

# Domains Formation Mediated by Electromagnetic Fields in Very Dilute Aqueous Solutions:

## 3. Quantum Electrodynamic Analyses of Experimental Data on Solutions of Weak Electrolytes and Non-electrolytes

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For a list of abbreviations, see **Table 2** on page 92.

*(This is the third part of a three-part series.)*

### Abstract

Molecular associates in very dilute solutions of weak electrolytes or non-electrolytes kept at ambient conditions are studied. Ambient electromagnetic fields affect some of the associates' types. For concentrations ( $C$ ) in the range of about  $10^{-7}$  -  $10^{-20}$  M, the associates were observed only for solutions with polar solvents, *e.g.*, water, chloroform. At such very low  $C$ , sizes of these associates reaching tens of micrometers imply that these mainly are composed of solvent molecules. Some associates' properties are correlated with the bioactivity of the solutions. As discussed in our two preceding publications in this journal's issue, electromagnetic fields affecting associates entail electrodynamic forces have to be explicitly described. We employ a quantum electrodynamic model for analyzing experimental data pertaining to the associates' properties and their

impact on the solutions physicochemical characteristics. Our analyses show that associates mediated by electromagnetic fields have the typical characteristics predicted by the model. Our analyses clarifies the prerequisites of polar solvents, serial dilutions and vigorous shaking after each dilution step for stabilizing associates in solutions with  $C$  below about  $10^{-7}$  M. Our analyses provide clues for the domains impact on bio-systems.

### Introduction

During the last decade  $10^{-7}$  -  $10^{-4}$  m sized molecular associates were observed in polar liquids and their very dilute solutions kept at ambient conditions. For example, in water or chloroform containing non-electrolytic, weak or strong electrolytic solutes, with concentrations ( $C$ ) in the range of  $10^{-7}$  -  $10^{-20}$  M. At such ultra low concentrations (ULC), the huge sizes of these associates imply these are mainly composed of sol-

vent molecules. Some of these solutions are bioactive, *e.g.*, these are antioxidants, plant growth regulators, neuromediators. For reviews see Konovalov and Ryzhkina (2014a) and Elia *et al.* (2015). Stabilization of associates in ULC solutions have been induced by several perturbative techniques, *e.g.*, serial dilutions with vigorous shaking after each dilution step, iterative filtering liquids, iterative agitating liquids with a Nafion membrane. Independent research groups with a wide variety of techniques evidenced presence of the associates. Therefore it can be ruled out that the relevant data is due to measurement errors. As to impurities released by containers, by filters or by membranes, these affect the perturbed liquids but cannot account for their typical properties -- see Elia and Niccoli (2004), Elia *et al.* (2013, 2015), Yinnon and Liu (2015a,b).

With water, other polar liquids and their solutions playing central roles in many physical, chemical and biological systems, knowledge about the associates' properties and the physics underlying these is important. Analyses of associates in serial diluted vigorously shaken aqueous strong electrolyte solutions (SDVSASES) we presented in the paper preceding this publication in Water journal. For analyses of iterative filtered water or water agitated with a Nafion membrane (so called Iterative Nafionized Water) see Elia *et al.* (2015), Yinnon and Elia (2013), Yinnon *et al.* (2015c in press). The foci of our current manuscript are associates in polar liquids' solutions which were serial diluted, vigorously shaken after each dilution step and contain weak or non-electrolytic solutes. We denote these Serial Diluted Vigorously Shaken Polar Liquids of Weak Electrolytes or Non-Electrolytes as  $\text{SDVSPL}^{\text{we-ne, a}}$ .

<sup>a</sup>  $\text{SDVSPL}^{\text{we-ne}}$  preparation involves serial decimal or centesimal diluting a "stock" solution. Hitherto, mainly aqueous  $\text{SDVSPL}^{\text{we-ne}}$  have been studied, but effects of other solvents also was investigated. Aqueous  $\text{SDVSPL}^{\text{we-ne}}$

Molecular association in ULC  $\text{SDVSPL}^{\text{we-ne}}$  initially was revealed with calorimetric, electric conductivity ( $\chi$ ) and pH measurements (Elia and Niccoli, 1999, 2000, 2004). Vigorous shaking after each dilution step is a crucial perturbation required for inducing such association. Serially diluting solutions while omitting the vigorous shaking do not lead to molecular association for  $C$  below a solute type dependent critical concentration ( $C_{\text{crit}}$ ), *i.e.*, these solutions have the customary infinite diluted solutions characteristics. Typically  $\sim 10^{-10} \text{ M} < C_{\text{crit}} < \sim 10^{-6} \text{ M}$ . Not all solutes facilitate molecular association in ULC  $\text{SDVSPL}^{\text{we-ne}}$  (Konovalov, 2013). The required solute attributes are not yet clarified.

The sizes of associates in ULC  $\text{SDVSPL}^{\text{we-ne}}$ , their electrokinetic potential ( $\zeta$ -potential) and their impact on the liquid's dielectric permittivity were first uncovered with dynamic light scattering (DLS), electrophoresis and dielcometric titrations (Konovalov *et al.*, 2008; Ryzhkina *et al.*, 2009a). Stabilization of the associates occurs during about 1-18 hours after preparation of  $\text{SDVSPL}^{\text{we-ne}}$ . For solute type dependent  $C$  ranges, correlation was observed between ULC  $\text{SDVSPL}^{\text{we-ne}}$ 's bioactivity,  $\chi$ , their associates' effective hydrodynamic diameter  $d_{\text{eff}}$  are prepared with freshly doubly distilled water or water purified by Simplicity® Water Purification Systems - Millipore, with specific electrical conductivity below  $2.5 \mu\text{S/cm}$ . Dust is removed. Stock solution is analyzed for absence of impurities. Concentration of stock solutions are in the  $10^{-1} - 10^{-3} \text{ M}$  range. After each dilution step,  $\text{SDVSPL}^{\text{we-ne}}$  are vigorously shaken, *e.g.*, with lab dancer shaker, by vertical vortexing or other methods. Plastic or glass vessels are used. Temperature and pressure are kept constant, typically, respectively, at 298 K and about 1 Atmosphere. As controls, the solvent, *e.g.*, water, is serial diluted and shaken after each dilution step, with all experimental parameters identical to those of  $\text{SDVSPL}^{\text{we-ne}}$  preparation.

and  $\zeta$ -potential; all these properties non-linear depend on  $C$  and are reproducible (Ryzhkina *et al.*, 2009a,b, 2010a,b, 2011a-e, 2012a-d, 2013; Konovalov *et al.*, 2014a-c).

The origins of the physical, chemical, catalytic and bio-active properties of associates in ULC SDVSPL<sup>we-ne</sup> are not yet clarified. As to forces underlying the molecular association in ULC SDVSPL<sup>we-ne</sup>, electro-dynamic ones play a crucial role. As first demonstrated in 2011 by Konovalov's group, on storing samples under hypo electro-magnetic conditions, *i.e.*, in a Permalloy container with residual field of 10 nano Tesla, no associates are observable for  $10^{-20}\text{M} < C < C_{\text{thr}}$ ; a weak electro-magnetic field (EMF) influences the associates for  $C_{\text{thr}} < C < \sim 10^{-4}\text{ M}$  (Ryzhkina *et al.*, 2011d, 2012a, c, 2013; Konovalov, 2013, 2014a-c). The threshold concentration  $C_{\text{thr}}$  is solute type dependent, typically  $10^{-10}\text{ M} < C_{\text{thr}} < 10^{-6}\text{ M}$ . These findings signify explaining ULC SDVSPL<sup>we-ne</sup> phenomena necessitates electrodynamic theory. The quantum electrodynamic (QED) model for SDVSPL<sup>we-ne</sup> proposed by Yinnon and Yinnon (2011) has provided consistent explanations for various phenomena, *e.g.*, SDVSPL<sup>we-ne</sup>'s heat of mixing,  $\chi$  and their dependence on time and volume (Yinnon and Elia, 2013).

The goals of this paper are: (a) employing the QED model of SDVSPL<sup>we-ne</sup> for explaining recently observed (and to the best of our knowledge yet unexplained) characteristics of the various associate types present in these liquids; (b) elucidating the associates' impacts on these liquids' properties, *e.g.*, their dielectric permittivity, spectra and bioactivity. This paper is the third of a series on associates in serial diluted solutions printed in this journal's issue. In the first paper of the series, we presented a summary of the main QED theory aspects of polar liquids in general, and water in particular, relevant to serial diluted solutions. Hitherto this theory mainly is employed for explain-

ing special phenomena and therefore readers may be unfamiliar with these aspects. In the second paper: we summarized the model proposed by Yinnon and Yinnon (2011) for serial diluted vigorously shaken aqueous strong electrolyte solutions (SDVSASES), and we showed that the model's predictions pertaining to the properties of the associates present in these liquids conform to the measured ones. In the current paper, in its Theory section we concisely summarize the SDVSPL<sup>we-ne</sup> model proposed by Yinnon and Yinnon (2011). The model is similar to that of SDVSASES, but varies from it in several aspects. For preventing duplication, we will refer to the two preceding papers for detailing and explaining some QED properties of associates in polar liquids. In the Discussion Section we focus on recently reported measured SDVSPL<sup>we-ne</sup> properties and show these conform to those predicted by the model. At the end of this section we conjecture possible mechanisms for serial diluted vigorously shaken solutions affecting biosystems. We stress we do not present any new experimental results -- our discussions pertain to previous reported experimental data. A list with abbreviations is presented at the end of this paper.

## Theory

Properties of ULC SDVSPL<sup>we-ne</sup> containing molecular associates considerably differ from those predicted by the customary theories of equilibrated polar liquids and their ULC solutions. According to the customary theories: below a solute type dependent concentration all solutes solvate; solvated solutes distribute homogeneously, move independently and randomly; polar solvent molecules (except solvation shells' solvent molecules) move randomly; for aqueous solutions, its water molecules ( $\text{H}_2\text{O}$ ) form flickering hydrogen-bond networks; electromagnetic radiation, serial dilutions or vigorous shaking do not affect ULC solutions (Horne, 1972). These customary mod-

els explicitly include electrostatic forces and *assume electrodynamic ones can be treated perturbatively*. In contrast, QED theory explicitly includes electrodynamic forces.

QED indicates that EMF interactions with electrolytic solutes, polar solute molecules or with solvent molecules with sufficiently large electric dipole moments may lead to formation of various QED domains types. Formation of the domains occurs only in specific  $C$  ranges (including ULC). These ranges depend on solute type. The domains were generally denoted as “CD” -- a shortening for “coherence domains”. CD may agglomerate into supra-domains (supra-CD). Supra-CD are not ensembles of molecules but agglomerates of domains, like domains in liquid crystals. Details of the QED theory of polar liquids and their various hitherto identified CD, their schematic pictures, their properties, the physics underlying CD formation’s dependence on concentration, citations of experimental data verifying aforementioned and the reason underlying the name “coherence domain” are presented in the first paper of this series (Yinnon and Liu, 2015a).

**Coherence domains** -- The CD types relevant to the SDVSPL<sup>we-ne</sup> model include CD<sub>plasma</sub>, IPD<sub>plasma</sub>, CD<sub>rot</sub> and CD<sub>elec</sub>. Below we concisely summarize their properties. Some of these vary from those for solutions of strong electrolytes, summarized in Yinnon and Liu (2015b).

- CD<sub>plasma</sub> -- these domains only form when the solvent molecules have a sufficiently strong electric dipole moment. CD<sub>plasma</sub> are composed of few solvated solutes and numerous solvent molecules (Del Giudice *et al.*, 2000). The plasma oscillations of these solvated solutes are coherent. Interactions between the solutes and tetra Herz to mega Herz EMF underlie the coherence. CD<sub>plasma</sub> are not micelles.<sup>b</sup> CD<sub>plasma</sub> are very stable

<sup>b</sup> A micelle is an aggregate of surfactant molecules. In aqueous solutions, its molecules’

domains. Energy gained by a solute particle on its incorporation in CD<sub>plasma</sub> amounts to a few eV. Weak electrolytes only organize in CD<sub>plasma</sub> for  $C_{trans}^{IPDplasma} < C < C_{trans}^{CDplasma}$ . [This is not the case for solutions of strong electrolytes, in which CD<sub>plasma</sub> form at all concentrations above  $C_{trans}^{IPDplasma}$ ]. Solute characteristics determine the transition concentrations  $C_{trans}^{CDplasma}$  and  $C_{trans}^{IPDplasma}$ . Typically:  $C_{trans}^{CDplasma} < 1M$  (Del Giudice *et al.*, 2000);  $C_{trans}^{IPDplasma}$  is of the order of  $10^{-4} - 10^{-6}$  M (Yinnon and Yinnon, 2012).  $C_{trans}^{IPDplasma}$  is the concentration at which the distance between identical nearest neighbor solutes equals the Debye length.<sup>c</sup> As to non-electrolytes, only those with sufficiently large permanent or by the solvent induced electric dipole moments can organize in CD<sub>plasma</sub>. Just as for weak electrolytes this only occurs for  $C_{trans}^{IPDplasma} < C < C_{trans}^{CDplasma}$ . The diameter of CD<sub>plasma</sub> is of the order of  $10^{-6}$  m. It is an inverse function of concentration, *i.e.*, when the concentration decreases, the size of CD<sub>plasma</sub> increases, the number of its solvated solutes diminishes and the number of its solute molecules enhances.

- IPD<sub>plasma</sub> -- these domains only form when the solvent molecules have a sufficiently strong electric dipole moment. As to solutions of non-electrolytic compounds, IPD<sub>plasma</sub> only form when the solvated solutes have sufficiently strong electric dipole moments. IPD<sub>plasma</sub> formation requires  $C$  is below the transition concentration  $C_{trans}^{IPDplasma}$

hydrophilic “head” regions are in contact with surrounding solvent, sequestering their hydrophobic single “tail” regions in the micelle’s centre. The critical concentration of the surfactant molecules above which micelles form and all additional surfactants added to the system go to micelles is denoted critical micelle concentration.

<sup>c</sup> The Debye length equals the distance beyond which the Coulomb electric field around a solute molecule is at any instant fully screened by all its neighboring molecules.



