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Temperature and impurity dependence of the electrical resistivity in dilute gallium-based alloys. I. *b*-axis crystals

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Experimental results on the deviations from Matthiessen's rule in the electrical resistivity are presented for a series of single-crystal gallium alloys oriented for current flow along the [010] (*b*) axis. Indium, tin, and zinc solutes were used to span a range of over three decades of variation in the residual resistivity. Comparison is made with a recent theory which relies on the isotropization of the electron distribution function as impurity scattering is increased as well as with the "momentum nonconservation" model of Campbell *et al*. Certain common features of the observed deviations in the metals in the III-*B* group of the periodic table lead to the conclusion that of the two, the latter theory is better able to account for the results.

I. INTRODUCTION

It has long been recognized that any attempt to determine the nature of the scattering mechanisms which contribute to the electrical resistivity of an ideal metal is complicated by the fact that in any real metal, no matter how pure, there always exists an appreciable contribution from "residual" scattering mechanisms such as impurity or boundary scattering. The empirical assumption implicit in Matthiessen's rule (MR) that impurity and boundary scattering contribute only a temperature-independent term to the resistivity has been shown in a preponderant number of cases not to be in agreement with the results of experiment. It has been observed that even in the simplest metals, the magnitude and variation of the temperature-dependent part of the resistivity are affected by the amount of impurity scattering present in the metal, especially for temperatures well below the Debye characteristic temperature (cf. Bass[1] for a review). Experimental studies of the size effect[2-5] in extremely pure metals provide similar evidence in the case of boundary scattering.

It is important, therefore, that care be exercised in attributing a specific measured temperature dependence to a given ideal scattering mechanism because the deviations from MR must be corrected for. For this reason much attention has been focused on obtaining data on the deviations from MR in a large number of metals so that a valid theoretical explanation of the effect might be formulated. At present it appears that no treatment based on a detailed account of the scattering process is adequate in explaining all the details of the existing data.

Progress has been made, however, along the lines of developing phenomenological descriptions which involve the use of adjustable parameters to fit the data. Notable among this type of description are the so-called isotropization[6] models which are based on a consideration of the anisotropic nature of umklapp electron-phonon scattering at low temperatures[7,8] and which have been used to fit the data for several alloy series. The agreement of such models with experiment is quite good in some respects, but unconvincing in others. It appears that any exact association of such a model with the detailed features of the scattering problem remains at best superficial.

The area of most recent experimental interest is the investigation of the deviations from MR in dilute alloys at low ($T \leq 20$ K) temperatures because as the temperature is lowered the anisotropy of the electron-phonon distribution function becomes increasingly more marked, and so comparison with various theoretical calculations based on this phenomenon is more readily accomplished. Systematic studies have been carried out on dilute alloys of aluminum,[9] zinc,[10] tin,[11] and several other metals having reasonably well-known band structures,[12] with the view that knowledge of the details of the Fermi surface can be used to advantage in constructing a theoretical model.

There is a growing body of evidence,[13,14] however, that band-structure properties play a minor role at best and that the deviations from MR in a number of metals might be caused by some totally unaccounted-for scattering mechanism which is essentially universal in nature and which is independent of those features specific to a particular metal, as long as the overall properties such as effective interaction strength and Fermi surface area are taken into account. The model proposed

by Campbell *et al.*[15] in connection with data[9] on aluminum, which assumes that crystal momentum is not conserved in the dirty limit, clearly predicts such behavior.

This evidence will be discussed in some detail later in light of the present experimental study of the deviations from MR in gallium-based single-crystal alloys oriented for current flow along the b [010] axis of the orthorhombic lattice. The study was carried out for the most part in the liquid-helium temperature range, but selected data were taken at temperatures ranging up to ~7 K and above. This study is the first of its kind to employ single-crystal (rather than polycrystalline) specimens with known amounts and types of impurity doping. This particular orientation in gallium exhibits the highest conductivity, and coupled with the fact that this metal is available in very pure form, the residual resistivity measured along the b axis for high-purity gallium is lower than that presently recorded for any other metal (~0.03 nΩ cm). This fact makes it possible to detect changes in the temperature-dependent part of the resistivity upon the addition of impurities on the part per million level, thus more than amply satisfying the criterion for a dilute alloy.

