Nonlinear dielectric response of low-energy excitations in glasses

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Abstract

The nonlinear dielectric response of low-energy excitations in glasses is investigated on the basis of the two-level tunneling model. It appears that the tunneling model has sufficient flexibility to explain the low-temperature dielectric response of glasses, at least qualitatively. An explicit analysis of experimental data obtained on borosilicate glasses leads to a low energy cutoff $\Delta_{0\min}/k \approx 3$ mK in the density of tunneling states.

Key words: glasses; tunneling states; dielectric response

The thirty years since Zeller and Pohl [1] first presented data showing the anomalous low-temperature behavior of glasses have seen considerable theoretical and experimental activities. Most of these anomalous properties have been successfully explained by means of the standard tunneling model [2,3]. In this model localized elementary excitations are postulated to be present in amorphous materials and phenomenologically described by non-interacting tunneling systems (TS). Such a TS can be represented approximately by a particle in an asymmetric double-well potential. In the pseudospin representation with the Pauli matrices $\boldsymbol{\sigma}^x$ and $\boldsymbol{\sigma}^z$ the Hamiltonian of an isolated TS is given by $\boldsymbol{H}_0 = (1/2)(\Delta\boldsymbol{\sigma}^z - \Delta_0\boldsymbol{\sigma}^x)$, where Δ and Δ_0 are the asymmetry energy and the tunneling matrix element, respectively. The tunneling model (TM) states a broad distribution of both parameters according to $P(\Delta, \Delta_0) = \bar{P}/\Delta_0$, where $\bar{P}$ is a constant.

The behavior of a TS is defined by specifying the complex amplitudes, $a_+(t)$ and $a_-(t)$, of the eigenfunctions, $\varphi_+(x)$ and $\varphi_-(x)$. The time-dependent wavefunction takes the form

$$\Psi(x,t) = \sum_{\alpha=+,-} a_\alpha(t)\varphi_\alpha(x)\exp(-iE_\alpha t/\hbar), \qquad (1)$$

where E_+ and E_- are the energies of states φ_+ and φ_-.

Variations of $a_+(x)$ and $a_-(x)$ are determined through the time-dependent Schrödinger equation by interaction with external fields. In particular, when a uniform electric field, $F(t) = F_0\cos\omega t$, is applied parallel to x, the interaction, $H_{\rm int} = -qxF(t)$, leads to a change of the amplitudes in accordance with

$$\dot{a}_\mp = {\rm i}\frac{F(t)}{\hbar}\left((\pm p_- a_\mp + p_\mp a_\pm \exp(\mp {\rm i}tE/\hbar)\right), \qquad (2)$$

in terms of the characteristic dipole moments $p_- = -p_+ = \langle\varphi_- \mid qx \mid \varphi_-\rangle$ and $p_\mp = p_\pm = \langle\varphi_- \mid qx \mid \varphi_+\rangle$.

The average dipole moment is given by

$$\langle p(t)\rangle = q\langle x\rangle = p_-\left(A_-^*A_- - A_+^*A_+\right) + p_\mp\left(A_+^*A_- + A_-^*A_+\right), \qquad (3)$$

in which $A_\pm = a_\pm\exp\left(-{\rm i}E_\pm t/\hbar\right)$ and $E = E_+ - E_-$.

Thus, the resonant dielectric response of TS to an electric field has to be treated by solving the coupled equations (2). The additional incoherent interaction with thermal phonons can be included by introducing both the relaxation time τ_1 for the population difference, $A_-^*A_- - A_+^*A_+$, and the dephasing time τ_2 for the off-diagonal coherence between φ_+ and φ_-. Formal methods for solving this problem are familiar from the dynamics of a spin-1/2 particle in a magnetic field, described by the Bloch equations, and can be profitably applied to TS in glasses [4,5].

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By solving the linearized Bloch equations for an oscillating electric field and integration of the TS response over the parameter distribution, $P(\Delta, \Delta_0)$, the linear dielectric response of the TS in a glass can be calculated and compared to experimental results.

Fig. 1 shows the measured temperature dependence of the real part of the permittivity $\Delta\epsilon(T)/\epsilon(T_0) = [\epsilon(T) - \epsilon(T_0)]/\epsilon(T_0)$ for a borosilicate glass sample.

