAS is prepared. At concentrations slightly lower than the transition concentrations $C_{trans}^{IPDplasma}$ or C_{crit}^{CDrot}, the solute molecules serve as substrates which may stabilize respectively, IPD_{plasma} or CD_{rot}. Vigorous shaking of the liquid excites the domains, *i.e.*, creates electric dipole aggregates. For $C < C_{crit}^{CDrot}$, these aggregates stabilize additional CD_{rot}. Thus for $C < C_{crit}^{CDrot}$, the creation of new electric dipole aggregates no longer depends on the solute molecules. The CD_{rot} may stabilize CD_{elec}^{H2O}. At $C \sim C_{crit}^{CDrot}$ or at lower C, the solute properties become imprinted in the CD_{rot} and CD_{elec}^{H2O}. On diluting the liquid, part of these domains get transferred to the dilute. Vigorous shaking of the dilute breaks up the CD_{rot} and thus creates new electric dipole aggregates. These aggregates stabilize new CD_{rot} and CD_{elec}^{H2O}. At C above the Avogadro limit but below C_{crit}^{CDrot}, the solutes locate within CD_{rot} and may orbit around CD_{elec}^{H2O}. At such low concentrations, solutes still may affect physical, chemical or biological processes, *e.g.*, through resonances (Del Giudice *et al.*, 2010b). For SDVSAS diluted below the Avogadro limit, the current data hint that the physicochemical and biological properties of these liquids are attributable to CD_{rot}, CD_{elec}^{H2O} and the imprint of the characteristics of the substrates (original solutes) in these domains.

The above analyses support the following

[g] Konovalov A. I., Ryzhkina I. S., Yinnon T. A. (2014). Molecular assemblies mediated by electromagnetic fields in highly diluted aqueous solutions: Accordance of experimental data and predicted by electro-dynamic theory data. *VIIth International Symposium "Design and Synthesis of Supramolecular Architectures, Kazan, Russia; Annual Conference on the Physics, Chemistry and Biology of Water, Bulgaria* — http://www.waterconf.org/participants-materials/2014/Presentations/Alexander_Konovalov_Water_Conference_2014.pdf

[h] Yinnon T. A. and Konovalov A. I. (2014). Domains formation mediated by electromagnetic fields in dilute solutions. *Annual Conference on the Physics, Chemistry and Biology of Water, Bulgaria* — see Tamar Yinnon Research Gate.

conclusions, which recently were drawn by Konovalov and Ryzhkina (2016): At concentrations below $C_{thr} \approx C_{crit}^{CDrot}$ but above the Avogadro limit, the characteristics of the nano-associates imply that these associates constitute a phase which is different from the medium, *i.e.*, SDVSAS are nano-heterogeneous. Consequently, these SDVSAS cannot strictly be regarded as solutions in the customary sense. Instead, these are nano-disperse systems. Nano-associates composed of H_2O constitute the disperse phase. According to QED, although not yet experimentally verified, the solute (substrate) molecules locate in the disperse phase. On serial diluting these systems and vigorous shaking after each dilution step, the solute molecules decrease in numbers and ultimately these are not any longer present in the liquid. However, even beyond the Avogadro limit, nano-associates composed of H_2O persist. Thus on serial diluting a SDVSAS beyond the Avogadro limit, in combination with vigorous shaking the liquid after each dilution step, a special kind of disperse system emerges — a disperse system of "water in water." The disperse phase consists of nano-associates composed of H_2O. The electronic structure and the ordering of the H_2O in the associates are determined by the characteristics of the substrate molecules. Therefore, the associates contain the molecular information of the substrate.

The above analyses in combination with previous ones show that the QED model of SDVSAS adequately describes many aspects of these liquids. A shortfall of the model is that it does not specify the relation between the characteristics of the substrates and the physicochemical, structural and bioactive properties of these liquids. Future research directed at such derivations is called for. The challenges involved are not just theoretical. The many experimental studies of SDVSAS hitherto have shed very little light on this relation. For highly diluted SDVSAS, until now, UV, IR, ULF or NMR spectra

