

# Universal behavior of the vibrational state density in glasses

M. G. Zemlyanov, V. K. Malinovskii, V. N. Novikov, P. P. Parshin,  
and A. P. Sokolov

*Institute of Automation and Electronics, Siberian Branch of the Academy of Sciences of the USSR, Novosibirsk*

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The vibrational state density of glassy materials of different chemical compositions was determined by the method of inelastic scattering of cold neutrons. In the low-energy region of the spectrum the behavior of the vibrational state density is found to be universal, though not of the Debye type, in various glassy materials.

One of the important unsolved problems in the physics of the amorphous state is the origin of the excess density (compared with the Debye density) of the vibrational states in the energy range  $E \sim 2\text{--}10$  meV. This density gives rise to excess heat capacity at  $T \approx 10\text{--}30$  K (Ref. 1) and to the appearance of a boson peak in the low-frequency Raman scattering of light.<sup>1,2</sup> It is also seen in neutron scattering spectra<sup>3,4</sup> and IR spectra.<sup>5</sup>

We used the neutron scattering method to study the vibrational state density in

glasses of various chemical compositions and various short-range orders. We discovered that the low-energy (2–10 meV) vibrational state density is universal in nature in various glassy materials.

The measurements were carried out at room temperature using a time-of-flight spectrometer with a cold neutron source. The results were analyzed in the usual way, with allowance for all the necessary corrections. The results of a neutron experiment for diatomic systems can be used to directly reconstruct the so-called generalized vibrational spectrum  $\Theta(E)$  which is related to the standard vibrational state density  $g(E)$  by the relation

$$\Theta(E) = \sum_{i=1,2} (\sigma_i c_i M_i^{-1} |e_i(E)|^2) g(E).$$

Here  $\sigma_i$ ,  $M_i$ ,  $c_i$ , and  $e_i(E)$  are the scattering cross section, mass, concentration, and the vibration vector of the  $i$ th component.

Figure 1 shows the results of the measurements of  $\Theta(E)/E^2$  for  $\text{As}_2\text{S}_3$  and  $\text{SiO}_2$  in the crystallographic and glassy states. In the state density of the crystal samples at low  $E$  we see a large region with a Debye behavior  $\Theta(E) \propto E^2$ . At  $E \gtrsim 7$  meV the spectra of the test crystals become clearly distinguishable. In glassy  $\text{SiO}_2$  and  $\text{As}_2\text{S}_3$  a Debye behavior is not seen even at the lowest energies that have been reached, 1–2 meV. These spectra, however, have a common feature: an asymmetric peak which lies at  $E_{\text{max}} \approx 5.1$  and 2.65 meV for  $\text{SiO}_2$  and  $\text{As}_2\text{S}_3$ , respectively. We have compared the spectra at the energy scale  $E_n = E/E_{\text{max}}$ , assuming  $E_{\text{max}}$  to be the energy unit (Fig. 2). Also shown in this figure are the data for metallic glass  $\text{Mg}_{70}\text{Zn}_{30}$  which were obtained previously.<sup>4</sup> As can be seen in Fig. 2, the shape of the peak in these materials is identical. Consequently, despite substantial difference in the chemical composition, in the short-range order, and in the electronic properties of the three glassy materials, the low-energy vibrational state density in them has a universal, though not a Debye, form. The result which we obtained shows that these anomalies stem from the universal laws governing the behavior of glassy materials in the structure.

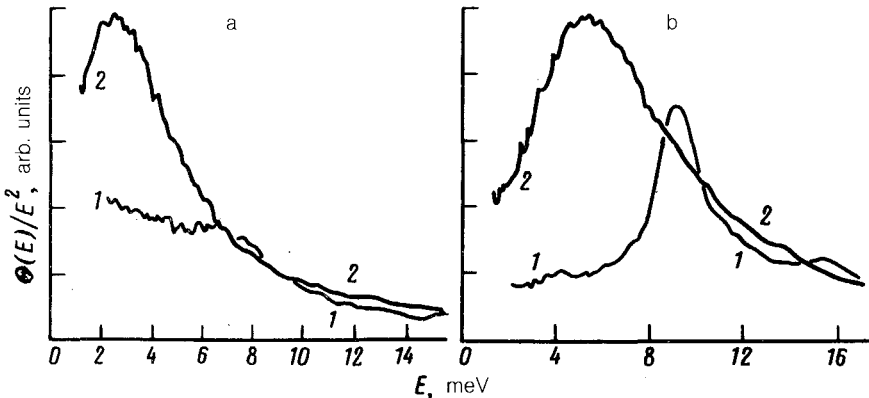


FIG. 1. The energy dependence  $\Theta(E)/E^2$ . (a) Crystalline  $\text{As}_2\text{S}_3$  (1) and glassy  $\text{As}_2\text{S}_3$  (2); (b) crystalline  $\text{SiO}_2$  (1) and glassy  $\text{SiO}_2$  (2).

