in molecular hydrogen. Therefore the mechanism considered in this paper can lead to loss of stability of a discharge produced in hydrogen at temperature corresponding to the decreasing branch of the thermal-conductivity coefficient.

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Nonlinear variation of the energy gap and the phonon spectrum of indium near the electron transition

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The method of electron tunneling is used to investigate the electron-phonon interaction and the phonon spectrum of indium in a wide range of pressures and tin-impurity concentrations. An anomalous behavior of the lattice-vibrational spectrum is discovered in the region topolgical changes in the Fermi surface. The possible mechanisms leading to such changes in the lattice are considered.

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1. INTRODUCTION

The electron transition, due to changes in the topology of the Fermi surface, leads to anomalies in the thermodynamic and kinetic characteristics of metals (the so-called phase transition of order $2\frac{1}{2}$).[1] Upon the attainment of some critical value by the Fermi energy of the metal, there should arise in the electron density of states a distinctive feature that is reflected in the electronic specific heat and other second derivatives of the thermodynamic potential. The presence of such anomalies in the behavior of the electron spectrum has been confirmed in experiments on specific heat[2a] and the de Haas–van Alphen effect.[2b] The method developed by Lazarev and his students[3–5] for observing the topological anomalies in the electron spectrum through the study of the combined effect of impurities and pressure on the superconducting-transition temperature T_c turned out to be effective. Underlying this method is Makarov and Bar'yakhtar's theoretical model,[4] which establishes a connection between the variation of T_c and the changes in the Fermi surface. From the investigations of the shape of the $dT_c(C, P)/dP$ curves arose the possibility of reestablishing the type of topological transition and of extracting quantitative information about a number of parameters of the electron spectrum.

At the same time, the question of the behavior of the phonon spectrum of a metal in the vicinity of a phase transition of order $2\frac{1}{2}$ was not rigorously analyzed. However, a correlation was observed between the nonlinear variation of the lattice parameters and the anomalies of the electronic characteristics in alloys for the same impurity concentrations.[6] Il'ina, Itskevich, and Titov[7] have shown that, under the action of high pressures, the anomalies in T_c and the critical magnetic field, H_c, are accompanied by irregularities in the lattice parameters.[8] All this points, it seems to us, to the possible appearance of anomalies in the phonon spectrum as well. A detailed critical analysis of the large amount of factual material, obtained by various groups of investigators, on the problem under discussion predetermined the necessity for the performance of a careful investigation of the vibrational spectrum of the lattice of a metal that clearly exhibits changes in the topology of the Fermi surface. In the present work we studied indium. The topological changes were realized through the introduction of a tin impurity, or the application of pressure. In both cases, as follows from the published data, there appear in indium new electron holes in the third Brillouin zone: first at the symmetry points W and then as α tubes in the [101] direction.[1] The choice of the tunneling method of investigation was dictated by its high resolving power and its high information-yielding capacity.

2. EXPERIMENT

1. *Sample preparation.* The Al-Al_2O_3-In tunnel junctions (or the alloys $In_{1-x}Sn_x$, $x = 0$–8 at.% Sn) were pre-

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pared by condensing the metals in a 10^{-6}-Torr vacuum onto a glass substrate cooled to liquid-nitrogen temperatures. The film thicknesses were ~100 Å for aluminum and ~3000 Å for indium. The regime of oxidation of the aluminum film in an atmosphere of dry oxygen at a pressure of ~100 Torr and a temperature of 350 K over a period of several minutes allowed us to obtain initial junction-resistance values of $R_T = 50$–$300\ \Omega/\text{mm}^2$. The alloy films were deposited by sputtering from large weighed portions. To homogenize the composition of the films, we annealed the prepared tunnel samples in a vacuum at 300–350 K over long periods of time—from a day to several weeks. All the junctions upon the initial heating had, like Josephson junctions,[10] an $R_T(T)$ maximum that shifted from 260 K for indium to 300 K as the tin concentration was increased.