did not reveal any *solute specific* effects on their molecules' oscillations or on the structure of their associates. The UV absorbance and fluorescence spectra of SDVSAS prepared from different substrates may vary. However, no specific features attributable to a particular substrate have been identified (Lo,1996; Lobyshev *et al.*, 2005; Wolf *et al.*, 2011; Klein *et al.*, 2013). The spectra consist of broad rather featureless bands. Also in IR and NMR spectra of SDVSAS, no solute specific features have yet been observed (Zubareva *et al.*, 2003a&b; Lo *et al.*, 2009; Demangeat, 2009, 2010; Elia *et al.*, 2015). The group of Elia *et al.* (2012) demonstrated that aqueous SDVSAS emit radiation. They demonstrated this for SDVSAS of fullerene which were 5 to 30 times centesimally diluted and for SDVSAS of carbon nanotubes which were 5 times centesimally diluted. Both these SDVSAS were prepared in glass vessels. However, their analyses did not reveal the impact of the original solute on the emitted radiation. As to ULF emission by SDVSAS, the currently available data are very limited. The data hint that only for certain types of DNA the emission is sufficiently strong to be observable with the method developed by Montagnier *et al.* (2009). The effects of excitation of SDVSAS by 7.85 Hz EM field, observed by D.A. Konovalov *et al.* (2015) and analyzed above, suggest that various non-linear optical processes occur in these liquids. Such processes pose challenges for revealing details of highly diluted SDVSAS with absorption or fluorescence spectroscopy.

So far, to the best of my knowledge, the sole significant findings pertaining to the relation between substrates' characteristics and the physicochemical and structural properties of SDVSAS are:

❖ The bioactivity, physicochemical and structural properties of SDVSAS are interrelated (Lobyshev *et al.*, 2005; Palmina *et al.*, 2009; Ryzhkina *et al.*, 2010, 2011b, 2012b, 2013; Konovalov

and Ryzhkina, 2014). The bioactivity is solute specific.

❖ The thermoluminescence spectra of 10^{-30} g/cm^3 SDVSAS of LiCl and SDVSAS of NaCl differ (Rey, 2003). The difference pertains to the hydrogen bond network in these SDVSAS. The hydrogen bond network in SDVSAS of LiCl is more suppressed than that in SDVSAS of NaCl. Electro-chemical research has shown that Li$^+$ ions attract more hydration H$_2$O than Na$^+$ ions, because the former have a smaller radius (Robinson and Stokes, 2002). As a result, the hydrogen-bond suppressing effect of LiCl salts is larger than that of NaCl salts. Thus, the thermoluminescence measurements revealed that the hydrogen bond suppressing effect of these salts even persists in their SDVSAS diluted to the Avogadro limit.

Uncovering the relation between the substrates' characteristics and physicochemical and structural properties of highly diluted SDVSAS is important for identifying the processes underlying their bioactivity. Several hypotheses concerning the active principle in SDVSAS have been forwarded:

♦ Yinnon and Liu (2015c) surmised that the CD_{elec}^{H2O} present in SDVSAS constitute the active principle, e.g., that the electronic structure of the substrate leaves an imprint on the pool of quasi-free electrons (QFE) of CD_{elec}^{H2O}. The substrate affects the vortices in the QFE which are induced by the vigorous shaking of the SDVSAS. Moreover, they conjectured that the polarity of the substrate affects the ordering of CD_{rot}. They based their conjecture on the experimental findings of Konovalov and Ryzhkina (2014) - for SDVSAS with ~10^{-18} M$<C<C_{thr}$, their bioactivity is related to the hydrodynamic diameter and the electrokinetic potential of their molecular associates with sizes of the order of 10^{-7} m. Yinnon and Liu (2015c) showed that these associates

have the typical properties of CD_{elec}^{H2O}. Based on their analyses, they surmised that these domains affect electron transport processes. Experiments have indicated that SDVSAS of Melafen [the melamine salt of bis (hydroxymethyl) phosphinic acid dihydrate] affect electron transport in the mitochondrial respiratory chain, even when the concentration of these solutions is as low as 10^{-18} - 10^{-22} M (Zhigacheva et al., 2009).

♦ Chakraborty et al. (2015) and Bell et al. (2015a) have surmised that the bioactivity of SDVSAS is related to traces of the original solute molecules. These molecules may organize in structures composed of compounds released by the containers. Traces of solutes in 30 and 200 times centesimally serially diluted, vigorously shaken solutions of several metals, with ethanol as solvent, have been reported (Chikramane et al., 2012). The solutions were prepared by pharmaceutical companies. However, a follow up study by Bell et al. (2015b) showed that inductively-coupled plasma mass spectroscopy could not detect traces of silver in 30 and 200 times centesimally serially diluted, vigorously shaken solutions of silver with ethanol as solvent. These studies and others also detected presence of compounds released by the glass containers, their cork stoppers and other impurities in extremely diluted solutions (Bell et al., 2015c; Witt et al., 2006; Elia and Napoli, 2010).