II. THEORETICAL BACKGROUND

The number of theoretical models which have been put forward in an attempt to explain the observed deviations from MR is quite large,[1] and it would serve no good purpose to present a detailed account of them all here. It is useful, nevertheless, to consider some of the general features exhibited by a number of these specific models to facilitate comparison with the experimental results. For this purpose we consider two broad categories of theoretical model for the deviations from MR: (i) deviations arising from the intrinsic anisotropy of electron-phonon scattering at low temperatures and (ii) deviations occurring as the result of some unaccounted-for additional contribution to the scattering.

The first of these groups is loosely classified under the heading of "isotropization." The basic idea underlying such a model is that the highly directional nature of low-temperature electron-phonon scattering with umklapp, and hence the highly anisotropic spatial variation of the electronic distribution function for phonon scattering, is increasingly ameliorated by isotropic impurity scattering as the impurity concentration is increased. This idea represents the main thrust of most of the variational calculations[16,17] which have been put forward to explain the results for multiband metals such as aluminum where umklapp can play an important role. It is also clear that certain calculations[18,19] based on the diffusion model of Klemens[20] rely on the same physical idea.

Schotte and Schotte[21] have considered the variational approach to calculating the deviations from MR from a very general standpoint, and have shown that at a fixed low temperature where there is an appreciable difference between the spatial variation of the phonon and impurity distribution functions, the deviation function [defined as $\Delta \equiv \rho(T) - \rho_{\text{ideal}}(T) - \rho_0$] must exhibit a "flattened" steplike behavior when plotted against the residual resistivity ρ_0, of the form $\Delta \sim \rho_0/(A + B\rho_0)$. This step can span 2–3 orders of magnitude of variation in ρ_0 and eventually saturates in the "dirty" limit value when $\rho_0 \gg \rho_{\text{ideal}}$. In this case, the step height and width apparently depend on the disparity in the spatial variation of the ideal and the impurity distribution functions.

The same general features are shared by any theory which is based on the premise that the isotropic distribution produced by impurity scattering eventually washes out the highly anisotropic nature of the distribution for electron-phonon scattering at low temperatures. It is not surprising, moreover, that the behavior expected on the basis of an isotropization model is similar to that predicted by the so-called two-band model, because again the same general idea of one group of electrons gradually dominating another group as the residual resistivity is increased is used. A convenient representation[1] of the two-band model's prediction for the deviation function is

$$\Delta = \frac{\rho_i(T)\rho_0(\beta - \alpha)^2}{\alpha(1+\beta)^2\rho_0 + \beta(1+\alpha)^2\rho_i(T)}, \tag{1}$$

where ρ_0 and $\rho_i(T)$ are the residual and ideal resistivities, respectively; α and β are adjustable parameters which characterize the ratio of ideal and of impurity scattering in the two "bands". The characteristic $\rho_0/(A + B\rho_0)$ dependence is evident as long as α and β are constants, and it is not inaccurate to state that most models which incorporate the isotropization idea could be represented in terms of a multiband model with suitably chosen parameters. A key feature of this type of theoretical explanation for the deviations from MR is the saturation of Δ in the "dirty" limit.

Dosdale and Morgan[8] have considered a detailed approach to the two-band idea as a prelude to a more rigorous calculation of the deviations from MR in the case of aluminum. The above authors distinguish the two bands by dividing the Fermi surface up into one region which is essentially free-electron-like where only small-angle phonon scattering is operative, and another region lying near zone boundaries where large-angle umklapp

scattering occurs in addition to the small-angle normal scattering. Because of the variation of the effective width of the umklapp region on temperature, the resulting expression for the deviation function yields a dirty limit value of

$$\Delta \simeq (\rho l/v_f)\eta B T^{(n+1)}, \tag{2}$$

where ρl is the product of the resistivity and the electron mean free path, v_f is the Fermi velocity in the free-electron approximation, BT^n is the umklapp contribution to the scattering frequency in the second band, and ηT is the fraction of the total Fermi surface area available for umklapp scattering.

This model, if correct, affords a means of arriving at an approximate assessment of the umklapp contribution to the resistivity. This implies that it can be demonstrated that the model fits the data over the entire range of variation of ρ_0 and of T. The above authors have, in addition, calculated the deviation function for aluminum in the dirty limit using a more detailed "multiband" model which takes many of the specific features of that metal's band structure into account. The agreement with the experimental results is fairly good in this limit, but detailed calculations on the ρ_0 dependence of Δ have not as yet been carried out on the basis of this model. It is clear that any such isotropization calculation will be highly sensitive to the precise details of the Fermi surface in question and so will undoubtedly yield different results for different metals.