(Yinnon and Yinnon, 2012).  $IPD_{\text{plasma}}$  are composed of few solvated solutes and numerous solvent molecules.  $IPD_{\text{plasma}}$  are not micelles. The plasma oscillations of the solutes in  $IPD_{\text{plasma}}$  are in phase *i.e.*, an  $IPD_{\text{plasma}}$  is a special CD -- an In-Phase Domain. Also the plasma oscillations of its solvent molecules are in phase. Interactions between its molecules and tetra Herz to mega Herz EMF underlie all these in phase plasma oscillations.  $IPD_{\text{plasma}}$  are crystalline structured. The dipole moments of their solvent molecules are spherical symmetric aligned around their crystalline ordered solvated solutes.  $IPD_{\text{plasma}}$  are very stable domains, slightly more stable than  $CD_{\text{plasma}}$ . On diluting below  $C_{\text{trans}}^{IPD_{\text{plasma}}}$ ,  $CD_{\text{plasma}}$  transform into  $IPD_{\text{plasma}}$ , *i.e.*, the coherent plasma oscillations of the domains' solvated solutes become in phase. The diameter of  $IPD_{\text{plasma}}$  equals that of  $CD_{\text{plasma}}$  at  $C_{\text{trans}}^{IPD_{\text{plasma}}}$ , *i.e.*, about  $10^{-6}$  m. In contrast to the case for  $CD_{\text{plasma}}$ , the diameter of  $IPD_{\text{plasma}}$  does not significantly change with concentration. On diluting solutions below  $C_{\text{trans}}^{IPD_{\text{plasma}}}$ , the number of  $IPD_{\text{plasma}}$  diminishes.

- $CD_{\text{rot}}$  -- these domains only form when the solvent molecules have a sufficiently strong electric dipole moment. These domains are composed of ferroelectric ordered solvent molecules (Del Giudice *et al.*, 1988; Del Giudice and Vitiello, 2006). The molecules constituting  $CD_{\text{rot}}$  coherently oscillate between two rotational states.  $CD_{\text{rot}}$  formation results from the dipole moments of their molecules interacting with Far Infra Red (FIR) EMF.  $CD_{\text{rot}}$  have an electric dipole moment due to the ferroelectric ordering of their solvent molecules. In bulk water and most other polar liquids at ambient conditions  $CD_{\text{rot}}$  do not auto-organize. However, immersing objects with sizable asymmetric charge distributions (*e.g.*, macromolecules, hydrophylic membranes) may induce their formation, resulting in a permanent time dependent polarization. Solute particles are pulled into  $CD_{\text{rot}}$ . Few solute particles can locate in

$CD_{\text{rot}}$  and do not wreck their host. Many solute molecules destroy  $CD_{\text{rot}}$ . Solute and solvent types determine critical  $C$  below which  $CD_{\text{rot}}$  persist ( $C_{\text{crit}}^{CD_{\text{rot}}}$ ).  $CD_{\text{rot}}$ 's diameter is of the order of  $10^{-4}$  -  $10^{-5}$  m.

- $CD_{\text{elec}}$  -- these domains are composed of electronically excited solvent molecules. Only for water the characteristics of these domains were derived (Arani *et al.*, 1995; Bono *et al.*, 2012). The  $CD_{\text{elec}}$  present in water were denoted by  $CD_{\text{elec}}^{H_2O}$ . These domains are composed of solvent molecules only.  $CD_{\text{elec}}^{H_2O}$  cannot contain solutes. Solvated solutes or aggregates of solutes (*e.g.*,  $CD_{\text{plasma}}$  or  $IPD_{\text{plasma}}$ ) locate adjacent to  $CD_{\text{elec}}^{H_2O}$ . The  $H_2O$  constituting  $CD_{\text{elec}}^{H_2O}$  coherently oscillate between their electronic ground  $|0\rangle$  state and an excited  $|b\rangle$  state.  $CD_{\text{elec}}^{H_2O}$  formation is mediated by ultraviolet (UV) EMF. One electron of an  $H_2O$  residing in its  $|b\rangle$  state is almost free (binding energy of about 0.4 eV). Hence, a  $CD_{\text{elec}}^{H_2O}$  is a pool of  $\sim 10^6$  quasi-free electrons located at their boundary, and correspondingly an ensemble of quasi-free protons (the partners of the quasi-free electrons). At ambient conditions, in bulk water: the fraction of  $H_2O$  included in  $CD_{\text{elec}}^{H_2O}$  is about 20 percent;  $H_2O$  continually adsorb on  $CD_{\text{elec}}^{H_2O}$  while simultaneously  $H_2O$  desorb, causing a  $\sim 10^{-14}$  s timescale flickering landscape. Thus  $CD_{\text{elec}}^{H_2O}$  observation requires fast resolution probes.  $CD_{\text{rot}}$ ,  $CD_{\text{plasma}}$ ,  $IPD_{\text{plasma}}$  or other aggregates of solutes may stabilize  $CD_{\text{elec}}^{H_2O}$ , *i.e.*, reduce their flickering and ease their observation.  $CD_{\text{elec}}^{H_2O}$  and supra- $CD_{\text{elec}}^{H_2O}$  may get encapsulated in  $CD_{\text{rot}}$  and supra- $CD_{\text{rot}}$ . Such assemblies we denote [ $\text{supra-}CD_{\text{rot}} <\text{supra-}CD_{\text{elec}}^{H_2O}>$ ]. The state of  $H_2O$  belonging to both  $CD_{\text{rot}}$  and  $CD_{\text{elec}}^{H_2O}$  is a superposition of the state typifying the  $H_2O$  constituting  $CD_{\text{rot}}$  and the state typifying the  $H_2O$  constituting  $CD_{\text{elec}}^{H_2O}$ . The diameter of  $CD_{\text{elec}}^{H_2O}$  is about  $10^{-7}$  m.

To give the reader an intuitive feeling for the relative sizes of the  $CD_{\text{rot}}$ ,  $CD_{\text{plasma}}$  and  $CD_{\text{elec}}^{H_2O}$ , we note their ratios are similar, re-

spectively, to those of the sun, earth and moon.

**Superfluidic domains** --  $CD_{rot}$ ,  $IPD_{plasma}$  and  $CD_{elec}^{H_2O}$  are superfluidic domains, *i.e.*, their molecules do not collide (see Yinnon and Liu, 2015a).  $CD_{plasma}$  are not superfluidic. The superfluidity of CD has implications for the liquid's properties, *e.g.*, its electric conductivity.

**Schematic pictures of aqueous solutions according to QED** --

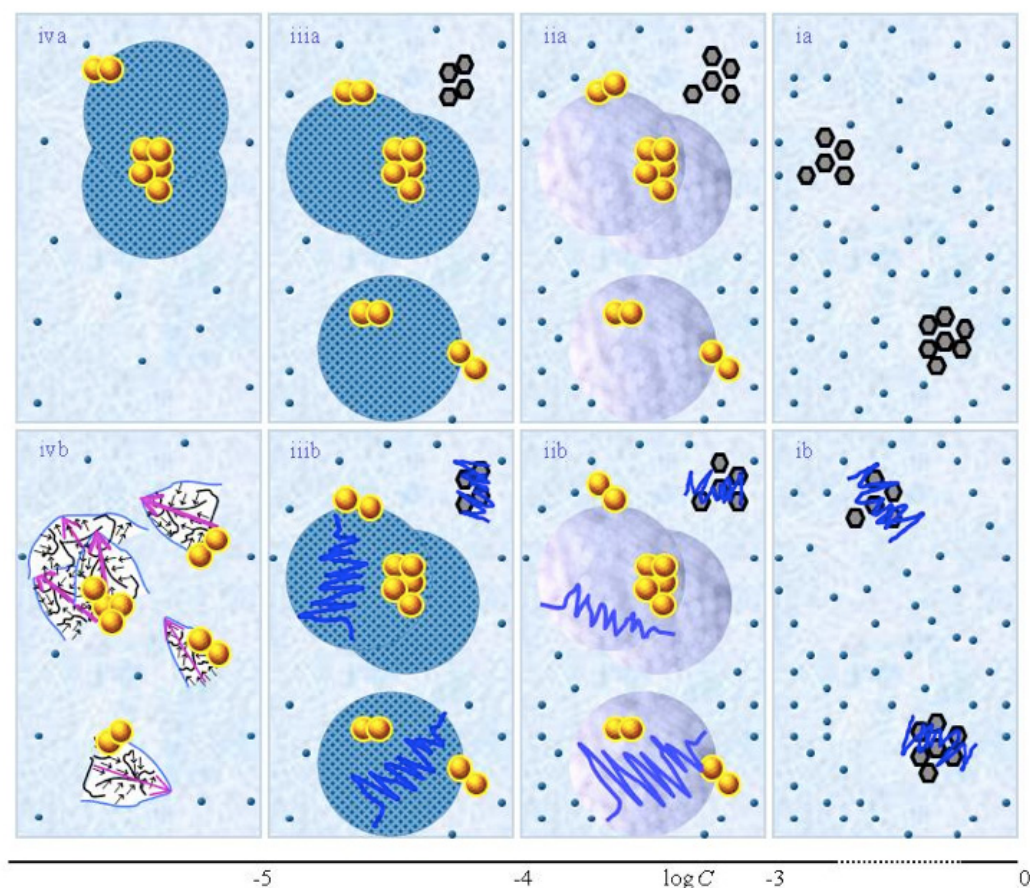
Figure 1 presents schematic pictures of serial diluted aqueous solutions of weak- or non-electrolytic compounds for  $10^{-20} M < C < 1 M$ . The (a) and (b) series pertain to solutions which, respectively, were not vigorously shaken and those which were vigorously shaken after each dilution step. Figure 1 is similar to the schematic picture of aqueous solutions of strong electrolytes presented in Figure 1 in the preceding paper (Yinnon and Liu, 2015b). The main differences are that in solutions of weak- or non-electrolytic compounds: none or only few solutes solvate for  $C$  above  $C_{trans}^{CD_{plasma}}$ ; solvated solutes only organize in  $CD_{plasma}$  for  $C_{trans}^{IPD_{plasma}} < C < C_{trans}^{CD_{plasma}}$ . Hence Figure 1ia communicates that for concentration above  $C_{trans}^{CD_{plasma}}$  the solvated solutes move randomly, *i.e.*, do not organize in  $CD_{plasma}$ , and aggregates of non-solvated solutes are present. Figure 1iia shows that for  $C_{trans}^{IPD_{plasma}} < C < C_{trans}^{CD_{plasma}}$  solvated solutes organize in  $CD_{plasma}$ , and  $CD_{elec}^{H_2O}$  and supra- $CD_{elec}^{H_2O}$  are stabilized by  $CD_{plasma}$ . Figure 1iia and Figure 1iiaa exhibit the transition of  $CD_{plasma}$  into  $IPD_{plasma}$ . On comparing Figure 1iiaa, Figure 1iva and Figure 1v, one discerns that the diameter of  $IPD_{plasma}$  does not significantly change with concentration. Instead on diluting, the number of  $IPD_{plasma}$  diminishes. On diluting also the number of non-solvated solutes diminishes. Figures 1vi-viia illustrate that in very dilute solutions,  $IPD_{plasma}$  do not form and all solvated solutes locate randomly.

**SDVSPL<sup>we-ne</sup> model**

Figures 1ib-viib present a schematic picture of the SDVSPL<sup>we-ne</sup> model proposed by Yinnon and Yinnon (2011), *i.e.*, the structure of SDVSPL<sup>we-ne</sup> for different concentration ranges. Its details we discuss in the following paragraphs. We emphasize that the model only holds when the solvent molecules have a sufficiently strong electric dipole moment.

i. For  $C$  above the transition concentration  $C_{trans}^{CD_{plasma}}$ , few or none weak- or non-electrolytic solutes solvate. Most solutes do not solvate. The few solvated solutes move randomly -- see Figure 1ib. Aggregates of unsolvated solutes may stabilize  $CD_{elec}^{H_2O}$  and may form supra- $CD_{elec}^{H_2O}$ . For  $C$  in between the transitions concentrations, *i.e.*,  $C_{trans}^{IPD_{plasma}} < C < C_{trans}^{CD_{plasma}}$ , solvated solutes organizing in  $CD_{plasma}$  enhances (drives) the solvation process. At these  $C$ , part of the solvated weak electrolytes organize in  $CD_{plasma}$  and supra- $CD_{plasma}$  (see Figure 1iib). As to solvated non-electrolytic compounds, only those having a sufficiently large electric dipole moment organize in such domains.  $CD_{plasma}$  and supra- $CD_{plasma}$  may stabilize  $CD_{elec}^{H_2O}$  and supra- $CD_{elec}^{H_2O}$ . The aforesaid holds independent of the solutions' preparation procedure, *i.e.*, not just for SDVSPL<sup>we-ne</sup> but also for solutions prepared without serial dilutions or vigorous shaking. Serial dilutions or vigorous shaking affect  $CD_{plasma}$ , mainly causing their breakup. However, after perturbations are over  $CD_{plasma}$  reform, as illustrated in Figure 1iib.