It is, however, doubtful that traces of the original solute could be the solute specific bioactive element in SDVSAS, as hinted by the following experimental results. As shown by Konovalov and Ryzhkina (2014), for SDVSAS with 10^{-18} M $<C<C_{thr}$, their solute specific bioactivity disappeared when samples were stored under hypo-electromagnetic conditions, (in Permalloy containers). Under such storage, their 10^{-7} m and 10^{-6}-10^{-5} m sized

associates, which mainly are composed of H_2O, also disappeared. Moreover, after such storage, the physicochemical properties of SDVSAS returned to those of serially diluted solutions which were not vigorously shaken after each dilution step. In other words, after storage in Permalloy containers, neither the very tiny amounts of solutes still present in solutions with 10^{-18} M $<C<C_{thr}$, nor the compounds released by the containers or other impurities, could trigger specific bioprocesses. Furthermore, Montagnier *et al.* (2011) demonstrated the following for a sample of SDVSAS of DNA kept in a closed plastic Eppendorf tube and a sample of SDVSW kept in a closed plastic Eppendorf tube, which were both placed in the same Permalloy container and exposed for 18 h to a very low frequency (7 Hz) magnetic field: The ULF radiation emitted by the SDVSAS induced solute specific properties in the SDVSW. In other words, even when there was no direct contact between the SDVSAS samples and the SDVSW samples, they had similar properties. Additionally, Davenas *et al.* (1988) showed that for serially diluted, vigorously shaken solutions of anti-IgE antibody diluted below the Avogadro limit, their bioactivity varied quasi-periodically with the number of dilution steps (NDS). It is hard to envision, within the context of the customary electro-static theories, that compounds released by the containers or other impurities can induce such quasi-periodic patterns in liquids.

Very recent findings raise another hypothesis for the bioactivity of highly diluted SDVSAS. Capolupo *et al.* (2016) showed that for DNA and enzymes embedded in water, the electric dipole-dipole interactions between the aromatic ring structures in DNA and the enzymes may get correlated by the QED interactions between rotating H_2O. In other words, the DNA radiative dipole field triggers collective dipole waves in its sur-

rounding water, *i.e.*, stabilizes CD_{rot}, which in turn couple with the enzyme's radiative dipole field. Based on these findings by Capolupo *et al.* (2016), it is plausible that by adding SDVSAS to the DNA-enzyme-water system, the CD_{rot} present in the former affect the CD_{rot} present in the latter. For SDVSAS, the relation between the substrate and the ferroelectric ordering of the H_2O constituting their CD_{rot} have not yet been derived. However, analogous to the imprint of information in magnetic materials, the substrates' properties can affect the ferroelectric orderings of CD_{rot}. Some experimental findings provide support for this hypothesis. Belov *et al.* (2004) showed that when only polar solvents were used for preparing serially diluted, vigorously shaken liquids with concentrations below 10^{-18} M and and even when such liquids were diluted beyond the Avogadro limit, these could affect microsomal membranes. CD_{rot} only form in polar liquids. It is thus possible that the ferroelectric ordering of the solvent molecules constituting CD_{rot} affects membranes. However, it is also possible that the presence of CD_{rot} in serially diluted, vigorously shaken polar liquids is required just for stabilizing the domains consisting of solvent molecules, which coherently transit between two of their electronic states, such as the CD_{elec}^{H2O} in water and their analogues in other polar liquids. In case the sole function of CD_{rot} is stabilization of CD_{elec}^{H2O}, the latter can be regarded as the unit carrying the substrates' information relevant for bio-processes.

Summary and Conclusions

The importance of SDVSAS for toxicology, pharmacology and agriculture mandates research directed at uncovering their bioactive principle. During the last decades, much information has been gathered on the physicochemical and structural properties of SDVSAS. For such properties that still require additional clarification, some

warranted research projects have been specified in the previous sections. During the last decades, much information has also been gathered on the physicochemical and structural properties of serially diluted solutions, which were prepared with ethanol or other polar solvents instead of water. The QED models of all these solutions are very similar. The models provide consistent explanations for the liquids' physicochemical and structural properties, as shown in previous publications and supplemented in the current paper.

Experimental data and QED theory, however, leave us in the dark in regards to the relation between the bioactivity of the aforementioned solutions and their physicochemical properties. Experimental data and QED theory also leave us in the dark about the relation between the characteristics of the substrates (solutes) and the physicochemical properties of the aforementioned solutions. Therefore, research projects are called for that aim to develop techniques capable for elucidating the relation between the substrates' characteristics and the structure, physicochemical and bioactive properties of highly diluted SDVSAS. At the current stage of knowledge, only few vague road signs are available for directing such research (*e.g.*, electron transport processes may be involved.) Yet, the success of the QED model for elucidating the physicochemical and structural properties of serially diluted polar liquids indicates that it can serve as a tool for identifying techniques which can reveal the information of the substrate's imprint in these liquids.