The second type of theoretical model to be considered here was first proposed by Campbell *et al.*,[15] and later elaborated upon by Caplin *et al.*[22] The model involves the rather drastic assumption that crystal momentum is not conserved in the regime where deviations from MR become important. It should be pointed out that this model also predicts a saturation in the deviations from MR in the dirty limit so that the mere fact that such a saturation occurs offers no means of distinguishing one explanation from the other. Based on a simple heuristic argument which implies that all phonon scattering events are equally effective in contributing to the resistivity, the model put forward by Campbell *et al.*[15] results in the following prediction for the value of temperature-dependent part of the resistivity in the dirty limit:

$$\Delta\rho = \frac{4.8}{\Theta_D^2}\frac{d\rho}{dT}\Big|_{T\sim\Theta_D} T^3. \tag{3}$$

Caplin *et al.*[22] have also considered the interference resulting from combined electron-phonon and electron-impurity scattering, and argue that the onset of the resulting contribution to the deviations from MR is governed by the condition

$$q_T \sim (4\pi/l\lambda_F)^{1/2}, \tag{4}$$

where q_T is the dominant phonon wave vector at temperature T, l is the electron mean free path, and λ_F is the de Broglie wavelength of electrons at the Fermi surface. This expression can be recast in terms of the critical value of the residual resistivity ρ_0' where the onset of the deviations occurs to give

$$\rho_0' = q_D^2 (T/\Theta_D)^2 \rho l \lambda_F/4\pi, \tag{5}$$

where q_D is the Debye wave vector.

As yet, no calculations have been made concerning the contribution to the deviations from MR to be expected in the region between the onset value and the dirty limit and so no comparison can be made between this model and the experimentally determined behavior of $\Delta\rho$ on residual resistivity. It is a characteristic of this kind of explanation that if correct it must be a general feature of the resistivity in a large number of metals without regard to their specific band structure.

III. EXPERIMENTAL DETAILS

Experimental data on a total of 19 different single-crystal gallium specimens oriented to within 1° of the [010] axis by Laue back-reflection x-ray techniques are presented. Three different impurity solutes, zinc, indium, and tin, were used to alloy with 99.9999+% pure gallium stock [23] with widely varying degrees of success. Of the three, zinc was the one solute which was most readily soluble in gallium for single-crystal growth, with a solubility limit of approximately 2000 at ppm. Beyond this level, the lack of solubility is readily indicated both by dendritic precipitation of nonuniform regions during crystal growth and by the departure from a monotonic increase in the residual resistivity of the specimens with increasing impurity concentration.

All of the zinc alloys studied were grown from the supercooled melt using translucent plastic molds and conventional seeding techniques,[24] with potential probes grown as an integral part of the specimen. As reported in a previous paper,[14] in the case of tin and indium solutes the precipitation limit was reached at much lower solute concentration levels, lying at approximately 20 at ppm in each case. In an effort to overcome this problem, a number of tin and indium alloys were grown as single crystals using a zone leveling technique with magnetic stirring of the molten zone.[25]

The resistance of each specimen was measured using a superconductive tunneling device to serve

