Effect of lead impurity on the temperature dependence of the electric resistance of indium

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An experimental study was made of the temperature dependence of the electric resistance in a zero field and of the transverse magnetoresistance of single-crystal indium samples containing from 10^{-3} to 1 at.% lead impurities at low temperatures. The measurements were performed with identical samples and special attention was paid to monitoring the amount of lead in the sample and uniformity of its distribution. The temperature-dependent part of the resistance in a zero field varies like $\beta(c)T^5$ at $T \leq 4.2$°K (c is the lead concentration). A strong and nonlinear dependence of β on c is observed. The value of $\beta(c)$ increases by 2.5 times when 10^{-2} at.% Pb is introduced. For the initial indium, $\beta = 2.2 \times 10^{-13}$ ohm cm/deg^5. A peak in the temperature dependence of the impurity part of the resistance is observed at $T = 8$°K. The temperature-dependent part of the transverse magnetoresistance varies like $\alpha(c)T^4$. For the initial indium, $\alpha = 3.3 \times 10^{-12}$ ohm cm/deg^4. The value of $\alpha(c)$ increases by 20% when 5×10^{-3} at.% Pb is introduced, whereas $\beta(c)$ is doubled in this case. It is shown that the experimental results are consistent with the Kagan–Zhernov theory.[2]

In experimental investigations of the influence of impurities on the temperature dependence of the resistivity of metals and of the deviation from the Matthiesen rule, the parameters varied in the experiment are usually the type and concentration of the impurity. In spite of the large number of experimental data obtained to date and the large number of proposed theoretical models, the question of the nature of the deviation from the Matthiesen rule remains unexplained[1]. It can be assumed that introduction into the experiments of some additional controllable parameters would cast light on the situation.

In connection with recent theoretical papers[2,3], in which the deviations from the Matthiesen rule are attributed to the isotropizing effect of the impurities on the nonequilibrium electron distribution function, arguments were advanced that the magnetic field should exert an effect similar to that of the impurities[3]. Regardless of the validity of these arguments, an investigation of the deviation from the Matthiesen rule in the presence of a magnetic field, and the comparison of these measurements with the results of experiments in zero field, are of great interest.

We have investigated the temperature dependence of the resistivity on the transverse magnetoresistance in a strong magnetic field, using the same indium samples with small lead content.

THE EXPERIMENT

In addition to introducing into the experiment a new parameter, namely the magnetic field, much attention was paid by us to the choice of the material, to the control of the homogeneity of the impurity distribution over the sample, and to a determination of the impurity concentration. The method of sample preparation and the components were chosen in such a way that the investigated impurity was dominant in the entire range of investigated concentrations, and the influence of all other impurities and crystal-structure defects in the samples was negligible. The single-crystal indium samples with lead impurity were prepared from indium with resistivity ratio $\rho(300°K)/\rho(0°K) = 150\,000$ and lead of 99.999 grade.

The choice of the lead as the investigated impurity in indium has the following advantages: 1) the In +Pb alloy is a substitutional solid solution up to 12% lead concentration[4], 2) the coefficient of the lead distribution in indium is close to unity[5], so that when single crystals are grown from the In + Pb melt the homogeneity of the lead distribution should not be disturbed.

The lead distribution over the ingot was made homogeneous by the method of zone equalization in opposing directions. To monitor the content and the distribution of the lead in the alloy we used an x-ray spectral method (sensitivity 10^{-2} at.%) and polarographic method (sensitivity 10^{-4} at.%). Simultaneously, the residual resistivity of the sample was monitored. The results of the analysis have shown that the lead is uniformly distributed ($\Delta c/c \leq 5\%$) in the central part (not less than 75% of the length) of the ingot, with dimensions $5 \times 6 \times 200$ mm.

The single-crystal samples in the form of spheres of 12 mm diameter were grown from the prepared alloys in a dismountable quartz mold with polished surfaces, by the method of Sharvin and Gantmakher[6]. The scatter of the diameter did not exceed 10^{-2}%.

The uniformity of the distribution and of the content of the lead in the single-crystal spheres was also determined by the x-ray spectral and polarographic methods. To this end, the spheres were cut after the measurements into 10 disks. The disks were investigated by the x-ray spectral method at 500 points of the surface.

It was established that the distribution of the lead atoms is uniform over the area of each disk ($\Delta c/c \leq 5\%$), and that the Pb content is the same in all the disks of a given batch and coincides with the amount of lead in the initial alloy.

The sample for the investigation of the magnetoresistance were single-crystal disks cut from the single-crystal spheres by the electric-spark method. The disks were 1 mm thick and 12 mm in diameter. The normals to the surface of the disk coincided with the [100] axis accurate to 1%. Resistance measurements in zero field on one of the disks have shown that the influence of the defects introduced by the cutting and of the changes in the dimensions are negligible.

The characteristics of the samples are listed in the table. Sample No. 0 was the initial indium.