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Table 1: *List of abbreviations in alphabetic order, followed by Greek symbols abbreviations*

Abbrev.	Explanation
A/m	Amperes per meter
C	Concentration
Cx	X times centesimally diluted
C_{crit}^{CDrot}	Critical concentration below which CD_{rot} may form
$C_{trans}^{CDplasma}$	Transition concentration for CD_{plasma} formation
$C_{trans}^{IPDplasma}$	Transition concentration for IPD_{plasma} formation
C_{thr}	Threshold concentration below which no domains are present in SDVSASES samples with concentrations above the Avogadro limit and screened by Permalloy.
CD_{elec}^{H2O}	Coherence domain composed of coherent electronically excited water molecules
CD_{plasma}	Coherence domain composed of few solvated solutes performing coherent plasma oscillation and numerous H_2O molecules

CD_{rot} — Coherence domains of ferroelectric ordered H_2O molecules

DLS — Dynamic light scattering

DNA — Deoxyribonucleic acid

E_{coh} — The energy a H_2O gains by participating in the coherent oscillations of the molecules in a domain.

EDA^{CDrot} — Excited or broken CD_{rot} piece, which has an electric dipole moment and therefore is denoted electric dipole aggregate (EDA)

$EDA^{IPDplasma}$ — Excited or broken IPD_{plasma} piece, which has an electric dipole moment and therefore is denoted electric dipole aggregate (EDA)

EM — Electro magnetic

ELF — Extremely low frequency

EZ — Exclusion zone

eV — Electron Volt

H_2O — Water molecule

H_3O^+ — Hydronium ion

Hz — Hertz

IPD_{plasma} — In phase domains composed of few solvated solutes and numerous solvent molecules performing in phase plasma oscillation.

IR — Infra red

LiCl — Lithium Chloride

M — Molarity in mol per liter

m — Meter

mg — Milli gram

ml — Milli liter

NaCl — Sodium Chloride

NDS — Number of dilution steps

ng — Nano gram

nm — Nano meter

NMR — Nuclear magnetic resonance

OH^- — Hydroxide ion

Q — Heat of mixing of SDVSAS with acids or bases

QED — Quantum electro-dynamics

QFE — Quasi free electrons

SDVSAS — Serially diluted vigorously shaken aqueous solution

$SDVSAS_{LB}$ — Serially diluted vigorously shaken aqueous solution kept on laboratory bench

$SDVSAS_{LB}^{a-b}$ — SDVSAS of anti-bodies kept at the laboratory bench

$SDVSAS_P$ — Serially diluted vigorously shaken aqueous solution kept in Permalloy container

$SDVSAS_P^{a-b}$ — SDVSAS of anti-bodies kept in Permalloy containers

Supra-CD_{elec}^{H2O} — Agglomerate of CD_{elec}^{H2O}

Supra-CD_{rot} — Agglomerate of CD_{rot}

TGA — Thermogravimetric analyses

ULF — Ultra low frequency (500-3000 Hz)

UV — Ultra-violet

v_c — Cyclotron frequency

v_p — Frequency of plasma of quasi free electrons of CD_{elec}^{H2O}

σ — Surface tension

χ — Electrical conductivity in $\mu S\ cm^{-1}$

Discussion with Reviewers

Reviewer A:

I am very curious if there is in the field any consideration on the possible role of gas bubbles inevitably included into the system during vigorous shaking procedure (necessary for manifestation of many of discussed properties of very diluted solutions). There are many of experimental evidences of existence of stable nanobubbles in water. If those gas inclusions are present and are filled with ambient air containing for example CO_2 gas and water vapor (as in the theory of vesicles in a liquid presented by Prof.

Pollack), both of those gases, CO_2 and H_2O, are so called greenhouse gases. Therefore, they absorb and re-emit infrared radiation and this radiation can stimulate formation of coherent domains of water. If we then cut-off external radiation we also cancel the effect of the gases. Could therefore gas bubbles be related to the effect of vigorous shaking on some properties of diluted solutions?

