transition from an atomically smooth to an atomically rough surface,[12] which manifests itself in the vanishing of the mirror, cannot be carried out in purely classical terms in the case of helium.

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*Addendum* (12 September 1980). We have recently learned of work by Landau *et al.*[14] in which it is indicated that flat sections are present in equilibrium He⁴ crystals below 1 K, and which reports also the results of measurements of $\bar{\alpha}$ for the rounded sections ($\bar{\alpha} = 0.1$–0.2 erg/cm² for different samples, and is independent of temperature in the range 0.4–1.45 K). These results, as seen from the text, are in full agreement with ours.

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1) We have no coherent explanation of this fact at present.
2) The reflectivity of the capacitor surface under grazing illumination changes abruptly immediately prior to formation of visible seeds. This is due, in all probability, to the formation of a large number of minute (1–10 μm) crystallites in regions with maximum field intensity.
3) We note that in this case, too, all the seeds have as a rule the same orientation, which differs in general from the orientation of the seeds prior to application of the electric field (see footnote 1).

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# Anharmonicity of local vibrations of Be in Cu–Be alloy

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The local-vibration (LV) spectra in Cu–Be alloys with Be impurity density from 0.5 to 9.5% are investigated in the 100–515 K temperature range by the methods of inelastic incoherent scattering of slow neutrons. The width of the spectral density of the LV is determined by the temperature and concentration broadenings. The weak temperature dependence of the LV frequency is due to the mutual cancellation of the volume expansion of the lattice (i.e., of the anharmonicity of the lattice vibrations) and the anharmonicity of the LV. It is shown that the principal process that determines the temperature broadening and the LV shifts is the third-order anharmonicity, namely the decay of the LV into two phonons of the main spectrum.

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## 1. INTRODUCTION

The appearance of anharmonicity of local vibrations (LV) of a light substitutional impurity is a little-investigated question even though it is just for the LV that the observation of the real and imaginary parts of the frequency shift is greatly facilitated because the LV are located in a spectral region forbidden to the main spectrum. An exception is the case of ionic crystals, for which the shifts and broadening of the LV of various impurity atoms were investigated by optical methods.[1,2] It was observed in particular that in alkali-halide crys-

tals with H⁻ and D⁻ impurities the width of the LV level of the hydrogen isotopes increases approximately tenfold when the sample temperature is raised from 100 to 400 K.[1] To our knowledge, no such investigations were made for metallic impurity systems to date.

The width of the LV in metallic systems is determined, besides by the anharmonicity, also by the interaction of the impurity atoms with one another and by the electron-phonon interaction. In a preceding study[3] the concentration dependence of the energy position $\varepsilon_{l,V}$ and of the width $\Delta\varepsilon_{l,V}$ of the LV level were investigated for a system

based on copper with Be impurity. At room temperature the width $\Delta\varepsilon_{LV}$ increases linearly with the impurity density. The impurity interaction, however, could not explain fully the experimentally obtained level widths. The value of $\Delta\varepsilon_{LV}$ extrapolated to zero impurity density remains finite: $(\Delta\varepsilon_{LV}/\varepsilon_{LV})_{c\to 0}\approx 0.05$. In the present paper, to determine the contribution of the anharmonicity to the width $\Delta\varepsilon_{LV}$, we investigated experimentally the influence of temperature on the character of the LV distribution in Cu-Be alloys.

## 2. MEASUREMENTS AND REDUCTION OF RESULTS

The spectral distributions of the LV were measured by neutron spectroscopy, which is the principal source of information on the LV spectra in metallic systems, at temperatures 100, 200, 295, 415, 475, 515 K. The samples investigated were those used to study the effect of the impurity density on the LV characteristics.[3] The upper limit of the investigated temperature interval is connected with the specifics of the samples, which are supersaturated $\alpha$-solutions obtained by quenching followed by annealing at 640 K. Prolonged measurements at temperatures close to the annealing temperature are impossible because of the abrupt increase of the $\alpha$-solution decay probability.[4]

