Exponential contribution to the electric resistance of the superconducting compound $Nb_3Al_{0.75}Ge_{0.25}$ and the anomalous temperature dependence of its crystal-lattice parameter

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Results are presented of measurements of the temperature dependence of the electric resistance (61–300 K) and of the crystal-lattice parameter (70–310 K) of a high-temperature superconducting compound $Nb_3Al_{0.75}Ge_{0.25}$ with A15 lattice. It is shown that an important contribution to the resistance is made by scattering of electrons by phonons that correspond to the low-frequency peak of the density of states. The scattering can be expressed by a term $\exp(-T_0/T)$, where $T_0 = 117.0 \pm 6.8$ K. The temperature dependence of the thermal expansion coefficient is found to change abruptly at $T \sim T_0$. No lowering of the crystal-lattice symmetry of $Nb_3Al_{0.75}Ge_{0.25}$ is observed.

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The $Nb_3(Al, Ge)$ compounds occupy a special place among the super-conductors with β–W (A15) lattice, in that unlike other high-temperature superconductors ($T_c \geqslant 20$ K) they are thermodynamically stable in a wide range of temperatures[1)] (Ref. 1). Even though the existence of the high-temperature $Nb_3(Al, Ge)$ superconducting compounds has been known for a long time[2] and the influence of various factors on the critical superconductivity parameters have been investigated in sufficient detail (see, e.g., Refs. 3 and 4), very little is known about the behavior of the electronic and lattice properties of these super-conductors at low temperatures. Thus, the presence of a maximum of the magnetic susceptibility of the alloy $Nb_3Al_{0.75}Ge_{0.25}$ at $T \sim 50$ K has been demonstrated.[5] For films of the alloy $Nb_{3-0.56}(Al_{0.28}Ge_{0.72})_{1+0.56}$, which is quite far from stoichiometry, the presence has been established of an exponential contribution of the form $d\exp(-T_0/T)$, where T_0 is a certain characteristic temperature, to the electric resistance at low temperatures.[6] In essence, what remains open is the question of the structural instability of $Nb_3(Al, Ge)$, since the published data on the possible loss of crystal-lattice stability are extremely contradictory: according to certain data the temperature T_m of the martensitic transformation of $Nb_3Al_{0.75}Ge_{0.25}$ is ~105 K (Ref. 7), and according to other $T_m < 24$ K (Refs. 8, 9).

The appearance of a contribution, with an exponential temperature dependence, to the resistance of superconducting compounds with A15 structure has recently atrracted much attention (see, e.g., Refs. 10 and 11). The point is that the empirical equation for the temperature dependence of the resistance of these superconductors[10]

$$R(T) = R_0 + b_n T^n + d_n \exp(-T_0/T) \qquad (1)$$

contains a temperature T_0 that characterizes a certain thermally activated electron-scattering process; this temperature practically coincides with the phonon energy in the vicinity of the low-frequency maximum of the phonon state density.[11] That the phonon spectrum really has a low-frequency peak follows directly from the results of tunnel investigations and experiments on inelastic neutron scattering. [We must note immediately that no such investigations were performed for $Nb_3(Al, Ge)$ alloys.] Following Ref. 10, the presence of an exponential term in the temperature dependence of the resistance can be attributed to intraband scattering of electrons by phonons or to electrons by phonons or to electron–phonon scattering with umklapp, while the temperature T_0 can be regarded as a measure of the energy of the transverse acoustic phonons in the ⟨110⟩ direction. The external character of the phonon spectrum of superconductors with A15 structure should obviously lead to the appearance of anomalies of not only the electronic but also the lattice properties.

To establish a correlation between the behavior of the electronic and lattice properties of superconducting compounds with A15 structure, whose phonon spectra have a low-frequency maximum, we have carried out precision measurements of the temperature dependences of the electric resistance and of the crystal-lattice parameters of the high-temperature superconductor $Nb_3Al_{0.75}Ge_{0.25}$ at low temperatures. On the basis of these measurements we were able also to ascertain finally whether the superconductors $Nb_3(Al, Ge)$ have or have not a structural instability in the temperature interval 60–300 K.