Yinnon TA:

Indeed, many researchers have considered the possibility that gas bubbles affect or even underlie the specific properties of SDVSAS. During preparation of SDVSAS, in particular when the vigorous shaking is carried out by succussions, not just nanobubbles are created but the formation of gas bubbles is observable with the naked eye. Of course, the bubbles affect some physicochemical properties of the liquid. Your conjecture that gas bubbles enhance formation of coherent domains indeed is plausible. However, by analyzing extensive data, researchers have shown that the specific physicochemical and bioactive properties of SDVSAS are not attributable to gas bubbles alone. Below, I suffice with presenting only a few arguments which reject the option that gas bubbles underlie all the measured physicochemical and bioactive properties of SDVSAS.

i. As noted in the introduction, for aqueous solutions of many but not all compounds which are bioactive at physiological concentrations (about 10^{-2} to 10^{-7} mol/liter), their serial dilutions beyond a transition concentration (C_{thr}), when combined with vigorous shaking after each dilution step, endow these with a specific bioactivity. C_{thr} depends on the solute. Solute specific bioactivity has been observed for SDVSAS diluted up to the Avogadro limit and beyond it. For such highly diluted

SDVSAS, their bioactivity is correlated with their various physicochemical properties. Moreover, their bioactivity non-monotonically changes with the number of dilution steps. Sometimes, the bioactivity of SDVSAS at concentrations above C_{thr} differs from that at concentrations below C_{thr}. For example, Ryzhkina *et al.* (2013) have demonstrated that SDVSAS of Mebicar at 1×10^{-3} mol/liter exhibits an anxiolytic effect in mice, *i.e.*, it provides a sedative effect combined with a decrease in exploratory activity while retaining the motion activity. However, at concentrations below 10^{-5} mol/liter, the liquid has a psychostimulating effect, *i.e.*, it increases the motion and exploratory activities of the mice. The physicochemical properties of this liquid show that $C_{thr} \approx 10^{-5}$ mol/liter (Ryzhkina *et al.*, 2013) — in other words, on screening the liquid from ambient EM fields, at concentrations below this C_{thr}, its molecular associates disappear and its physicochemical properties become those of the solvent. At 1×10^{-7} mol/liter, the psychostimulating effect of the SDVSAS of Mebicar has a maximum. At concentrations of 1×10^{-10} and 1×10^{-14} mol/liter, the psychostimulating effect decreases. However, at 1×10^{-14} mol/liter, the motion activity increases.

On shielding the SDVSAS of Mebicar from ambient EM fields, at concentrations below $C_{thr} \approx 10^{-5}$ mol/liter, Ryzhkina *et al.* (2013) observed: "the mean pH values of the solutions (pH 6) do not change, the $\Delta\varepsilon$ values[i] of the solutions approach zero; and α (the optical activity) does not reliably exceed the measurement error." Moreover, these researchers observed that the screening led to the disappearance of the 10^{-7} - 10^{-4} m sized molecular associates. Accordingly, Ryzhkina *et al.* (2013) pointed out: "*These findings serve as evidence that, in spite of the possibil-*

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[i] $\Delta\varepsilon$ is the difference between the dielectric constant of this SDVSAS and that of the pure solvent, *i.e.*, twice distilled water.

ity of the influence of such contaminations as CO_2 from the atmosphere or Ca^{2+} and silicate ions from glass, the true cause of the nonlinear concentration dependences of physicochemical properties is the formation and rearrangement of nanoassociates. If there are no nanoassociates, the properties of low-concentration Mebicar solutions correspond to the properties of distilled water." The phenomena observed by Ryzhkina *et al.* (2013) are repeatable. They carried out their experiments in triplicate.

ii. The UV absorbance spectra of SDVSAS have the typical features of structured water (Elia *et al.*, 2014c, 2015). Examples of structured waters are water adjacent to Nafion or other hydrophilic membranes [*i.e.*, exclusion zone (EZ) water], aqueous solutions, water iteratively perturbed by Nafion membranes or by filtrations (Zheng *et al.*, 2006; Chai *et al.*, 2008; Elia *et al.*, 2013a, 2014b-c). For aqueous solutions, *i.e.*, those which were vigorously shaken as well as those which were *not* vigorously shaken, dynamic light scattering in combinations with other analyses revealed H_2O ordered in sub-micron sized aggregates, *which are not due to gas bubbles* (Sedlak, 2006, 2013; Ryzhkina *et al.*, 2012c; Konovalov, 2013; Ryzhkina *et al.* 2013; Konovalov and Ryzhkina, 2014). Examples for such aqueous solutions are alkali halide, sugar or alcohol solutions with concentrations in the range of 1 M to 0.1 M and the SDVSAS referred to in this paper.

SDVSAS, just as all the above mentioned structured waters, absorb in the 200 - 400 nm wavelength range. Typically, these waters have a peak in the 205 - 210 nm range. The peak is similar to the one observed for magnetized water (Pang, 2014). In addition, these waters have a broad peak in the 225 - 325 nm range. This broad peak has a maximum around 260 - 280 nm. A similar broad peak was first observed in water by Larzul *et al.* in 1965. Subsequent extensive research showed that pure water (gas phase H_2O, bulk water, amorphous, hexagonal or cubic ice) does not absorb in the 200 - 400 nm range (Quickenden and Irvin, 1980; Mulliken and Ermler, 1981; Cabral do Couto and Chipman, 2012; Segarra-Martí, 2013).[j] Hence, the peaks in the spectra of water studied by Larzul *et al.* in 1965 have been attributed to some impurities.