The doubly differential cross sections of the neutron inelastic scattering were measured with two types of time-of-flight spectrometers of equal energy resolution. For the measurements at 200, 295, 415, and 515 K we used a spectrometer with a cold-neutron source.[5] At 100 K the measurements were made with a multidetector slow-neutron spectrometer with a crystal monochromator.[6] In this case we used the "dumping" method, wherein inelastic scattering of neutrons with initial energy $E_0 > \varepsilon_{LV}$ leads to "production" of phonons and it becomes possible to observe high-energy LV at low temperatures. The inelastic scattering of neutrons by pure copper were measured at the same temperatures.

In the entire investigated temperature and density interval it is easy to distinguish in the difference spectra

$$\Delta\frac{d^2\sigma}{d\Omega dt}=\left(\frac{d^2\sigma}{d\Omega dt}\right)_{Cu-Be}-\left(\frac{d^2\sigma}{d\Omega dt}\right)_{Cu}$$

one of which is shown by way of example in Fig. 1, two singularities. The first is at $\varepsilon\approx 40$ meV and is due to the LV, while the second, at $\varepsilon\approx 30$ meV, is due to the hardening of the spectrum of the acoustic oscillations in the alloy compared with the copper spectrum. Only at the lowest of the investigated impurity densities, $c=0.005$, when the LV density is low, does the singularity at $\varepsilon\approx 30$ meV exert a noticeable influence on the experimental LV spectrum, and leads to a filling of the gap between the two singularities with increasing temperature. Even in this case, however, inclusion, with the aid of the method developed in Ref. 7, of the resolution function for the entire experimental spectrum, makes it possible to reconstruct the spectral distribution of the LV in the form of a distinctly separated line.

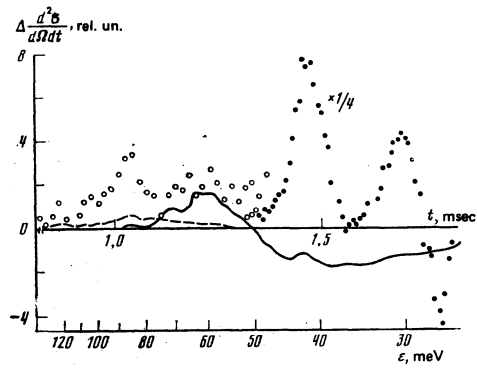To establish the real form of the spectral distribution



FIG. 1. Difference between the cross sections for neutron scattering by a Cu-Be alloy ($c = 0.027$) and by Cu at $T= 473$ K; points—experiment; soild line—contribution of two-phonon and second-order processes; dashed—contribution of three-phonon and third-order processes with participation of LV.

of the LV it is necessary to take into account, besides the resolution function, also the influence of the multiple-order interactions and multiphonon scattering of the neutrons. Calculations were made in this connection of the probabilities of two- and three-phonon as well as second- and third-order processes for copper and for the investigated alloys. The calculations were made under the following assumptions: the Be atoms participate only in the LV, while the Cu atoms take part in both the matrix vibrations and in the LV (the fraction of the Cu atoms taking part in the LV was determined in Ref. 3). The spectrum of the original matrix was taken to be the phonon spectrum reconstructed from the measured dispersion relations[8]; the LV spectra were approximated by Lorentz curves with widths $\Delta\varepsilon_{LV}$ and positions $\varepsilon_{LV}$ determined in the present paper. As seen from Fig. 1, which shows in addition to the experimental data also the differences between the calculated cross sections for the multiple-order and multiphonon processes for an alloy of 2.7 at. % Be in copper, at $\varepsilon< 50$ meV the cross sections for two-phonon and second-order processes on copper are larger than the same cross section for the alloy. At $\varepsilon\gtrsim 50$ meV these processes are seen to predominate for the alloy. At $\varepsilon> 75$ meV the contribution of the three-phonon and third-order processes becomes more substantial. It is of interest to note that in the energy region $\varepsilon > \varepsilon_{LV}$ the multiple-order and multiphonon processes do not fit exactly the experimental distributions (Fig. 1).

