Sound attenuation due to the decay of coherent concentration waves in a niobium-hydrogen solution

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The attenuation peaks of low-frequency (~ 1 kHz) sound have been detected near the points of stability loss of a solid $NbH_{0.04}$ solution relative to the formation of concentration waves. The diffusion coefficient of hydrogen $D \gtrsim 10^{-11}$ cm^2/sec at $T \sim 20$ K has been estimated. Despite the high hydrogen concentration, this value is three to five orders of magnitude higher than those determined by using other methods.

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It was first shown in Ref. 1 that under certain conditions the decomposition of solid solutions proceeds via the formation of periodic modulated structures. This rather common phenomenon has been studied in depth in a number of papers (see monograph[2]). The decomposition of metal-hydrogen solid solutions[3,4] involves the formation of two types of plane concentration waves--macroscopic waves with a wavelength λ of the order of the size d of the sample and microscopic waves with $\lambda \ll d$.

Using a cold-neutron-scattering method, Conrad *et al.*[5] have determined the stability limits of short-wave concentration waves in an Nb-H solution. It turned out that for hydrogen concentrations in the solution $c_H < 0.18$ two spinodals maintained at $T < 80$ K correspond to two distinct orientations of the wave vector $\vec{\kappa}, \vec{\kappa} \parallel [110]$, and $\vec{\kappa} \parallel [111]$.

In this study we have measured the attenuation of low-frequency (~ 1 kHz) sound in an $NbH_{0.04}$ solid solution in the temperature range 1.5–80 K; this range included the temperatures of the spinodals T_{s_1} and T_{s_2} for the composition used by us. The samples were comprised of $10 \times 3 \times 0.3$-mm plates, in which the transverse quarter-wave oscillation mode was excited. The measurements were carried out using an acoustic spectrometer that operated in the regenerative mode.[6] We used polycrystalline, zone-refined niobium; the sample was saturated with hydrogen at $T \approx 850$ K in a quartz ampoule that was filled with pure gas obtained by thermal decomposition of hydride.

The temperature dependence of sound attenuation (in units of reciprocal Q of an acoustic vibrator) is represented by curve 1 in Fig. 1; this curve has an attenuation peak at $T \approx 18$ K and a peak at $T \approx 55$ K, which is masked by a background attenuation. The background curve (curve 2 in Fig. 1) was obtained at a much lower hydrogen content in the solution ($c_H \approx 0.005$).

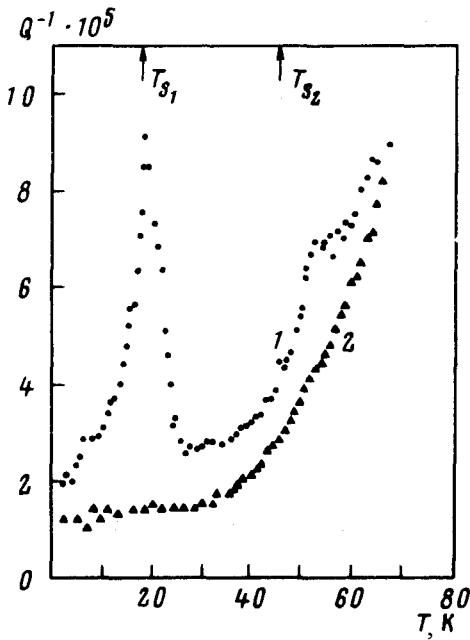The arrows in Fig. 1 indicate the temperatures T_{s_1} and T_{s_2} of the spinodals for

FIG. 1.

an $NbH_{0.04}$ solid solution, in accordance with Ref. 5. It can be seen that the temperature of the detected peaks corresponds to the temperatures T_{s_1} and T_{s_2}. We can conclude from this that the observed absorption peaks are associated with the loss of stability of the concentration waves that have been polarized in the [110] and [111] directions.

Since the systems are analogous from the thermodynamic point of view near the temperature of the coherent spinodal T_s and near the temperature of the second-order phase transition, we can describe the sound attenuation near T_s by using the relations which were first obtained in Ref. 7 for the λ point in He^4 (and which, as it turned out, can be used for second-order phase transitions in solids). In the context of the self-consistent field theory[7] the relaxational absorption of sound is determined by

$$\delta \sim \omega \tau / (1 + \omega^2 \tau^2) \tag{1}$$

with $\tau \sim (T_c - T)^{-1}$.

Here δ is the logarithmic damping factor, ω is the oscillation frequency of the sample, T_c is the phase-transition temperature, and τ is the relaxation time of the order parameter. The sound attenuation near the temperature of the spinodal is caused, just as in the case of second-order phase transitions, by the "critical slowing down"—the relaxation time in this case is determined by the following expression[8]:

$$\tau \approx \frac{\lambda^2}{\pi^2 D} \frac{T_s}{T_s - T}, \tag{2}$$

Here λ is the length of the concentration wave and D is the diffusion coefficient of hydrogen.

We note that the temperature of the spinodal was measured directly for the first time in these experiments without extrapolation from the region of a homogeneous solution.

The limiting value of the diffusion coefficient of hydrogen at $T \sim 20$ K was determined by monitoring the absorption peaks of sound near T_{s_1} and T_{s_2}, which are the points at which the stability of plane concentration waves is lost. Since $\tau \sim 1/\omega$ at the sound-attenuation maximum, it follows from Eq. (2) that $D \gtrsim \omega\lambda^2/\pi^2$ when $T \lesssim T_s$. Inserting $\omega \sim 10^4$ sec^{-1} and $\lambda \sim 10$ Å, we estimate

$$D \gtrsim 10^{-11} \text{ cm}^2/\text{sec}.$$

This value of the diffusion coefficient proved to be unexpectedly large compared with the known data. Thus the diffusion coefficient determined from reorientation of the coupled O–H pairs[9] at $T \sim 40$ K is equal to $\sim 10^{-16}$ cm^2/sec for weak H solutions in Nb, and the diffusion coefficient determined from the precipitation rate of hydrogen from a supersaturated solution at $T \sim 34$ K is equal to $\sim 10^{-14}$ cm^2/sec.[10]

In summary, we would like to emphasize the following important point. In spite of the fact that the amount of hydrogen in the solution was much larger in our experiments than in those of Refs. 9 and 10, the determined value of D was much higher, although it was shown in Refs. 11 and 12 that the subbarrier-diffusion coefficient decreases with increasing particle density.

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