Liquids Prepared by Serially Diluting and Vigorously Shaking of Aqueous Solutions: Unveiling Effects of the Solute on their Properties

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Abstract

Altering the bioactivity of aqueous solutions via serial dilutions is important for toxicology, medicine and wastewater treatment. Extensive research has shown that serially diluting aqueous solutions beyond a threshold concentration, combined with vigorous shaking after each dilution step, has several notable effects. (a) The solutes may serve as substrates, which facilitate association of water molecules (10^{-5} and 10^{-7} m sized associates). (b) The associates and the bioactivity of these liquids persist when these fluids are diluted below 10^{-24} mol/liter. (c) The associates and the bioactivity of these liquids disappear when such ultra-diluted fluids are screened from ambient electromagnetic radiation. (d) The structural and physicochemical properties of these liquids are explainable within the context of quantum electrodynamics (QED). Hitherto, relations between the physicochemical properties of these liquids and their substrates have not been unveiled. There are no reports of statistically significant data that reflect physicochemical aspects of these liquids attributable to specific characteristics of substrates. Moreover, the aforementioned relations have not been theoretically derived. In this study, the relations are analyzed within the context of QED. The analyses have helped elucidate the challenge of observing the impact of the substrate on the associates. The analyses also indicate that measuring the quantum Hall effect of these liquids holds the promise of unveiling the aforementioned relations.

Introduction

Over the last decades, the physicochemical and bioactive properties of serially diluted aqueous solutions have been extensively studied [see reviews by Konovalov *et al.* (2014), Bellavite *et al.* (2014) and Elia *et al.* (2015)]. These solutions are serially diluted from a solution with a concentration (*C*) of ~4 - 10^{-3} mol/liter (M). The dilutions continue up to and beyond 10^{-24} M. Such dilutions, combined with vigorous shaking after each dilution step, may affect these liquids' bioactive, structural and physicochemical properties. These properties may differ in a statistically significant way from those of liquids with the same chemical composition but which were not serially diluted and vigorously shaken after each dilution step. The differences only occur when the solutions are diluted beyond a solute dependent threshold concentration (C_{thr}) on the order of 10^{-6}-10^{-10} M. The differences have been observed for solutions of many solutes, but not for all kinds. Impurities released by containers, contaminants and bubbles affect the properties of serially diluted liquids but cannot fully account for the differences in properties. The denotation SDVSAS will be used for a serially diluted vigorously shaken aqueous solution for which its properties are *altered* by serial dilutions and vigorous shaking after each dilution step.

To the best of my knowledge, no experiments have produced statistically significant data revealing relations between the physicochemical properties of SDVSASs diluted beyond 10^{-24} M versus the characteristics of the solutes (substrates) with which these liquids were prepared.[a,b] Moreover, the relations have not yet been theoretically derived.

The goals of this paper are to analyze the relations and to propose an experimental technique that has the potential of revealing these relations. These goals are important, because SDVSASs are relevant for toxicology, medicine, wastewater treatment and power generation (Konovalov *et al.*, 2014; Ryzhkina *et al.*, 2011; van Wassenhoven *et al.* 2017, 2018; Bandyopadhyay *et al.*, 2017). To pursue these goals, firstly, I summarize the facets of the empirical and the quantum electrodynamic (QED) models of SDVSASs, *i.e.,* only those facets required for analyzing the abovementioned relations. Subsequently, I analyze these relations and propose an experimental technique that holds the promise of verifying them. A list with abbreviations is presented at the end of this paper.

Models of SDVSASs

According to the empirical model of Konovalov *et al.* (2016), SDVSASs are disperse systems. They are not liquids (solutions) in the customary sense. For 10^{-24} M<C<C_{thr}, associates composed of water (H_2O) molecules and a few solute (substrate) molecules constitute the disperse phase. At each dilution-shaking step, the prevalence of the substrate decreases but associates composed of H_2O molecules persist. Upon crossing the 10^{-24} M threshold, a disperse system of associates of H_2O molecules surrounded by water (the medium) emerges. The model is mainly based on the experimental data of Konovalov *et. al.* (2014, 2015), *e.g.*:

- ~10^{-5} m and ~10^{-7} m sized associates, mainly composed of H_2O molecules, are present in SDVSASs with C< C_{thr}. The associates are self-organized, substrate-induced, entities stabilized during the preparation of SDVSASs. These fluidic associates are surrounded by randomly moving liquid molecules.
- The associates only form in the presence of ambient or artificial EM radiation. On screening such radiation, the associates disintegrate.
- The electronic structure and ordering of the associates' H_2O molecules are determined by the substrate.
- The associates constitute a phase which is different from the bulk liquid.

The empirical model of Elia *et al.* (2014, 2015), besides emphasizing the molecular association, addresses the solid residue (phase) obtainable by isolating associates through evaporating of the SDVSASs. The

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[a] For a partial overview of the experimental techniques used for studying SDVSASs see Yinnon (2017).

[b] Lenger *et al.* (2014) observed that information indicating characteristics of the substrates of SDVSASs is contained in these liquids' megahertz resonant frequencies. However, these frequencies often are too close to enable differentiating between SDVSASs prepared with different substrates. Bázár *et al.* (2015) and Gowen *et al.* (2015) demonstrated that near infrared spectroscopy, coupled with aquaphotomics, can only reveal molecular changes in water caused by different solutes for solutions with C>~10^{-5} M.

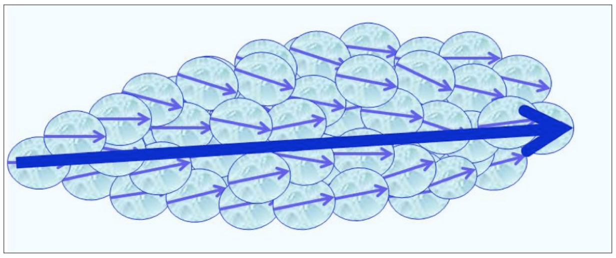

Fig. 1. *Schematic drawing of a CD_{rot} revealing its internal structure. Light-blue colored balls symbolize the H_2O molecules constituting the domain. Their blue arrows symbolize the electric dipole moments of the H_2O molecules. The large blue arrow symbolizes the electric dipole moment of the domain, resulting from the ferroelectric ordering of its molecules.*

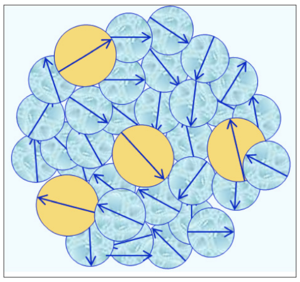

Fig. 2. *Schematic drawing of a $CD^{H_2O}_{elec}$ and its internal structure. Light-blue and yellow colored balls symbolize its H_2O molecules residing, respectively, in their ground and excited electronic states. The electric dipole moments of the H_2O molecules, symbolized by arrows, are randomly oriented.*

residue is soluble in water. After dissolving it in water, the liquid's physicochemical parameters are almost exactly like those of the original SDVSAS from which the solid was extracted.