The verification of the sample quality was carried out at liquid-helium temperatures by finding the values of the ratio

$$\sigma = \left(\frac{dI}{dU}\right)_N \Big/ \left(\frac{dI}{dU}\right)_S,$$

for $U=0$ at the barrier. The value of $\sigma_{1.5\ \text{K}}$ varied from 10^{-1} for Al-In to 10^{-2} for Al-$In_{0.9}Sn_{0.1}$, which indicated that the tunneling mechanism of electron transmission played the dominant role. The homogeneity and flawlessness of the films were estimated from the sharpness of the peaks of the (dI/dU) versus U curves at the energies $\Delta_1 - \Delta_2$ and $\Delta_1 + \Delta_2$.

The tin-impurity concentration was determined to within ~0.2 at.% from the T_c versus film composition curves[11] and from the residual resistance $\rho = R_{4.2}/(R_{300} - R_{4.2})$, whose concentration dependence in the α phase is well described by a straight line, $\rho = \rho_0 + kC$. The initial value of dI/dU for indium films deposited in a 10^{-6}-Torr vacuum is equal to ~0.05. It corresponds to the limiting value at which the process of isotropization of the effective electron-electron attraction parameter is essentially completed.[12] Therefore, this effect, which leads to nonlinearities in film systems, can be neglected.

2. *Measurement procedure.* In the experiment we measured the I–U, (dI/dU)-U, and (d^2I/dU^2)-U curves

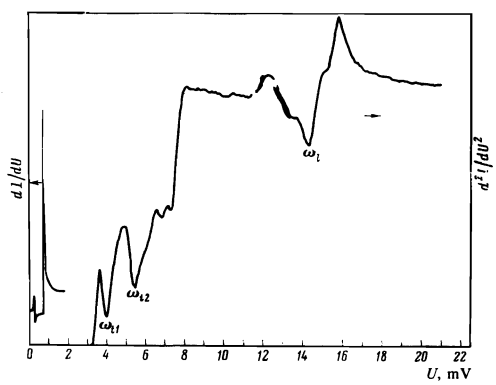

FIG. 1. Example of the recordings of the (dI/dU)-U and (d^2I/dU^2)-U tunnel characteristics for indium.

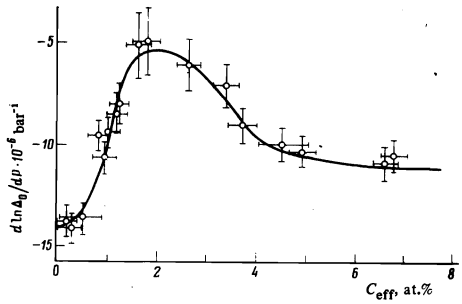

FIG. 2. Concentration dependences of the derivatives $d\ln\Delta_0/dP$ in the In–Sn system.

at a temperature of 1.4 K. For the purpose of increasing the measurement accuracy, we inserted in the circuit from which the first-harmonic signal was tapped a transformer with a 1:500 transmission coefficient, cooled down to liquid-helium temperatures. This allowed at a modulation amplitude of 1 μV a considerable enhancement of the useful signal and the reduction of the measurement error down to $(1-3)\times10^{-3}$ meV. From the I–U and (dI/dU)-U characteristic curves we determined the energy gap $\Delta(T)$, whose value was referred to $T=0$ (Δ_0).

The critical temperature, T_c, was found from the temperature dependences of the film resistance and the junction resistance, R_T, for $U=0$.[13] The latter procedure allows the measurement of the T_c of a small part $(1\times1\ \text{mm}^2)$ of the film, which precludes the smearing of the superconducting transition as a result of a possible inhomogeneity of the film along its length. The error in the determination of T_c was 0.005 K.

