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ORIGINAL PAPER

Nanosized solvent superstructures in ultramolecular aqueous dilutions: twenty years' research using water proton NMR relaxation

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Background: **Proton Nuclear Magnetic Resonance (NMR) relaxation times T1, T2, T1/T2 are sensitive to motion and organization of water molecules. Especially, increase in T1/T2 reflects a higher degree of structuring. My purpose was to look at physical changes in water in ultrahigh aqueous dilutions.**
Methods: **Samples were prepared by iterative centesimal (c) dilution with vigorous agitation, ranging between 3c and 24c (Avogadro limit 12c). Solutes were silica–lactose, histamine, manganese–lactose. Solvents were water, NaCl 0.15 M or LiCl 0.15 M. Solvents underwent strictly similar, simultaneous dilution/agitation, for each level of dilution, as controls. NMR relaxation was studied within 0.02–20 MHz.**
Results: **No changes were observed in controls. Increasing T1 and T1/T2 were found in dilutions, which persisted beyond 9c (manganese–lactose), 10c (histamine) and 12c (silica–lactose). For silica–lactose in LiCl, continuous decrease in T2 with increase in T1/T2 within the 12c–24c range indicated growing structuring of water despite absence of the initial solute. All changes vanished after heating/cooling. These findings were interpreted in terms of nanosized (>4-nm) supramolecular structures involving water, nanobubbles and ions, if any. Additional study of low dilutions of silica–lactose revealed increased T2 and decreased T1/T2 compared to solvent, within the 10^{-3}–10^{-6} range, reflecting transient solvent destructuring. This could explain findings at high dilution.**
Conclusion: **Proton NMR relaxation demonstrated modifications of the solvent throughout the low to ultramolecular range of dilution. The findings suggested the existence of superstructures that originate stereospecifically around the solute after an initial destructuring of the solvent, developing more upon dilution and persisting beyond 12c.** *Homeopathy* (2013) **102**, 87–105.

Keywords: Proton NMR relaxation; Water; Ultrahigh dilution; Silica; Histamine; Manganese; Superstructures; Nanobubbles

Introduction

A stimulating challenge for physicists involved in research in homeopathy consists in looking for the existence of signals in water in order to explain the storage of information – the so-called "*memory of water*" – in high dilutions, up to and even beyond the Avogadro limit of molecular presence of solute. The journal *Homeopathy* has devoted a whole issue to "*The Memory of Water*" in which the problem was debated from a scientific point of view.[1] The first hypothesis proposed the existence of specific polymer structures, like 'imprints', induced in water by the stepwise dilution process under mechanical agitation.[2] Such an induced organization, if any, should be characterized by a loss of freedom of water molecules. Nuclear Magnetic Resonance (NMR) relaxation of the proton is sensitive to dynamics and structural organization of water, and hence has been widely applied for decades to the study of water in numerous kinds of solutions and macromolecular

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systems. It has been applied since 1985 to high dilutions by some teams, without definitive proof of reproducible results (they were sometimes discrepant). Most were not published in journals of physical chemistry.[3–11] From a general point of view, there are numerous hypotheses regarding high dilutions, but too little reliable experimental data. Meta-analyses in the fields of clinical, biological or physical research could demonstrate an effect of high dilutions, but most experimental studies have been assessed of insufficient quality or reproducibility.[12–15]

Homeopathic remedies are prepared through iterative centesimal dilution (1c corresponding to a 10^2-fold dilution, and *n*c to a 10^{2n}-fold dilution), so that the initial solute is virtually no longer present beyond 12c (Avogadro = 6.02×10^{23}). Proton relaxation is very slow in quasi-pure water, thus particularly sensitive to contaminants, especially the paramagnetic atmospheric oxygen; contaminants even become predominant in high dilutions. Moreover, very minor signals are expected that are hardly distinguishable from random experimental fluctuations. Rigorous protocols are therefore required for preparations and measurements and for the reproducibility of results, if any. Furthermore, I have emphasized the need for publishing in specialized journals in such a high controversial field.[16] This has been my position and explains why my works have been spread over 20 years. This voluntary restriction has led to a longer path to publication. The underlying program — a thread where each paper was intended to confirm the preceding one and ask new questions — has proceeded over years. Consequently, the links between successive papers did not appear obvious. In addition, publishing in scientific journals led to limited access by the main target — homeopaths — who may not be familiar with pure physics. Through the present paper, I provide an easier-to-read summary report of my work.[17–21] For further details and references, please refer to original papers.

I would like to thank all the collaborators who followed me in this adventure of high dilutions, mainly Drs Pascal Gries and Bernard Poitevin.