The presence of ~10^{-5} m and ~10^{-7} m sized molecular associates in SDVSASs with $C < C_{thr}$, which only stabilize with EM radiation, signifies strong interactions between the molecules and the radiation. Therefore, modeling of SDVSASs necessitates a QED approach (Yinnon *et al.*, 2015a).[c]

Indeed, the associates have properties resembling those of two types of QED domains (Yinnon *et al.*, 2015b&c) — CD_{rot} and $CD^{H_2O}_{elec}$ (see *Fig. 1 & 2*). Stabilization of these domains, during the SDVSAS preparation procedure, is schematized in Appendix *Figs. A1 and A2*. The QED model of SDVSASs (*see Fig. 3*) is congruent with the abovementioned empirical models and data (Yinnon *et al.*, 2011, 2013, 2015b&c, 2017, 2018). Properties of CD_{rot} and $CD^{H_2O}_{elec}$ pertinent to the goals of this paper are summarized below. For more details, and recent references to the experimental verification

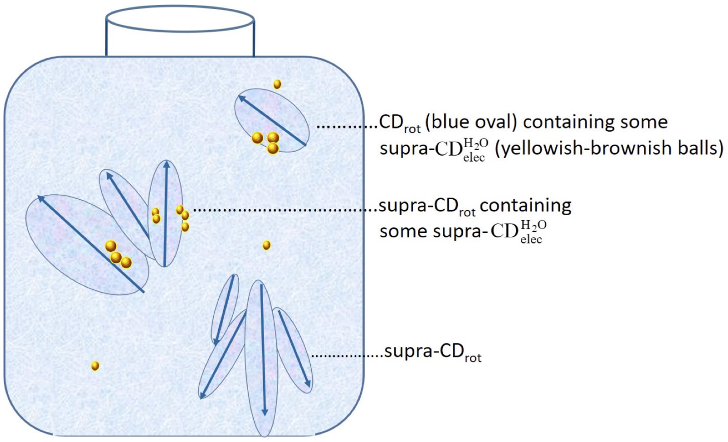

Fig. 3. *Schematic picture of a SDVSAS diluted beyond 10^{-24} M according to its QED model. CD_{rot} and $CD^{H_2O}_{elec}$ are dispersed in bulk water. CD_{rot}, with their electric dipole moments, are depicted as ovals with a blue arrow. $CD^{H_2O}_{elec}$ are depicted as yellowish-brownish balls. The domains' sizes are not presented according to their realistic scale ratios. Some of the domains are organized in supradomains: supra-CD_{rot}, supra-$CD^{H_2O}_{elec}$ and supra-CD_{rot} containing supra-$CD^{H_2O}_{elec}$ (see* Appendix*-paragraph ix & x). These supradomains are not ensembles of molecules but agglomerates of domains, like domains in liquid crystals.*

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[c] QED models explicitly describe QED forces, such as the interactions between the dipole moment or electrons of H_2O molecules and the external EM field (Del Giudice, 1988, Preparata, 1995). These forces may lead to association of part of the H_2O molecules in aqueous liquids. The various domains that may form and their concentration dependencies are summarized by Yinnon and Liu (2015a). The QED models of aqueous liquids considerably diverge from the customary ones, which explicitly describe electrostatic forces and assume that QED interactions can be treated as small perturbations (Horne, 1972; Robinson *et al.*, 2002). These models show that H_2O molecules, except for those in solvation shells, move randomly.

of these properties, see the Appendix.

CD_{rot} — These domains are composed of H_2O molecules that coherently oscillate between two of their rotational states (Del Giudice *et al.,* 1988, 2006). Their diameter may reach 10^{-4} m. A CD_{rot} has an electric dipole moment due to the ferroelectric ordering of its H_2O molecules. CD_{rot} may order in supradomains, wherein their electric dipoles are about parallel, anti-parallel or randomly aligned. CD_{rot} are superfluidic domains (*see Appendix, paragraph IX*). Just as in superfluidic Helium, whirlpools (vortices) may be created in CD_{rot}. Collisions between a CD_{rot} and its surrounding randomly moving molecules may cause part of its molecules to whirl into vortices. Also, EM radiation may excite H_2O molecules within a CD_{rot} and create vortices. The vortices cannot decay thermally, *i.e.*, they are cold. Therefore, their lifetimes are very long (months – years) (Yinnon *et al.*, 2013).

Information can be stored in supra-CD_{rot}. Due to their ferroelectric properties, information can be stored in a manner analogous to that in materials consisting of domains with magnetic moments. Moreover, information can be stored in their cold vortices.

$CD^{H_2O}_{elec}$ — These domains consist of H_2O molecules that coherently oscillate between their electronic ground state |0> and an excited electronic |b> state (Preparata, 1995). The energy of the |b> state is 12.07 eV. Since the ionization threshold of H_2O molecules is 12.62 eV, the electrons in the |b> state are weakly bound. Therefore, a $CD^{H_2O}_{elec}$ contains many coherent quasi-free electrons (QFEs). The diameter of a $CD^{H_2O}_{elec}$ is ~10^{-7} m.

$CD^{H_2O}_{elec}$ are superfluidic domains (see *Appendix, paragraph iv*). Therefore, interactions between $CD^{H_2O}_{elec}$ and the surrounding, randomly moving, molecules or EM radiation may excite the whole $CD^{H_2O}_{elec}$, *i.e.*, cause their molecules to whirl in *cold* vortices. QFEs do not collide with each other, as they are coherent, and a single collision would destroy their coherence. Thus, QFE are superconducting. Exciting a $CD^{H_2O}_{elec}$ may lead to *cold* coherent vortices in its pool of superfluidic QFEs. The energy spectra of these vortices are typified by a lowest lying excited state with a frequency of ~1000 Hz (Del Giudice, 1998, 2010). The energy spacing of the spectra is on the order of ~1000 Hz. The magnetic dipoles of these vortices may become aligned by external magnetic fields, *e.g*. the terrestrial magnetic field (Del Giudice *et al*., 2002; Montagnier *et al.,* 2011). The QFE's cold vortices constitute "long-term information storage devices." The same holds for the cold vortices of the H_2O molecules in $CD^{H_2O}_{elec}$.

Information on the Substrate Imprinted in SDVSASs

The processes leading to stabilization of CD_{rot} and $CD^{H_2O}_{elec}$ in SDVSASs diluted below C_{thr} have been specified by Yinnon *et al.* (2011, 2015 b & c). They pointed out that, for a SDVSAS at $C<C_{thr}$, vigorous shaking excites or breaks up its CD_{rot}. Subsequent dilution of the SDVSAS transfers excited or broken CD_{rot} pieces, as well as $CD^{H_2O}_{elec}$, to the diluted liquid. The excited or broken CD_{rot} pieces stabilize new CD_{rot} and $CD^{H_2O}_{elec}$. Hence, these domains are present in SDVSASs with $C<<C_{thr}$, including SDVSASs diluted beyond 10^{-24} M.

Information on the substrate imprinted in CD_{rot} — In SDVSASs with $C<C_{thr}$, the solutes are located within CD_{rot} (see *Appendix, paragraph X*). An object present in an ensemble of H_2O molecules adds a term $\boldsymbol{H}=-\mathbf{d}.\boldsymbol{E}$ to the potential energy of the ensemble [see *Appendix - Eq. (A1)*]. $\mathbf{d}$ is the electric dipole moment of an H_2O molecule. $\boldsymbol{E}$ is the electric field due to the object. Analogously, each solute molecule adds a term to the potential energy of such

an ensemble. The term is a function of the electric field $\boldsymbol{E}_s$ of the solute molecule. Thus, solutes affect the wavefunction describing CD_{rot} (Ψ_{CDrot}) and the polarization $\mathbf{P_n}$ of the liquid in direction **n** [*see Appendix –Eq. (A2)*].

For SDVSASs at 10^{-24} M<C<<C_{thr}, it can be assumed that just one solute molecule is present in each of its CD_{rot}. On vigorously shaking this SDVSAS, part of its broken CD_{rot} pieces will not contain any solutes, yet, after breakup of a CD_{rot}, the polarization within its broken pieces ($\mathbf{P_n}^{\text{broken-CDrot}}$) is reminiscent of the "mother domain." Moreover, after subsequent break up of broken pieces, $\mathbf{P_n}^{\text{broken-CDrot}}$ is reminiscent of its "grandmother domain," "great-grandmother domain" and so on. A broken piece's memory of the polarization of its "ancestors" is due to the extremely slow dynamics of the superfluidic CD_{rot} and their cold vortices. The dynamics occur over days and even years (see *Appendix-paragraph V*). Accordingly, solute reminiscent $\mathbf{P_n}^{\text{broken-CDrot}}$ pieces persist for macroscopic times scales. Thus, on serially diluting aqueous solutions and vigorously shaking after each dilution step, even in SDVSASs diluted beyond 10^{-24} M, CD_{rot} and their broken CD_{rot} siblings are present along with their substrate reminiscent polarization.

