# THE TEMPERATURE DEPENDENT PART OF THE IMPURITY RESISTANCE IN ALUMINUM ALLOYS WITH GOLD OR SILVER

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The electrical resistance of dilute solutions of Au in Al (0.041, 0.064 and 0.13 at.% Au) and of Ag in Al (0.048, 0.082, 0.153 and 0.58 at.% Ag) is measured in the 1.2—300°K temperature range. It has been discovered that the character of the low temperature behavior of the impurity resistance $\Delta\rho$ in alloys with a small content of impurities is determined by a term proportional to the concentration multiplied by the square of the temperature. At temperatures equal to about 100°K for the Al-Au alloys and at temperatures of about 60°K for the Al-Ag alloys, the function $\Delta\rho(T)$ passes through a maximum. Here, at high temperatures, the sign of the derivative $\partial\Delta\rho/\partial T$ in both groups of alloys is negative. A comparison is made of the experimental results obtained with the theory.[1] Singularities in the low temperature behavior of the resistance of the alloys are explained by the specific scattering of the electron by the vibrating impurity atom. The genesis and location of the maxima on the curves for $\Delta\rho(T)$ are connected with the features of the behavior of atoms in regions with dynamic disordering, which arise around the impurity atoms. The hypothesis is advanced that the sharp increase in the impurity resistance for $T \sim T_{max}$ is connected in Al-Ag with scattering of the electron principally by the impurity atom itself, and in Al-Au, with the scattering of the electron by the lattice vibrations, which are anomalously perturbed from the appearance of the so-called quasilocal frequency in the phonon spectrum.

## INTRODUCTION

$I$N a metal with impurity atoms, the amplitude of electron scattering changes from site to site, and statistical disordering of such a type leads to the appearance of residual resistance. Along with this, the impurity atoms vibrate, so that dynamic disordering of the system is present. This dynamic disordering can be significantly increased if for any reason the character of the oscillations of the impurity atom and its surroundings is appreciably different from the vibrations of atoms in the unperturbed lattice. As a result, chaotically situated regions appear, from which the electrons are scattered inelastically. Hence the momentum is not conserved in scattering from the impurity atom itself. Inelastic scattering of the electron from individual, randomly located regions with dynamic disordering leads to the result that the impurity resistance $\Delta\rho$ begins to depend on the temperature. It is important that the character of the temperature behavior of $\Delta\rho$ is determined simultaneously by the nonconservation of momentum and by the difference in the amplitudes of the scattering and of the specific oscillations of the region of atoms of the lattice isolated about the impurity (see[1]).

In a recent work,[2] undertaken by us for testing the conclusions of the theory proposed in[1], our concern in the experimental results was chiefly in the effect of an anomalous rearrangement of the phonon spectrum in Mg-Pb on the temperature behavior of $\Delta\rho$. Although the studied weak substitution solutions in the electron spectrum also changes, nevertheless we succeeded in isolating that part of the temperature range where the contribution to $\Delta\rho(T)$ from a rearrangement of the phonon spectrum is dominant. We emphasize that this effect is observed only for sufficiently small amounts of the second component, when one can speak of the dynamic disordering of the individual regions.

Unfortunately, probably because of traces of paramagnetic impurities, to which the temperature-dependent part of the impurity resistance $\Delta\rho$ for Mg is also as sensitive, as, for example, for Cu,[3] we did not succeed in making a comparison between the experiment and the theory in regions of very low temperatures. In the present work, therefore, we have selected Al as the matrix. As is known, it is practically insensitive to paramagnetic impurities, and we sought to find in the impurity resistance effects connected with the nonconservation of momentum in the scattering of the electron by the impurity oscillations, which would lead to the appearance in $\Delta\rho$ of a component proportional to $cT^2$, where c is the impurity concentration.

Special interest attaches to the behavior of $\Delta\rho(T)$ over a wide range of temperatures, up to room temperature. The point is that by measuring the residual resistance we obtain definite information on the character of the amplitudes of scattering by the atoms of the solution. The theory proposed in[1], in which $\Delta\rho(T)$ is determined by the dynamic disordering brought about by the introduction of the impurities, allows us to see how the temperature variation of $\Delta\rho(T)$ changes as a function of the amplitude of scattering by the atoms of the matrix and as a function of the scattering by the atoms of the impurity. Of course, the temperature variation of $\Delta\rho(T)$ depends also on the character of the oscillations of the atoms in the regions of dynamic disordering. The fact that the latter effect should be significant in Al alloys with heavy impurities (in particular, a quasilocal frequency should appear[4] in these alloys) follows indirectly from the results previously obtained from our researches, where the alloys Mg—Pb[5] and Ti-U[6] were studied (see also[7]). This also allowed us to hope for the possibility of the separation, in the impurity resis-

tance of alloys of Al with Au and Ag, of effects associated with the resonance rearrangement of the phonon spectrum in the region of low frequencies.