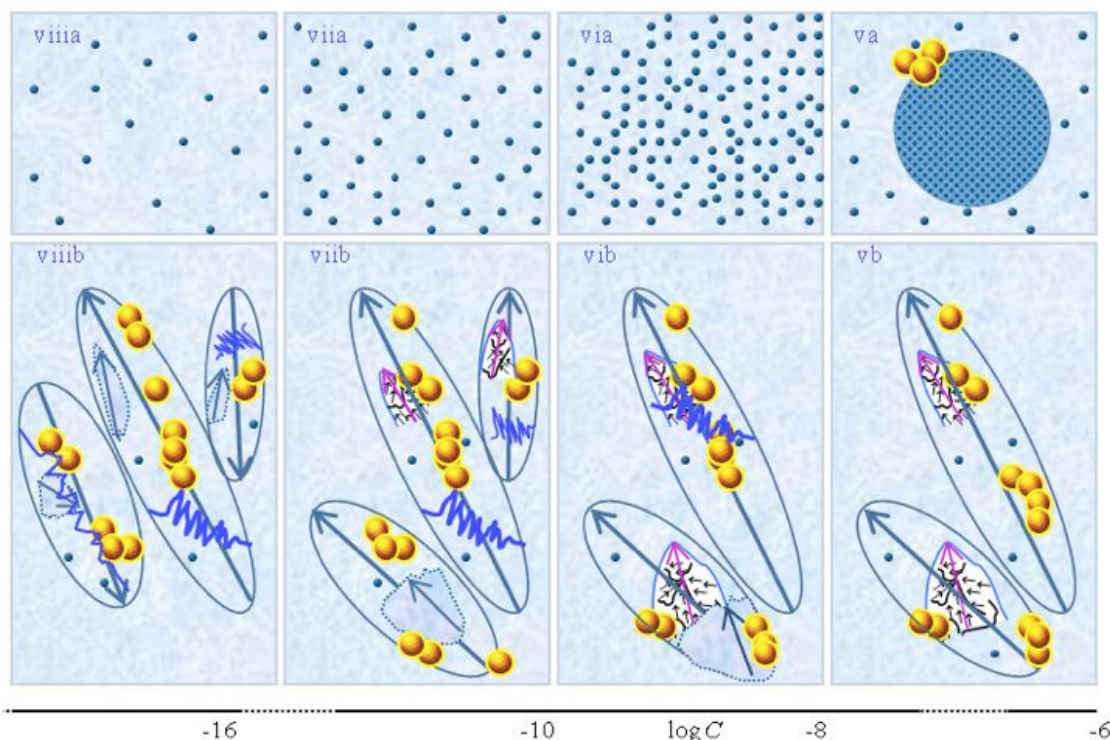
ii. At  $C \sim C_{trans}^{IPD_{plasma}}$ ,  $CD_{plasma}$  containing weak electrolytes transform into  $IPD_{plasma}$  (see Figs. 1iib-1iib). The transformation modifies electric conductivity and its dependence on  $C$  and time, because  $IPD_{plasma}$  are superfluidic and crystalline structured while  $CD_{plasma}$  are not superfluidic. As to solvated non-electrolytes, only those having a sufficiently large electric dipole moment organize in  $IPD_{plasma}$ . Differences between the transi-



**Figure 1:** This figure presents a schematic picture of serial 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  $C > C_{\text{trans}}^{\text{CDplasma}}$  all solvated solutes move randomly, i.e., do not organize in  $\text{CD}_{\text{plasma}}$ . The tiny blue balls represent randomly moving  $\sim 10^{-9} - 10^{-8}$  m solvated solutes. The irregular shaped bunches of black hexagons represent aggregates of non-solvated solutes. Figure iia illustrates that on dilution below  $C_{\text{trans}}^{\text{CDplasma}}$ , solvated solutes organize in  $\text{CD}_{\text{plasma}}$  (symbolized with purple-blue colored balls). The yellow-brown balls and their agglomerates represent, respectively,  $\sim 10^{-7}$  m  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  and supra- $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  stabilized by  $\text{CD}_{\text{plasma}}$ . Figures iia and iiaa illustrate the transformation of  $\text{CD}_{\text{plasma}}$  into  $\text{IPD}_{\text{plasma}}$  at  $C \sim C_{\text{trans}}^{\text{IPDplasma}}$ . Figures ia-iiia illustrate that on dilution the non-solvated solutes diminish, i.e., solvate. Figures iiaa-iva illustrate that on dilution the diameter of  $\text{IPD}_{\text{plasma}}$  does not change, but the number of  $\text{IPD}_{\text{plasma}}$  diminishes. Figures iiaa-iiia illustrate that below a certain concentration there are insufficient solutes to form  $\text{IPD}_{\text{plasma}}$ . The concentration below which no  $\text{IPD}_{\text{plasma}}$  form has yet not been theoretically derived. Figures iiaa-iiia illustrate that whenever there are too few solutes to form  $\text{IPD}_{\text{plasma}}$ , 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  $\text{CD}_{\text{plasma}}$ , which just as in Figure iiaa are represented with purple-blue colored balls. Figures iib and iiaa illustrate the transition from  $\text{CD}_{\text{plasma}}$  to  $\text{IPD}_{\text{plasma}}$ , with the latter pictured as blue-crystalline balls just as in Figure iiaa. Figures iiaa and iib illustrate that shaking excites or breaks up  $\text{IPD}_{\text{plasma}}$ . The excited or broken  $\text{IPD}_{\text{plasma}}$  pieces, which in the text we denoted electric dipole aggregates ( $\text{EDA}^{\text{IPDplasma}}$ ), are pictured as irregular shaped aggregates in (ivb). Their aligned black arrows orderings symbolize  $\text{EDA}^{\text{IPDplasma}}$ 's distorted ferroelectric  $\text{H}_2\text{O}$  orderings. The purple arrow in the  $\text{EDA}^{\text{IPDplasma}}$  symbolizes these domains' dipole moments. Figures ivb and iib illustrate that on diluting below a solute type dependent critical concentration ( $C_{\text{crit}}^{\text{CDrot}}$ )  $\text{CD}_{\text{rot}}$  get stabilized by  $\text{EDA}^{\text{IPDplasma}}$ , i.e., the irregular shaped  $\text{EDA}^{\text{IPDplasma}}$  are located within the elongated ovals representing  $\text{CD}_{\text{rot}}$ . The dark blue arrows symbolize the dipole moment of  $\text{CD}_{\text{rot}}$ . Figure iib shows that vigorous shaking excites or breaks up  $\text{CD}_{\text{rot}}$ , thus





**Figure 1**

creating  $EDA^{CDrot}$ . The chunk outlined with an irregular shaped broken curve and located at the bottom of one of the left  $CD_{rot}$  represents the  $EDA^{CDrot}$ . Figures vii b–viii b 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 viii b 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 viii b. Figures v b–viii b illustrate that  $CD_{rot}$  may align with their dipole moments parallel. Figure viii b 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.)

tion concentrations  $C_{trans}^{CDplasma}$  and  $C_{trans}^{IPDplasma}$  may be tiny, or solutes even might not organize in  $CD_{plasma}$ , i.e., these only organize in  $IPD_{plasma}$ . Incorporation of solvated solutes in  $IPD_{plasma}$  may be the drive behind the solvation process of solutes with low solubility product constants. Critical micelle concentrations may equal  $C_{trans}^{CDplasma}$  or  $C_{trans}^{IPDplasma}$ , but this has not yet been verified.  $IPD_{plasma}$  may aggregate in supra- $IPD_{plasma}$ . These domains may stabilize  $CD_{elec}^{H_2O}$  and supra- $CD_{elec}^{H_2O}$  (see Figure 1 ii b). Dilution below  $C_{trans}^{IPDplasma}$  diminishes the number of randomly moving solvated solutes as well as the number of solutes incorporated in  $IPD_{plasma}$ . At very low  $C$  the number of solvated solutes is too low for formation of  $IPD_{plasma}$ .

The aforesaid holds independent of the solutions' preparation procedure, i.e., not just for SDVSPL<sup>we-ne</sup>, but also for solutions prepared without serial dilutions or vigorous shaking.

iii. For solutions containing  $IPD_{plasma}$ , their vigorous shaking affects their properties. The effect is the same as that for SDVSASES containing  $IPD_{plasma}$ . That is, vigorous shaking excites or breaks up  $IPD_{plasma}$ , as pointed out by Yinnon and Yinnon (2011) and Yinnon and Liu (2015b) (see Fig.1 iv b). Excitations induce years-long lasting vortices in the superfluidic  $IPD_{plasma}$  (Yinnon and Elia, 2013). [For short discussion on these vortices see Yinnon and Liu (2015a).] The vortices partly destroy the spherical sym-



metric alignments of the dipole moments of the solvent molecules surrounding the crystalline ordered solvated solutes in  $IPD_{\text{plasma}}$ . Hence excited  $IPD_{\text{plasma}}$  and their broken pieces have electric dipoles, *i.e.*, are electric dipole aggregates, which we denote as  $EDA^{IPD_{\text{plasma}}}$  (see Figure 1ivb).  $EDA^{IPD_{\text{plasma}}}$  have the remnant crystalline structure of their “mother”  $IPD_{\text{plasma}}$  (Yinnon and Yinnon, 2011; Yinnon and Elia, 2013). (As to  $CD_{\text{plasma}}$ , only their solvation shells’ few solvent molecules are aligned, *i.e.*, their perturbation, for example vigorous shaking, does not create  $10^{-6}$  m-sized electric dipole aggregates.)

iv. For aqueous  $SDVSPL^{\text{we-ne}}$ ,  $EDA^{IPD_{\text{plasma}}}$  induce electric dipoles in the quasi free electron clouds of  $CD_{\text{elec}}^{H_2O}$ . The interactions between the dipoles of these clouds, as well as between these and the dipole moments of  $EDA^{IPD_{\text{plasma}}}$ , may stabilize  $CD_{\text{elec}}^{H_2O}$  and supra- $CD_{\text{elec}}^{H_2O}$  (see Figure 1 ivb).

v. For concentrations less than the critical concentration for  $CD_{\text{rot}}$  formation (*i.e.*,  $C < C_{\text{crit}}^{CD_{\text{rot}}}$ ), due to the interactions between the dipoles of  $EDA^{IPD_{\text{plasma}}}$  and the dipoles of the solvent molecules,  $EDA^{IPD_{\text{plasma}}}$  stabilize  $CD_{\text{rot}}$  and supra- $CD_{\text{rot}}$  (Yinnon and Yinnon, 2011; Yinnon and Elia, 2013) -- see Fig.1vb. In other words,  $EDA^{IPD_{\text{plasma}}}$ , due to their significant asymmetric charge distributions, stabilize  $CD_{\text{rot}}$ . Stabilization of  $CD_{\text{rot}}$  also may be induced by solutes with sufficient large electric dipoles. Thus even when no  $IPD_{\text{plasma}}$  and hence  $EDA^{\text{plasma}}$  form in  $SDVSPL^{\text{we-ne}}$ , some types of non-electrolytic solutes still might induce  $CD_{\text{rot}}$  stabilization for  $C < C_{\text{crit}}^{CD_{\text{rot}}}$ , and subsequent vigorous shaking might create  $EDA^{CD_{\text{rot}}}$  (and in aqueous solutions supra- $CD_{\text{rot}} < \text{supra-}CD_{\text{elec}}^{H_2O} >$ ) (Yinnon *et al.*, 2011, 2013).

Vigorous shaking excites or breaks up  $CD_{\text{rot}}$  (see Figure 1vb). Excitations induce years-long vortices in the superfluidic  $CD_{\text{rot}}$ . Due to ferroelectric ordering of the molecules constituting  $CD_{\text{rot}}$ , excited or broken  $CD_{\text{rot}}$

also are electric dipole aggregates, *i.e.*,  $EDA^{CD_{\text{rot}}}$  (see Figure 1vib). Unlike  $EDA^{IPD_{\text{plasma}}}$ ,  $EDA^{CD_{\text{rot}}}$  are not crystalline ordered. Due to interactions between the dipoles of  $EDA^{CD_{\text{rot}}}$  and the polar solvent molecules,  $EDA^{CD_{\text{rot}}}$  also stabilize  $CD_{\text{rot}}$ . Therefore, serial dilutions with vigorous shaking at each dilution step diminish  $EDA^{IPD_{\text{plasma}}}$  but  $EDA^{CD_{\text{rot}}}$  persist. These  $EDA^{CD_{\text{rot}}}$  stabilize  $CD_{\text{rot}}$  and supra- $CD_{\text{rot}}$  too. As a result  $CD_{\text{rot}}$  persist up to ULC and beyond (Yinnon and Yinnon, 2011; Yinnon and Elia, 2013) (see Figures 1vib-viiib). The aforementioned effects of vigorous shaking of  $SDVSPL^{\text{we-ne}}$  for  $C < C_{\text{crit}}^{CD_{\text{rot}}}$  are the same as those for  $SDVSASES$  described in Yinnon and Liu (2015b).

vi. In aqueous  $SDVSPL^{\text{we-ne}}$ ,  $CD_{\text{rot}}$  induce electric dipoles in the quasi free electron clouds of  $CD_{\text{elec}}^{H_2O}$ . The interactions between the dipole moments of these clouds, as well as between these and the dipole moments of  $EDA^{CD_{\text{rot}}}$ , may stabilize  $CD_{\text{elec}}^{H_2O}$  and supra- $CD_{\text{elec}}^{H_2O}$ , *i.e.*, cause formation of [supra- $CD_{\text{rot}} < \text{supra-}CD_{\text{elec}}^{H_2O} >$ ] (see Figure 1vb) (Del Giudice *et al.*, 2010). The aforementioned also holds for aqueous  $SDVSASES$  -- see Yinnon and Liu (2015b).

vii. QED domains affect physicochemical properties, requiring adjustment in customary equations. For example adjustments in equations describing electric conductivity are required because the following molecules do not collide: solvent molecules incorporated in the superfluidic domains (*i.e.*,  $CD_{\text{elec}}$ ,  $CD_{\text{rot}}$ ,  $IPD_{\text{plasma}}$ ), solvent molecules in the hydration shells of solvated solutes in  $CD_{\text{plasma}}$ ; solvated solutes incorporated in  $CD_{\text{plasma}}$  or in  $IPD_{\text{plasma}}$ . Hence in polar liquids and their solutions, the electric conductivity is an inverse function only of intermolecular collisions involving: the randomly moving solvent and solute molecules not included in the domains; solvent molecules incorporated in  $CD_{\text{plasma}}$  but not part of the solvation shells of the solutes

included in these domains. A decrease of the fractions of these colliding particles enhances the electric conductivity. Also the electric dipole moments of  $\text{EDA}^{\text{IPDplasma}}$ , of  $\text{CD}_{\text{rot}}$ , of  $\text{EDA}^{\text{CDrot}}$  and of the quasi free electron clouds of  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  reduce intermolecular collisions of randomly moving molecules neighboring on these domains. The reductions raise electric conductivity (Yinnon and Yinnon 2011; Yinnon and Elia, 2013). Currently, we are investigating quantitative implications.