The number of solute molecules that can be contained in a CD_{rot} is small (see *Appendix - paragraph X*). Hence, the impact of the solutes on the polarization of these domain ($\mathbf{\Delta_s P_n}$) is small. It can be treated as a small perturbation. Of course, $\mathbf{\Delta_s P_n}$ will have some effect on the physicochemical properties of SDVSASs, *e.g.,* its dielectric permittivity. For many types of solutes, the differences between their $\boldsymbol{E}_s$ are small. Therefore, the difference between their $\mathbf{\Delta_s P_n}$ will be tiny. Hitherto, measurements of physicochemical variables reflecting $\mathbf{\Delta_s P_n}$ have not revealed differences in SDVSASs prepared with different substrates. Seemingly, $\mathbf{\Delta_s P_n}$ is negligible compared to the effects of thermal fluctuations or ambient factors.

Vortices created in CD_{rot} by radiation will affect their $\mathbf{P_n}$. When the stimulating frequency does not match that of the system, and the radiation energy transferred to the domain is less than the cohesion energy of an H_2O molecule in a CD_{rot} (about ~0.025 eV), the effects of the vortices on $\mathbf{P_n}$ will just be tiny perturbations $\mathbf{\Delta_s P_n}$. The energy of these vortices is on the order, or less than that, of thermal fluctuations. Therefore, these vortices cannot have a statistically significant impact on the physicochemical variables of SDVSASs, like dielectric permittivity. However, when the frequency of the radiation matches that of the system, the impact on $\mathbf{P_n}$ may be significant.

The tininess of $\mathbf{\Delta_s P_n}$ suggests that we should continue searching for an observable variable which will enable distinguishing between SDVSASs prepared with different solutes.

Information on characteristics of the substrate imprinted in $\mathbf{CD}^{\mathbf{H_2O}}_{\mathbf{elec}}$ — Solutes cannot locate within $CD^{H_2O}_{elec}$ (Del Giudice *et al.*, 2000, 2010). In SDVSASs, solute molecules imprint their characteristics in $CD^{H_2O}_{elec}$ in the following ways:

1. Solutes affect $CD^{H_2O}_{elec}$ through their impact on $\mathbf{P_n}$. The electric field $\boldsymbol{E}_s$ of a solute molecule located in CD_{rot} impacts the polarization $\mathbf{P_n}$, as discussed above. $\mathbf{P_n}$ affects the polarization of H_2O molecules, as well as the cloud of QFEs of each $CD^{H_2O}_{elec}$. It is likely that thermal fluctuations blur the variations in $\mathbf{\Delta_s P_n}$ induced by different solutes. Such blurring would explain why no impact of solutes on $CD^{H_2O}_{elec}$ via $\mathbf{P_n}$ has been detected.

2. Ions or polar solutes are attracted by the fields of the photons condensed in $CD^{H_2O}_{elec}$ (Del Giudice *et al.*, 2002). The attraction affects the wavefunction describing $CD^{H_2O}_{elec}$, i.e., $\Psi CD^{H_2O}_{elec}$.

3. Whenever the motion of ions does not

get altered by collisions with other molecules, they orbit around their nearest neighbor $CD^{H_2O}_{elec}$ (Del Giudice *et al.*, 2002).[d] In SDVSASs, at 10^{-24} M<C< C_{thr}, solutes incorporate in CD_{rot} (see *Appendix – paragraph X*). The motion of these solutes does not get altered by collisions, because CD_{rot} are superfluidic. In SDVSASs, part of the $CD^{H_2O}_{elec}$ are localized in CD_{rot}. Hence, ionic solutes present in CD_{rot} may orbit around $CD^{H_2O}_{elec}$. The circular speed of ions orbiting around a $CD^{H_2O}_{elec}$ is proportional to the cyclotron frequency (ν_c) (Del Giudice *et al.*, 2002)[d]. ν_c is defined such that $\nu_c=(qB)/2\pi m$ where q and m are the electric charge and the mass of the ion, respectively, and B is the strength of the static magnetic field (*e.g.* the terrestrial magnetic field). An alternating magnetic field, with a frequency matching the orbit frequency, extracts the ions from their orbits (Del Giudice *et al.*, 2002). Due to the conservation of angular momentum, this extraction causes rotational excitation of the H_2O molecules and the QFEs of the $CD^{H_2O}_{elec}$, *i.e.*, the formation of coherent vortices. When the solute concentration is about uniform in a mesoscopic region, and the external field also spans over mesoscopic distances, the excitation energy of the vortices of many $CD^{H_2O}_{elec}$ may be uniform (Montagnier *et al.*, 2011, 2015). Since $\nu_c=(qB)/2\pi m$, the vortices are a function of the charge distribution of the ions.

4. Polar solutes in SDVSASs can indirectly affect the excitations of the QFE cloud. Such solutes affect the spontaneous dissociation of H_2O molecules. These solutes also affect the charge distribution of the hydronium or hydroxyl ions resulting from such dissociation. These ions orbit around $CD^{H_2O}_{elec}$, and the extraction of these ions by ambient radiation is thus affected by $\boldsymbol{E}_s$.

The effects described in paragraphs 1-4 all impinge on the wavefunction $\Psi_{CD^{H_2O}_{elec}}$. The general wavefunction Ψ, describing matter interacting with EM fields, has been derived by Preparata (1995, ch. 2 & 3). In these chapters, the excitations of Ψ due to perturbations are also presented. The details of Ψ relevant to an ensemble of H_2O molecules are presented in Arani *et al.* (1995).

Without going into detail about the effects of the solutes' $\boldsymbol{E}_s$ on $\Psi_{CD^{H_2O}_{elec}}$, it suffices to note that $\boldsymbol{E}_s$, at least perturbatively, affects the excited states of $CD^{H_2O}_{elec}$, *i.e.*, their molecular vortices and the vortices of their QFEs. Despite these vortices constituting memory devices for $\boldsymbol{E}_s$, extracting information on $\boldsymbol{E}_s$ by analyzing its impact on physicochemical properties of SDVSASs (*e.g.* electric conductivity) has failed. The failure is attributable to the energies of the vortices being on the same order, or less than that, of the thermal fluctuations of water at ambient conditions.[e] The latter is on the order of ~10^{-2} eV.

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[d] The orbit of noncolliding ions around $CD^{H_2O}_{elec}$ is an aspect of the explanation, provided by Del Giudice *et al.* (2002), for the Zhadin effect (Zhadin *et al.*, 1998), which has been confirmed by Pazur (2004).

[e] Spectroscopic data taken from SDVSASs diluted far beyond C_{thr} does not provide information $\boldsymbol{E}_s$. The intensities of some UV-visible bands of such SDVSASs are correlated with the prevalence of their nano-associates (Ryzhkina *et al.*, 2018). $CD^{H_2O}_{elec}$ affects the ultraviolet (UV), visible (vis) and infrared (IR) spectra of aqueous systems (Yinnon *et al.*, 2016, 2018; Elia *et al.*, 2017). The UV absorption and UV-vis fluorescence spectra of water containing $CD^{H_2O}_{elec}$ typically contain broad featureless bands. Differences have been observed in UV transmission intensities of SDVSASs with C<<C_{thr} and prepared with different solutes (Klein *et al.*, 2013). The featureless broad UV-vis spectral bands have not revealed any differences between SDVSASs at C<<C_{thr} attributable to specific characteristics of their substrates. Spectral measurements cause slight changes in the electronic, vibrational and rotational states of H_2O molecules. Likely, these changes blur the tiny differences between the excited states (vortices) of the QFEs. These tiny differences also cannot be imprinted, in a statistically significant manner, in those physicochemical variables of SDVSASs that are affected by thermal fluctuations. The energy of the vortices in QFEs is ~1000 Hz (10^{-12} eV), *i.e.*, many orders less than that of the thermal fluctuations of water at ambient conditions.