The integrated characteristics (position, width, intensity) of the LV spectral distributions obtained in the incoherent approximation were estimated by the methods described earlier.[3] Figures 2–4 show the temperature dependences of these characteristics. The greatest change in the width $\Delta\varepsilon_{LV}$ [Fig. 2(a)] is observed for alloys with small Be content. Thus, at $c = 0.01$ the value of $\Delta\varepsilon_{LV}$ is somewhat damped by the influence of the concentration broadening, which in accordance with Ref. 3 is described by the relation

$$\frac{1}{\varepsilon_{LV}}\frac{\partial\Delta\varepsilon_{LV}}{\partial c}=0.81. \tag{1}$$

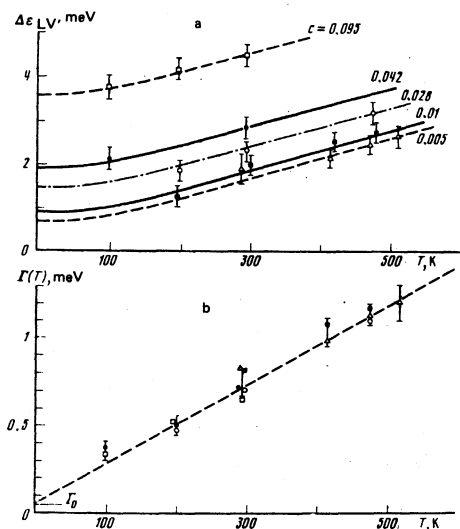Assuming that relation (1) is valid in the entire investigated range of temperatures $T$, we can separate

FIG. 2. Temperature dependences of the width $\Delta\varepsilon_{LV}$ (a) and damping $\Gamma(T)$ (b) for different Be densities: $\triangle$) $c = 0.005$; $\blacksquare$) 0.01; $\bigcirc$) 0.027; $\bullet$) 0.042; $\square$) 0.095. The curves on Fig. a were calculated from Eq. (7).

the temperature-dependent part of the LV width

$$2\Gamma(T_j) = \Delta\varepsilon_{LV}(c_i, T_j) - \frac{\partial\Delta\varepsilon_{LV}}{\partial c}c_i. \qquad (2)$$

The values of $\Gamma(T_j)$ obtained for alloys with different impurity densities $c_i$ agree with one another within the limits of the experimental errors, and have a linear temperature dependence in practically the entire measurement range [Fig. 2(b)].

The temperature shift of the LV (Fig. 3) should be determined primarily by the volume expansion of the lattice of the alloy as a whole. The thermal expansion can be treated as a manifestation of an external deforma-
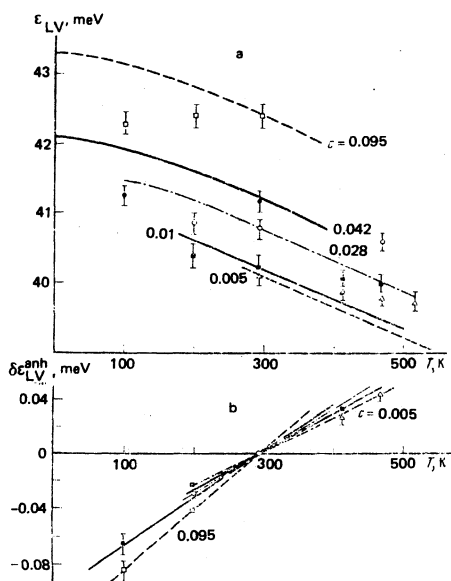


FIG. 3. Temperature dependences of the energies $\varepsilon_{LV}$ (a) and of their shifts $\delta\varepsilon_{LV}$ due to anharmonicity (b). The symbols for the experimental points are the same as in Fig. 2. Curves on Fig. a—contribution of the lattice thermal expansion calculated from Eq. (9).
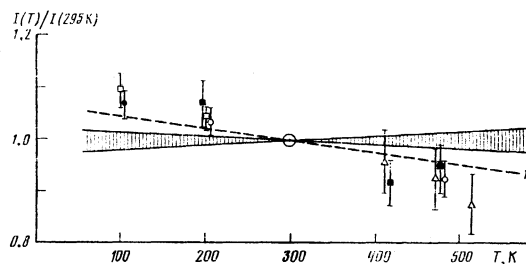