Rendering our above detailed qualitative  $\text{SDVSPL}^{\text{we-ne}}$  model into quantitative one requires numerous computations, which are beyond this paper's scope.

## Discussion

### **Correspondence between $\text{SDVSPL}^{\text{we-ne}}$ properties predicted by QED and observed ones**

Recently published experimental data, which evidence the characteristics of associates present in a  $\text{SDVSPL}^{\text{we-ne}}$  as predicted by its QED model (summarized in paragraphs i-viii above), we cite and discuss in paragraphs i-viii below.

i. For many weak electrolytes and non-electrolytes in various polar solvents, below a solute type dependent transition concentration of the order of  $1 - 10^{-3}$  M, part of the solvated solutes and numerous solvent molecules were observed to organize in  $\sim 10^{-7} - 10^{-6}$  m sized groupings. These groupings are not micelles. The groupings were observed with techniques like DLS, laser light scattering, static light scattering, nanoparticle tracking analysis, scanning electron microscopy and nuclear magnetic resonance (Li and Ogawa, 2000; Samal and Geckeler, 2001; Kononov *et al.*, 2002; Sedlak, 2006; Sedlak and Rak, 2013; Hagmeyer *et al.*, 2012; Konovalov, 2013; Ryzhkina *et al.*, 2013). These groupings are present in serial diluted vigorously shaken solutions, as well as in solutions not perturbed by these

techniques. These groupings have characteristics conforming to those of  $\text{CD}_{\text{plasma}}$  and  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  (Yinnon and Yinnon, 2009). It should be stressed, however, that for specific concentration ranges, numerous solutes form other cluster types which are not  $\text{CD}_{\text{plasma}}$ .

Identifying  $\text{CD}_{\text{plasma}}$  is difficult, as holds for  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  stabilized by  $\text{CD}_{\text{plasma}}$ . ( $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  may also be stabilized by other cluster types.) EMF screening induced disappearance of groupings promises to be an effective method for exposing their QED nature. Radio frequency screening of tetra Hz to mega Hz EMF destroying  $\sim 10^{-6}$  m domains and UV EMF screening by Permalloy destroying  $\sim 10^{-7}$  m domains we expect to be optimal probes for identifying, respectively,  $\text{CD}_{\text{plasma}}$  and  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  (Yinnon and Liu, 2015b). An exemplary study partly corroborating this expectation is that of aqueous  $\text{SDVSPL}^{\text{we-ne}}$  of 2,4,6,8-tetramethyl-2,4,6,8-tetraazabicyclo [3.3.0] octane-3,7-dione (the active ingredient in the tranquilizer Mebicar). For this  $\text{SDVSPL}^{\text{we-ne}}$ , Ryzhkina *et al.* (2013) measured its groupings' diameter under normal conditions ( $\mathcal{D}_{\text{lb}}$ ), i.e., for samples kept at the laboratory bench. Moreover, they measured the groupings' diameter ( $\mathcal{D}_{\text{p}}$ ) for samples kept in Permalloy containers. In samples kept at the laboratory bench, at  $\sim 10^{-2} \text{ M} < C < \sim 10^{-1} \text{ M}$ , clusters with  $\mathcal{D}_{\text{lb}} \approx 1.5 \times 10^{-9}$  m (hydrated solute molecules) and domains with  $\mathcal{D}_{\text{lb}} \approx 7 \times 10^{-6}$  m are present; on diluting to  $\sim 10^{-3}$  M, the  $\mathcal{D}_{\text{lb}} \approx 1.5 \times 10^{-9}$  m clusters disappear; at  $\sim 10^{-5} \text{ M} < C < \sim 10^{-3}$  M,  $\mathcal{D}_{\text{lb}} \approx 8 \times 10^{-8}$  m and  $\mathcal{D}_{\text{lb}} \approx 2 \times 10^{-7}$  m domains are dominant. Screening samples by Permalloy does not significantly affect the domains present at  $\sim 10^{-2} \text{ M} < C < \sim 10^{-1} \text{ M}$ , evoking the  $\mathcal{D}_{\text{p}} = \mathcal{D}_{\text{lb}} \approx 7 \times 10^{-6}$  m domains are  $\text{CD}_{\text{plasma}}$ . (Recall that the diameter of  $\text{CD}_{\text{plasma}}$  is of the order of  $10^{-6}$  m.) Radio frequency screening of samples is called for to verify this evocation. Screening by Permalloy reduces prevalence and dominance of the  $\sim 8 \times 10^{-8}$  m and  $\sim 2 \times 10^{-7}$  m domains present

at  $\sim 10^{-5} \text{ M} < C < \sim 10^{-3} \text{ M}$ ; due to their diminished dominance, the  $D_p \approx 7 \times 10^{-6} \text{ m}$  domains again become observable. Available electric conductivity data is insufficient for exposing a  $C_{\text{trans}}^{\text{IPD}_{\text{plasma}}}$ , i.e., currently it is impossible to designate the  $D_{\text{lb}} = D_p \approx 7 \times 10^{-6} \text{ m}$  domains present at  $10^{-2} \text{ M} < C < \sim 10^{-1} \text{ M}$  and at  $\sim 10^{-5} \text{ M} < C < \sim 10^{-3} \text{ M}$ , respectively, as  $\text{CD}_{\text{plasma}}$  or  $\text{IPD}_{\text{plasma}}$ . The fact that screening by Permalloy does destroy part but not all  $\sim 8 \times 10^{-8} \text{ m}$  and  $\sim 2 \times 10^{-7} \text{ m}$  domains implies some of these might be  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$ . (Recall that the diameters of  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  and supra- $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  are of the order of  $10^{-7} \text{ M}$ .) Those  $\sim 8 \times 10^{-8} \text{ m}$  and  $\sim 2 \times 10^{-7} \text{ m}$  domains which were not destroyed by the Permalloy screening might be  $\text{EDA}^{\text{plasma}}$  -- as perhaps radio frequency screening can reveal.  $\zeta$ -potential data provides additional evidence for presence of  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$ . At  $\sim 10^{-5} \text{ M} < C < \sim 10^{-3} \text{ M}$ , the  $\zeta$ -potential varies between  $-11.5 \text{ mV}$  and  $-13 \text{ mV}$ , which is compatible with the  $\zeta$ -potential of  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$ , as discussed below in paragraph vi.

For  $C < \sim 10^{-6} \text{ M}$ ,  $10^{-7} \text{ m}$  and  $10^{-6} \text{ m}$  domains are present, but these disappear on screening by Permalloy, i.e., these are not  $\text{CD}_{\text{plasma}}$  or  $\text{IPD}_{\text{plasma}}$ . A conclusion commensurate with the QED model, which predicts that at such low  $C$  the number of solutes is too small for forming a significant number of these domains. The domains present at  $C < \sim 10^{-6} \text{ M}$  we discuss below in paragraph v.d.

ii. The domains observed in numerous  $\sim 10^{-6} \text{ M} < C < \sim 10^{-4} \text{ M}$  weak- and non-electrolytes solutions, with properties differing from those of the domains present for  $C > \sim 10^{-4} \text{ M}$ , might be  $\text{IPD}_{\text{plasma}}$ . Albeit, these could be other cluster types, e.g., micelles. Radio frequency screening destroying the domains will be required for identifying their  $\text{IPD}_{\text{plasma}}$  nature. While we await such screening outcomes, for some exemplary solutions we present physicochemical data attributable to  $\text{IPD}_{\text{plasma}}$  presence and  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  stabilized by these domains:

(a) For aqueous  $\text{SDVSPL}^{\text{we-ne}}$  of Melafen [the melamine salt of bis(hydroxymethyl) phosphinic acid dihydrate] in water (with 1%  $\text{D}_2\text{O}$ ) Konovalov *et al.* (2008) found: their dilution from  $\sim 1 \times 10^{-3} \text{ M}$  to  $\sim 1 \times 10^{-4} \text{ M}$  reduces the UV absorption coefficient from 31900 to 30000, but on further dilution it increases to 34700 and 43500, respectively, for  $\sim 1 \times 10^{-5} \text{ M}$  and  $\sim 1 \times 10^{-6} \text{ M}$ ; other UV spectral features indicate at  $\sim 2 \times 10^{-3} \text{ M}$  the solution has a salt character; Infra Red (IR) spectra indicate for  $\sim 8 \times 10^{-2} \text{ M} < C < \sim 1.6 \times 10^{-1} \text{ M}$  Melafen dissociates into anions and cations with their hydration shell surrounded by a mobile  $\text{H}_2\text{O}$  network; with the ionic nature of aqueous Melafen for  $C > \sim 10^{-3} \text{ M}$ , the reduction in UV absorption observed on diluting from  $\sim 1 \times 10^{-3} \text{ M}$  to  $\sim 1 \times 10^{-4} \text{ M}$  corresponds to that typical for salt solutions, while the increase in UV absorption for  $C < \sim 10^{-4} \text{ M}$  is attributable to association of ions. The aforementioned concurs with the ionic associates present at  $C < \sim 10^{-4} \text{ M}$  being  $\text{IPD}_{\text{plasma}}$ , which stabilize  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  interacting with UV EMF. Also for  $\text{SDVSPL}^{\text{we-ne}}$  of hemin derivatives at  $\sim 1 \times 10^{-6} \text{ M} < C < \sim 1 \times 10^{-4} \text{ M}$  Ryzhkina *et al.* (2011c) measured UV absorbance. They concluded its enhancements with dilution are ascribable to various types of aggregates. As to their nature, e.g., are these  $\text{IPD}_{\text{plasma}}$  which stabilize  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$ , the current available data is insufficient for reaching at conclusions and EMF screening is called for.

(b) For aqueous  $\text{SDVSPL}^{\text{we-ne}}$  of the anti-oxidant Ichfan C-10 (a 2,6-dialkylphenol derivative), Ryzhkina *et al.* (2009a, 2011d) showed:

1. For  $C$  above the critical micelle concentration ( $\sim 1 \times 10^{-3} \text{ M}$ ), surface tension of samples kept at the laboratory bench ( $\sigma_{\text{lb}}$ ) or in Permalloy containers ( $\sigma_{\text{p}}$ ) indicate it is a functional cationic surfactant. Electrophoresis evidence their micelles'  $\zeta$ -potential varies from 40 to 15 mV.
2. On diluting from  $\sim 1 \times 10^{-3} \text{ M}$  to  $\sim 5 \times 10^{-4} \text{ M}$



<sup>7</sup> M,  $D_{lb}$  increases from  $\sim 1.0 \times 10^{-7}$  m to  $\sim 1.6 \times 10^{-7}$  m, the  $\zeta$ -potential of associates in samples kept at the laboratory bench ( $\zeta_{lb}$ ) drops to -12 mV, and  $\sigma_{lb}$  strongly increases approaching that of double distilled water thus indicating the domains are not micelles or pre-micelle aggregates.

3. For samples kept on the laboratory bench, domain size distributions are unimodal for  $\sim 10^{-6}$  M  $< C < \sim 10^{-3}$  M. However, for samples kept for 18 hours in Permalloy containers the distributions are not unimodal for  $\sim 10^{-9}$  M  $< C < \sim 10^{-5}$  M. Clusters with  $\sim 10^{-9}$  m  $< D_p < \sim 10^{-8}$  m and domains with  $\sim 10^{-7}$  m  $< D_p < \sim 10^{-6}$  m persist.

4. At  $\sim 10^{-7}$  M  $< C < \sim 10^{-5}$  M, the electric conductivity of samples kept at the laboratory bench ( $\chi_{lb}$ ) are slightly larger than the electric conductivity of samples kept in Permalloy containers ( $\chi_p$ ). For this concentration range also  $\sigma_{lb}$  is slightly larger than  $\sigma_p$ .

Paragraphs 2-4 mentioned changes in physicochemical variables observed on diluting from  $\sim 10^{-3}$  M to  $\sim 10^{-7}$  M indeed are attributable to micelles transforming to IPD<sub>plasma</sub> which in turn stabilize  $CD_{elec}^{H_2O}$ , *e.g.*:  $D_p \approx 10^{-6}$  m conforms to the  $\sim 10^{-6}$  m diameter of IPD<sub>plasma</sub>;  $D_{lb} \approx 10^{-7}$  m conforms to the  $\sim 10^{-7}$  m diameter of  $CD_{elec}^{H_2O}$ ;  $\zeta_{lb} = -12$  mV is ascribable to the quasi free electrons of  $CD_{elec}^{H_2O}$ ;  $\chi_{lb}$  versus  $\chi_p$  and  $\sigma_{lb}$  versus  $\sigma_p$  discrepancies are attributable to the  $\zeta$ -potential and the dipole moment of the distorted clouds of quasi free electrons of  $CD_{elec}^{H_2O}$ . The  $\chi_{lb}$  versus  $\chi_p$  discrepancies are also attributable to the superfluidity of  $CD_{elec}^{H_2O}$ . Studying the effects of radio frequency screening is required for evidencing the IPD<sub>plasma</sub> nature of the domains. Such screening is also desirable for other solutions, *e.g.*, aqueous SDVSPL<sup>we-ne</sup> of potassium phenosan,  $\alpha$ -tocopherol or membranotropic amphiphilic calix[4]resorcinarene with tris(hydroxymethyl)methylamide functional groups, because Ryzhkina *et al.* (2012a,c) observed these solutions

have physicochemical properties similar to aforementioned ones for Ichfan C-10.

iii. Creation of EDA<sup>IPDplasma</sup> by vigorous shaking of solutions requires IPD<sub>plasma</sub> presence, which as discussed above, still has to be unambiguous evidenced for SDVSPL<sup>we-ne</sup>. Radio frequency screening and Fourier transform analyses of the molar conductivity dependence on time for assessing EDA<sup>IPDplasma</sup> presence are called for. The benefits of such Fourier transform analyses is discussed in Yinnon and Liu (2015b).

