

Spectrum of low-energy (2–10 meV) vibrational excitations of glasses in a disclination model

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It is shown that vibrations localized on linear topological defects in glasses can cause the observed density of vibrational states (DVS), excessive in comparison to the Debye density, in the 2–10 meV energy range. In this approach, the spectra of DVS and of inelastic light scattering by vibrations are obtained in accordance with recent experimental results.

The vibrational spectra of crystals and glasses differ most drastically in the region of low energies $E < 8\text{--}10$ meV. In glasses at 2–10 meV, the density of vibrational states (DVS) exceeds the Debye density (calculated from experimentally determined sound velocity) severalfold.¹ This characteristic is manifested in an excessive heat capacity and plateau in thermal conductivity at $T \sim 10\text{--}30$ K, in spectra of IR absorption and inelastic light scattering.²

In the present article we propose a model for the vibrational excitations which are responsible for these anomalies.

It has recently been observed that the spectra of excess DVS, as well as the corresponding spectrum of inelastic light scattering, have a universal shape in a whole series of different glasses.^{3,4} This attests to the universal nature of the structural elements responsible for the anomalies discussed.

As will be shown here, the neutron and Raman spectra of glasses in the low-energy region are satisfactorily described on the assumption that additional vibrational excitations are localized on linear objects of size $L \sim 30 \text{ \AA}$ which are enclosed in a loop. The presence of such topological structural defects is the basis for a disclination model of glass,⁵ where they are associated with the line that passes through five- or seven-membered rings of bonds. It is well known that such lines (odd lines), which are

genetically related to disclinations, cannot break inside glass, and either form loops, or come to the surface.⁶ The elastic properties of the material near a disclination loop should obviously differ markedly from the average values, and this serves as the basis for the appearance of vibrations localized on the loop. We note that the intrinsic vibrational modes of a 2D defect—the surface of microcrystals 30–60 Å in diameter, grown within a glass matrix of the same or different composition are clearly visible in inelastic light scattering spectra.^{7,8}

As the initial model, we shall consider an ensemble of randomly arranged elastic loops of length $L \gg a$ (a is the characteristic atomic spacing). We shall find the DVS of the excitations localized on loops. The DVS of d -dimensional vibrations in the Debye approximation has the form $g(E) \propto E^{d-1}$. For 1D objects this constant in our case is

$$g_1^0 = \alpha/a^3 E_D \quad (1)$$

where E_D is the Debye energy, and α is the fraction of atoms that participate in the vibrations localized on loops. From the experimental data on DVS^{3,4,9} it follows that the integrally excessive DVS in the low-energy region amounts to 10–15% of the total DVS, i.e., $\alpha \sim 0.1$ – 0.2 . A characteristic and one of the most important features of the model in question is the fact that the vibrational spectrum of the loops is cut off from below by energy E_0 , where $E_0 \sim 2\pi v/L$, and v is the velocity of sound. Thus, the 1D DVS which we discussed is described by the step function $g_1(E) = g_1^0 \theta(E - E_0)$, which must be averaged with the Gauss distribution function for each mode in order to allow for the size and shape dispersion of the loops (the discreteness of the lowest-lying modes becomes unimportant):

$$\bar{g}_1(E) = \frac{1}{\sqrt{2\pi\Delta^2}} \int_{E_0}^{\infty} dE g_1^0 \exp \left[-\frac{(E - E_0)^2}{2\Delta^2} \right] \quad (2)$$

The function $\bar{g}_1(E)$ is shown in Fig. 1. The estimate for the dispersion $\Delta \sim E_0 \sqrt{L/a}$ is found to be close to the value $\Delta \approx \frac{1}{3} E_0$, obtained from the condition of best fit of the experimental curves. Since $\bar{g}_1(E)$ is ensured by only a small fraction ($\alpha \sim 0.1$) of the

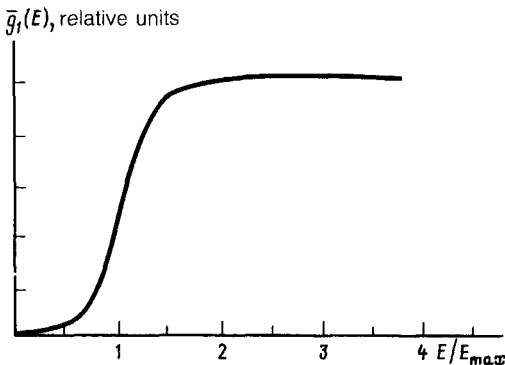


FIG. 1. Averaged DVS for 1D defects ($\Delta/E_0 = 1/3$).