Studying Quantum Hall Effects for Exposing Variations between SDVSASs Prepared with Different Substrates

The above presented analyses accentuate the need for directly measuring details of the vortices in the superfluidic QFEs. Vortices in ensembles of superfluidic electrons have been extensively studied during the last decades (Abrikosov *et al.*, 2002; Jain, 2015). Superfluidic electrons underlie the superconductivity of superconductors, and vortices in ensembles of superfluidic electrons can be studied by measuring the quantum Hall effect (Matsuda *et al.*, 2002).

The Hall effect is the voltage difference (V_H) across an electrical conductor, which appears transverse to an electric current (I) on applying a magnetic field (**B**) perpendicular to the current (Purcell, 1963). Immediately after applying the magnetic field, the charge carriers are deflected towards a rim of the conductor. As a result, at some distance from the current-introducing contacts, for example, positively charged particles pile up on the upper rim of the conductor and become depleted from its opposite lower rim. See *Fig. 4*. The piling up and depletion of these particles creates an electric field, which underlies V_H. At steady state, the induced electric force exactly cancels the magnetic force, so that the charge carriers are no longer deflected. The Hall coefficient (R_H) is the ratio of the magnitude of the induced electric field at steady state (E) to the product of the current density (j_x) and the magnitude of the applied magnetic field (B), that is, $R_H= E /(j_x B)$. The Hall coefficient is a characteristic of the material from which the conductor is made. R_H depends on the type, number, and properties of the charge carriers that constitute the current.

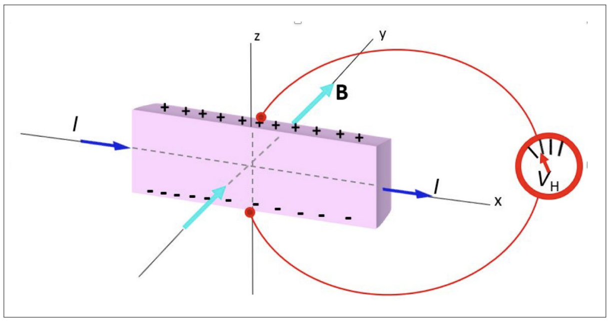

Fig. 4. *Schematic drawing of the Hall effect. A voltage* V_x *applied to a conductor (the pinkish rectangle) leads to the current z. After adding a magnetic field in the y-direction (***B***), the charge carriers become deflected towards an edge of the conductor. As a result, the charge carriers pile up on the upper rim of the conductor and become depleted from the lower rim, creating an electric field. This underlies the Hall voltage* V_H, *as indicated by the Voltmeter. At steady state, the induced electric force exactly cancels the magnetic force, so that the charge carriers are no longer deflected.*

The quantum Hall effect is the quantum analogue of the above described classical Hall effect. It appears in two-dimensional electron systems (Prange *et al.*, 1990)[f]. According to quantum physics, when a two-dimensional electron system is subjected to a magnetic field, its electrons follow circular orbits with discretely valued energy levels. As a result, the Hall conductance (σ) is quantized: $\sigma=I/V_H=\nu e^2/h$, where e is the elementary charge and h is Planck's constant. The dimensionless value of the filling factor ν is either an integer (1, 2, 3,...) or a fractional (1/3, 2/5, 3/7, 2/3, 3/5, 1/5, 2/9, 3/13, 5/2, 12/5,...) number. Correspondingly, the quantum Hall effect is denoted as the integer or fractional quantum Hall effect. The latter can be regarded as an integer quantum Hall effect, albeit not of electrons but of charge-flux composites (Jain, 2015). Interactions between electrons, resulting in highly correlated quantum states, underlie these composites. The quantum Hall effect reflects details of quantum states of the electron system and impurities present in

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[f] Until 2007, the quantum Hall effect was solely observed in systems at cryogenic temperatures (below ~30 K) and at sufficiently high magnetic fields (a few Tesla). In 2007, the effect was observed in graphene at room temperatures and magnetic fields of about 30 Tesla (Novoselov *et al.*, 2007; Parmentier *et al.*, 2016).

the conducting material.

Vortices present in ensembles of superfluidic electrons impact on their quantum Hall effect (Matsuda *et al.*, 2002). For example, a vortex motion perpendicular to the transport current corresponds to a dissipative longitudinal electric field, while parallel motion impacts the Hall electric field ($\boldsymbol{E}$). Measuring the quantum Hall effect enables extracting information on energy dissipation processes. Such processes reflect the electronic structure of the vortices, the characteristics of the ensemble of superfluidic electrons and the low energy excitations out of the condensate.

I propose to study the vortices of QFEs by measuring quantum Hall effects in SDVSASs,[g] or in the solid residue left over after evaporating these liquids. Some properties of SDVSASs are preserved in the residue (Elia *et al.*, 2014; Klein and Wolf, 2016). Such studies might reveal properties of SDVSASs diluted beyond 10^{-24} M, which are related to the $\boldsymbol{E}_\mathrm{s}$ of their substrate.

It is beyond the scope of this paper to specify techniques for measuring the quantum Hall effect of SDVSASs. Vortices in ensembles of superfluidic electrons are still extensively studied (Abrikosov, 2002; Jain, 2015). Assessing implications of the superfluidic QFEs, for detailing techniques, will be left to those specialized in quantum Hall effects. It is likely that the quantum Hall effects of SDVSASs will be more discernible after aging of these liquids. On aging of SDVSASs, their physicochemical properties become more distinct, because they are far from equilibrium self-organizing systems (Elia *et al.*, 2008).

Discussion and Conclusion

The above analyses indicate that for SDVSASs with $C<C_\mathrm{thr}$, the degree to which their substrates impact on their properties is similar or less than that of thermal fluctuations. Therefore, variables affected by such fluctuations cannot reveal the impact. The QED model indicates that the vortices in the superfluidic domains ($\mathrm{CD}^{\mathrm{H_2O}}_{\mathrm{elec}}$ and CD_rot) present in these liquids are a function of the electronic characteristics of their substrates but are not affected by thermal fluctuations. Accordingly, I propose to study the vortices of the superfluidic QFEs by measuring their impact on the Quantum Hall effect. Such a study promises to unveil variations between SDVSASs prepared with different substrates, even when their serial dilutions combined with vigorous shaking continued beyond 10^{-24} M.

Revealing substrate dependent quantum Hall effects in SDVSASs will support the QED interpretation of these liquids. It will also provide additional evidence that QED domains may play roles in aqueous systems. Moreover, it will show that outcomes of chemical or biological processes, which involve SDVSASs diluted close to or far beyond 10^{-24} M, (*i.e.*, outcomes attributable to properties of their substrates) are real phenomena. In addition, it would indicate that measuring the quantum Hall effects might be an effective method for studying other aqueous systems. Phenomena in various systems have been attributed to stabilization of QED domains at ambient conditions (Del Giudice *et al.*, 2010, 2013; De Ninno *et al.*, 2011, 2013, 2017; D'Emilia *et al.*, 2015; Elia *et al.*, 2015, 2017; Yinnon *et al.*, 2009). For example, the 10^{-5} -10^{-4} m wide exclusion zone (EZ), which forms adjacent to hydrophilic membranes (Zheng *et al.*, 2006), has been attributed to stabilization of $\mathrm{CD}^{\mathrm{H_2O}}_{\mathrm{elec}}$ and CD_rot (Del Giudice *et al.*, 2010, 2013; De Ninno, 2017). The attribution has been supported by experiments on water iteratively brought in contact with such membranes. The contact may rupture clumps of EZ and disperse these in the bulk liquid.