## METHOD OF MEASUREMENT

The Al-Au and Al-Ag alloys were weak substitution solutions. In the preparation of the samples, pure initial metals were used: Al (99.9999%), Ag (99.999%), and Au (99.999%). For the initial Al, the resistance ratio was $R(300°K)/R(4.2°K) = 10^4$. To avoid systematic errors associated with the different conditions of preparation of pure samples of Al and its alloys, the Al was prepared in the same crucible in which the alloys were prepared. After this, the resistance ratio became $R(300°K)/R(4.2°K) = 0.6 \times 10^3$. After smelting, the samples were extruded by means of a press through a hole of 0.8 mm diameter. Then they were forced through a series of Pobedit dies. As a result, the samples were produced in the form of wires of diameter 0.5 mm and length 60 mm. Then the samples were annealed in a helium atmosphere at ~550° C.

To determine the concentration of Au and Ag in the Al alloys, a method of activation by neutrons was used, which made it possible to carry out the analysis comparatively simply and with great accuracy. The results of the analysis are given in the table.

| Al — Au | | Al — Ag | |
|---|---|---|---|
| No. of sample | at . % Au | No. of sample | at . % Ag |
| 1 | 0.041±0.002 | 1 * | 0.048±0.005 |
| 2 | 0.064±0.003 | 2 * | 0,082±0.0008 |
| 3 | 0.130±0.007 | 3 | 0.153±0.004 |
|  |  | 4 | 0.580±0.006 |

*In these samples, traces of Ir were observed ( $1 \times 10^{-5}$ at.%)

For measurement of the electrical resistance of the samples studied, a method was used which has previously been described in[2,8]. The resistance of the samples and of the thermometers was measured by means of an ordinary potentiometer circuit with an R-308 potentiometer (sensitivity $10^{-8}$ V/mm). Here a double oil switch was used to switch the supply currents to the thermometers and the samples, in order to eliminate parasitic thermal emfs.

The quantities $\rho(T)$ and $\Delta\rho(T)$ were calculated from the data obtained, and also the relative change in the resistance of the alloys in comparison with the pure metal $[\Delta\rho(T) - \Delta\rho(0°K)]/c\rho_{Al}(273°K)$. In the calculation of $\rho$, the principal error lies in the inaccuracy of the determination of the geometric dimensions $(s/l)$, inasmuch as the error in the measurement of the value of the resistance R does not exceed ±0.1% for very low temperatures, and ±0.01% for high temperatures. In the calculation of the geometric factor $(s/l)$, the diameter and length were measured with the help of the horizontal comparator IZA-2. The total error in the measurement of the geometric factor $s/l$ amounted to ±0.4%. Thus the absolute resistance was measured with an accuracy greater than ±0.05%.

## RESULTS OF MEASUREMENTS AND THEIR DISCUSSION

We carried out measurements of the temperature dependence of the electrical resistance of pure Al and its alloys with Au and Ag of the composition shown in the region of temperatures 1.2—300°K. The results of these measurements are shown in Figs. 1—4.

Figure 1 gives the dependence of the impurity resistance on the concentration c of the second component. The impurity resistance $\Delta\rho(0)$ for Al-Ag is approximately an order of magnitude larger than $\Delta\rho(0)$ for the solution of Au in Al for the same values of concentration. This result is seen to be important for the analysis of the temperature behavior of the impurity resistance. As is seen from Fig. 1, in the range of values of concentration studied, the impurity resistance depends linearly on the concentration. It should be noted that, if the system of Al with Ag represents an unbounded region of solid solutions, then for Al with Au this region, from the side of Au, is small, as is known, less than 0.2 at.% Au.[9]

Figures 2—4 illustrate the temperature behavior of
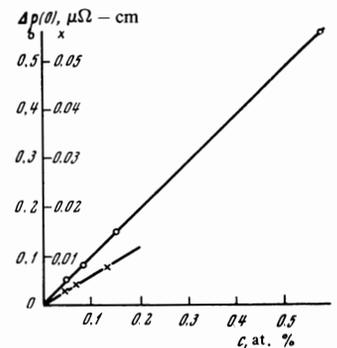
FIG. 1. Dependence of the residual impurity resistance on the percentage composition of the second component for the alloys: X — Al–Au and O — Al–Ag (at upper left, $\Delta p(0)$, $\mu\Omega$–cm).
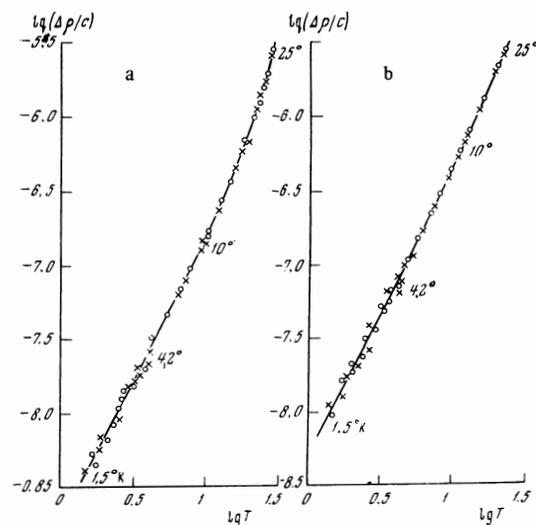
FIG. 2. Temperature dependence of the impurity resistance Al–Ag $\Delta p/c$ in the region of low temperatures: a — for Al–Ag alloys; O — c = 0.048 % at. % Ag, X — c = 0.082 at. % Ag and b — Al–Au alloys: O — 0.041 at. % Au, X — 0.94 at. % Au.