The information about the phonon spectrum, $F(\omega)$, was extracted from the (d^2I/dU^2)-U characteristic curves (Fig. 1), which indicate the characteristic frequencies, $\omega_i^{cr} = \omega(q_{cr})$, of the spectrum: $\omega_{t1} = 3.16$ meV, $\omega_{t2} = 4.6$ meV, corresponding to the transverse branches of the vibrational spectrum, and $\omega_l = 13.4$ meV, corresponding to the longitudinal branches. In a number of cases, especially for the alloys with high impurity concentrations, when the distinctive features of the transverse vibrations were washed out, to determine more accurately the locations of these features we measured the (d^3I/dU^3)-U curves.

To show the weak nonlinearities in the dependences $\Delta_0(C)$ and $\omega_i^{cr}(C)$, we also constructed the pressure derivatives of these quantities: $d\Delta_0(C)/dP$ and $d\omega_i^{cr}(C)/dP$:

$$\frac{d\Delta_0}{dP} \approx \frac{\delta\Delta_0}{\delta P} = \frac{\Delta(P)-\Delta(0)}{P-P_0}, \quad \frac{d\omega_i^{cr}}{dP} \approx \frac{\delta\omega_i^{cr}}{\delta P} = \frac{\omega_i^{cr}(P)-\omega_i^{cr}(0)}{P-P_0},$$

where $\delta P \sim 2$–3 kbar. The experiments were performed in a high-pressure chamber with the use of a kerosene-oil mixture as the transmitting medium at pressures of 0–12 kbar.[14] The pressure was determined from the temperature T_c in indium and tin with an error of 100 bar. The high degree of hydrostaticity could be judged from the width, which virtually did not change with increasing pressure, of the peaks of the (dI/dU)-U curves in the $(\Delta_{In} - \Delta_{Al})/e$ and $(\Delta_{In} + \Delta_{Al})/e$ voltage-potential

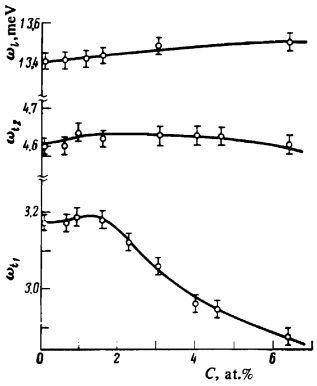

FIG. 3. Variation of the characteristic frequencies ω_i in the α phase of the In–Sn alloys.

region. The obtained values of the quantities were averaged over several samples and many measurements.

3. RESULTS OF THE EXPERIMENT

1. The In-Sn system

The energy gap. Upon the introduction of the tin impurity, the energy gap Δ_0 and $2\Delta_0/T_c$ increase within the boundaries of the α phase.[15] In the 2 at.% impurity region a nonlinear behavior of Δ_0 is observed (Fig. 2). The value $d\ln\Delta_0/dP = -14.2\times10^{-6}$ bar^{-1} for pure indium agrees with the data obtained in earlier experiments.[16] In the region of the anomalies the derivative is similar in character to dT_c/dP.[17]

The phonon spectrum. The dependence of the characteristic frequencies ω_i^{cr} on the concentration in the vicinity of the topological transition is shown in Fig. 3. The lowest frequency, ω_{t1}, of the transverse branch of the lattice vibrations undergoes a rapid decrease; ω_{t2} begins to decrease noticeably in the 5–6 at.% region; the longitudinal frequency within the boundaries of the α phase almost does not change. The values obtained for the derivatives $d\ln\omega_i^{cr}/dP$ are shown in Fig. 4. The initial values of the derivatives agree with the data of other authors.[16] Notice that the whiskers drawn on the presented curves parallel to the concentration axis indicate not the errors in the determination of C, but the effective shift in the latter upon the application of pressure, since the impurity and pressure have the same effect on the Fermi energy in the present case. The coefficient of conversion of pressure into an effective concentration was taken from the relation $C_c = aP_c$, where $C_c = 1.5$ at.% and $P_c = 6$ kbar are the critical values at which the $2\frac{1}{2}$-order phase transition occurs.