All the measurements were performed on samples cut from an ingot of $Nb_3Al_{0.75}Ge_{0.25}$ obtained by a metallurgical technique. (The samples were cut by the electric-spark method and subjected to chemical etching.) The x-ray patterns showed, besides the phase with the A15 structure, weak lines of the γ phase of $Nb_3(Al, Ge)_3$. The temperatures of the start (T_c^s) and the midpoint ($T^{1/2}$) of the superconducting transition were respectively 19.30 and 19.15 K.

The electric resistance was measured at 61–300 K by a standard four-probe technique.[12] The error in the measurement of the relative resistance ($R_T/R_{273\ K}$) did not exceed 10^{-4}, and the error in the measurement of

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the temperature with a platinum resistance thermometer was ~0.05 K. For the structure investigations at 77–310 K we used a low-temperature attachment to an x-ray diffractometer.[13] The diffraction line (622) from a cubic lattice of the β-W type was registered in Cu$K\alpha$ radiation at large diffraction angles ($2\Theta \approx 162°$). The relative error ($\Delta a/a$) in the measurement of the crystal-lattice parameter was in this case $\sim 2 \cdot 10^{-5}$. The temperature was measured with a chromel-alumel thermocouple accurate to ∓ 0.5 K.

The measured temperature dependence of the crystal-lattice parameter of $Nb_3Al_{0.75}Ge_{0.25}$ are shown in Fig. 1.[2)] No diffraction-line splitting which could indicate a lowering of the crystal-lattice symmetry were observed in the temperature interval 77–310 K. This indicates unequivocally that the A15 crystal lattice does not lose stability in the indicated temperature interval. What is unusual is the appearance of a characteristic "knee" of the $a(T)$ curve at temperatures below 120–130 K, which attests to a rapid decrease of the linear expansion coefficient when the temperature is lowered (inset in Fig. 1).

The temperature dependence of the relative electric resistance of the $Nb_3Al_{0.75}Ge_{0.25}$ sample is shown in Fig. 2. Outwardly, the plot of $R_T/R_{273\,K}$ resembles the analogous plots for the compounds Nb_3Sn, V_3Si, V_3Ge, and other,[6,10,14,15] whose resistance is described by an equation of the type (1) in which usually $1 \leq n \leq 2$. The reduction of our data by a functional-minimization computer method has made it possible to describe the temperature dependence of the resistance by the equation

$$R_T/R_{273\,K} = 0.476 \pm 0.010 + (7.61 \pm 0.25) \cdot 10^{-6} T^{1.8} + (0.521 \pm 0.007) \exp\left[-(117.0 \pm 6.8)/T\right]. \quad (2)$$

To find the exponent of T in the second term responsible for the electron-electron scattering,[10] the value of n in (1) was varied in a wide range: the parameters of Eq. (2) correspond to the value of n that minimizes the total deviation of the calculated curve from the experimental data.

The insets of Fig. 2 show the temperature dependence of the resistance temperature coefficient ($R^{-1}dR/dT$)

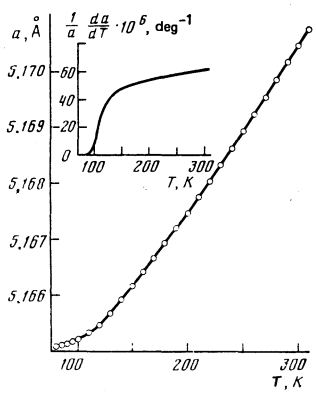

FIG. 1. Temperature dependences of the crystal-lattice parameter and of the linear-expansion coefficient of the compound $Ab_3Al_{0.75}Ge_{0.25}$.

