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Empirical Correlation between Impurity-Dependent and Size-Dependent Deviations from Matthiessen's Rule in Indium

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A study of published measurements on indium wires at liquid-helium temperatures indicates that the effects of boundary scattering and impurity scattering on the temperature dependence of electrical resistivity are equal, for equal values of $\rho_{0°K}$. This simple relation, and similar relations which may hold for other metals, appear to have important implications for proposed explanations of deviations from Matthiessen's rule.

IN recent years there has been a renewal of interest in the determination of the electrical resistivity of simple metals at low temperatures. This has been stimulated by the availability of high-purity materials and apparatus of improved sensitivity. Considerable attention has been paid to the effects of specimen size and specimen impurity on the temperature dependence of resistivity. Aside from having intrinsic interest, these "deviations from Matthiessen's rule" must be taken into account in nearly all studies, since they are usually a major source of uncertainty concerning the precise behavior of the ideal resistivity.

The primary purpose of this paper is to call attention to a remarkably simple empirical correlation for indium wires between deviations from Matthiessen's rule associated with boundary scattering and those associated with dilute impurity scattering. Figure 1, which exhibits the correlation, was constructed using the data from four major studies of resistivity in indium.[1–4] Each of the various points in the figure represents the quantity $\rho_{4.2°K}-\rho_{0°K}$ for an individual specimen from one of the studies. (The data of Garland and Bowers correspond to samples of differing purities but similar sizes.[2] The data of Blatt, Burmester, and La Roy correspond to samples of differing sizes but similar purities,[3] as do the data of Wyder.[4] Finally, the data of Olsen correspond to samples of differing sizes and differing purities.[1]) Despite the large range of sizes and impurities represented, $\rho_{4.2°K}-\rho_{0°K}$, whose variation may be taken as a measure of deviations from Matthiessen's rule, appears to be a function of $\rho_{0°K}$ alone. This is independent of whether the resistivity at 0°K is size-limited or impurity-limited.[5]

It would seem that unless the results of Fig. 1 are fortuitous, they establish severe constraints for proposed explanations of departures from Matthiessen's rule. In particular, it would seem that such departures could hardly result from the operation of separate and unrelated mechanisms in the size-limited and impurity-limited regimes. The experimental results appear to imply just the opposite—that effects in the two regimes ought to be susceptible to a unifying theoretical treatment.

A proposal subject to question on these grounds is that of Olsen[1] (elaborated by Blatt and Satz[6]), that the temperature-dependent *size effects* in indium are a consequence of the small-angle character of electron-phonon scattering at low temperatures. Since this factor is (presumably) not relevant to the effects connected with impurity scattering, it would appear unlikely that it plays a dominant role in any of the effects seen in indium.

We should note in passing that size-dependent deviations from Matthiessen's rule are implied by calcu-

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[1] J. L. Olsen, Helv. Phys. Acta **31**, 713 (1958). In this study, the range of sample diameters d was $0.06<d<2.54$ mm. Three of the ten samples were prepared from material for which the *bulk* residual resistivity was 1.7×10^{-10} Ω cm. For six others the corresponding value was 4×10^{-10} Ω cm. (The tenth sample, prepared independently, was apparently the least pure of the study.)

[2] J. C. Garland and R. Bowers, Phys. Rev. Letters **21**, 1007 (1968). The diameters of the four samples for which data are given ranged from 0.50 to 0.75 mm.

[3] F. J. Blatt, A. Burmester, and B. La Roy, Phys. Rev. **155**, 611 (1967). The *bulk* residual resistivity of the indium employed in this study was about 5×10^{-10} Ω cm. The range of sample diameters was $0.0156<d<0.642$ mm.

[4] P. Wyder, Physik Kondensierten Materie **3**, 263 (1965). Here, the range of sample diameters was $0.096<d<1.48$ mm. The *bulk* residual resistivity was $\sim5\times10^{-10}$ Ω cm. Wyder and others (see Ref. 2) have interpreted his data as evidence for the *absence* of size-dependent deviations from Matthiessen's rule. However, Blatt, Burmester, and La Roy (Ref. 3) pointed out that the scatter of Wyder's results is too great to justify such a conclusion. In fact, one can see from Fig. 1 that Wyder's values of $\rho_{4.2°K}-\rho_{0°K}$ are actually consistent, within scatter, with the other data.

[5] For clarity and simplicity, Fig. 1 does not include points from every published study of resistivity in indium wires. It was decided that the present discussion would be limited to those studies containing data for at least two indium samples with different values of $\rho_{0°K}$. For instance, we did not include one additional point obtainable from the work of B. N. Aleksandrov and I. G. d'Yakov, Zh. Eksperim. i Teor. Fiz. **43**, 852 (1962) [English transl.: Soviet Phys.—JETP **16**, 603 (1963)]. However, in this case, as in all other cases of which we are aware, there is no inconsistency with the result represented in Fig. 1. (The point corresponding to the work of Aleksandrov and d'Yakov would fall exactly on the line in the figure, at a value $\rho_{0°K}\sim3\times10^{-10}$ Ω cm.)

[6] F. J. Blatt and H. G. Satz, Helv. Phys. Acta **33**, 1007 (1960).