For the 225 - 325 range peak, computations have recently been carried out with a high level, well established, quantum **mechanical**, *ab initio* derived theory, *i.e.*, the Complete-Active-Space Self-Consistent-Field second-order perturbation theory (CASPT2). These computations indicate that the aggregates in structured waters are composed of excimers (Segarra-Martí *et al.*, 2014). In particular, CASPT2 analyses of EZ water adjacent to Nafion membranes have show that these excimers form networks of multilayer honeycomb ice-like layers [see Fig. 1 and 6 in Segarra-Martí *et al.*(2014)]. The formula of the excimers is $H_{38}O_{20}$. The formula of the two monomers constituting the excimer is $H_{19}O_{10}$. The H_2O forming the monomer are organized in two fused hexagons. In the $H_{38}O_{20}$ excimer, each of the two central (fused) H_2O of one monomer attracts its opposite central (fused) H_2O of the other monomer [See Fig. 6 in Segarra-Martí *et al.*(2014)]. The attractive interaction

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[j] For gas phase H_2O, its lowest-energy band of the electronic spectrum covers the 151 - 182 nm range, with a maximum at 168 nm (Segarra-Martí *et al.* 2013; Cabral do Couto and Chipman, 2012). In pure bulk liquid, this band is broader and blue-shifted (Quickenden and Irvin,1980; Segarra-Martí *et al.* 2013; Cabral do Couto and Chipman, 2012). Its maximum is at 151 nm and its absorbance falls off monotonically by about ten orders of magnitude in the 151 - 400 nm range [it is only about 0.0001 cm^{-1} at 320 nm -- see (Quickenden and Irvin,1980)]. Amorphous, hexagonal or cubic ice have absorbance features similar to those detailed in the previous sentence (Segarra-Martí *et al.* 2013; Cabral do Couto and Chipman, 2012).

is due to π-stacking. In the $H_{38}O_{20}$ networks, the π-stacked H_2O resonate between their ground and an excited electronic state. About 10 percent of the H_2O constituting the aggregates are simultaneously electronically excited.

The quantum *mechanics* CASPT2 results correspond to those obtained by the quantum *dynamic* model of structured waters, *i.e.*, the quantum electro-*dynamic* model (QED). According to QED, the aggregates underlying the UV absorption in the 200-400 nm in structured waters are composed of coherent domains of the type denoted CD_{elec}^{H2O} in this paper (Del Giudice *et al.*, 2013; Elia *et al.*, 2015 & 2017; Yinnon *et al*, 2015b&c, 2016). Within these domains, the H_2O get electronically excited by ambient radiation with wavelength λ. These H_2O deexcite by emitting photons which immediately get absorbed by H_2O located within a distance λ. The subsequent repetitive excitation-deexcitation processes associate these H_2O into an aggregate. About 10 percent of the H_2O within a CD_{elec}^{H2O} simultaneously reside in their excited electronic state (Del Giudice *et al.*, 2013).

A significant similarity between the quantum *mechanical* $H_{38}O_{20}$ and quantum *dynamic* CD_{elec}^{H2O} models is their predictions concerning the following property of the H_2O constituting the aggregates: About 10 percent of these H_2O simultaneously reside in their electronic excited state. One of the differences between the models is that the former predicts that the π-stacked H_2O resonate between two of their electronic states (Segarra-Martí *et al.* 2014), while the latter predicts that all of the about 10^6 H_2O composing a CD_{elec}^{H2O} resonate between two of their electronic states (Del Giudice *et al.*, 2013).

The aggregates in the structured waters are observable with fluorescence micros-copy (Elia *et al.*, 2013a, 2014c, 2017). Moreover, the aggregates are isolatable by evaporating in air drops of structured water or on lyophilizing (Lo *et al.*, 2009; Ryzhkina *et al.*, 2012b; Elia *et al.*, 2013a, 2014b&c). The isolated aggregates are observable with atomic force microscopy and scanning electron microscopy (Lo *et al.*, 2009; Ryzhkina *et al.*, 2012b; Elia *et al.*, 2013a, 2014b&c, 2017). The morphology of the isolated aggregates depends on the type of structured waters, their preparation techniques and concentration. State-of-the-art chemical analyses have indicated that these isolated aggregates mainly are composed of H_2O (Elia and Napoli, 2010; Elia *et al.,* 2017). On dissolving the isolated aggregates in Milli-Q water, the resulting liquid has physicochemical properties resembling those of the structured water from which it was prepared (Elia *et al.*, 2014c, 2017). For example, its UV (absorption, fluorescence and circular dichroism) spectra have similar properties, *e.g.*, the typical absorption peaks in the 200 - 400 nm range specified above. The fact that the chemical analyses have shown that the isolated aggregates are composed of H_2O, *implies that the aggregates are not a kind of isolated bubbles*. Of course, in the liquid phase, air bubbles may adhere to the aggregates.