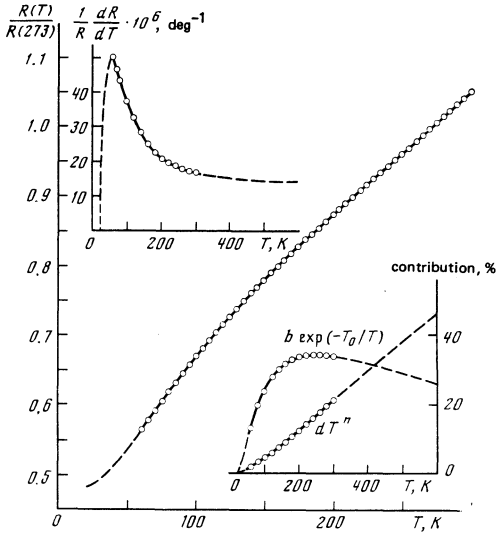

FIG. 2. Temperature dependences of the relative resistance, of the resistance coefficient $R^{-1}\,dR/dT$ (upper inset), and of the powerlaw and exponential contributions to the resistance (lower inset) of the compound $Nb_3Al_{0.75}Ge_{0.25}$.

and of the relative contributions made to the electric resistance of $Nb_3Al_{0.75}Ge_{0.25}$ by electron-electron (bT^n) and electron-phonon [$d\exp(-T_0/T)$] scattering. The shape of the $R^{-1}dR(T)/dT$ curve points to the possible appearance of a maximum of the resistance temperature coefficient at ~60 K. Although it was not possible to observe this maximum directly, the probability of its appearance is very high, since Eq. (2) describes with sufficient accuracy the temperature dependence of the resistance in a wide range of temperatures.

It is typical that the temperature of the hypothetical maximum of the temperature coefficient of the resistance is close to the temperature at which a maximum was observed in the paramagnetic susceptibility of $Nb_3Al_{0.75}Ge_{0.25}$ (Ref. 5). Since the appearance of a maximum in the temperature dependence of the thermal coefficient of resistance at 60 K is a natural result of the presence of power-law and exponential terms in the temperature dependence of the resistance, it must be assumed that the presence of the susceptibility maximum is likewise not connected with the strucural phase transition.

It is seen from the presented plots of the temperature dependences of the bT^n and $d\exp(-T_0/T)$ that at $T < 400$ K the electrons are scattered mainly by phonons.

Judging from the measured resistance of $Nb_3Al_{0.75}Ge_{0.25}$ at low temperatures, the phonon spectrum of this compound with A15 structure has a maximum at a phonon energy 117.0 ± 6.8 K. Moreover, since the exponential contribution to the resistance is sensitive to the height of the peak on the phonon spectrum,[6,11] the observed nontrivial effect that the exponential term prevails over the power-law term means that this peak is quite high. Comparison of the results of the electric-measurements and of the structural investigations can answer the question of how the ob-

served singularities of the phonon spectrum of the compounds $Nb_3Al_{0.75}Ge_{0.25}$ manifest themselves in its lattice properties.

As already noted, the dependence of the linear expansion coefficient on the temperature has an anomalous character; the high-temperature branch of the $\alpha(T)$ curve, for which a weak dependence of α on T is typical, goes over into the low-temperature dependence ($\alpha \sim T^n$) at temperatures close to T_0. This means that the thermal expansion of the investigated superconductor is due in the main to phonons with energy corresponding to the peak of the phonon state density, while the temperature T_0, which is a measure of this energy, plays the role of the low-temperature analog of the Debye temperature (the "true" Debye temperature is ~300 K, Ref. 16).

We note in conclusion that the observed presence of a strong electron-phonon interaction in $Nb_3Al_{0.75}Ge_{0.25}$, which manifests itself in the appearance of a "discernible" exponential term in the resistance and of characteristic singularities of the thermal expansion, agrees with the high values of the critical parameters of the superconductivity of this compound. No singularities whatever that might be due to the loss of stability of the crystal lattice at 61–310 K were observed in the temperature dependences of the thermal expansion and of the resistance of $Nb_3Al_{0.75}Ge_{0.25}$.

[1)] By thermodynamic stability is meant here the possible existence of a compound of stoichiometric composition A_3B down to the very lowest temperatures (for such superconductors as Nb_3Ge, Nb_3Ga, Nb_3Al, a phase composition A_3B with A15 lattice is realized only at quite high temperatures, while at low temperatures the stable compounds have a β-W lattice and are deficient in the B component).

[2)] Figure 1 (as well as Fig. 2) shows only part of the experimental points pertaining to one measurement run. The reproducibility of the $\alpha(T)$ curve was quite satisfactory.

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