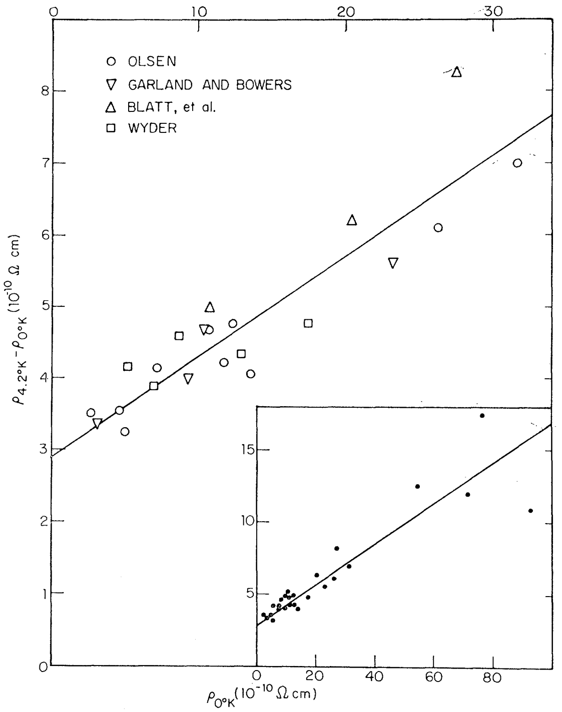

FIG. 1. Values of $\rho_{4.2°K} - \rho_{0°K}$ (whose variation serves as a measure of deviations from Matthiessen's rule) for a number of indium wires of various sizes and purities, plotted against residual resistivity. The inset covers a broader range of $\rho_{0°K}$, in order to include every sample from the size-effect study of Ref. 3. (The straight lines are drawn to facilitate comparison between the figure and the inset.)

lations based on very simple models. This is contrary to prevailing opinion. In particular, Dingle's often-quoted size-effect calculation[7] is invariably misinterpreted on this point; it actually implies substantial departures from Matthiessen's rule in size-limited specimens.[8] The source of confusion has been the fact that Dingle's values for the additional resistivity due to boundary scattering differ only slightly from those corresponding to a simple additive contribution (of the sort originally discussed by Nordheim[9]). However, the difference may in some cases amount to a substantial fraction of the *temperature-dependent* portion of the resistivity. The difference varies with temperature because of its dependence on l/d, leading to sizable deviations from Matthiessen's rule in the regime $l/d > 0.1$, where l is the bulk mean free path of electrons and d is the specimen diameter. We calculate that for Olsen's samples[1] the deviations implied by Dingle's calculation are $\sim 30\%$ of those observed, and in general have the same qualitative character. Of course, it should be emphasized that Dingle's calculation, in common with the calculation of Blatt and Satz,[5] is concerned only with size effects, and would not, on the face of it, explain the results shown in Fig. 1.[10]

Returning to the experimental results for indium, we note that they become of particular interest when taken together with the results of Reich and Forsvoll[11] for tin wires. For that metal also, there were indications that the magnitudes of impurity-dependent and size-dependent deviations from Matthiessen's rule are equal for equal values of $\rho_{0°K}$. This result of Ref. 11 might have been considered fortuitous—especially in light of the limited temperature range of the measurements (3.75–4.22°K) and the limited range of overlap between the impurity-effect and size-effect data ($0.6 < \rho_{0°K} < 2.3 \times 10^{-10}\,\Omega$ cm). However, it would appear that the present results for indium and the results for tin complement each other and, taken together, suggest that in neither case is the correlation between size effects and impurity effects accidental. Indeed, one is led to wonder whether such a relation might not be quite general.

There also exist published measurements of size effects and impurity effects on the resistivity of aluminum.[12] For various reasons, a comparison is difficult in that case and can only be made with experimental results for somewhat higher temperatures. Nevertheless, the results seem to be consistent with the possibility that (at least for $\rho_{0°K} \lesssim 30 \times 10^{-10}\,\Omega$ cm) the same type of correlation holds for aluminum as for indium and tin.

In all these metals, more precise and extensive experimental studies are required. It would clearly be of great interest to determine how nearly the relation discussed here is exact or universal. Even the present evidence, however, suggests the advisability of a more unified theoretical treatment of the effects of boundary scattering and impurity scattering on the temperature dependence of resistivity.[13]

[7] R. B. Dingle, Proc. Roy. Soc. (London) **A201**, 545 (1950).

[8] The situation is, in general, quite complex and deserves separate treatment. A short discussion of the relevant facts is given in J. E. Neighbor and R. S. Newbower (unpublished). This paper comprises a critical discussion of evidence for the importance of electron-electron scattering in the electrical resistivity of simple metals at low temperature.

[9] L. Nordheim, Act. Sci. Ind., No. 131 (Hermann & Cie., Paris, 1934).

[10] The deviations from Matthiessen's rule implied by Refs. 6 and 7 are associated with a variation of l/d. Therefore, both studies implicitly yield impurity-dependent deviations for small specimens of equal size, since l, in addition to its temperature dependence, is a function of impurity concentration. Neither calculation, however, would appear to give the simple behavior exhibited in Fig. 1.

[11] R. Reich and K. Forsvoll, Compt. Rend. **261**, 125 (1965).

[12] J. B. Van Zytveld and Jack Bass, Phys. Rev. **177**, 1072 (1969), and references cited therein.

[13] Some speculations on mechanisms consistent with the results of Reich and Forsvoll (Ref. 10) can be found in I. Holwech and J. Jeppesen, Phil. Mag. **15**, 217 (1967).