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[g] The electric conductivity of water is enhanced in the presence of $\mathrm{CD}^{\mathrm{H_2O}}_{\mathrm{elec}}$ (Yinnon *et al.*, 2013, 2016; Elia *et al.*, 2017).

The iteratively perturbed water (IPW) has physicochemical properties commensurate with its clumps containing $CD^{H_2O}_{elec}$ and CD_{rot} (Elia *et al*., 2015, 2017; Yinnon *et al*., 2016). Some of these properties, *e.g*., the optical activity or pH, are perturbing membrane specific (Elia *et al*., 2018, 2019). Accordingly, it is plausible that water flowing adjacent to various types of hydrophilic cellulose (a common phenomenon in nature) contains $CD^{H_2O}_{elec}$ and CD_{rot}, with perturbing membrane specific vortices. Studying perturbing membrane dependent quantum Hall phenomena in IPW may have implications for revealing effects of such liquids on organisms. It has been hypothesized that the aqueous phase in cells entirely consists of EZ water (Zheng *et al*., 2006). Perturbing membrane specific vortices in IPW might alter the $CD^{H_2O}_{elec}$ and CD_{rot} of cellular water. Alterations in the properties of these domains might affect biological processes. Modes of interactions between biological molecules and $CD^{H_2O}_{elec}$ and CD_{rot} have been proposed by De Giudice *et al*. (2010).

Appendix

Properties of CD_{rot} — The properties of CD_{rot} described below are derived *ab initio* by Del Giudice *et al*. (1988, 2013, 2006) and Sivasubramanian *et al*. (2005).

I. CD_{rot} are composed of H_2O molecules that coherently oscillate between two of their rotational states. Analyses of Raman and IR spectroscopic data by Elton *et al*. (2016) indicate the presence of coherent long-range dipole–dipole interactions in water. Dielectric dispersion measurements by Mahata (2013) indicate that association of molecules in SDVSASs involve the electric dipole rotations of the H_2O molecules (Yinnon, 2017).

II. Interactions between the electric dipole moment of H_2O molecules and EM radiation underlie the formation of CD_{rot}, as corroborated by experimental data on SDVSASs (Yinnon *et al*., 2015c).

III. The ferroelectric ordering of the H_2O molecules within CD_{rot}, as well as that of the CD_{rot}, are corroborated by dielectric permittivity measurements of SDVSASs (Yinnon *et al*., 2015c).

IV. The conditions for formation of CD_{rot} have been derived *ab initio* with classical electrodynamics and with QED. These conditions [summarized by Yinnon *et al*. (2016)] include: These domains cannot auto-organize, because the energy an H_2O molecule gains by joining a CD_{rot} (~0.025 eV) is on the same order as the energy of the water's thermal fluctuations. However, objects with sizable asymmetric charge distributions (*e.g*., polar solutes, polar macromolecules or hydrophilic membranes) may stabilize CD_{rot}. The presence of such an object adds a term

$$H=-\mathbf{d}.\boldsymbol{E} \qquad \text{(A1)}$$

to the potential energy of a H_2O ensemble. $\mathbf{d}$ is the electric dipole moment of a H_2O molecule. $\boldsymbol{E}$ is the electric field due to the object. $\boldsymbol{H}$ augments the energy an H_2O molecule gains on its inclusions within CD_{rot}. Hence, near the object, $\boldsymbol{H}$ may induce H_2O molecules to order into CD_{rot}. The ordering results in a permanent polarization $\mathbf{P_n}$ in direction $\mathbf{n}$ with magnitude:

$$P_{\mathrm{n}}(t) = \left(\frac{1}{\sqrt{3}}\right)(sin2\zeta)\{[A_0(t)]^2 - [A_1(t)]^2\} + \left(\frac{2}{\sqrt{3}}\right)A_0(t)A_1(t)(cos2\zeta)cos\,\{[\omega - (\omega_0 + 4H)^{\frac{1}{2}}]\mathrm{t}\} \qquad \text{(A2)}$$

where $\tan(\zeta) = \left[\omega_0 - (\omega_0^2 + 4H^2)^{\frac{1}{2}}\right]/(2H)$. Here $A_0(t)$ and $A_1(t)$ represent the amplitudes of the collective modes of the H_2O molecules in their ground state (angular momentum l=0) and their excited state (l=1) respectively at microscopic time t. ωt signifies the difference in phases of these modes. ω_0 is the frequency of the l=0 ↔ l=1 transition. The time average of the polarization is:

$$\overline{P_{\mathrm{n}}} = \left(\frac{1}{\sqrt{3}}\right)(\sin\ 2\zeta)(\overline{A_0^2} - \overline{A_1^2}) \qquad \text{(A3)}$$

with $\overline{A_0^2}$ and $\overline{A_1^2}$, respectively, the time average of $[A_0(t)]^2$ and $[A_1(t)]^2$. *Eq. (2)* shows that a net polarization implies a nonzero difference in the l=0 and l=1 populations, *i.e.*, a state different from the $[A_0(t)]^2 \approx [A_1(t)]^2$ typifying the equilibrium Boltzmann distribution of bulk water.

V. After CD_{rot} are stabilized, and the object is removed, these domains still may persist for macroscopic times (years) (Yinnon *et al.*, 2013, 2016). Water containing CD_{rot} is a far-out-of equilibrium dissipative system. Its CD_{rot} continuously realign on a macroscopic time scale.

VI. Thermogravimetry indicates that isolated CD_{rot} may persist up to ~650°C, which agrees with *ab initio* derived predictions (Yinnon *et al.*, 2016).

VII. CD_{rot} reaching dimensions of 10^{-4} m is commensurate with Atomic force microscopy revealing 10^{-5} m sized domains in SDVSASs with $C<C_{thr}$ (Ryzhkina *et al.*, 2012) and Fluorescence microscopy revealing 10^{-4} m sized domains in water perturbed by Nafion (Elia *et al.*, 2013). These domains have characteristics resembling those of CD_{rot} (Yinnon *et al.*, 2015c, 2016).

VIII. Serially diluting an aqueous solution with $C>10^{-3}$ M, and vigorously shaking it after each dilution, may stabilize CD_{rot} when C drops below C_{thr} (see *Figs. A1 & 2*) (Yinnon *et al.*, 2015c).

IX. CD_{rot} are superfluidic domains. A single collision between the H_2O molecules constituting a CD_{rot} would destroy their rotational coherence. The superfluidity implies that exciting CD_{rot} may create vortices within these domains. Collisions of CD_{rot} with surrounding molecules or EM radiation may excite CD_{rot}. When the excitation energy is larger than the energy an H_2O molecule gains by joining a CD_{rot} (~0.025 eV), an H_2O molecule desorbs. When the excitation energy is smaller, the whole domain becomes excited and vortices are created. These vortices cannot decay thermally. The coherence of the rotations of the H_2O molecules within the

CD_{rot}, which surround the vortices, prevents random thermal fluctuations, *i.e.,* the vortices are cold.

X. Solutes are pulled into CD_{rot}, yet many solutes destroy CD_{rot}. The critical concentration below which CD_{rot}- persist (C^{CDrot}_{crit}) depends on the solute. Typically, 10^{-10} M < C^{CDrot}_{crit} < 10^{-6} M. C^{CDrot}_{crit} equals C_{thr} (Yinnon *et al.*, 2015 b & c).

