Variation in the partial densities of Ni and B atom vibrational states with amorphization of Ni_2B

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The variation in the partial densities of the vibrational states of the Ni_2B system in a transition from the crystalline to the amorphous state was investigated. The inelastic neutron-scattering technique was used. The measurements were carried out on samples of two isotope compositions, $Ni_2^{nat}B^{11}$ and $Ni_2{}^{60}B^{11}$. It is determined that a significant portion of the vibrational state density of the light metalloid atoms is redistributed from the high-energy region to the low-energy region upon amorphization.

INTRODUCTION

The transition of matter from the crystalline to the amorphous state is accompanied by a significant change in its physical properties. As a result obtaining microscopic information on the spatial configuration of the atoms and on the behavior of the phonon and electron subsystems in such a transition is of indisputable interest.

The most reliable and complete information on the spectrum of thermal excitations of amorphous substances can be obtained by measuring inelastic neutron scattering. To date this technique has been used to investigate the thermal excitation spectra for a variety of systems (metal-metal, metal-metalloid) in the amorphous state (see, for example, Refs. 1-5). Since virtually all amorphous systems are multicomponent systems, the measured doubly-differential neutron scattering cross section is determined by the following expression[6]:

$$\frac{d^2\sigma}{d\Omega\, dE}=\frac{\mathbf{k}}{\mathbf{k}_0}\frac{Q^2}{2\omega}(1+n(\omega))$$

$$\times\sum_{\alpha,\beta}\frac{c_\alpha\sigma_\alpha}{m_\alpha}\langle(\mathbf{Q}\mathbf{e}_{\alpha,\beta})_E{}^2\exp(-2W_\alpha)\rangle\delta(E-E_{\alpha,\beta}),$$

where k_0 and k are the momenta of the incident and scattered neutrons; $\mathbf{Q}=\mathbf{k}-\mathbf{k}_0$; $(1+n(\omega))$ is the population factor; $2W_\alpha$ is the Debye-Waller factor; c_α, m_α, and σ_α are the concentration, mass and neutron-scattering cross section of atom α; $\mathbf{e}_{\alpha,\beta}$ is the polarization vector of atom α in vibrational mode β.

Without resorting to any model representations, it is possible to deduce from the measurement results the function

$$\theta(E)=\sum_{\alpha,\beta}\frac{c_\alpha\sigma_\alpha}{m_\alpha}\langle(\mathbf{Q}\mathbf{e}_{\alpha,\beta})_E{}^2\exp(-2W_\alpha)\rangle\delta(E-E_{\alpha,\beta}),$$

which is called the generalized frequency-distribution function. In turn, the generalized function is the sum of the partial functions

$$g(E)^\alpha=\sum_{\beta}\langle(\mathbf{Q}\mathbf{e}_{\alpha,\beta})_E{}^2\exp(--2W_\alpha)\rangle\delta(E-E_{\alpha,\beta})$$

with weights determined by $c_\alpha\sigma_\alpha/m_\alpha$. When the weight factors are equal for all components of the system, the derived function $\theta(E)$ qualitatively reflects the energy dependence of the spectral distribution of the thermal excitations. In the case of a significant difference between the values of the weight factors in the $\theta(E)$ function deduced from experimental data, the vibrations of the atoms for which $c_\alpha\sigma_\alpha/m_\alpha$ is greatest will predominate. Therefore by proper selection of the values of $c_\alpha\sigma_\alpha/m_\alpha$ it is possible to obtain data on the vibrations of any of a variety of atoms, i.e., to measure the partial vibration functions. Such information makes it possible to track variations in the vibrational properties of specific atoms when the material makes the transition from the crystalline to the amorphous state, and to provide a more reliable comparison of the experimental data with results of theoretical calculations.

The Ni_2B system was selected to investigate the influence of amorphorization on the vibrational properties of atoms on the partial-function level. The high mass difference $(m_{Ni}/m_B\approx 5.4)$ should contribute to a clearer identification of the partial densities of the vibrations, since they should be concentrated in different frequency ranges. Moreover, the fact that Ni has isotopes with strongly differing neutron-scattering cross sections will also make for a more reliable identification of the partial functions.

