EXPERIMENTAL INVESTIGATION OF SCATTERING OF ELECTRONS BY DISLOCATIONS IN COPPER

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Submitted June 20, 1972

Zh. Eksp. Teor. Fiz. 63, 1752–1757 (November, 1972)

The dependence of the electric resistance and amplitude of the radio frequency size effect lines on dislocation density in copper is investigated. The transport cross section for electron scattering by edge dislocations is derived from data on the electric resistance; the total cross section for two orbits—for that on the neck and that on the belly with $\mathbf{H} \parallel [111]$ is derived from data on the size effect. The transport scattering cross section is found to be several times larger than that quoted in the literature; it is suggested that this may be due to the fact that in all previous investigations the measurements were carried out for higher dislocation densities and a correlation between the dislocations existed. A comparison of the transport cross section with the total cross section indicates that the scattering occurs predominantly at small angles. The difference between the total scattering cross sections on different orbits can be ascribed to anisotropy of the deformation potential.

DISLOCATIONS occupy a special place among the different types of crystal-lattice periodicity disturbances that lead to electron scattering. First, they are sizable defects and extend over hundreds of thousands of interatomic distances. Second, the perturbations introduced into the lattice by a dislocation decreases very slowly in a direction perpendicular to the dislocation axis. These specific features of dislocations must undoubtedly be reflected in scattering. We have undertaken a cycle of investigations of this question, performing parallel experiments on the influence of dislocations on the electric resistivity and on the amplitude of the radio-frequency size effect. The object of the investigation was copper, for which the principal data on the influence of dislocations on the de Haas-van Alphen effect are already known[1,2].

Measurement of the electric resistivity yield the transport cross section σ_{tr} for scattering by dislocations, averaged over all the electrons; the scattering through small angles θ enters in this cross section with a weight factor $(1 - \cos\theta)$. If ρ_d is the change in the residual resistivity following introduction of N dislocations per cm^2, then

$$\sigma_{tr} = (Nl_{tr})^{-1} = ne^2\rho_d / p_F N \qquad (1)$$

(l_{tr} is the transport mean free path, p_F is the Fermi momentum, n is the density of the electrons in the metal, and e is the electron charge). Since the calculation is carried out per unit dislocation length, σ_{tr} has the dimension of length and not of area, and is thus the scattering diameter.

The scattering diameter obtained by measuring the amplitude of the size-effect lines differs from (1) in two respects. First, the size-effect line is formed by a relatively small group of electrons that are located on the Fermi surface near some extremal section[3]. Therefore all the scattering data pertain precisely to this group. Second, the line amplitudes are equally affected by scattering through small and large angles, since any collision that takes the electron out of the effective region on the Fermi surface is important. (There exists, of course, a certain minimum angle, to scattering by which the amplitude of the size-effect is sensitive. This angle can be estimated from the relative line width[3]. In our experiment it was of the order of 1–2°.) What is measured in the size effect is therefore not the transport scattering diameter but the total scattering diameter σ, and furthermore it is averaged not over the entire Fermi surface but only over one of its extremal sections.

In the measurements we used the size effect on closed trajectories, and the sample thickness were such that even prior to the introduction of the dislocations the mean free paths l_0 were sufficient for only half a revolution, for the path λ from one side of the plate to the other. Under these conditions, the amplitude of the line A (the change of the signal between two characteristic extrema of the line) is

$$A \propto e^{-\lambda/l}. \qquad (2)$$

Assuming additivity of the reciprocal free paths $1/l_d$ on the introduced dislocations and $1/l_0$ on all the remaining previously existing disturbances of the lattice periodicity, we find that the line amplitude after N dislocations [cm^{-1}] are introduced is $A(N) = A_0 \exp(-\lambda/l_d)$, where A_0 is the line amplitude prior to the introduction of the dislocations. Hence

$$l_d^{-1}(N) = (\ln A_0 - \ln A)/\lambda. \qquad (3)$$

SAMPLES

The samples for the measurement of the electric resistivity (R1 – R4) were prepared in the following manner:

a) A single-crystal plate measuring 0.6 × 12 × 200 mm was grown by the Bridgman method. A blank of brand OSCh (specially pure) copper (resistance ratio $\gamma = R(273°K)/R(4.2°K) \approx 200$) having the indicated dimensions was placed in a dismountable graphite mold, molten, and subjected to directional crystalliza-