iv. As to  $CD_{elec}^{H_2O}$  stabilized by EDA<sup>plasma</sup>, their presence in aqueous SDVSPL<sup>we-ne</sup> can be revealed by  $D_{lb}$ ,  $D_p$ ,  $\chi_{lb}$  and  $\chi_p$  data. However, clusters other than EDA<sup>IPDplasma</sup>, *e.g.*, micelles or pre-micelle aggregates, might also underlie  $CD_{elec}^{H_2O}$  stabilization. While we wait for verification of presence of EDA<sup>IPDplasma</sup> by the techniques mentioned in paragraph iii, in the meanwhile for some exemplary SDVSPL<sup>we-ne</sup> we present physicochemical data consistent with  $CD_{elec}^{H_2O}$  presence. For aqueous SDVSPL<sup>we-ne</sup> of Ichfan C-10, potassium phenosan, and  $\alpha$ -tocopherol, Ryzhkina *et al.* (2011d, 2012a, 2012c) showed EMF affect these SDVSPL<sup>we-ne</sup> differently at  $\sim 10^{-20}$  M  $< C < C_{thr}$  versus at  $C_{thr} < C < \sim 10^{-3}$  M with  $\sim 10^{-10}$  M  $< C_{thr} < \sim 10^{-6}$  M:

(a) For  $C_{thr} < C < \sim 10^{-3}$  M,  $D_{lb}$  distributions are unimodal with  $\sim 10^{-7}$  m  $< D_{lb} < \sim 3.5 \times 10^{-7}$  m. However,  $D_p$  distributions are not unimodal: clusters with  $10^{-9}$  m  $< D_p < 10^{-8}$  m and domains with  $5 \times 10^{-8}$  m  $< D_p < 4 \times 10^{-6}$  m are distinguishable. For  $C < C_{thr}$ , no domains are present in samples kept for 18 hours in Permalloy containers and the liquid loses its typical SDVSPL<sup>we-ne</sup> physicochemical properties. The aforementioned unimodality and values of  $D_{lb}$  are commensurate with supra- $CD_{elec}^{H_2O}$  dominating the domain distribution and mainly determining  $D_{lb}$ . The  $5 \times 10^{-8}$  m  $< D_p < 4 \times 10^{-6}$  m domains might be EDA<sup>IPDplasma</sup> and the  $10^{-9}$  m  $< D_p < 10^{-8}$  m clusters might be hydrated ions, which become distinguishable

with DLS when the more abundant  $CD_{elec}^{H_2O}$  are eliminated by the Permalloy container screening UV EMF.

(b) For  $C_{thr} < C < \sim 10^{-5}$  M  $\chi_P$  is about 10% lower than  $\chi_{lb}$ . The difference is solute type dependent. It is attributable to  $CD_{elec}^{H_2O}$  and supra- $CD_{elec}^{H_2O}$  stabilized by  $EDA^{IPDplasma}$  or other cluster types in samples kept at the laboratory bench. Recall that  $CD_{elec}^{H_2O}$  are superfluidic. Therefore on their disappearance the number of intermolecular collisions increases and  $\chi$  decreases.

v. For  $C < C_{crit}$ , fingerprints of ferroelectric orderings participating in dissipative dynamics were identified in  $\chi$ , heat of mixing and pH data of SDVSPL<sup>we-ne</sup> (Elia and Niccoli, 1999, 2004a). These fingerprints were attributed to  $CD_{rot}$  and  $EDA^{CDrot}$  by Yinnon and Yinnon (2011) and Yinnon and Elia (2013). In serial diluted solutions which were not vigorously shaken after each dilution step, no such fingerprints were observed. Also IR spectra pass band coefficient fluctuations (IR-SPBCF) of several aqueous SDVSPL<sup>we-ne</sup>, observed by Zubareva *et al.* (2003), are attributable to the dissipative dynamics of  $CD_{rot}$  and  $EDA^{CDrot}$ . These IR-SPBCF resemble those of SDVSASES. The latter were shown to conform with  $CD_{rot}$  and  $EDA^{CDrot}$  agglomeration and reorganization in supra-domains (Yinnon and Liu, 2015b).

*Dielcometric titrations<sup>d</sup> provided the first*

<sup>d</sup> Dielcometric titrations provide the dependence of the dielectric permittivity ( $\epsilon$ ) on  $C$ , *i.e.*  $\epsilon(C)$ . As to  $\epsilon(C)$ , its deviation from linearity is a measure of intermolecular interactions of the mixture's components, with slope ratios characterizing its polarization affinity. Usually, the slope ratio of  $\epsilon(C)$  changes upon formation of associates. A sharp positively sloped  $\epsilon(C)$  leveling off in a plateau is characteristic of linear ordering of subunits and formation of "chain associates" in which the dipole moments are more or less parallel oriented. On increasing

*unambiguous evidence for ferroelectric ordering in non-aqueous and aqueous SDVSPL<sup>we-ne</sup> and showed the type of ordering depends on concentration (Konovalov et al., 2008):*

(a) For SDVSPL<sup>we-ne</sup> of Melafen with the low-polarity chloroform as solvent, the dielectric permittivity ( $\epsilon$ ) dependence on  $C$ , *i.e.*,  $\epsilon(C)$  has two inflection points in the  $\sim 10^{-10}$  M  $< C < \sim 10^{-4}$  M range: for  $\sim 10^{-10}$  M  $< C < \sim 3.3 \times 10^{-8}$  M  $\epsilon(C)$  is linear with a steep slope; for  $\sim 3.3 \times 10^{-8}$  M  $< C < \sim 6.6 \times 10^{-6}$  M  $\epsilon(C)$  is linear too but the slope is about half that one for  $\sim 10^{-10}$  M  $< C < \sim 3.3 \times 10^{-8}$  M; at  $\sim 6.6 \times 10^{-6}$  M  $\epsilon(C)$  reaches a plateau which continues up to  $C \approx 1 \times 10^{-4}$  M (Konovalov *et al.*, 2008). Based on the well known  $\epsilon(C)$  characteristics: the steeply sloped  $\epsilon(C)$  is attributable to chain associates of  $CD_{rot}$  and  $EDA^{CDrot}$ ; the moderately sloped  $\epsilon(C)$  is attributable to these domains being organized in multipole aggregates; the plateau at  $\sim 6.6 \times 10^{-6}$  M  $< C < \sim 1 \times 10^{-4}$  M is attributable to  $EDA^{IPDplasma}$  organized with their dipoles "inverse-parallel" orientated or to other non-polar groupings. The aforementioned hints that  $C_{crit}^{CDrot} \approx 10^{-6}$  M. To verify this paragraph's attributions, EMF screening of SDVSPL<sup>we-ne</sup> of Melafen is called for, because association of the molecules constituting  $CD_{rot}$  is mediated by FIR EMF.

(b) Our model's assumption that domain formation in SDVSPL<sup>we-ne</sup> at low and ul-

$C$  above that of the plateau,  $\epsilon(C)$  may become negatively sloped and reach a new plateau due to additional association leading to multipole aggregates with associated particles' dipoles "inverse-parallel" orientated leading to formation of non-polar aggregates. The non-polar aggregates with the "inverse parallel orientation" can also form at the first step of aggregation, omitting the "chain aggregate" phase. The transition point at which the slope of  $\epsilon(C)$  changes corresponds to the critical aggregate concentration *e.g.*, critical micelle concentration, if the solute is a surfactant.

tralow concentrations involves the dipoles of the polar solvent molecules is supported by  $\varepsilon(C)$  of SDVSPL<sup>we-ne</sup> of  $\alpha$ -tocopherol in the non-polar carbon tetrachloride versus that in chloroform (Ryzhkina *et al.*, 2011b). For the former at  $\sim 10^{-24}$  M  $< C < \sim 10^{-3}$  M,  $\varepsilon(C)$  is linear and only slightly changes, indicating absence of association. For the latter at  $\sim 10^{-20}$  M  $< C < \sim 10^{-3}$  M,  $\varepsilon(C)$  is non-linear and has kinks at  $C \approx 1 \times 10^{-20}$  M,  $C \approx 1 \times 10^{-15}$  M and  $C \approx 1 \times 10^{-10}$  M. The kinks are related to formation and rearrangement of domains. For  $\sim 10^{-20}$  M  $< C < \sim 10^{-15}$  M, growth of  $\varepsilon$  with  $C$  is steepest, but lessens when  $C$  increases. For  $\sim 10^{-4}$  M  $< C < \sim 10^{-3}$  M,  $\varepsilon(C)$  is flat. The aforementioned  $\varepsilon(C)$  features indicate domains only form in SDVSPL<sup>we-ne</sup> with polar solvent molecules.

(c) For aqueous SDVSPL<sup>we-ne</sup> of membranotropic amphiphilic calix[4] resorcinarene with tris(hydroxymethyl)methylamide functional groups (a compound with structure and properties rendering it a simple synthetic model of natural polypeptide cluster glycoconjugates inducing cascades of physiological reactions when binding to cell membranes), Ryzhkina *et al.* (2012c) also employed dielectric to evidence ferroelectric ordering. They found for the concentration dependence of  $\Delta\varepsilon$  [i.e.  $\Delta\varepsilon(C)$  with  $\Delta\varepsilon$  the difference between  $\varepsilon$  of this SDVSPL<sup>we-ne</sup> and  $\varepsilon$  of double distilled water]: on diluting from  $\sim 10^{-4}$  M to  $\sim 10^{-5}$  M  $\Delta\varepsilon$  decreases from 7.5 to 0, on diluting from  $\sim 10^{-5}$  M to  $\sim 5 \times 10^{-7}$  M  $\Delta\varepsilon$  stays 0, on diluting from  $\sim 5 \times 10^{-7}$  M to  $\sim 5 \times 10^{-8}$  M  $\Delta\varepsilon$  increases from 0 to  $\sim 1$ , on diluting from  $\sim 5 \times 10^{-8}$  M to  $\sim 3 \times 10^{-9}$  M  $\Delta\varepsilon$  increases to  $\sim 1.5$ , on diluting from  $\sim 3 \times 10^{-9}$  M to  $\sim 10^{-10}$  M  $\Delta\varepsilon$  decreases to  $\sim 0.75$ , on diluting from  $\sim 10^{-10}$  M to  $\sim 5 \times 10^{-11}$  M  $\Delta\varepsilon$  increases to  $\sim 1$  and on diluting from  $\sim 5 \times 10^{-11}$  M to  $\sim 10^{-12}$  M  $\Delta\varepsilon$  decreases to  $\sim 0.5$  (see Ryzhkina *et al.* 2012c Figure 2). In addition they demonstrated the orderings induce optical activity. By combining their  $\Delta\varepsilon$ , optical activity and  $D_{lb}$  data, they evidenced different

types of groupings containing ferroelectric-ordered molecules cause this  $\Delta\varepsilon(C)$ : for  $\sim 10^{-6}$  M  $< C < \sim 10^{-3}$  M the  $D_{lb}$  distribution is bimodal with  $D_{lb} \approx 4 \times 10^{-9}$  m clusters and  $\sim 1.2 \times 10^{-7}$  m  $< D_{lb} < 1.8 \times 10^{-7}$  m domains (on dilution from  $\sim 1 \times 10^{-3}$  M to  $\sim 1 \times 10^{-5}$  M the fraction of the  $D_{lb} \approx 4 \times 10^{-9}$  m clusters decreases from 0.3 to 0.1 while that of the  $D_{lb} \approx 10^{-7}$  m domains increases to 0.9); for  $\sim 10^{-12}$  M  $< C < \sim 10^{-7}$  M the  $D_{lb}$  distribution is bimodal with  $D_{lb}$  of the order of  $10^{-7}$  m and  $10^{-6}$  m and their fractions respectively 0.7 and 0.3; for  $C < \sim 10^{-12}$  M the domain distribution is polymodal preventing assessment of  $D_{lb}$ . Their  $\sigma_{lb}$  data evidence the solute is not a surfactant at  $\sim 10^{-12}$  M  $< C < \sim 10^{-3}$  M. They supplemented their  $D_{lb}$  data with atomic force microscopy (AFM) imaging. AFM in contact mode for  $\sim 10^{-6}$  M  $< C < \sim 10^{-3}$  M revealed  $1 \times 10^{-7}$  m -  $6 \times 10^{-7}$  m wide up to  $3.5 \times 10^{-8}$  m high hemispherical discrete relief particles, with the largest particles composed of smaller ones and all particles randomly distributed on the substrate -- their prevalence strongly diminishes with concentration. At  $C \approx 10^{-7}$  M the discrete particles are still present, but AFM in tapping mode revealed another type of particles appears:  $\sim 8 \times 10^{-8}$  m wide  $1.3 \times 10^{-9}$  m -  $1.7 \times 10^{-9}$  m high chain oriented soft particles aggregated in a thin film covering the substrate (similar soft particles were observed for aqueous NaCl SDVSASES at  $C \approx 1.7 \times 10^{-7}$  M (Lo *et al.*, 2009). For  $\sim 10^{-11}$  M  $< C < \sim 10^{-8}$  M, AFM in tapping mode exposed  $\sim 10^{-6}$  m sized soft particles forming different complex structures, e.g., ribbon or rounded configurations (similar ribbons were observed with Transmission Electron Microscopy of aqueous NaCl SDVSASES at  $C \approx 1.7 \times 10^{-11}$  M (Lo, 1996). The aforementioned  $D_{lb}$  and AFM analyses of Ryzhkina *et al.* (2012c) expounded the groupings present at  $\sim 10^{-6}$  M  $< C < \sim 10^{-3}$  M are associates composed of solute and solvent molecules, while the domains present for  $C < \sim 10^{-7}$  M are mainly composed of solvent



molecules. Placing samples for 18 hours in Permalloy containers also enabled them to unambiguously demonstrate the different nature of the groupings present at  $C$  below and above  $10^{-7}$  M: for  $C < \sim 10^{-7}$  M no groupings are distinguishable, *i.e.*,  $C_{\text{thr}} \approx 10^{-7}$  M; at  $\sim 10^{-6}$  M  $< C < \sim 10^{-3}$  M samples' physico-chemical properties are slightly altered. While the alterations are attributable to  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  stabilized by other groupings types, radio frequency screening and examining its effect on  $\Delta\epsilon(C)$  are called for to investigate whether these groupings include  $\text{IPD}_{\text{plasma}}$  and  $\text{EDA}^{\text{IPDplasma}}$ . *With the  $10^{-7}$  m and  $10^{-6}$  m groupings present at  $C < \sim 10^{-7}$  M mainly being composed of  $\text{H}_2\text{O}$ , the disappearance of these on screening by Permalloy and the ferroelectric ordering of their molecules exposed by above cited  $\Delta\epsilon(C)$  data, the properties of these groupings match those of  $\text{CD}_{\text{rot}}$ ,  $\text{EDA}^{\text{CDrot}}$ ,  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$  and  $[\text{supra-CD}_{\text{rot}} < \text{supra-CD}_{\text{elec}}^{\text{H}_2\text{O}} >]$ , implying  $C_{\text{crit}}^{\text{CDrot}} \approx C_{\text{thr}} \approx 10^{-7}$  M.* As to the AFM data and the detailed  $\Delta\epsilon$  values, according to theoretical aspects of  $\Delta\epsilon(C)$  summarized in footnote d: for  $\sim 3 \times 10^{-8}$  M  $< C < \sim 5 \times 10^{-7}$  M these are ascribable to chain structures of  $\text{CD}_{\text{rot}}$  and  $\text{EDA}^{\text{CDrot}}$ , with these domains' electric dipole moments parallel oriented; for  $\sim 1 \times 10^{-10}$  M  $< C < \sim 3 \times 10^{-8}$  M the kinks in  $\Delta\epsilon(C)$  signifies changes in the arrangement of these electric dipole moments, *e.g.*, these are attributable to polar chain as well as less polar multi-pole structures; the maximum of  $\Delta\epsilon(C)$  at  $C \approx 10^{-11}$  M also is ascribable to polar chain structures dominating the groupings' distribution. This attribution is supported by the non-zero optical activity observed for this  $C$  range.