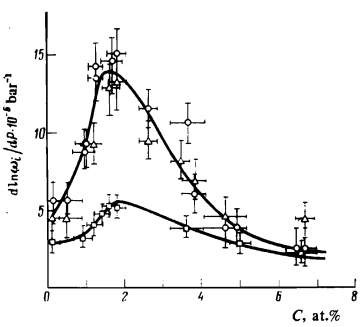

FIG. 4. Concentration dependences of the quantities $d\ln\omega_i/dP$ in the α phase of the In–Sn alloys: ○) ω_{t1}, △) ω_{t2}, □) ω_l.

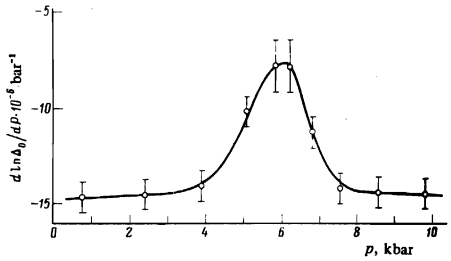

FIG. 5. Effect of pressure on the derivative $d\ln\Delta_0/dP$ for indium.

2. The effect of pressure

The same topological transition in the Fermi surface at 1.5 at.% Sn is also possible under the action of a 6-kbar hydrostatic pressure.[18] We observed a nonlinear variation of Δ_0 and ω_i^{cr} for pure indium in the 6-kbar region and a slower variation for the alloys in the region of zero pressure.[19] The derivatives $d\ln\Delta_0(P)/dP$ and $d\ln\omega_i^{cr}(P)/dP$ computed from these data for indium are shown in Figs. 5 and 6. Their values at low pressures agree with the data of other authors.[16,20]

4. DISCUSSION OF THE RESULTS OF THE EXPERIMENT

1. The linear components of the changes in the gap Δ_0 of indium upon the introduction of tin or the application of a hydrostatic pressure can be well explained within the framework of the theory of superconductivity with strong coupling, where changes in the electron-phonon interaction are connected first and foremost with the reconstruction of the phonon spectrum. The performed comparisons of the experimental values of $2\Delta_0/T_c$ [15] with the values computed from the well-known formula[21]

$$\frac{2\Delta_0}{T_c} = 3.52\left[1+5.3\frac{T_c^2}{\omega_0^2}\ln\left(\frac{\omega_0}{T_c}\right)\right], \qquad (1)$$

indicated a qualitatively correct explanation for the variation of the observable quantities. In the impurity case the coupling increases—the lattice "softens." Under the action of pressure the coupling decreases—the frequency energy increases.

The nonlinear changes in the gap Δ_0 are, in the first approximation, qualitatively explained by a theory[4] based on the concept of topological changes in the Fermi

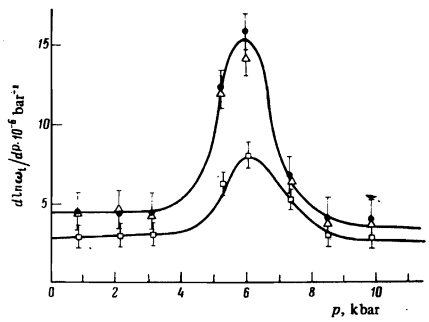

FIG. 6. Dependence on pressure of the quantities $d\ln\omega_i/dP$ for indium: ○) ω_{t1}, △) ω_{t2}, □) ω_l.

surface. The nonregular correction, $\delta\nu(\varepsilon)$, to the electron density that arises at the critical energy ε_c leads to the appearance of anomalies in T_c and the gap Δ_0 (and, accordingly, in dT_c/dP and $d\Delta_0/dP$). However, in the experimental $2\Delta_0/T_c$ dependence the nonlinearity is noticeable in the 2 at.% region,[15] which indicates a weakening of the coupling. The cause here may be the local shift of the phonon frequencies toward the region of higher energies.

