Influence of aluminum impurity atoms on the resistivity of V_3Ge

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The resistivity of polycrystalline V_3Ge, as well as of the alloys $V_3Ge_{0.88}Al_{0.12}$ and $V_3Ge_{0.75}Al_{0.25}$ was measured in the temperature range 4.2–300 K. The temperature dependence of the resistivity in the interval from T_c to 100 K is well described by the formula $\rho(T)=\rho_0+bT^2+d\exp(-T_0/T)$. The term quadratic in the temperature corresponds apparently to electron-electron scattering, while the exponential term corresponds to electron-phonon scattering. The coefficient b of the quadratic term increases when aluminum impurity atoms are introduced, meaning an increase in the density of the electron states. The value of T_0 correlates with the energy positions of the low-frequency maxima of the phonon state densities, which are obtained in measurements of inelastic neutron scattering and which change when the impurities are introduced. The impurity-induced resistivity change $\Delta\rho/\rho_0$ varies linearly with the impurity concentration. With increasing temperature, $\Delta\rho/\rho_0$ goes through a maximum at $T\sim 100$ K and has a negative derivative at higher temperatures. The causes of this particular behavior of $\Delta\rho/\rho_0$ are analyzed.

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1. INTRODUCTION

We have carried out systematic investigations of the changes that occur in the electron and phonon spectra of superconducting systems after alloying, and of the correlations of these changes with the superconducting transition temperature T_c (Ref. 1). In Refs. 1 and 2 were investigated the specific heat, the magnetic susceptibility, and the inelastic-scattering spectra of the superconducting compound V_3Ge, which has a type A-15 structure, and of its alloys with aluminum. It turned out that introduction of several percent of aluminum leads to appreciable changes of the properties of the alloyed system from the initial V_3Ge. Namely: the magnetic susceptibility and the electronic specific heat increase sharply, and with it the density of the electronic states on the Fermi surface. Local vibrational states connected with the light impurity Al appear in the phonon spectrum, but the spectrum as a whole becomes softer. The value of T_c is almost doubled. It was therefore of interest to investigate the influence of the restructuring of the phonon and electron spectra on the temperature dependence of the resistivity of V_3Ge. We note that there is still no universal accepted theory of the resistivity of compounds with A-15 structure, although some distinguishing features of the temperature dependence of the resistivity of such systems have already been noted.[3–6]

In the present study we measured the temperature dependence of the resistivity of V_3Ge and of the alloys $V_3Ge_{0.88}Al_{0.12}$ and $V_3Ge_{0.75}Al_{0.25}$ in the temperature range 4.2–300 K.

2. EXPERIMENT

The samples of V_3Ge and of its alloys with aluminum were prepared from electrolytic vanadium 99.8% pure, germanium 99.99% pure, and aluminum 99.999% pure, melted in an induction furnace in an inert atmosphere. The obtained ingots were annealed in quartz ampoules for 200 hours at 800°C. The composition cited is that of the initial charge. Metallographic and x-ray structure analysis has shown that the samples are practically single-phase (the content of the second phase does not exceed 2–3%). The superconducting transition temperatures T_c, the widths of the transitions ΔT_c, the resistivity ratio $\rho(300)/\rho(T_c)$, and the lattice parameters a of the investigated samples are listed in Table I. The samples for the measurement of the resistivity were cut by the electric-spark method and measured $1\times1\times8$ mm. The resistivity was measured by a four-contact method with direct current, the error not exceeding 0.4%. The accuracy of the measurement of the geometric factor s/l was ~1.5%; the resultant resistivity error was 2%.

3. MEASUREMENTS RESULTS AND THEIR DISCUSSION

The results of the measurements of the temperature dependence of the resistivity of pure V_3Ge and of the alloys $V_3Ge_{0.88}Al_{0.12}$ and $V_3Ge_{0.75}$ are shown in Fig. 1. As seen from the figure, the resistivity increases rapidly with increasing temperature in the interval up to 200 K, and at higher temperature the curve becomes flattened out.

It was shown in Refs. 4–6 that the resistivity of compounds with A-15 structure does not follow the usual low-temperature power law that holds for most metals and becomes linear at high temperatures. In Ref. 4 it was proposed to describe the temperature dependence of the resistivity at low temperatures by an expression of the form

TABLE I.