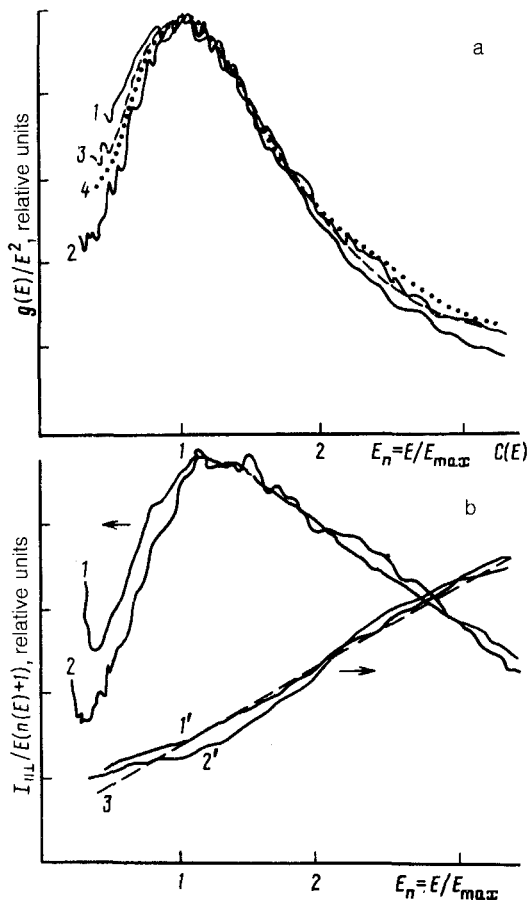


FIG. 2. (a) Ratio of low-energy DVS to Debye density on the energy scale E/E_{\max} . The amplitude of the peak is normalized to unity. 1, 2, 3—Experimental spectra for vitreous As_2S_3 , SiO_2 , and $\text{Mg}_{70}\text{Zn}_{30}$, respectively; 4—theoretical curve. (b) Low-energy spectra of DVS of the glasses As_2S_3 (1) and SiO_2 (2); dependence of $C(E)$ for As_2S_3 (1') and SiO_2 (2').⁴ Straight line—The linear dependence $C(E) \propto E$.

total number of atoms, as a first approximation, it enters additively into the total DVS: $g(E) = \bar{g}_1(E) + g_3(E)$, where $g_3(E) = 3E^2/E_D^3$ is the 3D Debye DVS. Figure 2a shows good agreement of this DVS model with the experimental curves.

We shall now turn to the data on inelastic light scattering from the vibrations we are considering. In the low-energy region the scattering intensity $I(E)$ shows a peak (so-called boson peak) which reflects the peak in DVS.² According to the Shuker-Gammon model,¹⁰ the light scattering intensity in glasses is given by

$$\frac{EI(E)}{n(E)+1} \approx C(E)g(E),$$

where $n+1$ is the Bose factor, and $C(E)$ is the square of the matrix element of the

interaction of light with vibrations. For scattering by phonons, $C(E) \propto E^2$ (Ref. 2). In the region of the boson peak, according to recent experimental results,⁴ $C(E) \propto E$ (Fig. 2b).

For light scattering by a vibration localized on a d -dimensional defect, the estimate of the metric element is given by

$$M = \int P_{ikjl} \epsilon_i \epsilon'_k e^{i\mathbf{k}\mathbf{r}} \nabla_j \psi_l(\mathbf{r}) e^{-i\mathbf{k}'\mathbf{r}} d^d r, \quad (3)$$

where P_{ikjl} are the elasto-optical contacts for the vibrations discussed, $\hat{\epsilon}, \mathbf{k}$ and ϵ', \mathbf{k}' are the vectors of polarization and momentum of the incident and scattered light waves, respectively, and ψ_l is the vibrational excitation wave function. In Eq. (3), $|k - k'|L \sim L/\lambda$ light and $\sqrt{\nabla} \psi \sim 1/L \psi$, where L is the characteristic size of the defect. Considering the normalization of ψ , $\int d^d r |\psi_l|^2 = 0$, so that $\psi_l \propto L^{-d/2}$. As a result, we obtain the estimate

$$M \propto \int d^d r \frac{1}{L} \psi \propto L^{\frac{d}{2} - 1} \quad (4)$$

Since the lower vibrational modes have energy $E \sim 2\pi\nu/L$, then $M \propto E^{1-d/2}$, and for incoherent vibrations, averaging over the volume, we obtain $\overline{|M|^2} \propto E^{2-d}$. The 1D case $d=1$ gives the experimentally observed dependence $C(E) \propto \overline{|M|^2} \propto E$ (Fig. 2b).

The model discussed thus correctly reproduces the position, amplitude, and spectral shape of the low-energy DVS in glasses, as well as the corresponding Raman spectrum.

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