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Influence of Voids on the Linear Magnetoresistance of Indium

C. J. Beers, J. C. M. van Dongen, (a) H. van Kempen, and P. Wyder

Physics Laboratory and Research Institute for Materials, University of Nijmegen, Toernooiveld,

Nijmegen, The Netherlands

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The influence of the presence of cylindrical voids on the linear magnetoresistance of indium wires is studied experimentally. We show that the voids increase the linear magnetoresistance drastically in accordance with a theoretical model.

The question of linear magnetoresistance in simple metals has attracted a great deal of interest in recent years and seems to be one of the still not clearly settled problems of electron transport in metals. The theory of Lifshitz, Azbel', and Kaganov¹ predicts for uncompensated metals without open orbits a magnetoresistivity (MR) which saturates at high magnetic fields; however, in the alkali metals and in simple polyvalent metals, like indium and aluminium, a linear magnetoresistance (LMR) has been found experimentally. In this Letter we present a

quantitative comparison of experiments and theories based on the presence of macroscopic imperfections. By introducing artificial voids in In samples, we show that the LMR increases drastically, and this increase is found to be in excellent agreement with theoretical predictions based on classical arguments.

Some of the observed LMR in the metals K.^{2,3} Al,⁴ and In^{5,6} has been attributed to intrinsic effects like magnetic breakdown or field-dependent relaxation times.^{7,8} However, the strong dependence of the LMR on sample handling and sample VOLUME 40, NUMBER 18

quality suggests that extrinsic effects, like sample inhomogeneities, dislocations, stacking faults, etc., should be considered. Other arguments⁹ point in this direction as well. Several theories were put forward lately to relate the LMR to different kind of sample inhomogeneities.^{2,10-13} In the approximation where the electron mean free path l is much smaller than the characteristic dimension d of the voids, Sampsell and Garland,¹¹ and Stroud and Pan,¹² have obtained theoretical expressions for the LMR for samples containing spherical voids, and cylindrical voids with the axes perpendicular to the directions of the magnetic field B and the current (classical, local approximation). In the high-field limit, their result can be written as

$$d[\Delta \rho(B)]/dB = \alpha f / n^{\circ} e , \qquad (1)$$

where $\Delta \rho(B) = \rho(B) - \rho(0)$ [with $\rho(B)$ the resistivity in a magnetic field B]; n^0 is the volume density of conduction electrons, *e* the electron charge, *f* the volume fraction of the voids, and α a numerical constant found to be $\alpha = 0.49$ for spherical and $\alpha = 1$ for cylindrical voids. An estimate by Lass² along the same lines leads to qualitatively the same result. A generalization of the theory of Stround and Pan¹² to more general shapes of the voids by van Gelder¹³ shows, that in this classical local approximation the LMR should vanish if the axis of the cylinder is along the direction of the magnetic field.

We have measured the LMR in In samples with artificial cylindrical voids where the axes of the cylinders are parallel to each other. The samples consisted of wires with a diameter of 1 mm. We report on ten samples, six were made of 6N-(99,9999%-) purity material [residual resistance ratio (RRR) \simeq 15000] and four from 4*N*-purity material (RRR \simeq 2000). After mounting and annealing, the MR was measured in the conventional way at 4.2 and 2.1 K in fields up to 7 T. Then cylindrical holes of 0.13 mm diameter were punched through the wires, using a glass fiber as a punch. The holes, 1.0 or 2.0 mm apart, were perpendicular to the axis of the wire and filled the whole length between the potential contacts. After annealing, the transverse MR was measured with the holes perpendicular and parallel to \vec{B} . In some cases, the longitudinal MR was measured as well. Since the samples were kept in the sample holder and all the leads were permanently attached during all manipulations (punching, annealing, etc.), any change in the MR can be attributed to the punching of the holes



FIG. 1. Resistivity ρ as a function of the magnetic field *B* for a sample without macroscopic voids (crosses), the same sample with one cylindrical void every millimeter (circles), and a sample with one cylindrical void every two millimeters (triangles).

alone. As shown in Fig. 1, the LMR is strongly increased by the presence of the holes. To illustrate more clearly the effect of the holes on the MR, Fig. 2 shows the difference in MR between samples with holes and the same samples without holes. Also shown in Fig. 2 is the striking difference of the LMR for different directions of the holes with respect to \vec{B} ; this indicates clear-



FIG. 2. Contribution of voids to the magnetoresistivity $\Delta \rho = \rho_{\text{with voids}}(B) - \rho_{\text{without voids}}(B)$ as a function of the magnetic field *B*. Open squares and circles, axes of the cylindrical holes perpendicular to \vec{B} for a 6N- (open squares) and 4N- (open circles) purity sample; filled squares and circles, same samples with axes of the cylindrical holes parallel to \vec{B} ; crosses, sample with indentations (see text).

ly that indeed the holes are giving rise to the increased LMR, and not crystal defects caused by the punching process. To check the influence of the thermal history on the LMR, one sample was mounted and heat treated together with other samples, however, without hole punching. For this control wire, no change in LMR was observed. In addition, no LMR change was observed for a wire which was indented every millimeter, also with the 0.13-mm punch, to check the influence of surface distortions (Fig. 2).