Reviewer A:

Yet another notion, when vigorously shaking my solutions of ionic liquids (not very diluted and not related to the discussed field of investigation) I found it virtually impossible to avoid contamination by airborne fungi. I have been using sterile containers and filtered my solutions through 0.22 μm filters, yet after some time I could always observe fungi growth in solutions where they had organic feeding (ionic liquid) material. Is it possible that in case of non-sterile air mixed with dilute vigorously shaken samples we introduce fungi spores

that can produce some artifacts in the analyzed samples?

Yinnon TA:

Paragraph ii of the Discussion with Reviewers Section partly is also relevant to this question. However, I would like to add the following remarks:

○ The group of Prof. Elia carried out many carefully controlled experiments to exclude the possibility that the differences between SDVSAS and aqueous solutions of equal chemical composition but prepared without serial dilutions and vigorously shaken are attributable to contaminants of any kind (Elia and Niccoli, 2004a&b; Ciavatta *et al.*, 2008; Elia and Napoli, 2010).

○ Highly controlled, blinded and randomized experiments of UV radiation transmission by SDVSAS were carried out by Wolf *et al.* (2011) and Klein *et al.* (2013). Statistical analyses of the data have shown that the UV radiation's transmission of these aqueous SDVSAS significantly differed from that of the controls. The controls were vigorously shaken solvents. These solvents were the various types of purified waters or the 99% water and 1% ethanol mixture with which the SDVSAS were prepared. The purified waters included distilled water, Quartz distilled water and de-ionized water. Part of the experiments were carried out in a metal-free class 100 High Efficiency Particulate Air (HEPA) filtered clean room. Clean flow boxes had class 5. These experiments showed that the UV radiation's transmission by SDVSAS of copper sulfate ($CuSO_4$), or aqueous SDVSAS of hypericum is lower than that of the control. However, for SDVSAS of sublimed sulfur (S_8), it is higher than the control.

Reviewer B:

How does the author correlates her de-scription of SDVSAS to the recent finding of V. Elia and collaborators concerning the supramolecular aggregates of water molecules remaining solid at ordinary pressure and room temperature both in homeopathic solution and also in other pure water physical treatments (iterative filtration, iterative exposure to Nafion…) presented in Elia *et al.* (2013a, 2014b, 2015, 2017) ?"

Yinnon TA:

In paragraph ii of the Discussion with Reviewers Section, I already mentioned that the group of V. Elia has shown that iteratively perturbing polar liquids (*e.g.*, water, alcohol or their solutions) may lead to stabilization of 10^{-7} - 10^{-4} m sized supramolecular aggregates, which mainly are composed of solvent molecules. Such aggregates may stabilize when such liquids are iteratively perturbed by:

☐ serial filtrations (Elia *et al.*, 2014b, 2015);

☐ repeated exposure to Nafion (Elia *et al.*, 2013a&b, 2014a, 2015, 2017).

☐ repeated shaking after each dilution step of serially diluted liquids, *i.e.*, the method used for preparing homeopathic liquids and SDVSAS (Lo *et al.*, 2009; Upadhyay and Nayak, 2011; Konovalov, 2013; Elia *et al.*, 2014c, 2015).

These aggregates are isolatable by evaporating-in-air drops of the perturbed liquid or by lyophilizing it. Such evaporation techniques leave a solid residue. For water perturbed by serial filtrations or repeated exposure to Nafion, the residue is composed of H_2O and contains minuscule amounts of impurities (organic contaminants and compounds released by the container or the Nafion).

Analyses of the physicochemical properties of the residues have revealed that their aggregates have characteristics of QED coherent domains (Elia *et al.*, 2015, 2017; Yinnon *et al.*, 2016). In particular, the ther-

mogravimetric analyses of the residue of water perturbed by repeated exposure to Nafion have revealed some of these characteristics. The thermogravimetric graph shows that the residue contains two types of H_2O aggregates. One type has the thermodynamic properties of CD_{rot} and the other type has the thermodynamic properties of CD_{elec}^{H2O} (Yinnon *et al.*, 2016). For example, the thermogravimetric graph reflects the critical temperatures below which these domains form. These measured temperatures are in good agreement with those predicted by classical electro-dynamic and quantum electro-dynamic (QED) models of aqueous systems.