FIG. 4. Dependence of integrated intensity of the LV spectrum on the temperature. The symbols for the experimental points are the same as in Figs. 2 and 3. Line 1—with allowance for the difference between the Debye-Waller factors for the Be and Cu atoms; lines 2—with allowance for the Debye-Waller factor and multiphonon and multiple-order processes. The funnel 2 determines the limiting values of the integrated intensity $I(T)$.

tion that leads to a decrease of the vibrational energy of the entire system, and it is possible to separate against this background the LV shift component due to the anharmonicity of the LV. For lattice excitations of copper, the temperature dependence of their energies[9] is described by the relation

$$\varepsilon_q(T) = \varepsilon_q(T_0) \left[ 1 - \gamma_G \int_{T_0}^{T} \beta(T)dT \right], \qquad (3)$$

where $\varepsilon_q(T)$ is the energy of the phonon at the temperature $T$; $\gamma_G$ and $\beta$ are respectively the Gruneisen and volume-expansion coefficients.

If it is assumed that $\gamma_G$ and $\beta$ are insignificantly changed by the introduction of the Be atoms ($c < 0.1$), then we can estimate the LV-position shift due to the temperature volume expansion. Figure 3(a) shows the calculated temperature dependences of $\varepsilon_{LV}(c_i, T)$ in accordance with relation (3) for different concentrations $c_i$ and for $T_0 = 295$ K. The difference between the experimental and calculated values of $\varepsilon_{LV}(c_i, T)$ determines the shift $\Delta\varepsilon_{LV}$ due to the LV anharmonicity whose temperature dependence is shown in Fig. 3(b).

Figure 4 shows the integrated intensities of the LV spectral $g(\varepsilon)$ reconstructed from the experimental data, neglecting the difference between the Debye-Waller factors of the impurity and matrix atoms (the isotropic model) and between the contributions of the multiphonon and multiple-order processes for the initial matrix and the atoms. Allowance for the influence of the relative change of the force constants in the investigated system on the Debye-Waller factor,[10] as well as of the multiphonon and multiple-order processes, makes the LV intensity practically independent of temperature.

## 3. DISCUSSION OF RESULTS

In the investigated Cu-Be system, the LV energy $\varepsilon_{LV}$ is lower than the doubled end-point energy $\varepsilon_{ep}$ of the spectrum of the acoustic vibrations of the lattice. Consequently the most probable process that leads to the temperature dependence of the LV width should be the third-order anharmonicity, i.e., the decay of the LV into two phonons of the main spectrum. In this case one can expect in the high-temperature classical limit

a linear dependence of the width on $T$, as is in fact observed in experiment at $T \geqslant 200$ K [Fig. 2(b)].

The angle of inclination of the experimental line determines the damping of the LV in the high-temperature limit:

$$\Gamma(T) = (0.027 \pm 0.004) \frac{T}{\Theta} \varepsilon_{ep} , \qquad (4)$$

where $\Theta$ is the Debye temperature of the impurity system.[1)]

It follows from the existing theoretical estimates of the anharmonic broadening of the LV[12] (there is no adequate quantitative theory of this phenomenon) that $\Gamma(T)$ should amount to 14% of $k_B T$. The result (4) shows that $\Gamma(T)$ is much less. Moreover, it follows from the same theoretical estimates that the anharmonic broadening of the LV should significantly exceed the anharmonic broadening of the phonons of the initial matrix. However, as follows from the results and from the data of Ref. 9, the values of the damping of the LV and of the acoustic phonons of copper have close temperature-dependence coefficients. Indeed, the damping of the longitudinal vibrations of copper[9] are well approximated by the relation

$$\Gamma_q = 0.025 \frac{T}{\Theta} \varepsilon_q, \qquad (5)$$

where $\varepsilon_q$ is the energy of the phonons with wave vectors $q$.