For a quantitative comparison of the experimental results with the theoretical calculations of Refs. 11 and 12, the finite size of the artificial voids should be taken into account. However, the relative small change in α when one goes from a spherical void to the limit of an infinite cylinder with axis perpendicular to \vec{B} shows that, in this configuration, α is not too sensitive to the exact shape of the void. Therefore, even for finite cylinders, a value of α of 1.0 seems to be a reasonable approximation. (However, one should not conclude from this that the shape of the void is unimportant for all configurations; the results with the cylinder axis along the direction of \vec{B} shows clearly the importance of the precise form of the void in this situation.)

In Fig. 3, the measured slopes $d\Delta\rho/dB$ are given for eight samples. For the lower-purity samples, a very good agreement with theory is found. The high-purity samples give results somewhat above the theoretical values. The linearity with respect to the void density cannot be determined



FIG. 3. Increase of the slope of the linear magnetoresistivity due to 1 void per millimeter, or 1 void per 2 mm length of wire. Triangles, 4N-purity sample; circles, 6N-purity sample. Line drawn, theoretical expression $d[\Delta \rho(B)]/dB = \alpha f/n^0 e$, using one electron per atom for n^0 and $\alpha = 1$; f is the volume fraction of voids (Refs. 11 and 12).

unambiguously for the pure samples. This could be due to mean-free-path effects ($l \simeq 0.2$ mm, therefore comparable to void dimensions and the distance between voids). which are neglected in the theory. Also in the purest samples and at the highest fields, a slight deviation from linearity with respect to the magnetic field is apparent; this becomes more pronounced when the temperature is lowered to 2.1 K. It appears, that independent of temperature and void-void distance, deviations from linearity occur above fields corresponding to about $\omega_c \tau = 40 (\omega_c, \text{ cyclotron reso-}$ nance frequency; τ , relaxation time). Again this might be due to nonlocal effects, but more experimental and theoretical studies are necessary to clarify this point.

The longitudinal LMR of a single void should be the same as the transverse LMR. However, the calculations of Refs. 11, 12, and 14 involve a coordinate transformation in which the axis along the field direction is shortened with a factor which is proportional to B in high fields. In our case, where the holes are lined up along the axis of the wire, this means that in the transformed system the holes become flat plates close to each other; therefore, the current profiles around the holes are no longer independent of each other. This picture is in agreement with our observations; the longitudinal LMR $[(0.26-0.36)\times10^{-12}]$ $\Omega m/T$ is much smaller than the transverse LMR and nearly independent of the void density. A full account of this longitudinal case will be published elsewhere.

As shown in Fig. 2, the LMR decreases strongly when the axes of the cylinders are directed along the B direction. However, even in this configuration there is a small but nonvanishing contribution of the voids to the LMR. This could be due to misalignments of the holes, or to other mechanisms contributing to the LMR. A quantum mechanical treatment of the problem by van Gelder¹³ shows a LMR which should also be present when the axes of the cylinders are along the field direction and which, in contrast to the classical calculations of Refs. 11 and 12, should be proportional to $\rho(0)$. We do find some indications that this is indeed the case (Fig. 2), but measurements on even less pure samples are needed to establish this effect unambiguously; this implies much higher fields in order to reach again the high-field limit.

Experiments on samples with melted in glass rods, carried out in our laboratory, did give qualitatively the same results as the experiments with the voids although with much larger scatter from sample to sample.¹⁵

In conclusion, we have shown that artificial cylindrical voids in a simple metal can induce a large linear magnetoresistance in quantitative agreement with classcial theory. The directional dependence shows clearly that indeed the influence of the voids has been studied, and not parasitic effects due to lattice imperfections or distortions.

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Fluctuation-Induced Tunneling Conduction in Carbon-Polyvinylchloride Composites

Ping Sheng, E. K. Sichel, and J. I. Gittleman RCA Laboratories, Princeton, New Jersey 08540 (Received 13 February 1978)

We present evidence that in carbon-polyvinylchloride composites, consisting of aggregates of carbon spheres (100-400 Å) dispersed in the insulating matrix, the electrical conductivity can be ascribed to a novel mechanism of tunneling with potential-barrier modulation by thermal fluctuations. Theoretical consideration of the tunneling-probability modification by thermal fluctuating electric field across tunnel junctions yields expressions for the temperature and the field dependences of the conductivity in excellent accord with experimental results.

In recent years hopping conduction in disordered materials has recieved considerable attention as the mechanism responsible for the characteristic $\exp(-b/T^{\alpha})$ form of temperature dependence of conductivity observed in amorphous semiconductors¹ ($\alpha = \frac{1}{4}$) and sputtered granular metal films² ($\alpha = \frac{1}{2}$). There are, however, materials such as some conductor-insulator composites³ and disordered semiconductors⁴ for which no definitive theory has been proposed to explain their transport properties. In this Letter we report the observation of a new tunneling conduction mechanism for disordered materials in which the modu-

lation of tunneling barriers by thermal fluctuations plays an important role in determining the dependence of the conductivity on temperature and electric field. In the following, the experimental results for the carbon-polyvinylchloride (PVC) composites are presented and compared with theoretical predictions based on the above mechanism. Application of the theory to other disordered systems will be published subsequently.

Carbon-PVC is a conductor-insulator composite consisting of carbon particles embedded in the insulating PVC matrix. Samples of this composite