2. Let us consider the causes of the anomalous variation of the phonon frequencies. The behavior of the linear components of the characteristic frequencies of the spectrum can be qualitatively explained within the framework of the adiabatic approximation of the electron-phonon interaction.[22] In principle, the nonlinear changes in the spectrum of the lattice vibrations can be explained in the same way as a direct consequence of an electron-phonon interaction that changes upon the appearance of a new electron cavity at some point p_c. Let us use the explicit expression for the renormalization of the phonon frequency[23]:

$$\Delta\omega = \int \frac{|M|^2(n_{\mathbf{p}}-n_{\mathbf{p}+\hbar\mathbf{q}})}{\varepsilon_{\mathbf{p}}+\hbar\omega-\varepsilon_{\mathbf{p}+\hbar\mathbf{q}}}d^3p, \tag{2}$$

where $\varepsilon_{\mathbf{p}}$ is the energy of an electron with momentum $\mathbf{p}$, $n_{\mathbf{p}} \equiv n(\varepsilon_{\mathbf{p}})$ is the Fermi distribution function, $|M|^2$ is the square of the modulus of the transition matrix element, in which are included all the constant factors. Noticeable changes in the frequencies will occur upon the fulfillment of the condition

$$\varepsilon(\mathbf{p}_c)-\varepsilon(\mathbf{p}_c+\hbar\mathbf{q}_c)\sim\hbar\omega(q_c). \tag{3}$$

Let us assess the character of the change $\Delta\omega$. Let there arise at the point p_c a new electron cavity with the dispersion law

$$\varepsilon_p=\varepsilon_c+p^2/2m^*; \tag{4}$$

here m^* is the effective electron mass in the newly formed hole. For the estimate let us set $|M|^2 \sim$ const. At $T=0$ we have

$$\Delta\omega\sim-\ln\left|\omega-q\left[\frac{2}{m^*}(\varepsilon-\varepsilon_c)\right]^{1/2}-\frac{\hbar q^2}{2m^*}\right|=-\ln|(\varepsilon-\varepsilon_c)^{1/2}-(\varepsilon^*-\varepsilon_c)^{1/2}|, \tag{5}$$

where

$$\varepsilon^*=\varepsilon_c+\frac{m^*}{2q}\left(\omega-\frac{\hbar q^2}{2m^*}\right)^2$$

is the energy at which the strongest singularity in the phonon spectrum arises. The asymptotic behavior of the derivative $d(\Delta\omega)/d\varepsilon$ near this point has the form

$$\frac{d(\Delta\omega)}{d\varepsilon}\sim\frac{\omega}{(\varepsilon-\varepsilon_c)^{1/2}[(\varepsilon^*-\varepsilon_c)^{1/2}-(\varepsilon-\varepsilon_c)^{1/2}]}. \tag{6}$$

Using the quantity

$$\partial(\varepsilon-\varepsilon_c)/\partial C_{\mathrm{Sn}}=1.8 \text{ mV/at.\% Sn}$$[24]

and the value $C_c=1.7$ at.% Sn at the maximum of the derivative $d\omega_t/dP \sim d(\Delta\omega)/d\varepsilon$, we see that $\varepsilon^*-\varepsilon_c \sim 1$ meV. For the fulfillment of this equality, as well as of the relation (3), we require a small value of the quantity $\mathbf{q}$, which value will not in this case correspond to critical frequencies. Under the vector $\mathbf{q}$ we should understand the difference $\mathbf{q}=\mathbf{q}_c-\mathbf{q}_0$ ($\mathbf{q}_0$ is a vector joining two equivalent critical points in reciprocal space). In indium such equivalent points will be the W points, the distance between which is close in magnitude to the vector $\mathbf{q}=[001]$. It is possible that for indium the three observed characteristic frequencies ω_{t1}, ω_{t2}, and ω_l correspond to one and the same $\mathbf{q}_c$ wave vector.