Extrapolation of the linear $\Gamma(T)$ dependence to $T = 0$ K permits an experimental estimate [Fig. 2(b)] of the value of $\Gamma_0$ connected with the electron-phonon interaction. For the investigated alloys, $\Gamma_0$ turned out to be lower by one order of magnitude than the contributions of the concentration and temperature broadenings, and equal to $(0.05 \pm 0.05)$ meV or $(1.3 \pm 1.3) \times 10^{-3} \varepsilon_{LV}$. The obtained value of $\Gamma_0$ agrees with the theoretical estimate[13] of the damping of the LV in a metal on account of interaction with the conduction electrons: $\Gamma_0/\varepsilon_{LV} \approx 2 \times 10^{-3}$.

The observed linear dependence of $\Gamma(T)$ at $T \geqslant 200$ K agress with the theoretical conclusions in the high-temperature limit. At low temperatures, the anharmonicity of the zero-point vibrations may come into play. To estimate the LV lifetime near $T = 0$ we use the ratio of the LV damping obtained within the framework of perturbation theory. To this end, we transform the expression for $\Gamma(T)$, obtained for LV with the aid of the third-order anharmonicity Hamiltonian [(39.7) in Ref. 14] to a form convenient for numerical integration:

$$\Gamma(T) = \frac{\pi}{2\hbar^2} \int_{\varepsilon_{LV}-\varepsilon_{ep}}^{\varepsilon_{ep}} d\varepsilon g(\varepsilon) g(\varepsilon_{LV} - \varepsilon) |V_{\varepsilon, \varepsilon_{LV}-\varepsilon}|^2$$
$$\times \left( 1 + \left[ \exp \frac{\varepsilon}{k_B T} - 1 \right]^{-1} + \left[ \exp \frac{\varepsilon_{LV} - \varepsilon}{k_B T} - 1 \right]^{-1} \right), \qquad (6)$$

where $V_{\varepsilon_1 \varepsilon_2}$ is the potential of the interaction of the LV and of the phonons with energies $\varepsilon_1$ and $\varepsilon_2$. For comparison with the experimental data, Fig. 2(a) shows the temperature dependences of the calculated functions $\varphi(c_i, T)$:

$$\varphi(c, T) = 2[\Gamma_0 + \Gamma_c + \Gamma_T], \qquad (7)$$

where $\Gamma_c$ describes the concentration dependence of the

half-width in accord with relation (1), and $\Gamma_T$ is the temperature part of the broadening.

The value of $\Gamma_T$ was calculated from formula (6) in the simplest model for $V_{\varepsilon_1 \varepsilon_2}$:

$$V_{\varepsilon_1 \varepsilon_2} = A(\varepsilon_1 \varepsilon_2)^{1/2}, \qquad (8)$$

where $A$ is a fit parameter.

The entire aggregate of the experimental values of $\Delta \varepsilon_{LV}$ [Fig. 2(a)] is described by a single value of the parameter $A$. This is evidence of the additivity of the contributions of the impurity interaction and of the anharmonicity to the broadening of the LV spectrum in the entire investigated range of concentrations and temperatures.

According to calculations with the aid of relations (6) and (8), the interaction of the oscillations at $T = 0$ K limits the lifetime of the LV of an isolated impurity to 200 vibrations. The damping corresponding to this lifetime is relatively not small: it amounts to 0.3 of $\Gamma_T$ at room temperature and is comparable with the contribution made to the LV damping by the interaction of the impurity centers ($\Gamma_T \approx \Gamma_c$ at $T = 0$ K and $c = 0.012$).

