Study of the spectral distribution of vibrational states in metallic glasses $Mg_{70}Zn_{30}$ and $Cu_{33}Zr_{67}$

M. G. Zemlyanov, G. F. Syrykh, and N. A. Chernoplekov
I. V. Kurchatov Institute of Atomic Energy

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The generalized distribution functions of the oscillation frequencies $\Theta(\epsilon)$ are measured by the method of inelastic scattering of cold neutrons by metallic-glass samples $Mg_{70}Zn_{30}$ and $Cu_{33}Zr_{67}$ and by their single-phase crystalline analogs. In contradiction of the results of Refs. 1 and 2 [Suck *et al.*, J. Phys. C: Solid State Phys. **13**, L167 (1980); J. Phys. C: Solid State Phys. **14**, 2305 (1981)], it is shown that the oscillation spectrum of the amorphous state has an extended region in which $\Theta(\epsilon) \sim \epsilon^2$.

Suck *et al.*[1,2] have studied by making use of inelastic scattering of thermal neutrons the deformation of the oscillation spectra of $Mg_{70}Zn_{30}$ and $Cu_{46}Zr_{54}$ alloys during their transition to the metallic-glass state. Aside from observing an overall blurring of the structural features of the oscillation spectra of the crystals and an increase in the state density of their low-energy part, they also noticed that the Debye region ($\sim \epsilon^2$) was missing in the energy interval $4 \leqslant \epsilon \leqslant 7$ meV. For the metallic glass $Mg_{10}Zn_{30}$, the exponent turned out to be 1.75, while for $Cu_{46}Zr_{54}$ it was 1.33. At the same time, the vibrational-state density of polycrystals followed the Debye ϵ^2 law in the indicated energy range.

To obtain more accurate data on the change in the vibrational-state density at low energies during the transition from the crystal state to the metallic-glass state, we have carried out an additional measurement of inelastic scattering of neutrons by samples of $Mg_{70}Zn_{30}$ and $Cu_{33}Zr_{67}$ alloys in the glassy state and single-phase crystal state (in the case of $Mg_{70}Zn_{30}$ the single-phase crystal state is metastable).

The measurements were carried out at room temperature using a time-of-flight spectrometer with a cold-neutron source.[3] A standard procedure was used to analyze the data, with allowance for all the necessary corrections. By using in the spectrometer[3] a primary neutron line with an energy 15 times lower than that in Refs. 1 and 2 we were able to not only lower the lower boundary of the spectrum under study but also to eliminate the effect of coherent neutron scattering in the study of the low-energy part of the spectrum, which is of greatest interest. A valid comparison of the vibrational spectra of metallic glasses and crystals can thus be made by using single-phase crystal analogs of the glasses studied and by eliminating the coherent effects. We should keep in mind that the so-called generalized oscillation spectrum $\Theta(\epsilon)$, which is related to the standard vibrational-state density $g(\epsilon)$, can be reconstructed directly from a neutron experiment for diatomic systems[4]