The currently available analyses of water iteratively perturbed by Nafion or serial filtrations indicate that their aggregates, *i.e.*, their CD_{rot} and CD_{elec}^{H2O}, are clumps of EZ water (Elia *et al.*, 2015, 2017; Yinnon *et al.*, 2016). These aggregates became stabilized by Nafion or the filter, *i.e.*, constitute the interfacial EZ water. The iterative perturbations tore clumps of these aggregates from the interfacial zone and dispersed these in the liquid.

My QED model of SDVSAS points out that serially diluting polar liquids below a transition concentration ($C_{trans}^{IPDplasma}$) may create a QED domain, which I denoted IPD_{plasma}. In the paper by Yinnon and Yinnon (2012), the possibility for formation of IPD_{plasma} was derived for the first time. In that paper, experimental data in support of this possibility was cited. (After the paper was published, the late Prof. Del Giudice wrote me that he had been one of the reviewers.) Also the current paper's analyses of experimental data, as well as the analyses by Yinnon and Liu (2015b&c), show that the characteristics of IPD_{plasma} conform to those observed for aggregates present in aqueous solutions. The effects of vigorous shaking of a solution on their IPD_{plasma} are first analyzed in the paper

by Yinnon and Yinnon (2011). The central finding of this paper is that shaking may cause creation of aggregates with an electric dipole moment (EDA). In the Yinnon and Yinnon (2011) paper, it also is pointed out that on further diluting below a critical concentration C_{crit}^{CDrot} the EDA or solutes with a sizable electric dipole moment may stabilize CD_{rot} and CD_{elec}^{H2O}. As discussed in the current paper and my previous ones on SDVSAS, the CD_{rot} and CD_{elec}^{H2O} underlie these liquids' physicochemical properties. In other words, it was the theoretical discovery of IPD_{plasma}, $C_{trans}^{IPDplasma}$ and EDA, in combination of my identification of the extensive experimental data in the literature confirming the presence of aggregates with characteristics of IPD_{plasma}, $C_{trans}^{IPDplasma}$, EDA, CD_{rot}, CD_{elec}^{H2O} and C_{crit}^{CDrot} which enabled me to provide consistent explanations for the many observed physicochemical properties of serially diluted vigorously shaken polar liquids. Here, I emphasized the importance of the theoretical identification of IPD_{plasma}, $C_{trans}^{IPDplasma}$ and EDA for leading the way to understand SDVSAS. Just claiming that the QED model of aqueous systems is more accurate than the customarily electrostatic models is not sufficient for explaining the properties of SDVSAS. The effects of vigorous shaking on aggregates resulting from QED interactions have to be carefully assessed.

The previous paragraphs accentuate that the current available analyses all show that the CD_{rot} and CD_{elec}^{H2O} QED domains may become stabilized by iteratively perturbing water by serial filtrations or by repeated exposure to Nafion, as well as by repeated shaking after each dilution step of serially diluted liquids. Stabilization of such domains implies breaking of the symmetry of the liquid. The group of G. Vitiello has extensively studied such symmetry breaking. Examples are the following excerpts in the paper "Classical Limit and Spontaneous

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Breakdown of Symmetry as an Environment Effect of Quantum Field Theory" by Celghini, Graziano and Vitiello *Phys Lett* A 145:1-6 (1990):

➔ "Small reaction fields arising from the system-environment coupling are shown to drive the system into an asymmetric ground state in the group contraction limit. This limit acts as the theory of classical limit."

➔ We believe it is therefore worthwhile to move the discussion on small reaction fields from the quantum mechanics setting to QFT[k] which is the natural framework for phase transition and spontaneous breakdown of symmetry, and ..."

➔ "Every very tiny perturbation arising from the *system-environment* interaction (reaction field) may trigger the breakdown of symmetry thus producing long range correlations in the system."

➔ "The system order parameter does not depend on the details of the environment state."

➔ "Finite volume effect set a threshold above which the intensity of the reaction field must be in order to have symmetry breakdown."

➔ "The discussion presented in this paper shows that in QFT small reaction fields from the environment may play a crucial role in triggering the breakdown of continuous symmetry with the formation of ordered patterns in the system."

The above implies that the effects of iterative perturbations of water, polar liquids and their solutions, as analyzed by the group of V. Elia, A. Konovalov and others in the years 1999 - 2017 indeed conform to those predicted by the group of G. Vitiello in 1990.