Nearly Constant Loss Spectra in Glasses: Dipolar Interaction Effects

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The high-frequency/low-temperature dielectric response of many disordered materials generally is characterized by a nearly frequency-independent imaginary part of the dielectric susceptibility, $\chi''(\omega)$. In contrast to dc- and low-frequency ac-transport, which show thermal activation, this so-called "nearly constant loss" (NCL)-behavior is non-activated and displays an only weak temperature-dependence [1]. Experimental observations of this phenomenon include ion-conducting glasses, highly-defective ionic crystalline materials, polaron conductors protonic, conductors as well as highly-viscous melts of organic and inorganic glasses. While there is general agreement that the origin of the NCL-response lies in local, nonhopping motions of charged defects, the identification of a microscopic mechanism is an open question.

After a brief review of existing ideas for the interpretation of constant loss, we focus in this work on a microscopic many-body model that describes relaxation of charged defect centers located at random positions in the material [2, 3]. With each defect we associate an electric dipole moment that can undergo stochastic orientational moves. The probability of a selected dipole to reorient is governed by the instantaneous local electric field produced by other dipoles. The basic ingredients of the model therefore are electrostatic interactions in connection with positional disorder. At low temperatures it turns out that clusters of defects with small relative distances get nearly frozen and hence give rise to slow relaxation. Both from kinetic Monte Carlo simulation of a discretized system, the "random dipolar lattice gas", and from analytical calculations based on scaling arguments we demonstrate the emergence of NCL-type spectra with associated cut-off frequencies that can be related to experiment. An example of a simulated loss spectrum $\chi''(\omega)$ is shown in Fig. 1.

In both approaches the collective nature of the underlying relaxational mode spectrum is emphasized. This is in clear distinction to the familiar "asymmetric double well potential" (ADWP)-model, frequently used in the literature to represent an NCL-spectrum in terms of static distribution functions for local barriers and asymmetry parameters.

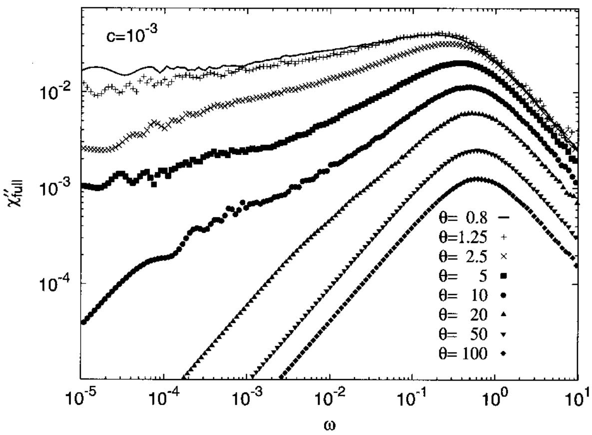

Figure 1: Double-logarithmic plot of the dielectric loss spectrum χ'', obtained from kinetic Monte Carlo simulation of a dipolar lattice gas. The concentration of defects is $c = 10^{-3}$, and $\theta = k_B T/V_{\rm dip}$ is the temperature normalized by the dipole-dipole interaction parameter. A gradual transition from Debeye-like to NCL-behavior is clearly observed when temperature is lowered.

References

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