A satisfactory description of the experimental data with the aid of relations (6)–(8) leads to the conclusion that the contribution of the anharmonicity of order higher than the third, and having a temperature dependence $T_n$ ($n > 1$) are relatively small. On this basis, an attempt was made of the shift $\delta \varepsilon_{LV}$ due to the third-order anharmonicity. The shift of the LV under this assumption turned out to be very sensitive to singularities of the phonon spectrum of the deformed lattice of the matrix and to the relative change of the impurity–matrix force interaction. It is noteworthy that in the case of the simplest form (8) of the interaction potential $V_{\varepsilon_1 \varepsilon_2}$ and neglecting the change of the matrix phonon spectrum, the calculated values of the shift $\delta \varepsilon_{LV}$ can be positive already at a small deviation (2–3%) of the ratio $\varepsilon_{LV}/\varepsilon_{ep}$ from the corresponding value for the case of an isolated impurity. It is quite probable that the observed [Fig. 3(b)] change of the slope of the temperature dependence of the displacement for different concentrations is due to a restructuring of the interactomic force interaction and consequently to changes in the ratio $\varepsilon_{LV}/\varepsilon_{ep}$.

It would undoubtedly be of interest to carry out the calculation with allowance for the real form of the vibrational spectrum of the matrix of the atoms in the alloy, as well as of the change in for force interaction of the impurity and of the matrix atoms with increasing temperature.

Arguments have already been advanced[3] that at low impurity densities the interaction of the impurity centers with one another in Cu-Be are predominantly indirect. Our present temperature investigations have shown that such an interaction of the impurity centers can be due, in particular to the anharmonicity of the LV. The LV are, as it were, delocalized near the impurity and capture some of the surrounding matrix atoms. Formation of bound phonons with participation of LV on account of the anharmonicity is therefore probable. The fact that the experimental cross section
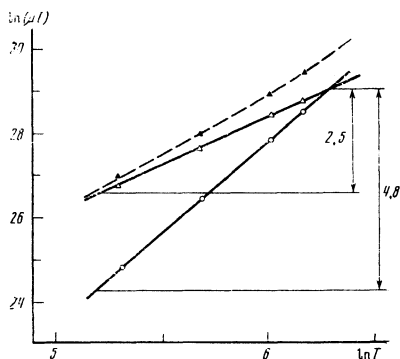
FIG. 5. Temperature dependence of the integated intensity of the integrated intensity of the difference spectra $\Delta d^2\delta/d\varepsilon d\Omega$ at $\varepsilon > \varepsilon_{LV}$; ▲) experimental points; ○) calculation of the contribution of the multiphonon and multiple-order processes; △) contribution of scattering by bound vibration, defined as the difference between the experimental data (△) and the calculation results (○).

of the scattered neutrons exceeds the cross section of the multiphonon and multiple processes at $\varepsilon > \varepsilon_{lv}$ (Fig. 1) can in princple be due to the scattering of the neutrons by the bound phonons, in analogy with the situation with optical spectra for impurity sytems.[15] The cross section for neutron scattering by such phonons, due to third-order anharmonicity, should have a linear dependence on $T$, in contrast to the quadratic dependence of the cross sections for two-phonon processes.[16] The experimentally observed integral intensity of scattered neutrons at $\varepsilon > \varepsilon_{lv}$ (Fig. 5) can be actually interpreted as a sum of the two mentioned processes, whose temperature dependences are in a ratio 1:2.

In conclusion, it is our pleasant duty to thank Yu. I. Zaĭtsev, V. G. Fedorov, and A. M. Finogin for help during the various stages of the measurements, and to A. P. Zhernov for constant interest and for remarks.

[1)]According to Ref. 11. the Debye temperature of the impurity system is

$$\Theta(c) = \Theta(0) \left[ 1 - 0.5c \left( \frac{2\tau}{1 - K\tau} - \xi \right) \right],$$

where the coefficients $\tau$ and $\xi$ characterize the relative change of the force constants and masses of the impurity and matrix atoms, and $K$ is connected with the structure of the matrix lattice. At $\xi = 0.86$, $\tau = 0.47$, and $K = 0.16$, which hold for Cu-Be, the value of $\Theta$ remains practically constant in the entire range $c < 0.1$.

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