Properties of $CD^{H_2O}_{elec}$ — The properties of $CD^{H_2O}_{elec}$ described below are derived by Preparata (1995 ch. 10), Arani *et al.* (1995), Del Giudice *et al.* (1998, 2000, 2002, 2007, 2010), Montagnier *et al.* (2011, 2015), Bono *et al.* (2012) and Sen *et al.* (2015).

i. Interactions between H_2O molecules and ultraviolet (UV) radiation underlie formation of $CD^{H_2O}_{elec}$, as corroborated by experiments of SDVSASs and other structured waters (Elia *et al.*, 2017; Ryzhkina *et al.*, 2018; Yinnon *et al.*, 2017, 2018).

ii. The ~10^{-7} m dimension of $CD^{H_2O}_{elec}$ is corroborated by experiments (Yinnon *et al.*, 2015b&c).

iii. The energy an H_2O molecule gains by joining a $CD^{H_2O}_{elec}$ (E_{coh}) results from its participation in the coherent oscillations of the domain's molecules. At ambient conditions, E_{coh}≈10^{-1} eV, as corroborated by experiments (De Ninno *et al.*, 2013).

iv. $CD^{H_2O}_{elec}$ are superfluidic domains. A single collision between the H_2O molecules constituting a $CD^{H_2O}_{elec}$ would destroy their coherence. The impact of a randomly moving liquid molecule bordering on a $CD^{H_2O}_{elec}$ colliding with it depends on the collision energy. When the collision energy is larger than E_{coh}, an H_2O molecule may desorb from the $CD^{H_2O}_{elec}$. Otherwise, superfluidity implies that the collision excites the whole $CD^{H_2O}_{elec}$, *i.e.*, vortices are created. These excited $CD^{H_2O}_{elec}$ states may also be induced by irradiation.

v. The formation of $CD^{H_2O}_{elec}$ is a central aspect upon condensation of a liquid from its vapor.

vi. $CD^{H_2O}_{elec}$ only form at temperatures below 500 K (at ambient pressures), as corroborated by experiments (Yinnon *et al.*, 2016).

vii. In bulk water at ambient conditions, the energy of thermal fluctuations is about an order of magnitude smaller than E_{coh}. Therefore, in bulk water, $CD^{H_2O}_{elec}$ are metastable. The competition between QED interactions and collisions resulting from thermal noise causes a continuing adsorption and desorption of H_2O molecules from $CD^{H_2O}_{elec}$. In other words, in bulk water there are everlasting transitions of H_2O molecules between a coherent and a noncoherent phase. The transitions are like those in superfluidic Helium. The transitions produce a continuously changing space distribution of the coherent and noncoherent fractions of H_2O molecules. At room temperature, ~20 percent of the H_2O molecules belong to the former (Preparata, 1995 ch. 10), as corroborated by experiments (Yinnon *et al.*, 2012). The time scale of the transitions is below 10^{-14} seconds. Therefore, observation of $CD^{H_2O}_{elec}$ requires very fast probes.

viii. Ensembles of $CD^{H_2O}_{elec}$ can be stabilized by CD_{rot}, membranes, macromolecules, CD_{plasma} or IPD_{plasma}. The latter two are QED domains consisting of H_2O molecules and solvated solutes (Del Giudice *et al.*, 2000; Yinnon *et al.*, 2012). Solvated solutes organize in CD_{plasma} for C larger than a solute dependent transition concentration ($C^{IPDplasma}_{trans}$). For C < $C^{IPDplasma}_{trans}$, solvated solutes may organize in IPD_{plasma}. The solutes in CD_{plasma} perform coherent plasma oscillations, while those in IPD_{plasma} participate in in-phase plasma oscillations. Experimental data indicating stabilization of

$CD^{H_2O}_{elec}$ by CD_{rot}, membranes, CD_{plasma} or IPD_{plasma} have recently been forwarded (Yinnon *et al.*, 2015b&c, 2016, 2017, 2018; Elia *et al.*, 2015, 2017, De Ninno, 2017).

ix. When a $CD^{H_2O}_{elec}$ is stabilized by a CD_{rot}, the former is located within the latter. The H_2O molecules of a $CD^{H_2O}_{elec}$ located within a CD_{rot} reside in a superposition state. These H_2O molecules simultaneously participate in the coherent electronic and coherent rotational oscillations typifying the molecular ensembles constituting these domains.

x. $CD^{H_2O}_{elec}$ agglomerate into supra-$CD^{H_2O}_{elec}$, as corroborated by experiments (Yinnon *et al.*, 2015c).

xi. Fingerprints of the QFEs, rendering $CD^{H_2O}_{elec}$ electrically charged, have been revealed by experiments (Yinnon *et al.*, 2015c).

xii. The ~1000 Hz spacing of the spectra of the vortices of QFEs has been corroborated by experiments (Montagnier *et al.*, 2015).

xiii. $CD^{H_2O}_{elec}$ cannot contain solutes.

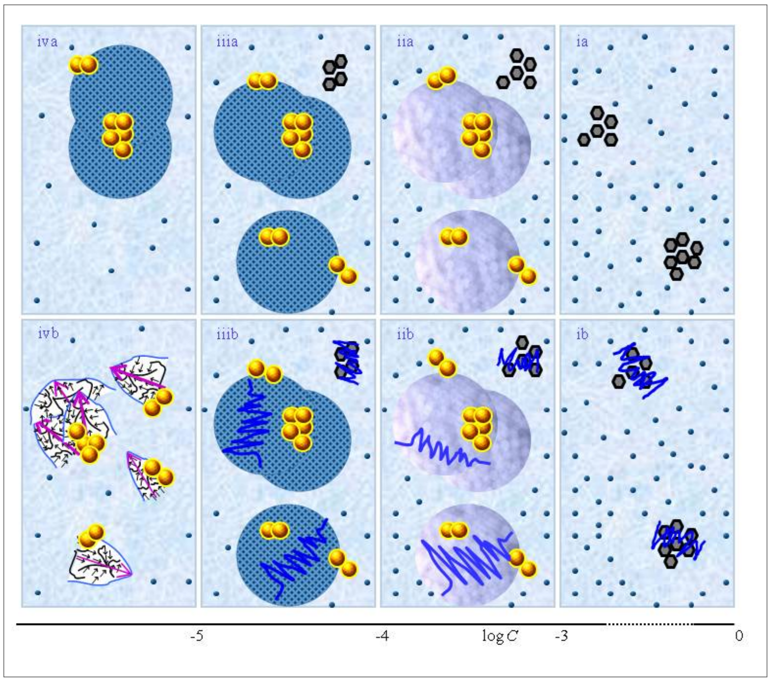

Fig. A1. Schematic picture of serially diluted solutions of weak electrolytes or nonelectrolytic compounds. Solutions diluted onto a solute dependent critical concentration C^{CDrot}_{crit} are pictured. *The top rows (a) and bottom rows (b) series pertain to solutions which, respectively, were not vigorously shaken and those which were vigorously shaken after each dilutions step.*[h]

Figure ia illustrates that, for C larger than a transition concentration $C^{CDplasma}_{trans}$, all solvated solutes move randomly, i.e., do not organize in a QED domain. The tiny blue balls represent randomly moving ~10^{-9} - 10^{-8} m solvated solutes. The irregularly shaped bunches of black hexagons represent aggregates of nonsolvated solutes. Figure iia illustrates that, on dilution below $C^{CDplasma}$, solvated solutes organize in a QED domain type denoted CD_{plasma} (symbolized with purple-blue colored balls). The yellow-brown balls and their agglomerates represent, respectively, ~10^{-7}m sized QED domains denoted $CD^{H_2O}_{elec}$ and supra-$CD^{H_2O}_{elec}$, which are both stabilized by CD_{plasma}. Figures iia and iiia illustrate the transformation of CD_{plasma} into another type of QED domain denoted $IPD_{plasm.a.}$ The transformation occurs at the transition concentration $C^{CDplasma}_{trans}$. Figures ia-iiia illustrate that on dilution the nonsolvated solutes diminish, i.e., solvate. Figures iiia-iv illustrate that, on dilution, the diameter of IPD_{plasma} does not change, but the number of IPD_{plasma} diminishes.

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[h] 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. The picture is based on the analyses presented by Yinnon *et al.* (2011, 2013, 2015b&c). The figure is a reprint of *Fig.1* in Yinnon and Liu (2015c). The schematic picture of serially diluted solutions of strong electrolytes is similar. It appeared in *Fig. 1* of Yinnon and Liu (2015b).