(d) For  $\sim 10^{-18}$  M  $\leq C \leq C_{\text{thr}} \approx 10^{-6}$  M aqueous  $\text{SDVSPL}^{\text{we-ne}}$  of 2,4,6,8-tetramethyl-2,4,6,8-tetraazabicyclo[3.3.0] octane-3,7-dione (Mebicar), Ryzhkina *et al.* (2013) also demonstrated ferroelectric ordering. It disappears on screening by Permalloy. With DLS they observed  $D_{\text{lb}} \approx 10^{-7}$  m and  $D_{\text{lb}} \approx 10^{-6}$  m domains, which disappear on

screening by Permalloy. With dielcometry they evidenced  $\Delta\epsilon$  of samples kept at the laboratory bench ( $\Delta\epsilon_{\text{lb}}$ ) strongly depends on concentration. It varies between about -1.5 and +1, indicating ferroelectric ordering of the domains depends on concentration. Negative  $\Delta\epsilon_{\text{lb}}$  values signifying domains form low-polar multipole structures, thus decreasing  $\epsilon$  of the solution below that of double distilled water. The maximal positive  $\Delta\epsilon_{\text{lb}}$  at  $\sim 1 \times 10^{-10}$  M is attributable to domains forming chain (ribbon) structures wherein the dipole moments are arranged uni-directionally.  $\Delta\epsilon(C)$  for samples screened by Permalloy, *i.e.*,  $\Delta\epsilon_{\text{p}}(C)$ , equals 0. With  $D_{\text{lb}} \approx 10^{-6}$  m domains present at  $C < \sim 10^{-6}$  M, these mainly being composed of  $\text{H}_2\text{O}$ , their disappearance on screening by Permalloy and the ferroelectric ordering of their molecules exposed by dielcometry, the properties of these domains match those of  $\text{CD}_{\text{rot}}$  and  $\text{EDA}^{\text{CDrot}}$ , implying  $C_{\text{crit}}^{\text{CDrot}} \approx C_{\text{thr}} \approx 10^{-6}$  M. Characteristics of the  $\sim 10^{-7}$  m domains, which we discuss below in paragraph vi, indicate these are  $\text{CD}_{\text{elec}}^{\text{H}_2\text{O}}$ .

(e)  $\sigma_{\text{lb}}$  dependence on  $C$  [*i.e.*  $\sigma_{\text{lb}}(C)$ ] supplements the  $\Delta\epsilon_{\text{lb}}(C)$  reflecting the inverse-parallel oriented polar domains versus polar chain domains orderings for  $10^{-18}$  M  $< C < C_{\text{crit}}^{\text{CDrot}} \approx C_{\text{thr}}$  aqueous  $\text{SDVSPL}^{\text{we-ne}}$ : Ryzhkina *et al.* (2009, 2011c-d) for various aqueous  $\text{SDVSPL}^{\text{we-ne}}$  observed in this  $C$  range (which is below these solutions' critical micelle concentration), that  $\sigma_{\text{lb}}$  equals the surface tension of double distilled water, with the exception of two broad minima, one near  $C \approx C_{\text{crit}}^{\text{CDrot}}$  and the other near  $C \approx 10^{-13}$  M for aqueous  $\text{SDVSPL}^{\text{we-ne}}$  of Ichfan C-10 or near  $C \approx 10^{-16}$  M for a Hemin derivative.

This section's cited experimental data indicate: domains are present for  $C < C_{\text{crit}}$  for  $\text{SDVSPL}^{\text{we-ne}}$  of certain solute and solvent types only when during their preparation after each dilution step the liquid is vigorously shaken; for  $C < C_{\text{crit}}$  domains only

| BAC                           | Electric dipole moment (Debye) | Theory predicts domain formation | Reference to electric dipole moment of bioactive compound                          |
|-------------------------------|--------------------------------|----------------------------------|--|
| Amino benzole acid            | 3.3                            | ?                                | Kondrashina <i>et al.</i> (1994)   |
| Sodium chloride               | 9.0                            | V                                | Nelson <i>et al.</i> (1967)  |
| Acetosalicic acid             | 1.9                            | -                                | Cabezas, <i>et al.</i> (2012)  |
| Glycine                       | 14.0                           | V                                | Raabe (2004).  |
| Adenine                       | 3.1 (in water)                 | ?                                | Li <i>et al.</i> (1999)  |
| $\beta$ -Sitosterol           | 2.5                            | -                                | Kurban <i>et al.</i> (2010)  |
| Thyrotropin-releasing hormone | 5.7                            | V                                | Protirelin - bioinformatics. charite.<br>de/main/content/databases: cas=024305279. |
| B-Cyclodextrin                | ~3                             | -                                | Botsi <i>et al.</i> (1996)   |
| Cholesterol                   | 2.0                            | -                                | McClellan (1963)   |
| Isoniazid                     | 2.0                            | -                                | Rajalakshmi <i>et al.</i> (2011)   |
| Ibuprofen                     | ~4.8                           | V                                | Huq (2006)   |
| Lysine                        | 4.0                            | V                                | Kumazaki and Shintaro (1972)   |
| Vancomycin                    | 16.                            | V                                | Felder an Sussman (2014)   |
| Uracil                        | 6.0                            | V                                | Li <i>et al.</i> (1999)  |

**Table 1:** SDVSPL<sup>we-ne</sup> of bioactive compounds and predictions concerning these liquids containing domains at ULC. The symbol “V” indicates that our model predicts presence of domains in ULC SDVSPL<sup>we-ne</sup>. The symbol “?” indicates that according to our model it is doubtful that domains are present in ULC SDVSPL<sup>we-ne</sup>. The symbol “-” indicates that our model predicts no domains are present in ULC SDVSPL<sup>we-ne</sup>.

are present in polar solvents; the numerous solvent molecules constituting these domains are ferroelectric ordered; these domains aggregate into chain or multipole structures; these domains disappear on screening by Permalloy. These indications evoke the domains include  $CD_{rot}$  and  $EDA^{CD_{rot}}$  and  $C_{crit} = C_{crit}^{CD_{rot}}$ . (Our discussion as to these domains also including  $CD_{elec}^{H_2O}$  at  $C < C_{crit}$ , we present in paragraph vi.) Aforementioned evocation replaces the enigma “Why does SDVSPL<sup>we-ne</sup> of some solutes at ULC contain domains while others have the customary infinite diluted solutions characteristics?” to the query “Which entity stabilize  $CD_{rot}$  at  $C_{crit}^{CD_{rot}}$ ?”. With the current fuzziness concerning  $IPD_{plasma}$  and  $EDA^{IPD_{plasma}}$  presence in SDVSPL<sup>we-ne</sup>, we cannot assess whether solvated solutes with a sufficiently large asymmetric charge distribution or only  $EDA^{IPD_{plasma}}$  stabilize  $CD_{rot}$  at  $C_{crit}^{CD_{rot}}$ . Since  $IPD_{plasma}$  and  $EDA^{IPD_{plasma}}$  formations both require solvated solutes with a sufficiently large asymmetric charge distribution, we contend that only SDVSPL<sup>we-ne</sup> of such solutes contain  $CD_{rot}$  at ULC. As a first step towards verifying our contention, in Table 1 we present a variety of bioactive

compounds and their measured or calculated electric dipole moment reported in the literature. We randomly choose these compounds from those listed by Konovalov (2013). Based on our SDVSPL<sup>we-ne</sup> model, we predict that only in ULC SDVSPL<sup>we-ne</sup> of bioactive compounds which have a dipole moment larger than the dipole moment of the solvent molecules,  $CD_{rot}$  and  $EDA^{CD_{rot}}$  are present for  $C < C_{crit}^{CD_{rot}} = C_{thr}$ . Since for  $H_2O$  in its gas phase, the electric dipole moment is 1.85 D and in liquid phase its values has been estimated as  $2.9 \pm 0.6D$  (Kemp *et al.*, 2008), we predict that only when the dipole moment of the bioactive compound is above  $\sim 3D$ , their ULC aqueous SDVSPL<sup>we-ne</sup> contain  $CD_{rot}$  and  $EDA^{CD_{rot}}$ . Table 1 presents our predictions. Measurements confirming these predictions are called for.

vi.  $CD_{elec}^{H_2O}$  stabilized by  $CD_{rot}$  cannot be evidenced by examining differences between physicochemical properties of samples kept at the laboratory bench or in Permalloy containers. Such differences we analyzed in paragraphs i, ii and iv for uncovering  $CD_{elec}^{H_2O}$  stabilization by  $CD_{plasma}$ ,  $IPD_{plasma}$  or  $EDA^{plasma}$ . However, screening by Permalloy destroys both  $CD_{elec}^{H_2O}$  and  $CD_{rot}$ . With UV

absorbance sensitive to  $CD_{elec}^{H_2O}$  (Del Giudice *et al.*, 2010; Yinnon *et al.*, 2015c), gathering UV absorbance data for various SDVSPL<sup>we-ne</sup> with  $C < C_{thr} \approx C_{crit}^{CDrot}$  is called for. While awaiting such measurements, we focus on  $CD_{elec}^{H_2O}$  fingerprints in  $\zeta$ -potential data, because many physicochemical properties of dispersed systems are determined by their dispersed particles' surface charge. Ryzhkina *et al.* (2010a, 2011d, 2012a-d, 2013) observed:  $\zeta_{lb}$  of the  $D_{lb} \approx 1 \times 10^{-7}$  m to  $D_{lb} \approx 4 \times 10^{-7}$  m sized domains, present in aqueous SDVSPL<sup>we-ne</sup> at  $\sim 10^{-20}$  M  $< C < C_{thr}$  (which we denote  $\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$ ) typically varies from -2 to -20 mV, non-linear changes with concentration and is related to their  $D_{lb}$  (which we denote  $D_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$ );  $\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$  also is related to the  $\Delta \epsilon_{lb}$  and pH of samples kept at the laboratory bench (pH<sub>lb</sub>);  $\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$  correlates with  $X_{lb}$  at solute type dependent concentration ranges. (In the aforementioned and the rest of this paragraph, we only cited data for  $\sim 10^{-20}$  M  $< C < C_{thr}$  samples kept at the laboratory bench, because at these concentrations no domains are present in samples screened by Permalloy.) With the diameter of  $CD_{elec}^{H_2O}$  and the size of supra- $CD_{elec}^{H_2O}$  of the order of  $10^{-7}$  m (similar to  $D_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$ ), we conjecture: the negative charged domains are  $CD_{elec}^{H_2O}$  and supra- $CD_{elec}^{H_2O}$ ;  $\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$  is due to the quasi free electrons located at their boundary; pH<sub>lb</sub> (typically varying from  $\sim 5.8$  to 7) result from the quasi free protons (which are the partners of the quasi free electrons) and are dispersed in the solvent surrounding  $CD_{rot}$ . As to the reasoning underlying this conjecture, for an  $H_2O$  ensemble with finite dimensions, ferroelectric ordering implies addition of an electrostatic term to its potential energy [the 2nd term in Eq. 4 of Iitaka (2010)], representing the effect of the macroscopic electric field due to the charge distribution at the boundary. This term's contribution to the potential energy is about hundred times larger than the energetic differences of various hydrogen bond configurations. Alike charged parti-

cles, dispersed within the ensemble, screen the boundary polarization and reduce this term. Hence accumulation of alike charged particles in  $CD_{rot}$  and expulsion of opposite charged ones reduce polarization at the  $CD_{rot}$  boundary and stabilize these domains. Apparently, the energy gained from  $CD_{rot}$  stabilization is higher than that required for separating the quasi free electrons of  $CD_{elec}^{H_2O}$  from their quasi free protons. Thus on  $CD_{rot}$  stabilization: some  $H_2O$  incorporated in  $CD_{elec}^{H_2O}$  dissociate (*i.e.*, their dissociation constant shifts); those quasi free protons becoming hydroxonium ions ( $H_3O^+$ ) disperse in between the  $CD_{rot}$  causing the solvent's slightly acid character; their counterions, the negatively charged  $CD_{elec}^{H_2O}$ , locate within the  $CD_{rot}$  and screen their boundary polarization. With calculation of the energetic changes involved in the mechanisms outside the scope of this paper, we turn to other data supporting our conjecture.