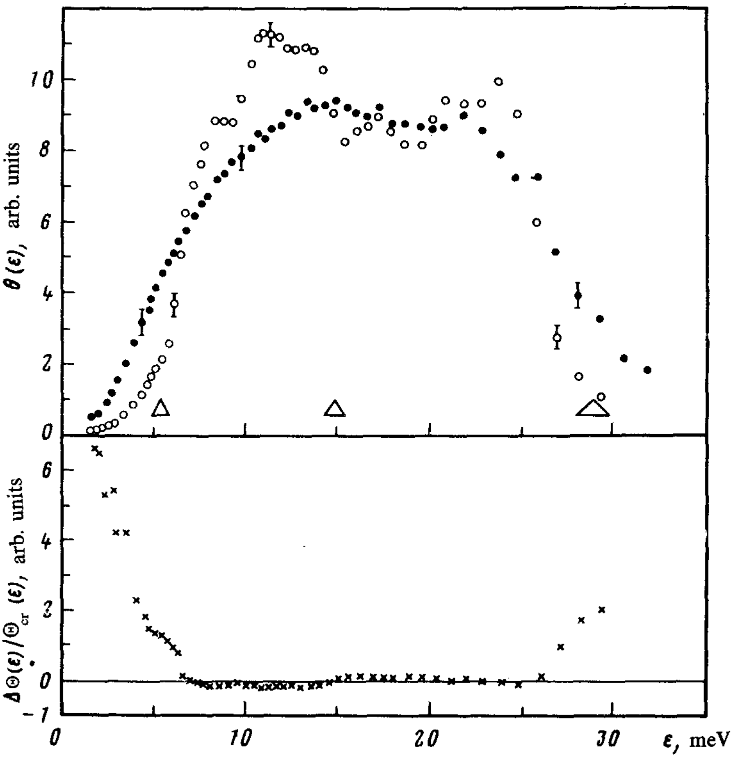

FIG. 1. Generalized frequency-distribution functions $\Theta(\epsilon)$ for $Cu_{33}Zr_{67}$ in the crystalline and amorphous states. O—$\Theta_{cr}(\epsilon)$; ●—$\Theta_{am}(\epsilon)$; + —$\Delta\Theta(\epsilon)/\Theta_{cr}(\epsilon)$. The triangles indicate the resolution of the spectrometer.

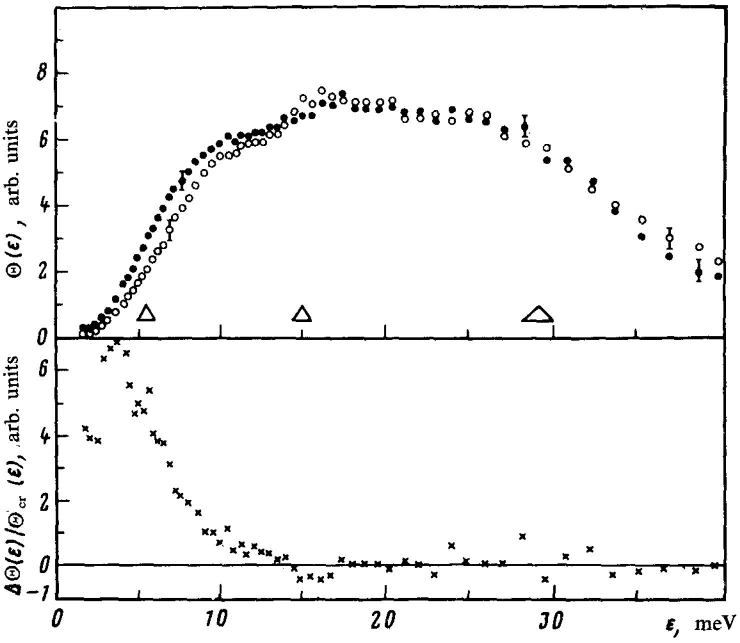

FIG. 2. Generalized frequency-distribution functions $\Theta(\epsilon)$ for $Mg_{70}Zn_{30}$ in the crystalline and amorphous states. ○—$\Theta_{cr}(\epsilon)$; ●—$\Theta_{am}(\epsilon)$; +—$\Delta\Theta(\epsilon)/\Theta_{cr}(\epsilon)$. Triangles—resolution of the spectrometer.

$$\Theta(\epsilon) = \sum_{i}^{2} \sigma_i c_i M_i^{-1} | \mathbf{e}_i(\epsilon) |^2 g(\epsilon).$$

Here σ_i, M_i, c_i, and $\mathbf{e}_i(\epsilon)$ are respectively the scattering cross section, mass, density, and the oscillation vector of the i-th component.