MEASUREMENTS AND DISCUSSION OF RESULTS

Specimens in the amorphous state were obtained by fast quenching on the surface of a rotating disk in an inert atmosphere. Two Ni_2B specimens were fabricated; the specimens had a different Ni isotope composition. A natural isotope mix was used in one specimen while the Ni^{60} isotope was used in the other. The B^{11} boron isotope was used to reduce neutron absorption in both specimens. The Table I lists the weight factors $c_\alpha\sigma_\alpha/m_\alpha$ of the specimens in relative units. It is noteworth that, owing to the small neutron scattering cross-section by the Ni^{60} isotope, information on the vibrational spectrum of the B atoms was predominantly obtained (~94%) in the experiments on the $Ni_2{}^{60}B^{11}$ specimen. Measurements of the inelastic neutron-scattering spectra were carried out with a time-of-flight spectrometer with a cold neutron source on the IR-8 reactor at the Kurchatov

TABLE I.

	$Ni_2^{NAT}B^{11}$	$Ni_2{}^{60}B^{11}$
$c_{Ni}\sigma_{Ni}/m_{Ni}$	0.203	0.011
$c_B\sigma_B/m_B$	0.123	0.123

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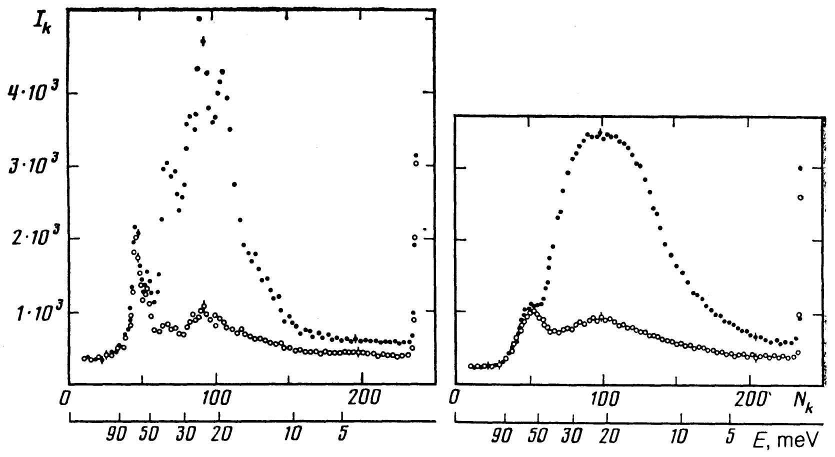

FIG. 1. Time-of-flight spectra of inelastically-scattered neutrons. Left: Crystal specimen ●—$Ni_2^{nat}B^{11}$, ○—$Ni_2{}^{60}B^{11}$. Right: Amorphous specimen ●—$Ni_2^{nat}B^{11}$, ○—$N_2{}^{60}B^{11}$.

Institute of Atomic Energy. The incident neutron energy E_0 was 4.8 meV, and $\Delta E_0/E_0 \sim 10\%$[7]. The neutrons scattered by the specimen were recorded simultaneously at angles of 15°, 30°, 45°, 60°, 75° and 90°. He^3-counter cassettes were used as the detectors.

Figure 1 shows a time plot of the doubly-differential neutron-scattering cross section in the Ni_2B system of the two isotope compositions in the crystalline and amorphous states. It is clear from the figure that both the crystalline-to-amorphous state transition and nickel-isotope substitution produce significant changes in the energy dependence of the doubly-differential cross section.

After the procedure corrections, assuming that the Debye-Waller factors are not substantially different for the different atoms, the partial vibrational state densities of the Ni atoms, $g(E)^{Ni}$ and the B atoms, $g(E)^B$ (Fig. 2) were obtained for the crystalline and amorphous states. In the crystalline state $g(E)^{Ni}$ and $g(E)^B$ are primarily concentrated in different frequency ranges, although some minor overlapping does occur. For the Ni atoms the primary density of states lies in the low-frequency range, while for the B atoms it lies in the high-energy portion of the spectrum.