For aqueous SDVSPL<sup>we-ne</sup> of 2,4,6,8-tetramethyl-2,4,6,8-tetraazabicyclo [3.3.0] octane-3,7-dione at  $\sim 10^{-17}$  M  $< C < C_{thr}$  (Mebicar), Ryzhkina *et al.* (2013) found: the molarities at which the dependence of pH<sub>lb</sub> and  $D_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$  on concentration have maximum coincide; for  $\sim 10^{-16}$  M  $< C < \sim 10^{-10}$  M, pH<sub>lb</sub> and  $D_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$  statistically significantly correlate. The findings indicate domains reorganization involves  $H_3O^+$ . Indeed according to QED,  $CD_{elec}^{H_2O}$  organizing in supra- $CD_{elec}^{H_2O}$  affects the energy levels of their quasi free electrons and quasi free protons (Preparata, 1995 chapters 3 and 10), altering the dissociation energy of their  $H_2O$ . To the best of our knowledge, the aforementioned correlation is its first identified manifestation. However, such correlation was not observed for all SDVSPL<sup>we-ne</sup> for which pH<sub>lb</sub> data were collected (Ryzhkina *et al.*, 2012c). With the still rudimentary understanding of the effects of  $CD_{elec}^{H_2O}$  agglomeration on the quasi free electrons and the quasi free protons, detailed calculations and comparing their results with pH<sub>lb</sub> and  $D_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$  data



analyses are called for.

With  $\zeta_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}$  and  $\mathcal{D}_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}$  for many SDVSPL<sup>we-ne</sup> at  $C < C_{thr}$  related but not statistically significantly correlated, the effects of  $CD_{elec}^{H_2O}$  organizing in supra- $CD_{elec}^{H_2O}$  seems to be influenced by quasi free electrons as well as by other factors, *e.g.*, the polarization density in their neighborhood. Indeed, the extremums in the absolute values of  $\zeta_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}$  (*i.e.*  $|\zeta_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}|$ ) coincide with those of  $\Delta\epsilon_{lb}$  for aqueous SDVSPL<sup>we-ne</sup> of Mebicar at  $C < C_{thr}$ : the local minimum of  $\Delta\epsilon_{lb} \approx -0.05$  at  $C \approx 10^{-7} \text{ M}$  coincides with a local minimum of  $|\zeta_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}|$  -- according to dielectric theory, for concentration ranges at which  $\Delta\epsilon < 0$ , polar domains likely are lined up in low polar multipole structures, thus decreasing  $\epsilon$  of the solution compared to that of the solvent; the global maximum of  $\Delta\epsilon_{lb} \approx 1$  at  $C \approx 10^{-10} \text{ M}$  coincides with a global maximum of  $|\zeta_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}| \approx 12 \text{ mV}$ , which is attributable to polar domains organized in chain (ribbon) structures with the dipole moments arranged unidirectionally; the global minimum of  $\Delta\epsilon_{lb} \approx -1.5$  at  $C \approx 10^{-14} \text{ M}$  coincides with a local minimum in  $|\zeta_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}| \approx 4 \text{ mV}$  -- the global nature of this minimum of  $\Delta\epsilon_{lb}$  indicates inverse-parallel orientation of the polar domains at this concentration range. As to the agglomeration of  $CD_{elec}^{H_2O}$  into supra- $CD_{elec}^{H_2O}$  at these extremums, we cite the local minimum of  $\mathcal{D}_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}} \approx 2.5 \times 10^{-7} \text{ m}$  at  $C \approx 10^{-7} \text{ M}$ , the local minimum of  $\mathcal{D}_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}} \approx 2.7 \times 10^{-7} \text{ m}$  at  $C \approx 10^{-10} \text{ M}$  and the global maximum of  $\mathcal{D}_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}} \approx 4.2 \times 10^{-7} \text{ m}$  at  $C \approx 10^{-14} \text{ M}$ . The aforementioned  $\Delta\epsilon_{lb}$  and  $\mathcal{D}_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}$  values suggest that for inverse-parallel oriented  $CD_{rot}$ , supra- $CD_{elec}^{H_2O}$  sizes are maximal. However, the opposite was observed for aqueous SDVSPL<sup>we-ne</sup> of membranotropic amphiphilic calix[4]resorcinarene: maxima in its  $\mathcal{D}_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}$  coincide with the polar domains mainly organizing in chain structures (Ryzhkina *et al.*, 2012c). Hitherto, details of  $\Delta\epsilon(C)$  only were gathered for the SDVSPL<sup>we-ne</sup> cited in this paragraph and theoretical aspects of  $CD_{rot}$

orderings,  $CD_{elec}^{H_2O}$  agglomerations, their dependence on concentration and solute type has not been investigated; therefore research directed at studying these concentration dependencies is called for.

vii. As to the observed effects of the various QED domains on the electric conductivity ( $\chi$ ) of SDVSPL<sup>we-ne</sup>:

(a) For  $C > C_{thr}$ , we cannot yet explain the dependence of  $\chi$  on  $C$ , *i.e.*,  $\chi(C)$ . As noted above existing data is insufficient for assigning domains to  $CD_{plasma}$ ,  $IPD_{plasma}$  or  $EDA^{IPD_{plasma}}$ , micelles or other aggregate types. Hence attributing  $\chi$  values to any of these is not yet doable.

(b) For  $\sim 10^{-18} \text{ M} < C < C_{thr}$ , Ryzhkina *et al.* (2011c, 2011d, 2012a, 2012c, 2012d, 2013) found:  $\chi(C)$  for samples kept on the laboratory bench [ $\chi_{lb}(C)$ ] is much larger than  $\chi(C)$  of samples kept in Permalloy containers [ $\chi_P(C)$ ]. For example for aqueous potassium phenosan SDVSPL<sup>we-ne</sup> at  $C \approx 10^{-14} \text{ M}$   $\chi_{lb} \approx 25 \text{ } \mu\text{S/cm}$  while  $\chi_P$  is an order of magnitude smaller.  $\chi_P$  very slowly decreases with concentration and typically is  $\sim 2.5 \text{ } \mu\text{S/cm}$  (slightly larger than  $\chi$  of double distilled water). Moreover,  $|\zeta_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}|$  and  $\chi_{lb}$  are significant statistical correlated -- an enhancement in  $|\zeta_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}|$  causes larger  $\chi_{lb}$ . Also  $\chi_{lb}(C)$  is related to  $\mathcal{D}_{lb}^{1 \times 10^{-7}-4 \times 10^{-7} \text{ m}}(C)$ ,  $\Delta\epsilon_{lb}(C)$  and  $\sigma_{lb}(C)$ . As to explanations for these phenomena:

1.  $\chi_{lb}(C) > \chi_P(C)$ , together with paragraphs v-vi, evoke the inequality results from Permalloy destroying  $CD_{rot}$  and  $CD_{elec}^{H_2O}$ .  $\chi$  is an inverse function of intermolecular collisions. For  $C < C_{thr}$  intermolecular collisions solely involve randomly moving  $H_2O$  and solvated solutes. Recall that the  $H_2O$  in the superfluidic  $CD_{rot}$  or  $CD_{elec}^{H_2O}$  do not collide. Thus the transformation of these domains' coherent oscillating  $H_2O$  into randomly moving  $H_2O$  (induced by the EMF screening) enhances the number of intermolecular collisions and thus reduc-

es  $\chi$ . Also the electric dipoles of  $CD_{rot}$  and the clouds of quasi free electrons of  $CD_{elec}^{H_2O}$  affect collisions of randomly moving  $H_2O$  neighboring on  $CD_{rot}$  (Yinnon and Yinnon, 2011, 2012; Yinnon and Elia, 2013).

2.  $\chi_{lb}(C)$  being correlated to  $|\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}|(C)$  and related to  $\mathcal{D}_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}(C)$ ,  $\Delta \varepsilon_{lb}(C)$  and  $\sigma_{lb}(C)$ , together with the strong concentration dependence of these variables, indicate domains rearrangements are reflected in  $\chi$ . However, the current fuzziness concerning the effects of  $CD_{rot}$  orderings on  $\mathcal{D}_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}(C)$  and  $|\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}|(C)$  (see paragraph vi), hinders explaining these relations.

3. Paragraphs (1-2) imply modeling  $\chi(C)$  for  $C < C_{thr}$  SDVSPL<sup>we-ne</sup> requires adequate description of alternating electric field interactions with: randomly moving solvent and solvated solute molecules; as well as with the dipoles of  $CD_{rot}$ ,  $EDA^{CD_{rot}}$  and the quasi free electron clouds of  $CD_{elec}^{H_2O}$ .

Experimental data cited in the above paragraphs i-vii and their analyses support most of the QED model of SDVSPL<sup>we-ne</sup>'s aspects i-vii, respectively.

### **Interactions between bio-systems and SDVSPL<sup>we-ne</sup> or SDVSASES**

$\sim 10^{-20}$  M  $< C < \sim 10^{-10}$  M laboratory-bench-kept serial diluted vigorously shaken polar liquids of a wide variety (thousands) of bioactive compounds, in a reproducible quantified manner, affect bio-systems, *e.g.*, bio-macromolecules, cells, organs, organisms or populations (Palmina *et al.*, 2009; Konovalov *et al.*, 2014a-c). The bioactivity of these solutions persists even when their concentrations are much lower than those of the bio-systems. Rhyzkina *et al.* (2010a, 2011b, 2013), and Konovalov *et al.* (2014a-c) observed: the bioactivity of these solutions disappears on screening by Permalloy; the concentration dependence of the bioactivity is related to the solutions' electric conductivity dependence on concentration  $\chi_{lb}(C)$ ; it also is related to the absolute value

of the  $\zeta$ -potential dependence on concentration  $|\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}|(C)$  of the  $\mathcal{D}_{lb} \approx 1 \times 10^{-7}$  m to  $\mathcal{D}_{lb} \approx 4 \times 10^{-7}$  m sized domains present in these solutions. Thus evidently these domains play a role in the bioactivity. Contemplating the effects of SDVSPL<sup>we-ne</sup> and SDVSASES on biosystems is hampered by the many unsolved puzzles pertaining to biochemical reactions. However,  $|\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}|(C)$  provides a clue.

Most biochemical reactions are redox electron transfer reactions, involving bio-molecules with high electronic excitation energies. Electron transfer occurs over distances beyond van der Waals contact, *i.e.*, direct donor-to-acceptor electronic interactions are negligible and customarily water is assumed to act as a go-between (Balabin *et al.*, 2008). Since water molecules have high ionization energy (12.6 eV), as a matter of course these are not regarded as a source of nearly free electrons. Their usual conjectured roles include electron tunneling facilitation, solvation of counter-ions, dielectric screening, proton coupling.

Supra- $CD_{rot} < \text{supra-}CD_{elec}^{H_2O}$  stabilized at biological interfaces playing a central role in electron transfer was conjectured by Del Giudice *et al.* (2010, 2013). Water in bio-systems can be considered interfacial water -- it is but a fraction of a micron from an interface (cell membranes, macromolecules, etc). The quasi free electrons of the supra- $CD_{elec}^{H_2O}$  in interfacial water constitute a pool of nearly free electrons. The superfluidity of  $CD_{elec}^{H_2O}$  entails its quasi free electrons reside in a collective state with a narrowly (peV) spaced spectrum. The pool's excited states persist for macroscopic times, *i.e.*, a supra- $CD_{elec}^{H_2O}$  is an efficient energy storage device. A  $CD_{elec}^{H_2O}$  attracts molecules containing in their spectrum a frequency close to one of its own, *e.g.*, a bioactive molecule. This bio-particle becomes a guest participant in the coherent dynamics of the  $H_2O$  constituting  $CD_{elec}^{H_2O}$ . It settles on the boundary of the

$CD_{elec}^{H_2O}$ . The difference in frequencies of the guest and the  $CD_{elec}^{H_2O}$  perturbs the domain's dynamics. Few guests do not harm their  $CD_{elec}^{H_2O}$ , but numerous guests wreck their host. Whenever the energy stored in supra- $CD_{elec}^{H_2O}$  matches the guest's activation energy, energy discharge of the supra- $CD_{elec}^{H_2O}$  and onset of the chemical array occur simultaneously. With the  $\sim 10^{-7}$  m diameter of  $CD_{elec}^{H_2O}$ , the  $H_2O$  and quasi free electrons in supra- $CD_{elec}^{H_2O}$  all residing in collective states, QED possibly may elucidate biosystems' fast electron transfer over long distances.

Experimental data supporting the last paragraph's conjecture of Del Giudice *et al.* (2010, 2013) has not yet been presented. Our analyses of  $SDVSPL^{we-ne}$  strengthen it. The relation between the concentration dependency of  $SDVSPL^{we-ne}$ 's bioactivity and  $|\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}|(C)$ , together with paragraph's vi's attribution that quasi free electrons underlie  $\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}$ , evokes  $SDVSPL^{we-ne}$ 's  $CD_{elec}^{H_2O}$  with their pool of superfluidic quasi free electrons and their guest bioactive molecules influence bio-systems. For example, the  $CD_{elec}^{H_2O}$  in  $SDVSPL^{we-ne}$  together with their bioactive guests molecules might directly affect membranes or interact with their interfacial water's  $CD_{elec}^{H_2O}$ . Indeed enhancement of bio-systems' biological activity or plasmatic membranes' lipid order parameter, induced by  $SDVSPL^{we-ne}$ , was observed to depend on these liquids' concentration in a manner similar to that of their  $\chi_{lb}(C)$  and  $|\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}|(C)$  (Palmina *et al.*, 2009; Konovlov, 2013; Konovalov *et al.*, 2014a-c). Moreover as mentioned above, the concentration at which numerous  $SDVSPL^{we-ne}$ 's bioactivity,  $|\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} m}|$ ,  $\chi_{lb}$  and IR-SPBCF have extremums fall in small ranges ( $\sim 10^{-9}$  M -  $10^{-10}$  M;  $\sim 10^{-13}$  M -  $10^{-15}$  M). Guest bioactive molecules' oscillations resonating with the coherent oscillations of  $H_2O$  in  $CD_{elec}^{H_2O}$ ,  $CD_{rot}$  and [supra- $CD_{rot}$  < supra- $CD_{elec}^{H_2O}$  >], and these bioactive molecules' prevalence being low enough not to wreck

their host, might underlie the extremums. As a first step for researching aforementioned conjecture, we study characteristics of  $CD_{elec}^{H_2O}$  and [supra- $CD_{rot}$  < supra- $CD_{elec}^{H_2O}$  >] adjacent to membranes (Yinnon *et al.*, 2015c WATER Journal in press; Liu *et al.*, 2015). In the current absence of detailed knowledge on interactions between biomolecules and  $CD_{elec}^{H_2O}$  or  $CD_{rot}$  their study too is called for.