Sample No.	c, at.% calc.	c, at.% meas.	$10^{10}\rho_0$, Ω-cm	$10^{13}\beta$, Ω-cm/deg^5	$10^{10}\rho_H(c, 0)$, Ω-cm	$10^{12}\alpha$, Ω-cm/deg^4
0	$1\cdot10^{-4}$	—	0.65	2.2	2.5	3.3
1	$1\cdot10^{-3}$	—	6.4	3.15	16.5	4.0
2	$3\cdot10^{-3}$	$3.5\cdot10^{-3}$	18	3.9	48	4.1
3	$5\cdot10^{-3}$	$5.4\cdot10^{-3}$	29	4.6	81	4.0
4	$8\cdot10^{-3}$	$8.2\cdot10^{-3}$	43	4.8	—	—
5	$1\cdot10^{-2}$	$1.1\cdot10^{-2}$	56	5.5	—	—
6	1.0	1.1	5300	—	—	—

Measurements of the resistivity in zero field were carried out in the temperature interval 3.4–11°K by the Sharvin and Zernov contactless induction method[7]. The sample was suspended on a torsion balance in such a way that the [001] axis was parallel to the axis of rotation of the magnetic field, and the quantity $\rho = (\rho_{\parallel} + \rho_{\perp})/2$ was measured, where $\rho_{\parallel}$ and $\rho_{\perp}$ correspond to the resistivities along the [001] axis and in the perpendicular direction.

The resistivity-measurement accuracy in the interval 3.4–4.2°K was 0.5%. When the measurements were made in the interval 4.2–11°K, the sample was in helium vapor inside a thick-wall copper vessel communicating with liquid helium through a copper cold finger. An Au + Fe–Cu thermocouple was used to measure the temperature of the copper vessel. The heat rise of the sample did not exceed 0.01°K. The temperature of the copper vessel was stabilized with accuracy ~0.1%.

The transverse magnetoresistance was determined from the helicon damping. The helicon resonance was recorded by the crossed-coil method (see, e.g., [8]). In all the measurements, the magnetic field was normal to the sample surface coinciding with the [100] axis. The helicon damping was independent of the wavelength excited in the sample, thus indicating a negligible contribution of collisionless damping (Landau damping)[9], which is a parasitic effect in similar measurements.

The transverse magnetoresistance was calculated from the measured Q of the resonance of the standing waves in the sample, using the formula

$$\rho_H(c, T) = RB[4Q^2(cT) - 1]^{-1/2}, \quad (1)$$

where R is the Hall constant, which is independent of the temperature T and of the lead concentration c in a strong magnetic field; B is the magnetic field at resonance[10].

The magnetoresistance was measured at temperatures 1.3–4.2°K, at frequencies 10^2 Hz, and in fields 3–20 kOe. In our fields we were unable to rise above 4.2°K and above a lead concentration 5×10^{-3}%, for then the strong-field condition $\omega_c\tau > 1$ ceased to be satisfied (ω_c and τ are the cyclotron frequency and the relaxation time). The accuracy with which the absolute magnetoresistance was measured was 1–2% for the purest samples and 10% for the contaminated ones.

RESISTIVITY MEASUREMENT RESULTS IN ZERO FIELD

The results of the measurement of the resistivity in zero field are shown in Figs. 1 and 2 and in the table. At 3.4–4.2°K, the resistivity of all samples satisfies the relation

$$\rho(c, T) = \rho_0(c) + \beta(c)T^5. \quad (2)$$

Above 4.2°K, the T^5 law holds only for pure indium without lead impurity, up to 7°K. The residual resistivity depends linearly on the impurity content in the entire range of concentrations from 10^{-3} to 1 at.%:

$$\rho_0(c) = Ac, \quad A = 0.51\cdot10^{-6}\ \Omega\text{-cm/at.\%}$$

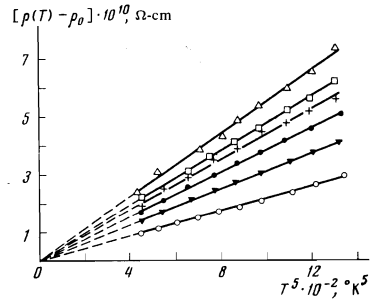

FIG. 1

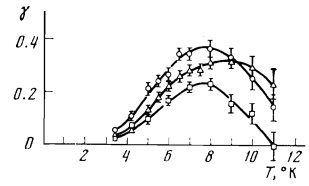

FIG. 2

FIG. 1. Temperature-dependent part of the resistivity of indium in zero field in the interval 3.4–4.2°K. ○–sample No. 0; ▽–1; ●–2; +–3; □–4; △–5.

FIG. 2. Temperature dependence of γ in zero field. ○–sample 2; △–3; □–4.

We observed a strong and nonlinear concentration dependence of the coefficient β(c) of Eq. (2) (see the table). Introduction of 10^{-2} at.% lead caused β(c) to increase by 2.5 times, as against $\beta = 2.2 \times 10^{-13}$ Ω-cm/deg^5 for the initial indium. Figure 2 shows the temperature dependence of the quantity

$$\gamma = \frac{\Delta\rho(c, T)}{\rho(c, 0)} = \frac{\rho(c, T) - \rho(c, 0) - \rho(0, T)}{\rho(c, 0)} \quad (3)$$

where ρ(c, T) is the total resistivity of the alloy, ρ(c, 0) ≡ ρ$_0$(c) is the resistivity at T = 0 and is due to the scattering of the electrons by the impurity, and ρ(0, T) is the resistivity of the pure (initial) indium and is due to scattering by photons. We see that γ goes through a maximum at 8°K.