The observed deformation of the functions $g(E)^{Ni}$ and $g(E)^B$ was different in the transition to the amorphous state. Smoothing of the principal peaks, an increase in the low-frequency vibration density, and shifts of the frequency boundary towards higher values were all observed for Ni. The restructuring of the partial function is more significant for the B atoms. There is a significant increase in the energy of the upper spectral boundary and a redistribution of the vibrational state density, namely, a steep increase in the density of the low-energy vibrations.

The noted changes in the spectral characteristics of the Ni atoms are qualitatively similar to the changes determined previously for a number of metal-metal systems, and can be attributed to fluctuations of the interatomic potentials. In

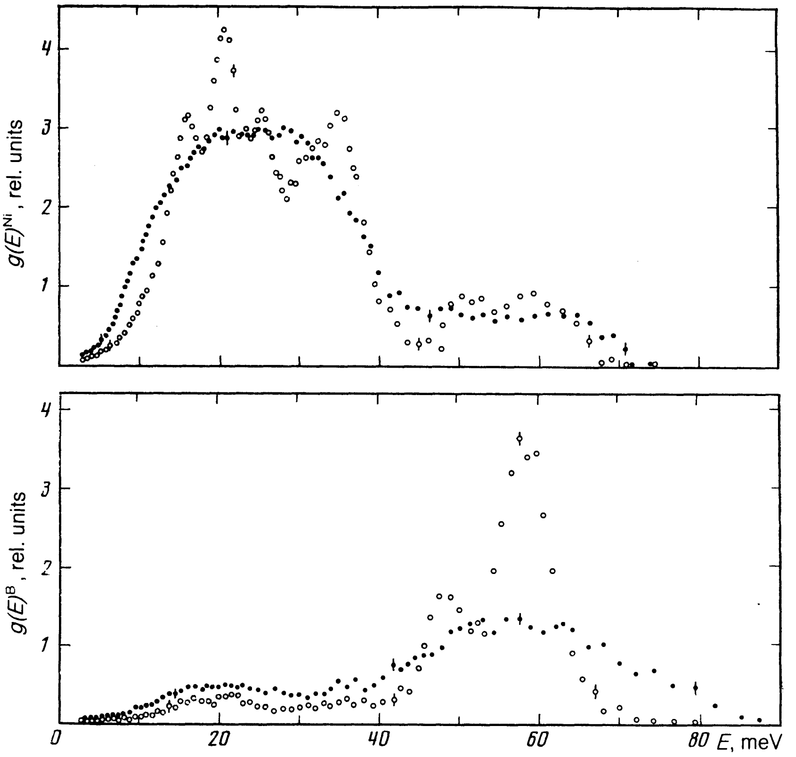

FIG. 2. Partial phonon state densities of Ni atoms (above) and B atoms (below): ○—crystalline specimen, ●—amorphous specimen.

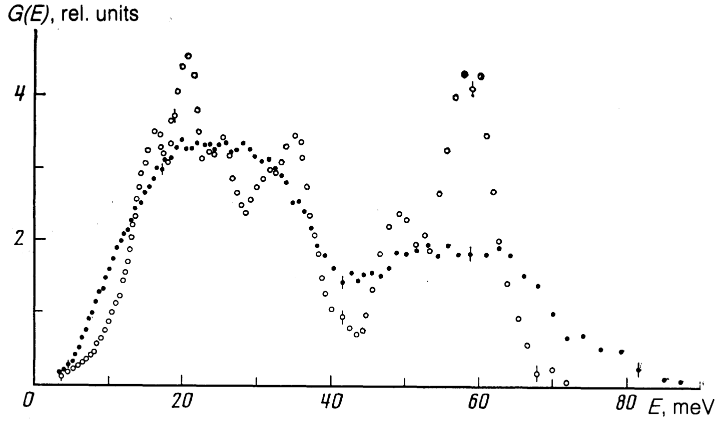

FIG. 3. Phonon state density of Ni_2B system: ○—crystalline specimen, ●—amorphous specimen.