Samples	T_c, K	ΔT_c, K	$\frac{\rho(300)}{\rho(T_c)}$	a, Å
V_3Ge	6.9	0.1	8	4.781
$V_3Ge_{0.88}Al_{0.12}$	9.4	0.2	3.3	4.789
$V_3Ge_{0.75}Al_{0.25}$	12.8	0.3	2.4	4.795

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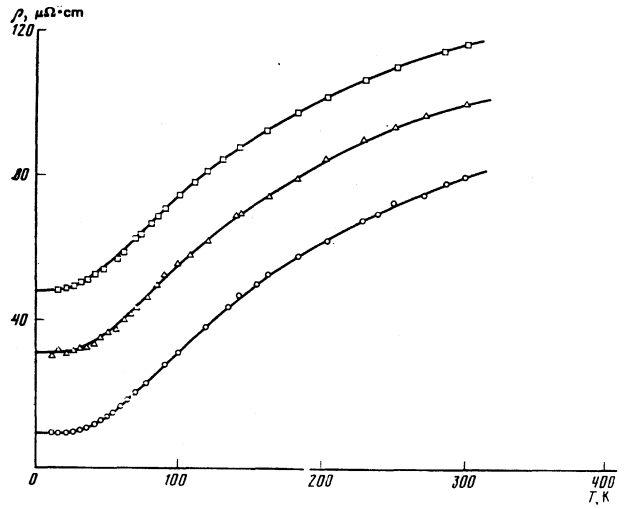

FIG. 1. Resistivity of V_3Ge_0(○), $V_3Ge_{0.88}Al_{0.12}$ (△) and $V_3Ge_{0.75}Al_{0.25}$ (□) in the range from T_c to 300 K.

$$\rho(T)=\rho_0+bT^2+d\exp(-T_0/T), \quad (1)$$

where ρ_0 is the residual resistivity, and b, d, and T_0 are empirical parameters.

An analysis of our data has shown that the resistivity of pure V_3Ge and of the alloys is described in the interval from T_c to 100 K by expression (1), with a mean squared deviation 0.5%. The values of the parameters ρ_0, b, d, and T_0 are listed in Table II. The error of the coefficients ρ_0 and T_0 is 2%, that of the coefficient d is 10%, and that of the coefficient b is 30%.

At low temperatures, the dominant term in the temperature dependent part of the resistivity is bT^2, which is usually attributed to electron-electron scattering.[4] In the present case the validity of this assumption is confirmed by the fact that when aluminum impurity atoms are introduced the coefficient b increases appreciably, a fact that correlated with the increase of the electronic-state density obtained from measurements of the specific heat and of the magnetic susceptibility of the same systems.[1] We call attention to the fact that in A-15 compounds the density of the electronic states has an anomalous temperature dependence and decreases rapidly with increasing temperature. As a result one can expect the contribution of the electron-electron scattering to decrease at high temperatures.

The exponential term in the resistivity corresponds apparently to electron-phonon scattering with umklapp.[4] Golovashkin[7] has shown that in intermetallides with A-15 structure a correlation exists between the energy position ω^* of the low-frequency maximum of the phonon state density and the parameter T_0 that characterizes the exponential term of the resistivity. The suggested cause of this correlation is the anomalously large contribution of the scattering of electrons by phonons with energy $\omega\sim\omega^*$ and wave vector $q\sim\pi/a$, which results from the singularities of the Fermi surface of compounds with A-15 structure. In the systems investigated by us, the parameter T_0, which characterizes the exponential term of the resistivity, decreases with increasing aluminum content, in accord with the softening of the phonon spectrum and the shift of the maxima of the density of the phonon states into the region of lower temperatures, as observed in experiments on inelastic neutron scattering.[3]

We have estimated the changes of the parameter ω^*. As follows from Ref. 2, in the system $V_3Ge_{0.75}Al_{0.25}$ the position of the low-frequency maximum of the phonon state density shifts by 4–5% compared with V_3Ge. In addition, the relative frequency shift between 300 K and 4.2 K for V_3Ge amounts to 3% and should amount to 6% for $V_3Ge_{0.75}Al_{0.25}$, if account is taken of the correlation between T_c and the anharmonicity.[8] As a result, the relative change of the impurity shift of the maximum of the phonon density of states is $\Delta\omega^*/\omega^*\sim 7$–8%, which correlates with the change of the characteristic temperature T_0 in Eq. (1), (see Table II).

Figure 2 shows the temperature dependence of the relative resistivity change $\Delta\rho/\rho_0$ connected with introduction of the impurity:

$$\frac{\Delta\rho}{\rho_0}=\frac{\rho_a(T)-\rho_a(0)-\rho_p(T)}{\rho_a(0)},$$

where $\rho_a(T)$ and $\rho_p(T)$ are respectively the resistivities of the alloy and of the pure compound V_3Ge at the temperature T, while $\rho_a(0)\equiv\rho_0$ is the residual resistivity of the alloy.