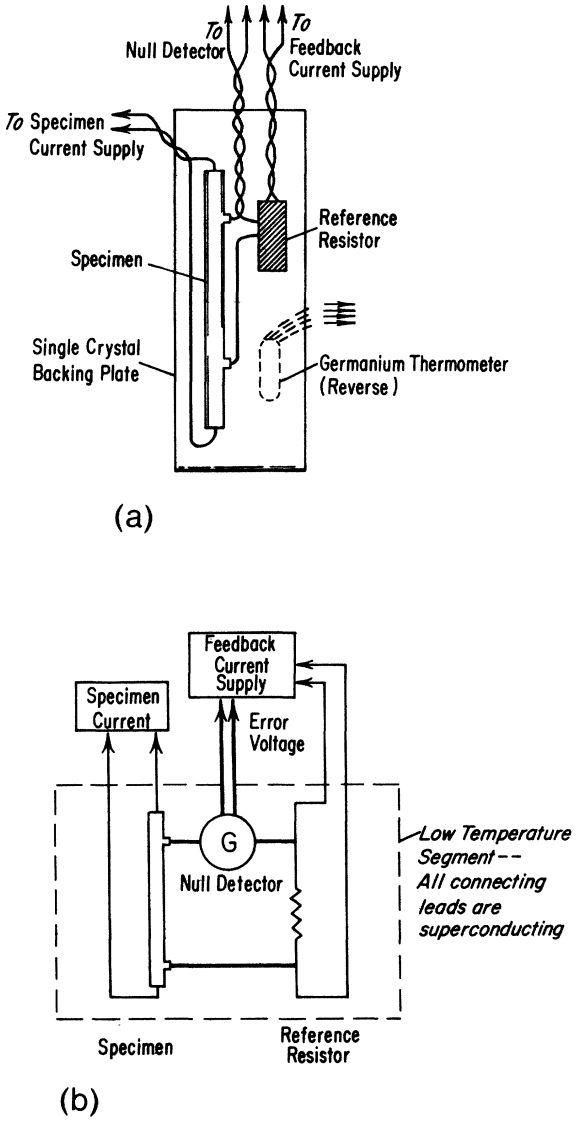

FIG. 1. (a) Detail of the specimen holder. (b) Schematic diagram of the measurement system.

as a galvanometric null detector in a Lindeck bridge circuit with feedback,[26] which is represented schematically in Fig. 1, along with a detailed drawing of the specimen mounting arrangement. In the course of the investigation, two types of device were used as null detectors, a Clarke[27] solder-drop SLUG and later, a commercially built rf SQUID. The use of such a sensitive detection system enables the resulting resistance measurements to be carried out with a relative precision of 0.01% or better, even though the specimen current is at all times kept at or below 10 mA in order to minimize the galvanomagnetic effects of the field produced by the measuring current. In the purest samples measured, the voltage developed under these conditions is on the order of 10^{-10} V.

The reference resistors used were fabricated from brass flat stock which is coated with a superconducting layer on each face where contact to the current and voltage leads is made. The reference resistors were calibrated first by measuring the absolute resistance at 4.2 K using a conventional room-temperature measurement system which incorporates an absolute resistance standard, and then by measuring the relative temperature dependence of the reference resistor utilizing the high-precision superconducting circuit and another dummy resistor which is held at a fixed low temperature. A reference resistor design was achieved wherein the total temperature dependence from 1.2 to near 6 K was limited to less than 0.1%. Tests were also conducted to check for the possibility of the variation of the resistance with time and thermal cycling with negative results. It should be pointed out that the uncertainty in the temperature dependence of the reference resistor easily becomes the dominant source of error in the measurement of $\Delta\rho \equiv \rho(T) - \rho_0$ in highly impure specimens where the temperature dependent part of the resistance at 4.2 K is only on the order of 1 part in 10^3 of the total resistance.

In order to obtain resistivity values from the measured specimen resistance values, the shape factor $1/A$ was determined by direct measurement and also compared against the value calculated by measuring the specimen resistance at room temperature and using published[28] values of the resistivity for b-axis gallium at this temperature. Correction for thermal contraction was made using published data.[29] The error connected with this procedure leads to an absolute uncertainty of ~0.5% in both the ρ_0 and the $\Delta\rho$ values to be presented since the difference in resistance is determined from measurements having much greater relative precision. Table I summarizes the physical properties of the specimens including solute type and nominal concentration, the method of growth, and the value of the residual resistivity extrapolated from the measurements.

IV. RESULTS AND DISCUSSION

A. Low temperature ($T \lesssim 4.2$ K) results

It was previously pointed out that the key assumption in the isotropization model was that the temperature must be low enough to ensure that the distribution function for phonon scattering is sufficiently anisotropic to produce significant deviations from MR. In an attempt to satisfy this assumption, the measurements undertaken in this investigation were concentrated in the liquid-helium temperature range (4.2 K and below), where

TABLE I. Physical properties of the specimens.