FIG. 3. Temperature dependence of the relative change in the resistance for Al−Au alloys: $O$ − c = 0.041 at. % Au, X − c = 0.064 at % Au, $\square$ − c = 0.131 at. % Au.
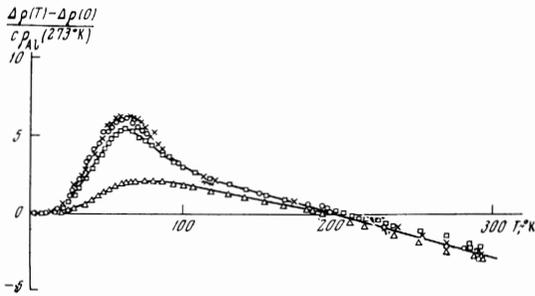


FIG. 4. Temperature dependence of the relative change in the resistance for Al−Ag alloys: $O$ − c = 0.048 at. % Ag, X − c = 0.082 at. % Ag, $\square$ − c = 0.153 at. % Ag, $\triangle$ − c = 0.58 at. % Ag.

the impurity resistance $\Delta\rho(T)$. Here, for alloys with a small impurity content (0.041; 0.064 at.% Au and 0.048; 0.082 at.% Ag), within the limits of accuracy of the measurements, $\Delta\rho(T)$ changes linearly with the concentration. The latter result makes it possible, in particular, to compare the corresponding experimental curves with the theory developed in[1], where, in the free electron approximation, linear effects in the impurity resistance (with respect to the concentration) are isolated. Later in the present work, we shall concern ourselves with such effects alone.

Figure 2 shows the low-temperature behavior of $\Delta\rho(T)$. By using the experimental results obtained by us and by defining the exponential power in the temperature in the expression for the resistance as the slope of the curve $\lg\rho = f(\lg T)$, we found that, in the temperature range $1.5-10°$K, $\rho_{Al}$ changes proportionally to $T^3$, while the impurity part of the resistance, more precisely, $\Delta\rho/c$, where $\Delta\rho/c = [\Delta\rho(T) - \Delta\rho(0)]/c$, varies like $T^2$. This is also seen from Fig. 2. In metals of the Al type, the correction to the Bloch resistance because of electron-electron collisions is assumed to be small. Therefore, it is difficult to assume that the behavior of a small number of impurities changes the character of the Coulomb interaction so strongly. Keeping this in mind, we associate the component in $\Delta\rho$ discovered in our present research (which is proportional to the concentration multiplied by the square of the temperature) with another mechanism, namely, with the scattering of the electron by the impurity oscillations. The noncon-

servation of momentum in the scattering of the electron by the impurity oscillations, and hence the possibility of electron scattering in such inelastic collisions and at large values of the angle as $T \to 0$, again leads to the appearance in $\rho$ ($T \to 0$) of a significant component, which is proportional to $cT^2$ (see[1]).

From the data we obtained in measurements of the electrical resistance of Al and its alloys, we calculated the relative change in the resistance for Al-Au and Al-Ag alloys with different contents of the second component in the range $1.2-300°$K. The corresponding curves are shown in Figs. 3 and 4. As is seen, both in Al-Au and Al-Ag alloys, the curve $\Delta\rho(T)$ passes through a maximum, but the location of the maximum is different: for Al-Au, $T_{max} \sim 100°$K and for Al-Ag, $T_{max} \sim 60°$K, and then the decrease is monotonic.

Proceeding to a comparison of these curves with theory,[1] it should be noted that from the viewpoint of theory, the most interesting region is that of intermediate temperature. At intermediate temperatures, effects should appear most directly in the impurity resistance, in correspondence with[1] that are connected with the presence in the phonon spectrum of a quasilocal frequency (QLF).