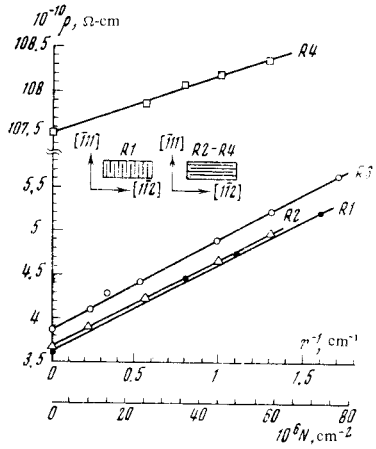

FIG. 1. Change of residual resistance following bending of the samples. T = 4.2°K. The different designations pertain to different samples.

tion, using an oriented primer, at a rate ~200 mm/h in a vacuum of 10^{-5} Torr. In all the samples, the normal to the surface of the plate was close to [110] (the deviations did not exceed 3°).

b) The obtained single crystals were annealed in an oxygen stream at a pressure ~5×10^{-4} Torr and a temperature 1030°C for eight hours. After such a treatment, the value of γ increased to 4000—4500 (sample R4 was not annealed).

c) The single crystal was cut by the electric-spark method into several parts, approximately 30 mm long each, on each of which a system of parallel grooves was cut (see the insert of Fig. 1). The final sample was an aggregate of series-connected plates with cross sections 0.5×2 mm^2, the total length of which was ~150 mm. A work-hardened layer ~0.2 mm thick, produced during the cutting, was removed by dissolution in a chemical-polishing reagent consisting of two parts of HNO_3, one part of H_3PO_4, and one part of acetic acid.

d) The contacts were secured to samples with a non-superconducting solder (40% Cd and 60% Bi).

e) To introduce the dislocations, the sample was bent on a cylindrical form of the required radius. The bending axis was always directed along [$\bar{1}$11]. To anneal the vacancies, the samples were soaked at ~100°C for one hour. After measuring the residual resistance, the operation (e) was repeated using a form of smaller radius.

The sample R1, where the bending axis was parallel to the long axis of the plate, was first bent and then grooved and etched. Therefore each point on the plot of Fig. 1 for R1 corresponds to an individual sample subjected to single bending, and the index R1 pertains, strictly speaking, to the initial single crystal from which all these samples were cut.

The samples for the observation of the radio-frequency size effect (S1—S3), in the form of plane-parallel plates of area ~1 cm^2, were cut by the electric spark method from a single-crystal ingot grown at Centre d'études de Chimie Métallurgique (France)[1].

[1]The authors take the opportunity to express deep gratitude to J. LeHerissy for growing and supplying this crystal.

The roughnesses on the sample surfaces were removed by mechanical polishing with silicon-carbide powder M-10, followed by etching in the reagent mentioned above. The ratio γ for these samples was ~6000, and increased after annealing in oxygen to ~14000. The sample thicknesses were 0.25 (S1), 0.58 (S2), and 0.71 mm (S3), the normals to the surfaces coincided with the [110] axis within one degree, and the bending axis was directed along [$\bar{1}$11].

According to estimates based on the etch pits, the initial dislocation density in all the investigated samples did not exceed 10^6 cm^{-2}.

RESULTS

The electric resistance was measured at 4.2°K by the usual potentiometer method (current through samples 2—5 A, voltage sensitivity of circuit 1×10^{-8} V). The measurement results are shown in Fig. 1. The ordinates show the residual resistivity $\rho = \gamma^{-1} \times 1.56 \times 10^{-6}$ Ω-cm, and the abscissas show the reciprocal bending radius 1/r and the density of the introduced dislocations, calculated from the formula

$$N = (rb\cos\varphi)^{-1}, \qquad (4)$$

where $b \cos \varphi = 2.55$ Å $\times \cos 30°$ is the projection of the most probable Burgers vectors, at the given bending, on a line perpendicular to the bending axis on the plane of the sample. Strictly speaking, this is the minimum possible number of introduced dislocations, determined from geometrical considerations; pairs of edge dislocations of opposite signs, not taken into account in formula (4), could be simultaneously produced. However, as shown by Livingstone's experiments[4], the overwhelming majority of dislocations obtained in single crystals of pure copper by such bending are of the same sign, and their number practically coincides with that calculated from formula (4). Livingstone[4] has also shown that in bending about the [111] axis the dislocations are distributed quite uniformly, without clusters. Our control experiments in which the dislocations were displayed by etching yielded the same results.