Principles of NMR relaxation

Theoretical and technical considerations

Let me briefly explain the basic phenomenon of NMR and how relaxation is related to molecular motion. In the water molecule H_2O, only protons ^1H have a spin (nuclear magnetic dipole, the moment of which is μ), so the motion of the water molecule will only be 'seen' through the motion of its protons (Figure 1a). When submitted to a magnetic field B_0, the magnetic nuclear dipole aligns itself with respect to B_0, and precesses at the Larmor frequency

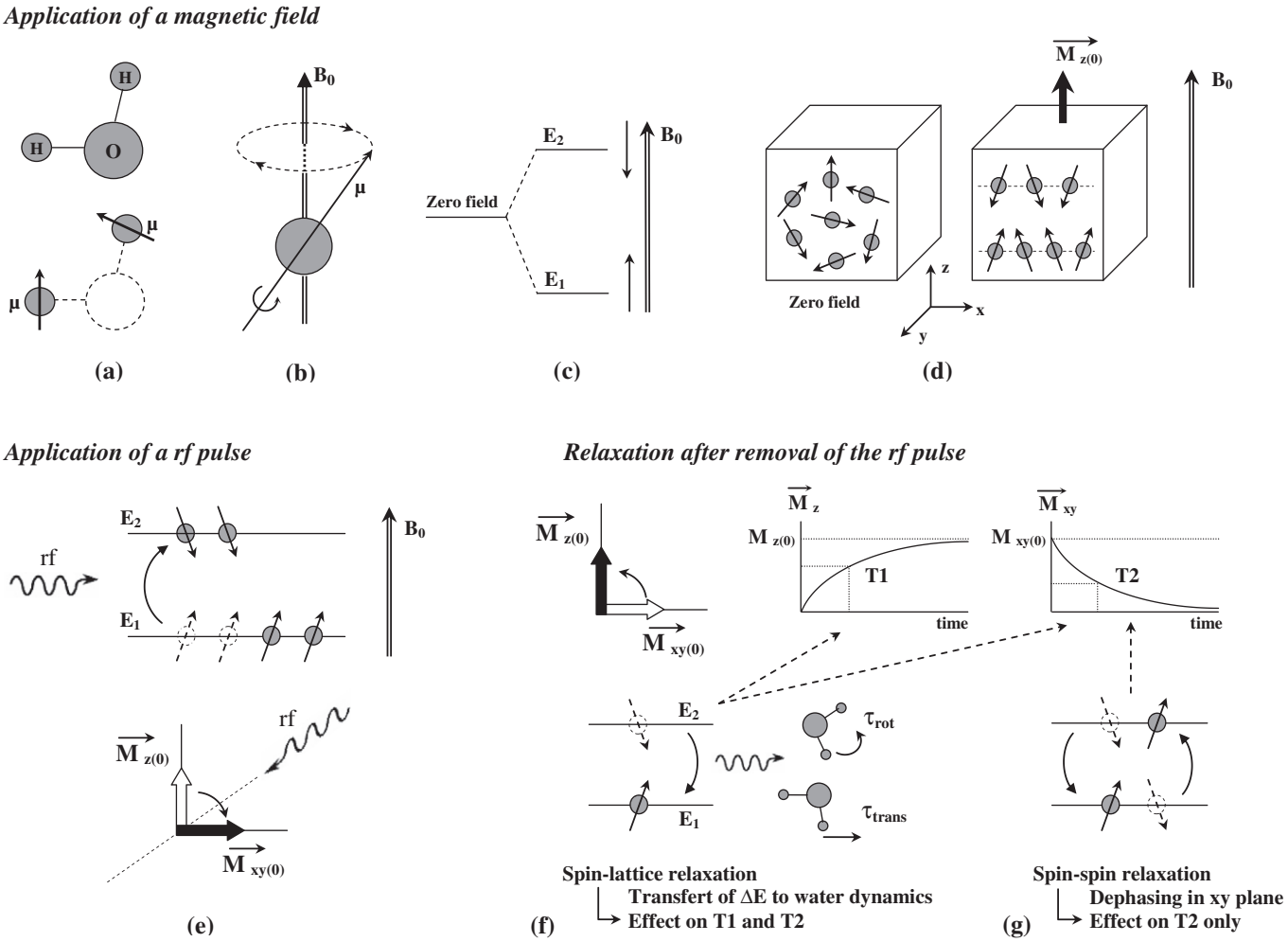

Figure 1 Principles of the NMR relaxation of the water proton (see text).