In carrying out numerical calculations within the framework of[1], we have assumed that the value of the parameter $(\Delta a)^2_{eff}$, where $\Delta a = a_1 - a_0$ is the difference in the scattering amplitudes on the impurity in the given alloy and on an atom of the matrix, can be determined approximately from the data on the residual resistance. The parameter $(\Delta a)^2_{eff}$ obtained from these data was used for an estimate of the contribution to $\Delta\rho(T)$ associated with the scattering on the oscillating impurity. As follows from the results of our measurements, $\Delta\rho_{Al-Ag}(0)/\Delta\rho_{Al-Au}(0) \gtrsim 10$, i.e., the effect of scattering on the oscillating impurity should be more sharply expressed in the Al-Ag alloys than in the Al-Au alloys.

The computed curve shown in Fig. 5a illustrates
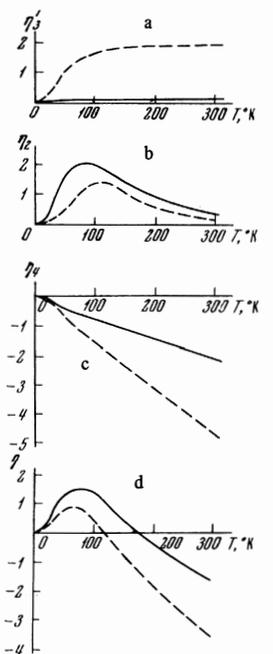


FIG. 5. Calculated theoretical curves of the relative contributions to the impurity resistance of different mechanisms of electron scattering: a) $\eta'_3 = \eta_3(T) - \eta_3(0)$ − scattering by impurity oscillations; b) $\eta_4$ − scattering on the deformed phonon spectrum, c) $\eta_4$ − interference between the scattering on the impurity and on atoms of the matrix, d) $\eta$ − computed curves of the temperature behavior of the impurity resistance.

what has been pointed out. In the present work, the contributions to the impurity resistance from various mechanisms of scattering has been marked in the same way as in[1], so that $\eta = \rho(T)/c\rho_0(2/3)\Theta$; here $\Theta$ is the Debye temperature of Al. We note that the character of the behavior of the curves in Fig. 5a does not change even with consideration of the rearrangement of the electron spectrum from the introduction of the impurity (see[10]). So far as the calculated curves are concerned, here and below we shall use the Debye approximation for the phonon spectrum of the matrix.

From the scattering of electrons by oscillations of the impurity, we proceed to the scattering of the electron from the region around the impurity atom, including the impurity itself. In our case, the properties of atoms in such regions differ sharply from the properties of the remaining atoms of the matrix. This difference and, as a consequence, the resonant increase in the density of phonon states in the region of low frequencies, and the appearance of QLF, should be especially clearly evident in solutions of Au in Al, which is also demonstrated by the curves of Fig. 5b. Here, we have used the data given in[11] for the calculations of the scattering amplitudes on the Al atoms. In the region of large transmitted momenta, the pseudopotentials are determined with limited accuracy. Therefore, it was assumed for all q (q characterizes the scattering angle of the electron) where $a_0(q) > 0$, that the corresponding values of $a_0(q)$ are only half those given in[1].

Allowance for the effect of a change in the force constants in the transition from the matrix to the alloy in the impurity resistance due to deformations of the phonon spectrum does not qualitatively change the character of the curves under discussion in the temperature region where scattering from the dynamically disordered regions around the impurities predominate.[10] By taking the above into account, we assume that the strong increase in the impurity resistance in the intermediate region of temperatures $T \lesssim T_{max}$ is connected in the Al-Ag alloys with the preferential scattering by the oscillations of the impurity itself, and in the Al-Au alloys with the scattering of the electron on the deformed phonon spectrum.

So far as the behavior of the impurity resistance in the classical temperature limit is concerned, we, in accord with[1], limit ourselves here to an account of the interference between the scattering of the electron by the impurity and by the atom of the matrix. Taking the product $\Delta a(q)a(q)$ as some effective constant, the value and sign of which are dictated by the experimentally observed quantities and by the sign of $\Delta\rho(T)$ as $T \gtrsim \Theta$, we have for the corresponding contributions of

$\eta_4$ to the impurity resistance the curves of Fig. 5c.

By summing the contributions to the impurity resistance associated with the different mechanisms, such that $\eta = \eta'_3 + \eta_2 + \eta_4$, we obtain computed curves that describe the temperature variation of the impurity resistance over the entire temperature range in both groups of alloys. By varying the numerical values of the parameters $a_0^2$ $(\Delta a)^2_{eff}$ and $(a\Delta a)_{eff}$ within known limits, one can achieve better agreement between experimental and theoretical curves. It should be noted that the significant quantitative divergence between these curves, in particular for Al-Ag, can be connected with the necessity of a more complete account than in[1] of the features of the Fermi surface of Al in constructing the solution of the kinetic equation.

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