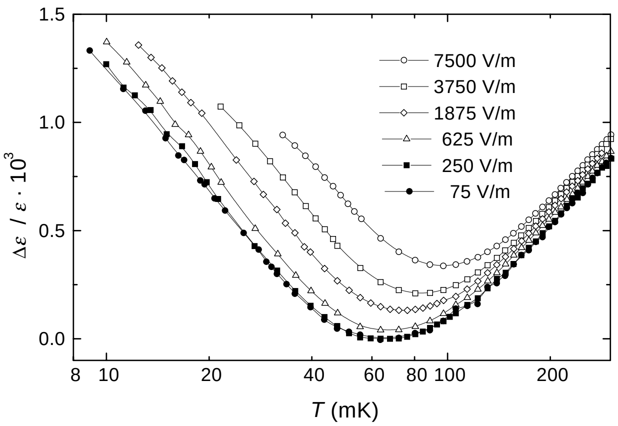

Fig. 1. Permittivity for a borosilicate glass (BK 7) as function of temperature measured at 1 kHz for several excitations.

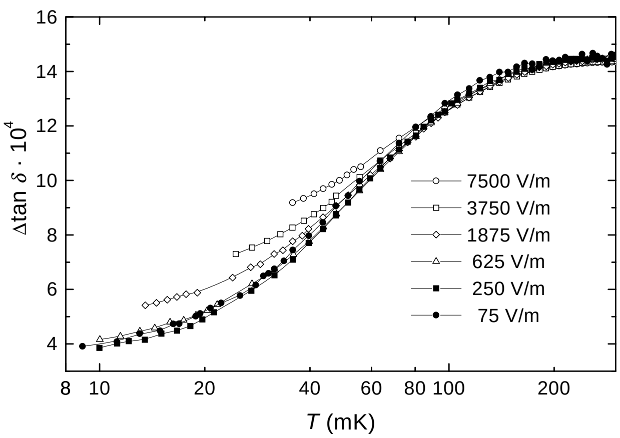

Fig. 2. Loss angle for a borosilicate glass (BK 7) as function of temperature measured at 1 kHz for several excitations.

The data show a linear behavior only for small amplitudes of excitation ($F_0 < 250\ \mathrm{Vm}^{-1}$). In qualitative agreement with the TM in the linear response approximation the permittivity decreases logarithmically with decreasing temperature, passes a minimum at T_0, and increases logarithmically again. The low-temperature logarithmic variation is caused by the resonant response of coherently driven TS in the low frequency limit, and is given by

$$\frac{\Delta\epsilon}{\epsilon} = \frac{\bar{P}p_0^2}{3\epsilon_0\epsilon} \int_{\Delta_{0\min}}^{\infty} \frac{\mathrm{d}E}{E} \tanh\frac{E}{2kT} \sqrt{1 - \left(\frac{\Delta_{0\min}}{E}\right)^2}. \quad (4)$$

The scale factor $\bar{P}p_0^2$ and the cutoff energy $\Delta_{0\min}/k$ were determined to be $1.28{\cdot}10^{-13}\mathrm{C}^2\mathrm{J}^{-1}\mathrm{m}^{-1}$ and 3 mK, respectively. The high value of the energy cutoff $\Delta_{0\min}$, which was also found in dielectric measurements on other glasses [6], describes the resonant and the relaxational part of the linear dielectric response including the observed ratio of their slopes (near -1:1) quite well.

The data of the dielectric loss, $\Delta\tan\delta(T) = \tan\delta(T) - \tan\delta(10\mathrm{mK})$, shown in Fig. 2, can be also described by the TM in the linear response regime. Both the scale factor given by the plateau value, $\pi\bar{P}p_0^2/6\epsilon_0\epsilon$, and the energy cutoff correspond exactly to that values determined by fitting (4) to the permittivity data.

It is obvious that the observed excitation dependencies of ϵ and $\tan\delta$ for amplitudes $F_0 > 250\ \mathrm{Vm}^{-1}$ cannot be explained in a linear response theory. The dependence of the dielectric response on the amplitude of the ac field has been treated theoretically by Stockburger *et al.* [7]. In contrast to the observations presented in Fig. 1, however, their model predicts a change of the $\log(1/T)$-slope and a field independent plateau at very low temperature.

In order to account for the observed excitation dependence of the dielectric response, we solve numerically the nonlinear Bloch equations and integrate the single TS response over the parameter distribution $\bar{P}/\Delta_0$, and over the dipole orientation angle. In this way, the observed dependence of the dielectric response on the amplitude of the ac field can be qualitatively described.

The interpretation of the nonlinear dielectric response in the framework of the TM leads to a low energy cutoff $\Delta_{0\min}/k \approx 3$ mK, which is due to the approximation of a constant density of state in the TM, and should not be microscopically related to the largest tunneling barriers present in glasses.

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