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Extended Abstract

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Remake of the non-linear dielectric effect in investigations of structure of liquids Abstract

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Structure of matter is determined by electrical interactions between molecules: dipole-dipole, dipole-ion, ion-ion interactions. Impact of an electric field on a matter allows to investigate these interactions and to understand the structure and dynamics of the investigated system. When a dielectric (in this case a liquid) is influenced by an external electric field, it undergoes polarization. For low-intensity field, the polarization is proportional to the field. However, if we increase the intensity of the electric field saturation effects could be expected. The measure of the so-called "non-linear dielectric effect" (NDE) is a nonlinear dielectric increment defined as the difference of the electric permittivity measured in a strong and in a weak electric field intensity:. According to Debye classic theory, in liquids consisted of dipolar molecules, the increment should be negative and proportional to the square of the electric field. In many liquids, these requirements are fulfilled. Interestingly, deviations from the classical behaviors are sometimes observed. This happens when a strong external electric field affects conformation of molecules, disturbs association equilibria, significant deviations are also observed in the vicinity of phase transformations. NDE experiments were popular in the last decades of the 20th century. Measurements are difficult and commercial equipment for these studies is scarce. This may explain the recent decrease of interest in this particular technique. The report will present the NDE investigations of association phenomena in alcohols and critical phenomena in the vicinity of phase transformations. The details of the NDE experiment will be also presented. I firmly believe that a proper presentation of possibilities offered by a non-linear dielectric effect will cause an increase of interest in these studies and restore their rightful place in investigations of chemistry and physics of matter. Recent Publications 1. Nowak J and Malecki J (1985) Dielectric studies of conformational equilibria in acetylphyiridines. Chem. Phys. Lett. 116(1):55-57. 2. Kosmowska M and Orzechowski K (2010) Non-linear dielectric effect and critical phenomena in a ternary mixture cyclohexane + acetonitrile + p-xylene. J. Non Cryst. Solids. 356(1):815-817. 3. Orzechowski K et al. (2014) Shift of the critical mixing temperature in strong electric fields: theory and experiment. J. Phys. Chem. B. 118(25):7187-7194.

Dielectric relaxation measurements probe how the polarization of a material responds to the application of an external electric field, providing information on structure and dynamics of the sample. In the limit of small fields and thus linear response, such experiments reveal the properties of the material in the same thermodynamic state it would have in the absence of the external field. At sufficiently high fields, reversible changes in enthalpy and entropy of the system occur even at constant temperature, and these will in turn alter the polarization responses.

The resulting nonlinear dielectric effects feature field induced suppressions (saturation) and enhancements (chemical effect) of the amplitudes, as well as time constant shifts towards faster (energy absorption) and slower (entropy reduction) dynamics. This review focuses on the effects of high electric fields that are reversible and observed at constant temperature for single component glass-forming liquids. The experimental challenges involved in nonlinear dielectric experiments, the approaches to separating and identifying the different sources of nonlinear behavior, and the current understanding of how high electric fields affect dielectric materials will be discussed. Covering studies from Debye's initial approach to the present state-of-the-art, it will be emphasized what insight can be gained from the nonlinear responses that are not available from dielectric relaxation results obtained in the linear regime.

Nonlinear dielectric effect (NDE) describes changes of dielectric permittivity induced by a strong electric field in a liquid dielectric. The most classical finding related to this magnitude is the negative sign of NDE in liquid diethyl ether (DEE), recalled by Peter Debye in his Nobel Prize lecture. This article shows that the positive sign of NDE in DEE is also possible, in the supercritical domain. Moreover, NDE on approaching the gas-liquid critical point exhibits a unique critical effect described by the critical exponent $\psi \approx 0.4$ close to critical temperature (TC) and $\psi \approx 0.6$ remote from TC. This can be linked to the emergence of the mean-field behavior in the immediate vicinity of TC, contrary to the typical pattern observed for critical phenomena.

The multi-frequency mode of NDE measurements made it possible to estimate the evolution of lifetime of critical fluctuations. The new way of data analysis made it possible to describe the critical effect without knowledge of the non-critical background contribution in prior.

Bottom Note: *This work is partly presented at 5th International Conference on Physics*