The generalized oscillation spectra $\Theta(\epsilon)$ reconstructed from the data on inelastic scattering of cold neutrons and the relative deformation of these spectra produced as a result of the transition of the alloys from the crystal state to the amorphous state, $\Delta\Theta(\epsilon)/\Theta_{cr}(\epsilon) = [\Theta_{am}(\epsilon) - \Theta_{cr}(\epsilon)]\Theta_{cr}^{-1}(\epsilon)$, are shown in Figs. 1 and 2. It follows from Refs. 1 and 2 that the rendering of the crystals amorphous blurs the structural features of the crystal spectra and increases the state density at low energies. The boundary of the spectrum of $Cu_{33}Zr_{67}$ is displaced toward higher energies and the boundary of the spectrum of $Mg_{70}Zn_{30}$ remains essentially unchanged. The energy dependence of the relative deformation of the spectra, $\Delta\Theta(\epsilon)/\Theta_{cr}(\epsilon)$, should be singled out. As was pointed out by Panova *et al.*,[5] the low-energy part of this energy dependence resembles the deformation of the vibrational spectrum of a crystal consisting of light atoms, caused by the introduction into it of heavy impurities. At this stage of research, it is difficult to determine the extent to which this low-energy peculiarity corresponds to that predicted by Karpov and Parshin[6] for amorphous systems on the basis of a model which takes into account the special role of the anharmonicity of local atomic potentials under conditions of fluctuation of the structure parameters. The model used by Kar-

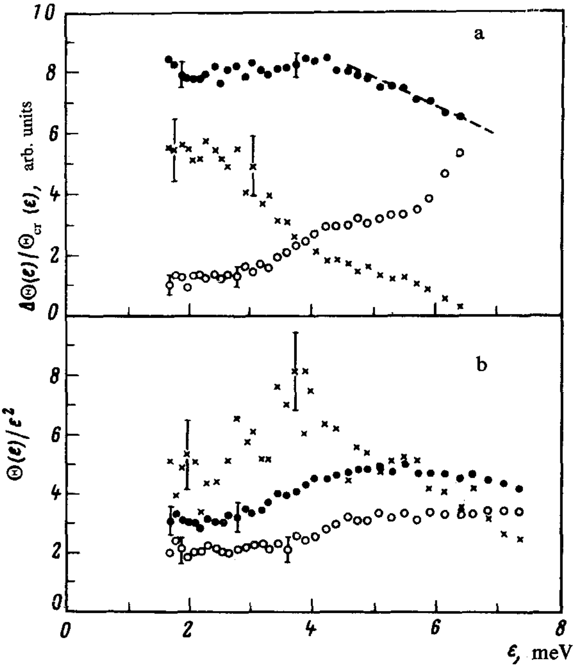

FIG. 3. Energy dependence of the ratios $\Theta(\epsilon)/\epsilon^2$ and $\Delta\Theta(\epsilon)/\Theta_{cr}(\epsilon)$. a—$Cu_{33}Zr_{67}$, ○—$\Theta_{cr}(\epsilon)/\epsilon^2$; ●—$\Theta_{am}(\epsilon)/\epsilon^2$; +—$\Delta\Theta(\epsilon)/\Theta_{cr}(\epsilon)$; - - - —data taken from Ref. 1; b—$Mg_{70}Zn_{30}$, ○—$\Theta_{cr}(\epsilon)/\epsilon^2$; ●—$\Theta_{am}(\epsilon)/\epsilon^2$; +—$\Delta\Theta(\epsilon)/\Theta_{cr}(\epsilon)$.

pov and Parshin[6] does not seem to reflect the actual situation well enough to describe the full range of low-frequency oscillations in an amorphous system.

Figure 3 shows a more detailed plot of the low-energy part of the ratios $\Theta(\epsilon)/\epsilon^2$ and $\Delta\Theta(\epsilon)/\Theta_{cr}(\epsilon)$ for both systems in the crystalline and amorphous states. Sufficiently extended regions, in which $\Theta(\epsilon)\sim\epsilon^2$, can be seen for both the metallic glasses and their crystal analogs. In the case of $Cu_{33}Zr_{67}$, this region is larger for the amorphous state than for the crystalline state and in the case of $Mg_{70}Zn_{30}$ the opposite situation occurs. We can infer from this that the inaccurate conclusion of Suck *et al.*,[1,2] that there are no $\Theta(\epsilon)\sim\epsilon^2$ regions in metallic glasses stems from the incorrect extrapolation of the results obtained by them to the energy region below 5 meV and also (possibly) from the invalid comparison of the data on $\Theta(\epsilon)$ for metallic glasses with the data on their stable crystalline states, which constitute the two-phase systems.

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