TRANSVERSE MAGNETORESISTANCE

It was shown earlier that the magnetoresistance of single-crystal plates of indium in strong fields is independent of the field within the limits of the measurement error and varies with temperature like

$$\rho_H(c, T) = \rho_H(c, 0) + \alpha(c)T^4,$$

and that the scattering from the sample boundaries can be neglected[9, 11]; ρ_H(c, 0) depends linearly on the impurity concentration

$$\rho_H(c, 0) = A'c, \quad A' = 1{,}4\cdot10^{-6}\ \Omega\text{-cm/at.\% Pb.}$$

We investigated the dependence of the coefficient α(c) on the impurity content. The measurement results are shown in Fig. 3 and in the table. The change in the coefficient α(c) following introduction of 5×10^{-3} at.% lead did not exceed 20%, whereas the accompanying change of β(c) is 100% (see the table). The initial indium had $\alpha = 3.3 \times 10^{-12}$ Ω-cm/deg^4.

DISCUSSION OF RESULTS

The experimental results can be explained within the framework of the theory of Kagan and Zhernov[2], where the anisotropy of the nonequilibrium increment to the electron distribution function, resulting from the aniso-

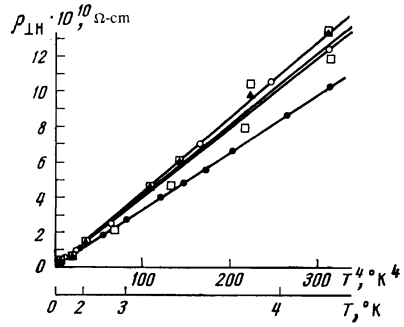

FIG. 3. Temperature-dependent part of the transverse magneto-resistance of indium in the interval 1.3–4.2°K. Field H = 11 kOe; ●–sample 0; ○–1; ▲–2; □–3.

tropy of the electron-photon scattering, was taken into account in the calculation of the electric conductivity of the metal with the aid of a variational procedure. It is the dependence of the anisotropy of the distribution function on the contribution of the isotropic impurity scattering which leads, according to this theory, to deviations from the Matthiesen rule. The Fermi surface of indium at individual point comes close to the boundaries of the Brillouin zone[12]. Thus, according to the experimental data on cyclotron resonance[13], the width of the gap between the second and third zones in the middle of the edge lying in the (001) plane is 7°K. According to[12], this gap should be even smaller at the corners W and W′ of the Brillouin zone. All this indicates that in the temperature interval in which our measurements were made one should expect a large contribution of the electron-phonon scattering processes with Umklapp, having large anisotropy.

According to Kagan and Zhernov, in a certain temperature interval the dependence of $\rho(c, T)$ on T can be expressed in the form $\rho(c, T) = \rho_0(c) + B(c)T^5$, where B(c) increases with increasing c. The form of the function B(c) coincides qualitatively with the $\beta(c)$ dependence observed by us. The theoretically predicted maxima in the dependence of γ on T agree qualitatively with experimentally observed one with respect to the amplitude and the position on the temperature scale. The measurement accuracy did not enable us to trace the dependence of the position of the maxima on the impurity concentration ($T^* \sim c^{1/5}$).

The presence of a maximum in the temperature dependence of γ cannot be attributed in this case to the appearance, in the phonon spectrum of the metal, of a quasilocal level connected with the presence of a heavy impurity in the lattice[14]. Estimates show that for the In +Pb alloy the value of γ at the maximum, at a lead concentration 10^{-2} at.%, is 300 times smaller than the experimentally obtained values.

The observed amount of the deviation from the Matthiesen rule, the dependence of β on $\rho(c, 0)$ for the In + Pb alloy, coincides with the dependence of β on $\rho(c, 0)$ obtained earlier for indium with an unknown qualitative impurity composition[16], but is more than double the deviation from the Matthiesen rule observed in[16] for the In +Bi alloy. The latter can be explained by assuming that the scattering by the Bi impurity atoms in the indium lattice is not isotropic.

We interpret our experimental results on the magnetoresistance also by assuming an appreciable contribution of Umklapp processes in those sections of the Fermi surface that come close to the boundaries of the Brillouin zone. The possibility that the magnetoresistance can have a T^4 temperature dependence under such conditions was pointed out already by Pippard[17].

In a magnetic field, just as in the presence of an impurity, the anisotropy of the nonequilibrium increment to the distribution function should decrease[3]. The reason is that the probability of scattering with Umklapp is the same, when averaged over the cyclotron period, for all the electrons on a given intersection of the Fermi surface and a plane perpendicular to the magnetic field. This leads to isotropization of the nonequilibrium increment to the distribution function and to a decrease of the concentration dependence of the temperature-dependent part of the magnetoresistance in comparison with the resistance in a zero field (see the table).

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