In the above-presented analysis the electron-phonon interaction matrix element was assumed to be a constant quantity. In reality, however, there can be anomalies in its structure.[25] Let us write it in the Bloch representation:

$$|M_{pp',\lambda}|^2=\frac{N|I_{pp',\lambda}|^2}{M\omega_{q,\lambda}\varepsilon(q,0)}, \tag{7}$$

where N is the number of ions in the metal, M is the ion mass, $\varepsilon(q,0)$ is the permittivity, $I_{pp',\lambda}$ is the matrix element of the electron transition from the state $\mathbf{p}$ into the state $\mathbf{p}'$ in the perturbing ionic field:

$$I_{pp',\lambda}=\int\Psi_{p'}^*[\hat{e}_{q,\lambda}\nabla V(q)]\Psi_{p'}d^3r; \tag{8}$$

here $\hat{e}_{q,\lambda}$ is the phonon polarization vector, $V(q)$ is a pseudopotential, Ψ_p^* and $\Psi_{p'}$ are the electron wave functions in the states $\mathbf{p}$ and $\mathbf{p}'$. For the electron states at the Brillouin-zone boundary the wave functions should be chosen in the form of a sum of several orthogonalized plane waves. As a result of the mixing of the plane waves the magnitude of the matrix element $I_{pp',\lambda}$ in the Bragg planes decreases abruptly,[26] which can lead to the renormalization of the frequencies. Let us note that this contribution coincides in sign with the observed change.

Finally, as a result of the interaction of the Fermi surface with the Brillouin-zone boundaries, a lattice instability can develop. This was pointed out by Galkin *et al.*,[27] and a rigorous theoretical analysis was recently published by Dagens.[28] The experimental data for indium[24] show that the values of the interband gaps at the critical points, where the change in the topology of the Fermi surface occurs, are smaller than the Debye energy. Such a situation leads to considerable interband transitions between the "new" and the "old" bands transitions which can lead to changes in the phonon dispersion.[27,29] According to Dagens, there arises a complete Kohn-anomaly surface at the critical energy ε_c for each $\mathbf{q}$ joining the new electron cavity with the old. The singularity in the polarization operator leads to logarithmic singularities (of the form $a\ln a$) in the phonon spectrum. Here a is a dimensionless parameter proportional to the energy difference $z=\varepsilon_c-\varepsilon_F$. Allowance for these corrections to the free energy of the phonons leads to the dependence $\delta E_{\mathrm{ph}} \sim -z^{3/2}$. The presence of this anomaly even at low temperatures can lead to a lattice instability, which again develops as the critical energy is approached. Since there are already in indium very soft transverse modes in the phonon spectrum,[30,31] this can manifest itself in an instability of the elastic moduli, first and foremost of the shear modulus $\frac{1}{2}(c_{11}-c_{12})$. The

observed nonlinear variation of the lattice parameters in indium alloys[6] at impurity concentrations when topological reconstructions of the Fermi surface occur are, possibly, a consequence of this. Although there are no comprehensive x-ray structural analysis data on the In-Sn alloys investigated by us (i.e., in the region of low concentrations), a tendency towards a nonlinear variation of the lattice parameters can be seen from a detailed analysis of the results of the measurement of the lattice parameters for the alloys InCd and InSn[6] in the α phase.

3. Our data agree with the behavior, observed in specific-heat measurement experiments, of the Debye temperature of these alloys.[32] In both cases the behavior of the lattice is reflected at large values of the **q** vector after its reconstruction.

In the case of experiments on the effect of pressure on the behavior of the lattice, the corresponding quantitative connection between the nonlinear variation of the frequency and the volume can be estimated, using the Grüneisen relation

$$\gamma = -d\ln\omega/d\ln V, \tag{9}$$

from which $\Delta\omega/\omega = -\gamma\Delta V/V$. Using the values $\gamma = 2.5$ and $\Delta V/V = -0.01$, computed from x-ray structural measurements of the lattice parameters of indium under pressure[8b] for the pressure range from 5 to 8 kbar, where the anomalous changes in the lattice are observed, we have $\Delta\omega/\omega = 0.025$, which is close to the values observed in tunneling experiments and thus indicates a change in the compressibility. The anomalous behavior of the lattice under pressure was also observed in ultrasound velocity measurements on coarse-grained indium samples[33] in the 15-kbar region. The overestimated pressure value is clearly due to the highly nonhydrostatic conditions that obtained in the chamber, a fact which was noted by the authors.