The additional resistance ρ_d that appears when dislocations are introduced, normalized to one dislocation, is found from the three lower straight lines (samples R1—R3) to be equal to $\rho_d/N = 2.2 \times 10^{-18}$ Ω-cm^3, i.e., it is approximately 10 times larger than in all the preceding measurements[5-7]. In[5-7] they measured the change produced in the residual resistance by tension or rolling; this always introduced appreciable dislocation densities, $N \gtrsim 10^9$ cm^{-2}. In a series of control experiments, we measured the change of the resistance of a single crystal with normal to the [110] surface, rolled along the [1$\bar{1}$2] direction; the number of introduced dislocations was determined with an electron microscope. We obtained a value $\rho_d/N = 1.8 \times 10^{-19}$ Ω-cm^3, which practically coincides with the results of[5-7].

We propose that the reason for the observed discrepancy is that at high densities the dislocations are correlated to a large degree, their slowly-decreasing fields become superimposed, and the total action of these fields decreases. It is possible that in[5-7] the

dislocation correlation was enhanced by the very method of introducing the dislocations.

Another possible reason for the discrepancies is connected with the presence of impurities. It is known that impurities tend to concentrate along the nuclei of dislocations. Therefore, if the number of impurities is large, their capture by the dislocation nuclei, which leads to a decrease in the number of independent scattering impurity centers, can become manifest in an apparent decrease of ρ_d. That such an effect exists is evidenced by our experiments with sample R4, which was not annealed in oxygen. For this sample, the value of ρ_d/N turned out to be 35% less than for R1—R3, namely $\rho_d/N = 1.5 \times 10^{-18}\ \Omega\text{-cm}^3$.

The amplitude of the radio-frequency size-effects was measured with an autodyne radio spectrometer operating at ~3 MHz, using the plots of the first derivatives of the active point of the surface impedance against the magnetic field (the field modulation frequency was 18 Hz, and the sensitivity was calibrated with a Pound-Knight calibrator[8]). To avoid the harmful effect of disorientation of the sample surface and of the crystallographic axes relative to the direction of the magnetic field as a result of bending, the measurements were made only in a field H parallel to the bending axis, i.e., at $\mathbf{H} \parallel [\bar{1}11]$. At field directions that did not coincide with the bending axis of the sample, the lines changed in shape and became smeared out.

We chose for the measurement two size-effect lines, one on the central section (cyclotron mass $m_b = 1.398m_0$, average velocity $v_b = 1.16 \times 10^8$ cm/sec), and the other on the neck ($m_n = 0.46m_0$, $v_n = 0.65 \times 10^8$ cm/sec). The measurements of the first were performed with the bending of the thick samples S2 and S3, and those of the second with the bending of the thin sample S1. The measurement results, reduced in accord with formula (3), are shown in Fig. 2.

DISCUSSION

On the basis of the arguments advanced in the preceding section, we propose that among all the experimental values of ρ_d/N it is precisely the value for samples R1—R3 of the present paper, and the quantity

$$\sigma_{tr} = 3.3 \cdot 10^{-7}\text{cm} \approx 13\ b \quad (b = 2.55 \cdot 10^{-8}\text{cm}) \qquad (5)$$

ensuring from it on the basis of Eq. (1), which correspond to scattering by a single dislocation that does not interact with anything in the copper crystal. It follows both from the value of σ_{tr} itself and from the dislocation-resistance values at larger densities N, which were discussed in the preceding section, that the value of σ_{tr} observed by us is determined mainly by scattering from the slowly decreasing field around the dislocation.

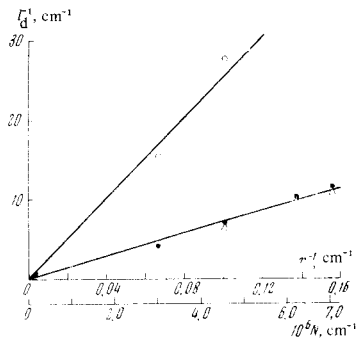

FIG. 2. Change of electron mean free path due to bending of the samples, obtained from measurements of the amplitude of the size-effect lines at T = 1.6°K. ○—line from neck (sample S1), △ and ●—lines from central section (belly) (samples S2 and S3).