Such a universality is also seen in the Raman spectra of glass. According to Malinovsky *et al.*,<sup>6</sup> the intensity of scattered light in amorphous substances is  $IE/(n+1) = C(E)g(E)$ , where  $C(E)$  characterizes the relationship between the light and the vibrational excitation, and  $n+1$  is the Bose factor. The excess  $g(E)$  gives rise to the appearance of the so-called boson peak in the scattered-light spectra.<sup>2</sup> It was shown in Refs. 2 and 6 that the shape of the boson peak in the coordinates normalized to the frequency of the maximum is identical in many oxide, chalcogenide, and low-molecular organic glasses (Fig. 2b). Taking into account the data of our study, we assume that the universality also applies to the vibrational state density of these materials.

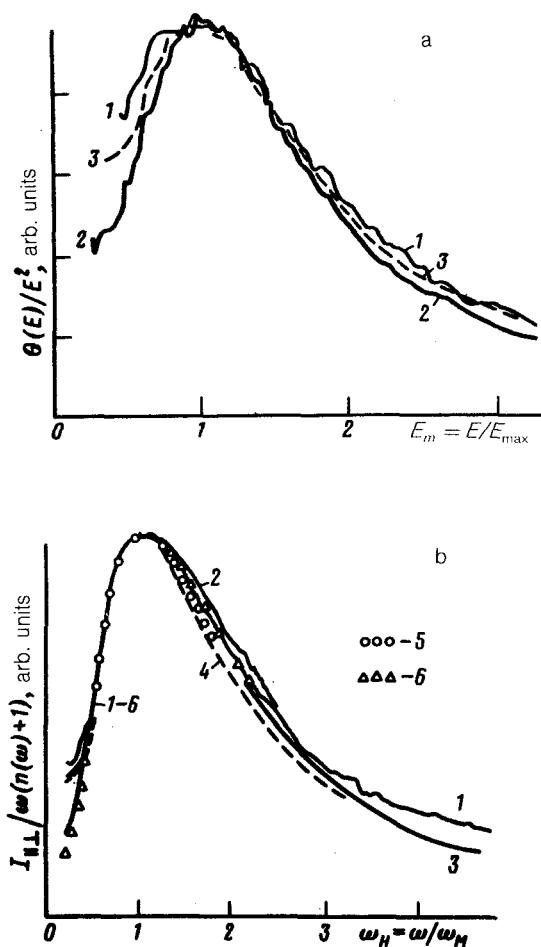


FIG. 2. Low-energy vibrational spectra of glassy materials at the scale normalized to  $E_{max}$  (a) 1— $\Theta(E)/E^2$  of  $As_2S_3$  ( $E_{max} = 2.65$  meV); 2— $SiO_2$  (5.1 meV); 3—metallic glass  $Mg_{70}Zn_{30}$  (5.5 meV); (b) 1—normalized intensity of the Raman spectrum of  $As_2S_3$  ( $\omega_m = 26$   $cm^{-1}$ ); 2— $Bi_4Si_3O_{12}$  (34  $cm^{-1}$ ); 3— $SiO_2$  (52  $cm^{-1}$ ); 4— $B_2O_3$  (28  $cm^{-1}$ ); 5— $C_3H_5(OH)_3$  (42  $cm^{-1}$ ); 6— $B_2O_3 \cdot 0.7Li_2O$  (88  $cm^{-1}$ ).

Several different models were proposed to describe the low-energy excitations in glasses and amorphous solids. In Refs. 3 and 7 these excitations are linked with the vibrations of some structural fragments characteristic of samples with a particular chemical composition, for example, the rotation of a group of  $\text{SiO}_4$  tetrahedra in the case of  $\text{SiO}_2$  or the vibrations of the residues of a layered structure in the case of chalcogenide glasses. It would be difficult to reconcile these models with the general behavior of the vibrational state density observed in our study.

The weak-potential model,<sup>9</sup> the fractal models,<sup>10</sup> and the models<sup>2,11</sup> with structural correlations at the level of 10–20 Å apparently share the required common features. A discussion of whether these models are in agreement with our results is, however, beyond the scope of this short letter.

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