As seen from the figure, $\Delta\rho/\rho_0$ initially increases with temperature and has a maximum at $T\sim 100$ K, and then decreases and becomes negative at $T\sim 200$ K. It is most important than in the investigated alloys the temperature dependence of the resistivity is practically linear in the residual resistivity. This result contradicts the hypothesis advanced in a number of articles

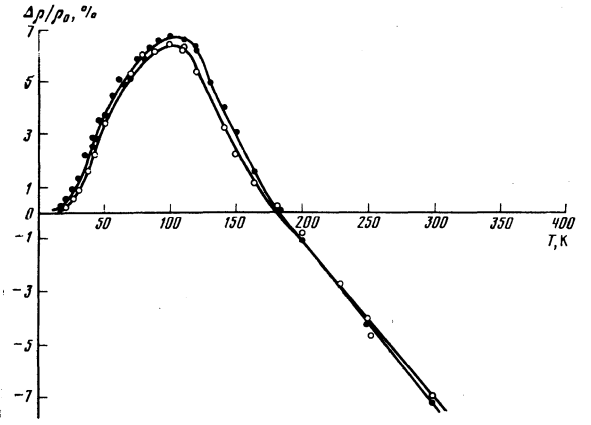

FIG. 2. Temperature dependence of the relative change $\Delta\rho/\rho_0$ of the impurity resistivity for the alloys $V_3Ge_{0.88}Al_{0.12}$ (●) and $V_3Ge_{0.75}Al_{0.25}$ (○)

TABLE II.

Samples	ρ_0, μΩ·cm	$b\cdot 10^{-3}$, μΩ·cm/K²	d, μΩ·cm	T_0, K
V_3Ge	8.92	0.4	90	158
$V_3Ge_{0.88}Al_{0.12}$	30.6	0.7	76	143
$V_3Ge_{0.75}Al_{0.25}$	48.4	0.9	63	132

(see, e.g., Ref. 9) that the resistivity "saturates" with increasing temperature on account of the smallness of the mean free paths of the electrons in type A-15 compounds.

It should be noted that in the investigated systems the mechanism that increases the resistivity and is connected with isotropization of the distribution of the electrons when scattered by impurities,[10] does not seem to contribute greatly to the observed change of the resistivity as a result of the large defect concentration.

We now discuss the possible causes of the negative resistivity change $\Delta\rho/\rho_0$ at high temperatures.

One of them, as shown in Ref. 11, is the anharmonicity of the lattice vibrations, and reflects directly the temperature dependence of the mean squared phonon frequency. We note that the correlation of T_c with the temperature shift of the phonon frequencies, which was noted in Ref. 8, allows us to conclude that the anharmonic effects are increased in the impurity system.

A second cause is the increase of the density of the electronic states upon introduction of the aluminum atoms. In contrast to the low temperatures, where electron-electron scattering predominates and the resistivity increases with increasing state density, at high temperatures the increase of the electron density leads to a decrease of the resistivity, since the number of carriers increases and the mean free path is determined by the electron-phonon scattering and is dependent of the number of electrons.

Furthermore, when a germanium atom is replaced by aluminum, the difference between the amplitudes of electron scattering by the matrix and impurity atoms, according to our estimates based on the data of Ref. 12, decreases the resistivity of the impurity system.

4. CONCLUSION

The temperature dependence of the resistivity of the investigated V_3Ge and of the alloys $V_3Ge_{0.88}Al_{0.12}$ and $V_3Ge_{0.75}Al_{0.25}$ in the interval up to 100 K is described by a sum of a quadratic term and an exponential term, the former apparently corresponding to electron-electron scattering processes, and the latter to electron-phonon scattering. The coefficient b of the quadratic term increases with increasing aluminum content in accord with the increase of the density of the electron states. The characteristic temperature T_0 in the exponential term correlates with the position of the low-frequency maximum in the density of the phonon states. In V_3Ge with impurity, the parameter T_0 decreases, a fact explained by the softening of the phonon spectrum. The negative sign of the impurity-induced resistivity change $\Delta\rho/\rho_0$ and of the change of its derivative with respect to temperature at high temperatures is explained by the increase of the density of the electronic states, by the enhancement of the anharmonicity, and by the negative difference between the amplitudes of the scattering of the electrons by the impurity atoms in the atoms of the initial crystal.

Thus, introduction of impurity aluminum atoms in V_3Ge, which causes a significant restructuring of the electron and phonon spectrum, does not change the character of the temperature dependence of the resistivity. All that increases quantitatively is the role of the electron-scattering processes, to which the lowering of T_c in impurity systems is usually attributed. In this case, however, T_c increases substantially (doubles) upon introduction of the impurity, and consequently the strengthening of the electron-phonon scattering does not lead apparently to "three-dimensionalization" of the electron spectrum in the impurity system.

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