In the b series, the blue zigzag curves symbolize that shaking excites or cracks domains and aggregates. Figure iib illustrates that excitations or cracking does not significantly alter the internal structure of CD_{plasma}, which just as in Figure 1iia are represented with purple-blue colored balls. Figures iib and iiib illustrate the transition from CD_{plasma} to IPD_{plasma}, with the latter pictured as blue-crystalline balls just as in Figure iiia. Figures iiib and ivb illustrate that shaking excites or breaks up IPD_{plasma}. The excited or broken IPD_{plasma} pieces are electric dipole aggregates ($EDA^{IPDplasma}$). These are pictured as irregularly shaped aggregates in figure ivb. Their aligned black arrows orderings symbolize $EDA^{IPDplasma}$'s distorted ferroelectric H_2O molecules orderings. The purple arrow in the $EDA^{IPDplasma}$ symbolize these domains' dipole moments.

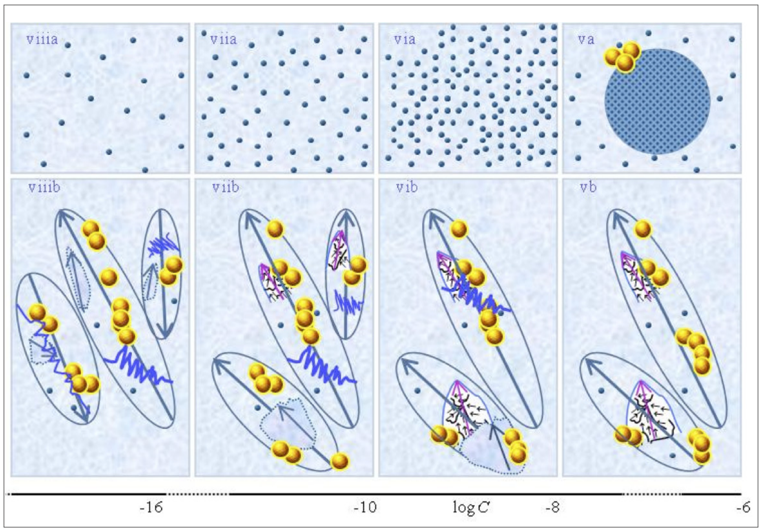

Fig. A2. Schematic picture of serially diluted solutions of weak electrolytes or nonelectrolytic compounds. Solutions diluted beyond a solute dependent critical concentration C_{crit}^{CDrot} are pictured.

The top rows (a) and bottom rows (b) series pertain to solutions which, respectively, were not vigorously shaken and those which were vigorously shaken after each dilutions step.[h]

Fig. va illustrates that at very low concentrations, the prevalence of IPD_{plasma} becomes negligible. Figures via-viiia illustrate that below a certain concentration there are insufficient solutes to form IPD_{plasma}. The concentration below which no IPD_{plasma} form has not yet been theoretically derived. Figures via-viiia illustrate that, whenever there are too few solutes to form IPD_{plasma}, the solution has the characteristics predicted by the customary models, i.e., all solvated solutes move randomly and their number diminishes on dilution.

In the b series, the blue zigzag curves symbolize that shaking excites or cracks domains and aggregates. Figure vb illustrates that on diluting below a solute type dependent critical concentration (C_{crit}^{CDrot}), a QED domain denoted CD_{rot} becomes stabilized by $EDA^{IPDplasma}$, i.e., the irregular shaped $EDA^{IPDplasma}$ are located within the elongated ovals representing CD_{rot}. The dark blue arrows symbolize the dipole moment of CD_{rot}. Figure vib shows that vigorous shaking excites or breaks up CD_{rot}, thus creating entities (electic dipole aggregates) denoted EDA^{CDrot}. The lump outlined with an irregular shaped broken curve, located at the bottom of one of the left CD_{rot}, represents the EDA^{CDrot}. Figures vib-viib show that at certain concentrations both $EDA^{IPDplasma}$ and EDA^{CDrot} are present within CD_{rot}, though the sizes of $EDA^{IPDplasma}$ diminish with concentration. Figure viiib shows that on diluting further, no $EDA^{IPDplasma}$ persist, i.e., there are too few solute particles to sustain $EDA^{IPDplasma}$. At these concentrations, vigorous shaking just breaks up CD_{rot} and creates new EDA^{CDrot}. These, in turn stabilize, new CD_{rot}, as pictured in Figure viiib. Figures vb-viib illustrate that CD_{rot} may align with their dipole moments parallel. Figure viiib illustrates that at certain concentrations their dipoles may be aligned antiparallel.

Dedication

I dedicate this paper to Dr. Suzanne S. D. Kleinberger. Her many years' long devoted patient support, combined with her gift of distinguishing between the medical efficacy of SDVSASs prepared with different substrates, made this study possible.

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Table 1

List of abbreviations in alphabetic order, followed by Greek symbols abbreviations

Abbreviation	Explanation
B	Magnetic Field
C	Concentration
C_{crit}	Critical concentration
C^{CDrot}_{crit}	Critical concentration below which CD_{rot} may form
$C^{CDplasma}_{trans}$	Transition concentration for CD_{plasma} formation
$C^{IPDplasma}_{trans}$	Transition concentration for IPD_{plasma} formation
C_{thr}	Threshold concentration below which no domains are present in SDVAS samples screened by Permalloy
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 domain composed of ferroelectric ordered polar solvent molecules
EDA^{CDrot}	Excited or broken CDrot piece, which has an electric dipole moment and therefore is denoted electric dipole aggregate (EDA)
$EDA^{IPDplasma}$	Excited or broken IPD_{plasma} piece, which has an electric dipole moment and therefore is denoted electric dipole aggregate (EDA)
EM	Electro-magnetic
eV	Electron Volt
E	Electric field
E_s	Electric field of solute molecule
H_2O	Water molecule
Hz	Herz
IPD_{plasma}	In phase domains composed of few solvated solutes and numerous solvent molecules performing in phase plasma oscillation.
I	Electric current
IR	Infra red
j_x	Current of density in x direction
M	Molarity in mole per liter
m	Meter
mol / l	Mole per Liter
NaCl	Sodium Chloride
QED	Quantum electro-dynamics
QFE	Quasi free electrons
R_H	The Hall coefficient

Abbreviation	Explanation
SDVSAS	Serially diluted vigorously shaken aqueous solution
Supra-CD	Agglomerate of coherence domains
UV	Ultra-violet
V_H	Hall Voltage
Vis	Visible light
Σ	Hall conductance
Ψ	Quantum wavefunction describing matter
$\Psi_{CD_{elec}^{H2o}}$	Quantum wavefunction describing $CD_{elec}^{H_2o}$
$\Psi_{CD_{rot}}$	Quantum wavefunction describing CD_{rot}

Discussion with Reviewers

Reviewer

In the paper, the acronym SDVSAS refers to a process originating in the work of Samuel Hahnemann devoted to the preparation of homeopathic remedies. A dog should be named and identified as a dog and not as a barking bunch of hair (BBH). If authors are afraid of speaking frankly of "homeopathic preparations," the best is to publish on other less controversial subjects. Accordingly, other groups have taken their responsibility by publishing very good scientific papers on the science involved behind the preparation of homeopathic remedies, a subject that is in direct relationship with SDVSAS.

TA Yinnon

"SDVSAS" is just an abbreviation, as customarily in scientific papers. There are several reasons for me sticking to this custom and not denoting such liquids as homeopathic remedies.