4. Let us estimate the contribution of the phonons to the nonlinear part of the variation of the derivative dT_c/dP. For this purpose let us differentiate the well-known MacMillan formula

$$T_c = \frac{\langle\omega\rangle}{1.2}\exp\left[\frac{-1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right] \tag{10}$$

with respect to pressure as a parameter. Here

$$\lambda = 2\int\frac{g(\omega)}{\omega}d\omega$$

is the electron-phonon coupling constant, μ^* is a Coulomb pseudopotential, and $g(\omega) = \alpha^2(\omega)F(\omega)$ is the electron-phonon interaction function. We have

$$\frac{d\ln T_c}{dP} = 2\left\{1 - \frac{1.04(1+0.38\mu^*)}{[\lambda-\mu^*(1+0.62\lambda)]^2}\right\}\left\langle\frac{d\ln\omega}{dP}\right\rangle - \left(\frac{\overline{d\ln\omega}}{dP}\right) + \text{electronic contribution}, \tag{11}$$

where

$$\left\langle\frac{d\ln\omega}{dP}\right\rangle = 2\int\frac{d\ln\omega}{dP}\frac{g(\omega)}{\omega}d\omega/\lambda, \tag{12}$$

$$\left(\frac{\overline{d\ln\omega}}{dP}\right) = \int\frac{d\ln\omega}{dP}g(\omega)d\omega \Big/ \int g(\omega)d\omega. \tag{13}$$

Using the values $\lambda = 0.82$ and $\mu^* = 0.125$,[16] we have

$$\frac{d\ln T_c}{dP} = -2.4\left\langle\frac{d\ln\omega}{dP}\right\rangle - \left(\frac{\overline{d\ln\omega}}{dP}\right) + \text{electronic contribution}. \tag{14}$$

For the linear part $(d\ln T_c/dP)_0 = -15.6\times10^{-6}$ bar^{-1} [24] this relation is fulfilled when the generalized Grüneisen parameter

$$\gamma^* = \left\langle\frac{d\ln\omega}{dP}\right\rangle = \left(\frac{\overline{d\ln\omega}}{dP}\right) = 4.6\cdot10^{-6}\ \text{bar}^{-1}.$$

Since its value agrees with the experimental data, the basic formula can be used to estimate the nonlinearities in $d\ln T_c/dP$. The phonon-induced change in this quantity is negative, and attains at the maximum (as far as can be judged from the tunneling data) a value lying within the limits from -0.8×10^{-5} bar^{-1} to -3.4×10^{-5} bar^{-1}. At the same time it is experimentally observed that $\delta(d\ln T_c/dP) = 0.17\times10^{-5}$ bar^{-1}. It follows from this that the $\delta(d\ln T_c/dP)$ curve that is measured in an experiment in In-Sn alloys and in indium under the action of pressure is, possibly, determined by the contributions of two mechanisms: a positive-electronic, and a negative-phonon, mechanism.

5. CONCLUSION

We have investigated the electron-phonon interaction and the lattice dynamics of indium undergoing the $2\frac{1}{2}$-order phase transition. The nonlinear variation of the superconducting energy gap confirms the theoretical predictions.[4] The observed behavior of the phonon frequencies and the data of other authors on the measurement of the various physical characteristics reflect the complex nature of the behavior of the lattice of this metal, the quantitative description of which meets with certain difficulties. Besides the changes in the spectrum, changes which are of the Kohn character and which were pointed out by Makarov,[34] a contribution by the anomalies in the matrix element of the electron-phonon interaction is possible. It can also be surmised that, in indium, there arises in the low-frequency region of the phonon spectrum at the time of the topological transition an essential singularity that gives rise to a local lattice instability.

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