Another conjecture for the bioactivity of  $SDVSPL^{we-ne}$  and  $SDVSASES$ , forwarded by Pershin (2014), bases on the quantum differences between ortho- and para- $H_2O$  spin isomers. Pershin's approach seemingly complements ours, *i.e.*, magnetic moments of vortices in  $CD_{rot}$  and quasi free electrons interacting with the  $H_2O$  spin isomers likely affect these solutions' properties. Hitherto, interactions between spin isomers and QED domains have not been studied and such studies are called for. While such interactions likely affect domains in  $SDVSPL^{we-ne}$  and in  $SDVSASES$ , at least these cannot play a decisive role in  $CD_{rot}$  formation, because these also form in non-aqueous serial diluted vigorously shaken solutions.

## Conclusions

This paper shows that the in 2011 presented QED model for serial diluted solutions which were vigorously shaken after each dilution step enables consistent explanations also for recently observed characteristics. For example for the characteristics of molecular associates in serial diluted vigorously shaken polar liquids containing weak electrolytes or non-electrolytic compounds ( $SDVSPL^{we-ne}$ ) with concentrations down to  $10^{-20}$  M. In the past the model explicated various physicochemical properties of  $SDVSPL^{we-ne}$ . The closely related QED model for serial diluted vigorously shaken aqueous solutions of strong electrolytes ( $SDVSASES$ ) also has provided consistent explanations for many phenomena (Yinnon and Yinnon, 2011; Yinnon and Elia, 2013;



Yinnon and Liu, 2015b). Our above presented analyses of experimental results, in particular those of Konovalov and Ryzhkina (2014a), for the first time enabled verification of the following central features of the SDVSPL<sup>we-ne</sup> model:

a) Interactions between EMF and molecules mediate several types of association, resulting in presence of various types of domains in specific concentration regimes. Screening samples from EMF by placing them in Permalloy containers verified this feature.

b) The typical characteristics of three domain types, *i.e.*, CD<sub>rot</sub>, CD<sub>plasma</sub> and CD<sub>elec</sub><sup>H<sub>2</sub>O</sup>, predicted by the model agree with observed ones. For example:

- Their sizes being of the order of 10<sup>-5</sup> m, 10<sup>-6</sup> m and 10<sup>-7</sup> m, respectively, and their organization in supra-domains was demonstrated with DLS and AFM.
- The macroscopic times (1 - 18 h) required for their stabilization, which was demonstrated with kinetic measurements.
- The ferroelectric ordering of the solvent molecules constituting CD<sub>rot</sub>, endowing these domains with a permanent electric dipole moment, are reflected in dielcometry and surface tension data. These domains were observed to agglomerate into supra-domains. Chain associates in which the dipole moments are more or less parallel oriented may compose these supra-CD<sub>rot</sub>. Multipole aggregates with the dipoles of CD<sub>rot</sub> “inverse-parallel” orientated, leading to formation of non-polar aggregates, were also observed.
- The dissipative dynamics of CD<sub>rot</sub> and CD<sub>elec</sub><sup>H<sub>2</sub>O</sup> are reflected in the IR spectra pass band coefficient fluctuations of SDVSPL<sup>we-ne</sup>.
- The critical concentration ( $C_{crit}^{CD_{rot}}$ ) below which 10<sup>-5</sup> m sized CD<sub>rot</sub> can persist over macroscopic times in SDVSPL<sup>we-ne</sup> was identified with a combination of techniques, *e.g.*, dielcometry, DLS, screening samples

from ambient EMF by placing them in Permalloy containers.

- CD<sub>elec</sub><sup>H<sub>2</sub>O</sup> being negatively charged has been substantiated by microelectrophoretic measurements of their electrokinetic potential ( $\zeta$ -potential).

- Stabilization of CD<sub>elec</sub><sup>H<sub>2</sub>O</sup> or CD<sub>rot</sub> has been demonstrated with electric conductivity data, which reflect their superfluidic properties.

For one domain type, *i.e.*, the ~10<sup>-6</sup> m sized IPD<sub>plasma</sub> predicted to be present at 10<sup>-6</sup> M < C < 10<sup>-4</sup> M in solution of weak electrolytes and non-electrolytes, we pointed out the kind of experiments required for unambiguous distinguishing these among the various types of observed domains. [Formation of IPD<sub>plasma</sub> in aqueous strong electrolytes has been verified, as discussed by Yinnon and Yinnon (2012), Yinnon and Liu (2015b)].

c) The QED model's prediction that only in solutions with polar solvents, their preparation by serial dilutions and vigorous shaking after each dilutions step induces molecular association at very low concentrations is supported by the experimental findings of Ryzhkina *et al.* (2011b). In SDVSPL<sup>we-ne</sup> of  $\alpha$ -Tocopherol dissolved in chloroform or dissolved in water they observed domains at 10<sup>-20</sup> M < C < 10<sup>-3</sup> M. However, for SDVSPL<sup>we-ne</sup> of  $\alpha$ -Tocopherol dissolved in the non-polar tetrachloride, they could not discern molecular association at such low concentrations.

d) Only in solutions of certain solute types, serial dilutions and vigorous shaking the liquid after each dilution step perpetuate presence of domains at very low concentrations, *e.g.*, at 10<sup>-20</sup> M < C < 10<sup>-10</sup> M. The model predicts that the required characteristic of the solute is that it has a sufficiently large permanent or induced electric dipole moment. Experiments indeed verified that only for certain solutes, their serial diluted vigorous shaken solutions contain molecular associates at very low concentrations

(Konovalov, 2013). However, these solutes and their specific characteristics have not yet been listed in the literature.

All these verifications support an important result attained with the QED model of polar liquids. The model uncovered the dynamics causing serial diluted solutions which were vigorously shaken after each dilution step at ultra low concentrations to have characteristics differing from those predicted by the customary electrostatic theories. The features of the QED model of SDVSPL<sup>ne-se</sup> substantiated in this paper and in previous ones, together with the corroborations of the QED model of SDVSASES published in previous articles, lead to the conclusion: “stabilization of  $CD_{rot}$  at  $C \approx C_{crit}^{CDrot}$  is crucial for inducing the distinctive characteristics of SDVSPL<sup>ne-se</sup> and SDVSASES at ultra low concentrations.” According to QED,  $CD_{rot}$  can get stabilized by entities with a sufficiently large asymmetric charge distribution, *e.g.*, solvated solutes or molecular associates with a large permanent or induced electric dipole moment. Once such an entity stabilizes  $CD_{rot}$ , subsequent vigorous shaking excites these domains or breaks them up. Due to the ferroelectric ordering of the solvent molecules constituting  $CD_{rot}$ , excited  $CD_{rot}$  as well as their broken pieces also have an electric dipole moment, *i.e.*, are electric dipole aggregates ( $EDA^{CDrot}$ ). Successive dilution, diminishes  $CD_{rot}$  and  $EDA^{CDrot}$ . However, these domains are molecular associates with an electric dipole moment, and as such stabilize new  $CD_{rot}$ . Thus serial dilutions with vigorous shaking after each dilution step perpetuate presence of  $CD_{rot}$  after these first got stabilized at  $C \approx C_{crit}^{CDrot}$ . In aqueous solutions  $CD_{rot}$  also may stabilize  $CD_{elec}^{H_2O}$  and supra- $CD_{elec}^{H_2O}$ .

As to the characteristics of entities stabilizing  $CD_{rot}$  in serial diluted solutions, hitherto only for SDVSASES is there some experimental evidence. Below a solute type dependent transition concentration ( $C_{trans}^{IPDplasma}$ ), ions and

the polar  $H_2O$  organize in  $IPD_{plasma}$ . Typically these  $IPD_{plasma}$  are present at  $\sim 10^{-6} M < C < \sim 10^{-4} M$ . These domains have no asymmetric charge distribution. However, excitation of  $IPD_{plasma}$  or their break up induced by vigorous shaking transforms  $IPD_{plasma}$  into molecular associates with an asymmetric charge distribution, *i.e.*, these are electric dipole aggregates ( $EDA^{IPDplasma}$ ). Accordingly in SDVSASES,  $EDA^{IPDplasma}$  stabilize  $CD_{rot}$  at  $C \approx C_{crit}^{CDrot}$ . Such stabilization requires that the electrolyte solution is first diluted below  $C_{trans}^{IPDplasma}$  and subsequently vigorously shaken after each additional dilution step. For SDVSPL<sup>we-ne</sup> no sufficient experimental data exist for determining presence of  $IPD_{plasma}$  in dilute solutions, and their transformation to  $EDA^{IPDplasma}$  induced by vigorous shaking. Moreover, for SDVSPL<sup>we-ne</sup>, experimental data unambiguously verifying solutes with sufficiently large electric dipole moments stabilize  $CD_{rot}$  at  $C \approx C_{crit}^{CDrot}$  is also lacking. The kinds of experiments required for revealing the presence of  $IPD_{plasma}$  are detailed in the discussion section.

The abovementioned implies that important qualitative aspects of the structure and physicochemical properties of SDVSPL<sup>ne-se</sup> and SDVSASES have now been explained. As to future research, in addition to that detailed in the Discussion section, we point to the following desirable projects:

- Quantitative verification of the structural and physicochemical properties of SDVSPL<sup>ne-se</sup> and SDVSASES predicted by QED.
- Research directed at elucidating the effects of SDVSPL<sup>ne-se</sup> and SDVSASES on bio-systems;
- Deriving equations for physicochemical properties of liquids containing the various kinds of QED domains; computing the values of the physicochemical variables and showing these agree with measured ones.

**Table 2:** List of abbreviations in alphabetic order, followed by Greek symbols abbreviations.

| Abbreviations  | Explanation   |
|--|---|
| AFM  | Atomic force microscopy   |
| $C$  | Concentration   |
| $C_{crit}$   | Critical concentration  |
| $C_{crit}^{CDrot}$   | 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   |
| CD   | Coherence domain  |
| $CD^{H_2O}_{elec}$   | Coherence domain composed of coherent electronically excited water molecules  |
| $CD_{plasma}$  | Coherence domain composed of few solvated solutes performing coherent plasma oscillation and numerous polar solvent molecules                         |
| $CD_{rot}$   | Coherence domains of ferroelectric ordered polar solvent molecules  |
| $\mathcal{D}$  | Effective hydrodynamic diameter of molecular associate  |
| $\mathcal{D}_{lb}$   | Effective hydrodynamic diameter of molecular associates of samples kept on the laboratory bench   |
| $\mathcal{D}_p$  | Effective hydrodynamic diameter of molecular associates of samples kept in Permalloy containers   |
| $\mathcal{D}_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} \text{ m}}$ | Effective hydrodynamic diameter of $1 \times 10^{-7} - 4 \times 10^{-7} \text{ m}$ sized molecular associates of samples kept on the laboratory bench |
| DLS  | Dynamic light scattering  |
| $EDA^{CDrot}$  | 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)                  |
| EMF  | Electro-magnetic fields   |
| eV   | Electron Volt   |
| FIR  | Far Infra Red   |
| $IPD_{plasma}$   | In phase domains composed of few solvated solutes and numerous solvent molecules performing in phase plasma oscillation.                              |
| IR   | Infra red   |
| IR-SPBCF   | Infra red spectra pass band coefficient fluctuations  |
| $pH_{lb}$  | pH of samples kept at the laboratory bench  |
| QED  | Quantum electro-dynamics  |
| SDVSASES   | Serial diluted vigorous shaken aqueous strong electrolyte solution  |
| SDVSPL <sup>we-ne</sup>  | Serial diluted vigorous shaken polar liquids of weak electrolytes or non-electrolytes   |
| Supra-CD   | Agglomerate of coherence domains  |
| $[Supra-CD_{rot} < supra-CD_{elec}^{H_2O} >]$                      | Agglomerate of $CD_{rot}$ containing agglomerates of $CD_{elec}^{H_2O}$   |
| ULC  | Ultra low concentration   |
| UV   | Ultra-violet  |
| $\epsilon$   | Dielectric permittivity   |
| $\epsilon(C)$  | Dielectric permittivity as function of concentration  |
| $\Delta\epsilon$   | Difference between $\epsilon$ of SDVSPL <sup>we-ne</sup> and $\epsilon$ of double distilled water   |
| $\Delta\epsilon(C)$  | $\Delta\epsilon$ as function of $C$   |
| $\Delta\epsilon_{lb}$  | $\Delta\epsilon$ of samples kept at the laboratory bench  |
| $\Delta\epsilon_p(C)$  | $\Delta\epsilon$ for samples screened by Permalloy as function of concentration   |
| $\zeta_{lb}$   | Electrokinetic potential of molecular associates in samples kept at the laboratory bench  |
| $\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} \text{ m}}$       | Electrokinetic potential of $1 \times 10^{-7} - 4 \times 10^{-7} \text{ m}$ sized molecular associates in samples kept at the laboratory bench        |
| $ \zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} \text{ m}} $     | Absolute value of $\zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} \text{ m}}$  |
| $ \zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} \text{ m}} (C)$  | $ \zeta_{lb}^{1 \times 10^{-7} - 4 \times 10^{-7} \text{ m}} $ as function of $C$   |
| $\zeta_p$  | Electrokinetic potential of molecular associates in samples kept in Permalloy containers  |
| $\zeta$ -potential   | Electrokinetic potential  |
| $\chi$   | Electric conductivity in $\mu\text{S cm}^{-1}$  |
| $\chi(C)$  | Electric conductivity as function of concentration  |
| $\chi_p$   | Electric conductivity of samples kept in Permalloy containers   |
| $\sigma_{lb}$  | Surface tension of samples kept at the laboratory bench   |
| $\sigma_{lb}(C)$   | Surface tension of samples kept at the laboratory bench as function of concentration  |
| $\sigma_p$   | Surface tension of samples kept in Permalloy containers   |



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