- The study of serially diluted liquids is important for advancing basic science. For example, it enabled discovering concentration dependent phase transitions, which seems to imply a paradigmatic shift in chemical reactivity and solution structure research (Kononov *et al.*, 2002, 2015).
- Some scientists use the denotations "highly diluted" or "extremely diluted" solutions for SDVSASs. I deem it important to emphasize that the liquids are vigorously shaken.
- SDVSASs are not just used in homeopathy. SDVSASs are of importance for other technological fields, *e.g.*, wastewater treatment or power generation (Ryzhkina *et al.*, 2011; Bandyopadhyay *et al.*, 2017).
- Homeopathic remedies generally contain alcohol. As emphasized by Van Wassenhoven *et al.* (2018): "Following the European pharmacopoeia, *[…] to avoid precipitates, first dilutions of soluble MT must be prepared using the same alcohol concentration as the MT, but alcohol is not required for higher dilutions when producing homeopathic medicines. When a homeopathic medicine is made for bulk storage in a pharmacy, 62% w/w alcohol is added as a preservative, but intermediate dilutions are always prepared in pure water and then discarded. All Korsakov intermediate preparations are similarly prepared using pure water, and alcohol is added only for the final dynamisations. The homeopathic manufacturing tradition expects that the homeopathic information is carried by the water and not by the alcohol."*
- To advance knowledge on the physicochemical properties of serially diluted liquids, I deem it wisest to stick to customarily scientific methods and to publish in peer reviewed physics or chemistry journals. I trust that science-

based evidence will eventually erase the controversies around homeopathy.

Reviewer

The EZ-water concept developed by Gerald Pollack's research group (Zheng *et al*., 2006) obviously has a relationship with SDVSASs.

TA Yinnon

In my open access paper (Yinnon, 2018 p. 27), I pointed out:

(a) EZ water might be a source of CD_{rot} and $CD_{elec}^{H_2O}$ in SDVSASs prepared in vessels of glass or other hydrophilic materials.

(b) The available experimental data were insufficient for delineating the relative contribution of EZ water or that of other mechanisms to the prevalence of CD_{rot} and $CD_{elec}^{H_2O}$ in SDVSASs.

After I submitted the manuscript of the current paper, new relevant data have been published. So, I thank you for your remark, which provides me with an opportunity to readdress the issue.

Montagnier *et al*. (2009, 2011, 2015, 2017) have shown that aqueous SDVSASs, with some types of DNA as substrate, may emit 500-3000 Hz radiation. They prepared and kept the SDVSASs in Eppendorf plastic tubes. They digitally recorded the radiation, sent the record to a distant laboratory, transformed it into an analog form and sent the electric vector of the analog signal to a solenoid, which generated a magnetic field in a test tube of water. They added enzymatic proteins to the water, to allow polymerase chain reaction processes to occur. DNA molecules identical to the DNA substrate materialized in the water. They explained their results with the QED formalism of aqueous systems developed by Kurian *et al*. (2018). Recently, Tang *et al*. (2019) examined factors affecting the aforementioned processes. For example, they showed that on using *hydrophilic* quartz cuvettes instead of the *hydrophobic* plastic Eppendorf tubes, the success rate of the processes increased (a rate of 38.5% for the quartz cuvettes versus 8.7% for the plastic tubes). EZ water forms adjacent to hydrophilic membranes (Zheng *et al*., 2006), but is not known to stabilize adjacent to hydrophobic surfaces. The finding that the success rates of the processes are significantly less on using the hydrophobic tubes than on employing the hydrophilic cuvettes indicates that also in the absence of EZ water, SDVSAS have the properties discovered by the group of Montagnier. However, the results also hint that entities in EZ water (likely its CD_{rot} and/or $CD_{elec}^{H_2O}$) contribute to the aforementioned processes.

Reviewer

My concern is how energy generated by mechanical shaking can be high enough to excite water domains? As far as I understand, excitation requires electronic transitions. How can such energy be provided by shaking? And what is the cohesion energy of the coherent domain? Is mechanical shaking able to induce its breakage?

TA Yinnon

Since the water domains CD_{rot} and $CD_{elec}^{H_2O}$ are superfluidic, these are excitable by collisions with surrounding molecules (see *Appendix – paragraphs IX and iv*). When the collision energy is larger than the energy a H_2O molecule gains by joining a domain (the cohesion energy), a H_2O-molecule desorbs from the domain. When the excitation energy is smaller, the whole domain becomes excited and vortices are created. The cohesion energy of the H_2O molecules constituting $CD_{elec}^{H_2O}$ is ~0.1 eV and that of CD_{rot} is ~0.025 eV. Since the latter is of the order of that of thermal fluctuations of water at ambient conditions, CD_{rot} cannot auto-organize in bulk water. However, once these domains become stabilized by objects with sizable asymmetric charge distributions, these may persist for macroscopic times (see *Appendix - paragraphs IV and V*).

In water at ambient conditions, collisions between H_2O molecules do not excite these from their ground electronic states to their excited electronic states. In section "*Properties of* $CD^{H_2O}_{elec}$ " on page 4 is written *"These domains consist of H_2O molecules that coherently oscillate between their electronic ground state |0> and an excited electronic |b> state …. The energy difference between the |0> and |b> states is 12.07 eV."* Collisions among molecules, even induced by vigorous shaking, *cannot* excite H_2O molecules from their |0> to their |b> state. Instead, QED processes underlie the coherent oscillations between the |0> and |b> states of the molecules constituting $CD^{H_2O}_{elec}$.

According to the QED model of SDVSASs, for $C<C_{thr}$ break-up of CD_{rot} is a crucial process in the preparation of these liquids (see the main text's first paragraph in section *"Information on the substrate imprinted in SDVSASs"*). As to whether mechanical shaking of SDVSASs may break up its CD_{rot}, Yinnon *et al.* (2015b footnote g) have pointed out that the energetics of processes induced by vigorous shaking of SDVSASs have not been quantified. Therefore, these authors roughly estimated the energy available for break-up of molecular associates in an aqueous liquid after it has been shaken. They followed the approach of Raine *et al.* (1993). They noted: On stirring water in a glass vessel, more than 95% of the applied energy heats the system and dissipates to the surroundings. After stirring one liter of water for one minute, the energy available for excitation or break-up of molecular associates is of the order of a few Joule, *i.e.*, 10^{18} - 10^{19} eV. Elia *et al.* (2010) estimated that in a 45-times decimally diluted SDVSASs, about 10^{23} - 10^{24} molecules are associated. Thus, the stirring provides maximal ~10^{-4} eV per associated molecule. In other words, the stirring cannot lead to a total disintegration of CD_{rot}, but only to some local break up.

I thank the anonymous Reviewer-1 for pointing out: *"From a fluid dynamics viewpoint, the afore-mentioned approach is not convincing, as it does not take into account the Richardson-Kolmogorov cascade for energy dissipation through turbulence. Taking into account the well-known laws of turbulence shows that for manual shaking the Kolmogorov dissipation scale cannot be lower than about 10 μm (Henry, 2016)."* Henry (2016) has estimated: Vortexing water in a manual Wasserwirbler with a height of 30 cm and capacity of about one liter, the Richardson-Kolmogorov cascade enables the applied energy to split up entities with sizes larger than 21 μm; while the most efficient mechanical commercial machines, (which can vortex 20 liters of water over a height of 50 cm), enable the applied energy to split up entities with sizes larger than 343 nm.

CD_{rot} may reach dimensions of hundreds of μm. The group of Konovalov has observed molecular associates with sizes of the order of 10^{-5} m in SDVSAS with $C<<C_{thr}$, which were prepared with a lab dancer or by vortexing. The properties of these associates resemble those of CD_{rot} (Yinnon *et al.*, 2015b&c). Thus, on vortexing SDVSASs, the Richardson-Kolmogorov cascade should enable the applied energy to split up these associates. The 0.1 μm sized $CD^{H_2O}_{elec}$ are smaller than the Kolmogorov dissipation scale. However, in the QED model of SDVSASs, neither is it assumed nor concluded that $CD^{H_2O}_{elec}$ break up.