Sample no.	Solute	Concentration (nominal) (at. ppm)	Method of growth (M: mold) (ZL: zone leveled)	Cross-sectional dimension (square) (mm)	ρ_0 ($n\Omega$ cm)
1	In	5	M	2.5	0.0521
2	Sn	2	M	2.5	0.0540
3	In	...	ZL[a]	...	0.0873
4	In	15	M	2.5	0.0865
5	In	...	ZL	...	0.0962
6	In	60	M	2.5	0.1256
7	Zn	50	M	1.0	0.1401
8	Zn	75	M	1.0	0.1697
9	Sn	...	ZL	...	0.1732
10	Sn	10	M	2.5	0.1941
11	Sn	...	ZL	...	0.3809
12	Sn	...	ZL	...	0.4072
13	Zn	100	M	1.0	0.5568
14	Sn	...	ZL	...	0.7116
15	Zn	200	M	1.0	2.700
16	Zn	500	M	1.0	11.47
17	Zn	500	M	1.0	11.40
18	Zn	1000	M	1.0	51.80
19	Zn	2000	M	1.0	57.05

[a]All zone-leveled specimens had a round cross section with a nominal diameter of 2.5 mm.

the ratio T/Θ_D is on the order of $\frac{1}{100}$ for gallium. A plot of the temperature-dependent part of the resistivity at 4.2 K, $\Delta\rho(4.2)$, versus the logarithm of the residual resistivity extrapolated from data taken at 1.2 K and above is presented in Fig. 2 for all of the specimens investigated. The indium and tin alloys which were mold grown had cross-sectional dimensions of 2.5 mm sq are plotted as open circles. All of the zinc alloys had nominal cross-sectional dimenions of 1 mm sq and are plotted as full circles. As can be seen, to within the scatter, no clear distinction can be drawn between the data for the various solutes used. This result is in essential agreement with the aluminum results obtained by Caplin and Rizzuto,[9] Krsnik *et al.*,[30] and Babić *et al.*[31] Also included on this plot are two points $P1$ and $P2$ representing samples grown from "pure" stock (as received) with square cross-section and nominal dimension of 2.5 and 1.0 mm, respectively. The pure sample

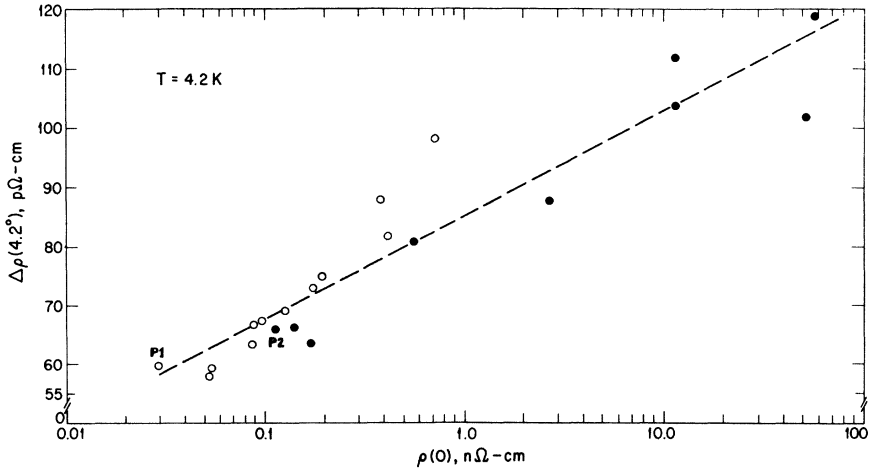

FIG. 2. Temperature-dependent part of the resistivity $\Delta\rho$ at $T = 4.2$ K vs $\log(\rho_0)$ for gallium alloys oriented along the b axis. Open circles are Ga-In and Ga-Sn alloys: Full circles are Ga-Zn alloys.