At such large a diameter σ_{tr}, one should expect the scattering to be anisotropic, since small-angle scattering should predominate. Actually, the total scattering cross sections obtained from the size effect for the electrons on the belly part of the Fermi surface σ_b and on the neck σ_n are much larger than σ_{tr}:

$$\sigma_b = 16 \cdot 10^{-7}\text{cm} = 63\ b, \quad \sigma_n = 56 \cdot 10^{-7}\text{cm} = 220\ b.$$

It is more correct to compare σ_{tr} with σ_b, since the greater part of the electrons is located on the belly of the Fermi surface, and it is precisely these electrons that determine mainly the electric conductivity. The relation $\sigma_b \approx 5\sigma_{tr}$ reflects the difference between the total and transport cross sections, thus confirming the predominantly small-angle character of the electron scattering by the dislocation field.

The difference between σ_b and σ_n agrees qualitatively with the known fact that the deformation potential for electrons on the neck is much larger than on the remaining part of the Fermi surface of copper[9]. From measurements of the temperature dependences of the amplitudes of the size-effect lines we obtained for the ratio of the frequencies of the collisions with the phonons $\tau_n^{(ph)}/\nu_b^{(ph)} \approx 10$, corresponding to a scattering cross section ratio

$$\sigma_n^{(ph)}/\sigma_b^{(ph)} = v_n^{(ph)}\bar{v}_n/v_b^{(ph)}\bar{v}_b \approx 20.$$

A similar result was obtained also by other methods[10,11]. We note, however, that the ratios of the scattering cross sections should be compared with extreme caution, since different components of the deformation-potential tensor probably make the principal contributions to the scattering by phonons and to the scattering by dislocations.

All the presently existing theories of electron scattering by dislocations[12-14] yield for the transport scattering diameter a value $\sigma_{tr} \lesssim b$. It is difficult to explain the reasons for such a discrepancy between the theory and our result (5). We note only the idea advanced by Kravchenko[13], that the influence of the dislocations on the electric resistance may be due not only to scattering as such, but also to local dislocation-induced distortions on the electron dispersion. In any case, as shown by Watts[15], the amplitude of the de Haas-van Alphen dislocations decreases when dislocations are introduced precisely because of the resultant spatial inhomogeneity of the parameters, and not because of scattering. Indeed, were we to interpret the decrease of the amplitude of the oscillations observed in[1,2] as the result of scattering, then in a field $\mathbf{H} \parallel [111]$ we would obtain for the two orbits discussed by effective scattering diameters 25 times larger than our σ_b and σ_n.

The authors thank V. Ya. Kravchenko for a discussion of the results and L. S. Kokhanchik and S. P. Gasparova for help with the experiments.

[1] P. T. Coleridge and B. R. Watts, Phil. Mag., 24, 1163 (1971).
[2] D. W. Terwilliger and R. J. Higgins, Phys. Lett., 31A, 316, 1970; Bull. Am. Phys. Soc., 17, 280 (1972).
[3] V. F. Gantmakher, Progr. in Low Temp. Phys., 5, Ed. C. J. Gorter, North-Holland, Amsterdam, 1967, p. 181.
[4] J. Livingstone, Direct Observation of Imperfection in Crystals, Ed. J. B. Newkirk,and J. H. Wernick, 1962.
[5] T. H. Blewitt, R. R. Coltman, and J. K. Redman, Rep. of the Conf. on Defects in Cryst. Solids (London, The Phys. Soc.), 1955, p. 369.
[6] J. G. Rider and C. T. B. Foxon, Phil. Mag., 16, 1166,(1967).
[7] Z. S. Basinski and S. Saimoto, Canad. J. Phys., 45, 1161 (1967).
[8] R. V. Pound and W. D. Knight, Rev. Sci. Instr., 21, 219 (1950).
[9] D. Nowak and M. J. G. Lee, Phys. Rev. Lett., 28, 1201 (1972).
[10] P. Häussler and S. J. Welles, Phys. Rev., 152, 675 (1966).
[11] J. F. Koch and R. E. Doezema, Phys. Rev. Lett., 24, 507 (1970).
[12] S. C. Hunter and F. R. N. Nabarro, Proc. Roy. Soc., A, 220, 542 (1953).
[13] V. Ya. Kravchenko, Fiz. Tverd. Tela 9, 836 (1967) [Sov. Phys.-Solid State 9, 653 (1967)].
[14] A. B. Bhatia and O. P. Gupta, Phys. Rev., B, 1, 4577 (1970).
[15] B. R. Watts, Phil. Mag., 24, 1151 (1971).

Translated by J. G. Adashko
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