the case of B, such substantial changes cannot be attributed solely to fluctuations of the interatomic states. It is most likely that the observed changes in the nature of the partial function of B could be related to the significant rearrangement of the nearest-neighbor configuration of this atom. Indeed, Ref. 8 has established that the radial distribution function of Ni_2B in the amorphous state is not an analog of the Ni_2B crystalline structure. Moreover the B-Ni closest distance diminishes upon amorphization.

The total vibrational state densities $G(E)^{cr}$ and $G(E)^{am}$ of the Ni_2B system in the crystalline and amorphous states (Fig. 3) was also deduced from the experimental data. The vibrational spectrum has two pronounced regions separated by a minimum at ~ 43 meV for both states. The low-energy region is related to modes caused principally by the Ni-Ni interaction, while the high-energy region is determined by B-Ni interaction. The transition to the amorphous state is accompanied by a smoothing of the sharp peaks characteristic of the vibrational spectrum of the crystalline phase, by a rise in the low-energy state density, and by a shift (larger than in metal-metal systems) of the boundary frequency towards higher values. In Ref. 5 was investigated the deformation of the vibrational spectrum of the Ni_2B system in the crystalline-amorphous state transition. A setup of a different type was used in this case, together with specimens of a single isotopic composition: $Ni_2^{nat}B^{11}$. A comparison of the results reveals that the position of the principal peaks of the vibrational spectra accords with the nature of their deformation in the crystal-amorphous state transition. However, the data presented here show more clearly the fine structure of the low-energy region (due to the better energy resolution of the setup), with the low-energy region investigated down to ~ 2 meV (to ~ 7 meV in Ref. 5). In this range the spectral distribution of the vibrations of the crystalline and amorphous states is satisfactorily described by the Debye approximation $G(E) \sim E^2$.

CONCLUSIONS

1. The partial and total vibrational spectra of the Ni_2B system in the crystalline and amorphous states were deduced for the first time from experimental inelastic neutron scattering data using the Ni isotope contrast technique.

2. The behavior of the state density for Ni atoms is analogous to the previously-established deformation of the generalized vibrational state function of metallic systems with close masses.

3. The transition to the amorphous state for the B atoms is accompanied by the onset of vibrational modes beyond the upper spectral boundary of the crystalline state and by a redistribution of the vibrational modes from the high-energy region to the low-energy region of the spectral distribution.

[1] J. B. Suck, H. Rudin, H. J. Guentherodt *et al.*, Phys. C. **13**, L167 (1980).
[2] J. B. Suck, H. Rudin, H. J. Guentherodt, and H. Beck, J. Phys. C. **14**, 2305 (1981).
[3] M. G. Zemlyanov, G. F. Syrykh, and N. A. Chernoplekov, Pis'ma Zh. Eksp. Teor. Fiz. **42**, 176 (1985) [JETP Lett. **42**, 217 (1985)].
[4] N. Lusting, J. S. Lannin, J. M. Carpenter, and R. Hasegawa, Phys. Rev. B. **32**, 2778 (1985).
[5] N. Lusting, J. S. Lannin, and R. Hasegawa, Phys. Rev. B. **34**, 6725 (1986).
[6] N. A. Chernoplekov, M. G. Zemlyanov, E. G. Brovman, and A. G. Checherin, Fiz. Tverd. Tela. **5**, 112 (1963) [Sov. Phys. Solid State. **5**, 78 (1963)].
[7] M. G. Zemlyanov, A. E. Golovin, S. P. Mironov *et al.*, Prib. Tekh. Eksp. 5, 34 (1973).
[8] S. N. Ishmaev, S. L. Isakov, I. P. Sadikov *et al.*, J. Non-Cryst. Sol. **94**, 11 (1987).

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