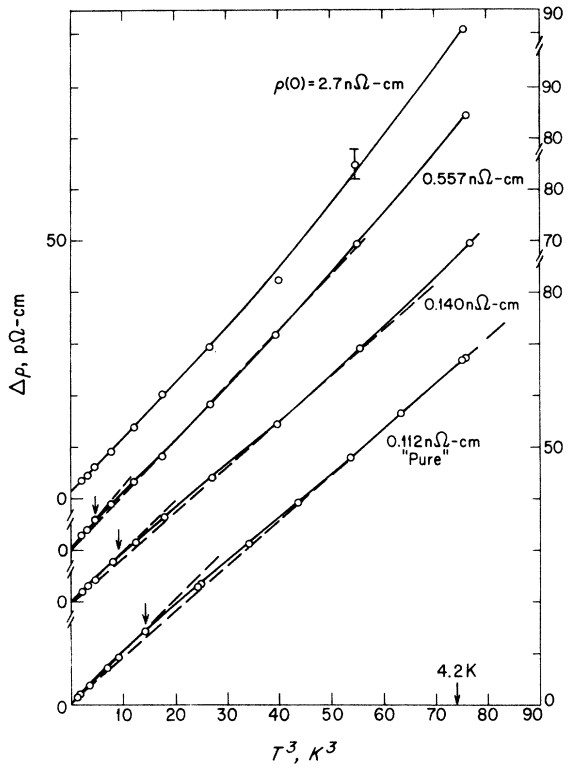

FIG. 3. Temperature dependent part of the resistivity $\Delta\rho(T)$ vs T^3 for a pure specimen and for alloys with increasing residual resistivity.

results illustrate the fact that a considerable contribution to the residual resistivity of a pure specimen arises from boundary scattering, on account of the fact that the electron mean free path in this material is quite long. It has been well established experimentally[24] that not only does boundary scattering contribute to the residual resistivity in this case but also to the temperature dependent part of the resistivity. It is therefore not feasible to simply subtract the resistivity of the pure specimen from that obtained for the alloys to determine the deviation function $\Delta(T) = \Delta\rho(T) - \rho_{\text{ideal}}(T)$ as it is usually defined, on the assumption that $\Delta\rho_{\text{pure}}(T) \simeq \rho_{\text{ideal}}(T)$. It is conceivable that such a procedure would become valid at some higher temperature where the importance of boundary scattering is reduced, but then the basic requirement for low-temperature phonon scattering would have to be relaxed.

In Fig. 3 the temperature dependent part of the resistivity for several Ga-Zn alloy specimens and a nominally pure sample of the same cross-sectional dimensions (1 × 1 mm) is plotted versus T^3 to facilitate comparison of the behavior of $\Delta\rho(T)$ with and without additional impurity scattering. It is apparent that the distinctive inflection (see arrows) which appears in $\Delta\rho(T^3)$ for the pure specimen gradually becomes washed out, and that the temperature dependence of $\Delta\rho$ very closely approaches a simple cubic power law for T less than ~4.2 K as ρ_0 is increased by introducing additional impurity scattering.

Based on the assumption that both boundary and impurity scattering affect the temperature-dependent part of the resistivity, and, hence, both mechanisms produce deviations from MR, one can assume that the temperature-dependent part of the resistivity is composed of the following contributions:

$$\Delta\rho(T) = \rho_{\text{ideal}}(T) + \Delta'(T, c, d/l)\,, \tag{6}$$

where the deviation function Δ' is explicitly assumed to be dependent upon both the impurity concentration c and the ratio of the specimen dimension to the electron mean free path. The simplest assumption which can be made about the contributions to Δ' from impurity and boundary scattering is that they are additive. Thus, we assume that the quantity Δ' consists of two terms,

$$\Delta'(T, c, d/l) = \Delta_1'(T, c) + \Delta_2'(d/l[c, T])\,. \tag{7}$$

As indicated by the explicit dependence of the mean free path both on T and on c, each of these deviation contributions is intrinsically interrelated.

It is possible to gain a qualitative idea of what the deviations due to boundary scattering might look like by considering the specific calculation of Dingle[32] for round wires in the free-electron approximation. The resistivity of a specimen of diameter d, $\rho_d(T)$, can be represented by the expression

$$\rho_d(T) = \rho_b(T) + (\rho l/d)G(d/l)\,, \tag{8}$$

where $\rho_b(T)$ is the resistivity of the bulk material which can be taken to be equal to

$$\rho_b(T) = \rho_b(0) + \rho_{\text{ideal}}(T) + \Delta_1'(T, c)\,,$$

and $G(d/l)$ is a function which is model dependent and which in the case of Dingle's calculation has the form shown in Fig. 4, where $G(d/l)$ is plotted versus d/l.

The temperature-dependent part of the resistivity is thus given by

$$\Delta\rho(T) = \rho_{\text{ideal}}(T) + \Delta_1'(T, c) + \frac{\rho l}{d}\left[G\left(\frac{d}{l(c,T)}\right) - G\left(\frac{d}{l(c,0)}\right)\right], \tag{9}$$

where the last term represents Δ_2' in this model. First, we consider the temperature dependence of a very pure sample with $l(0)$ greater than d. The

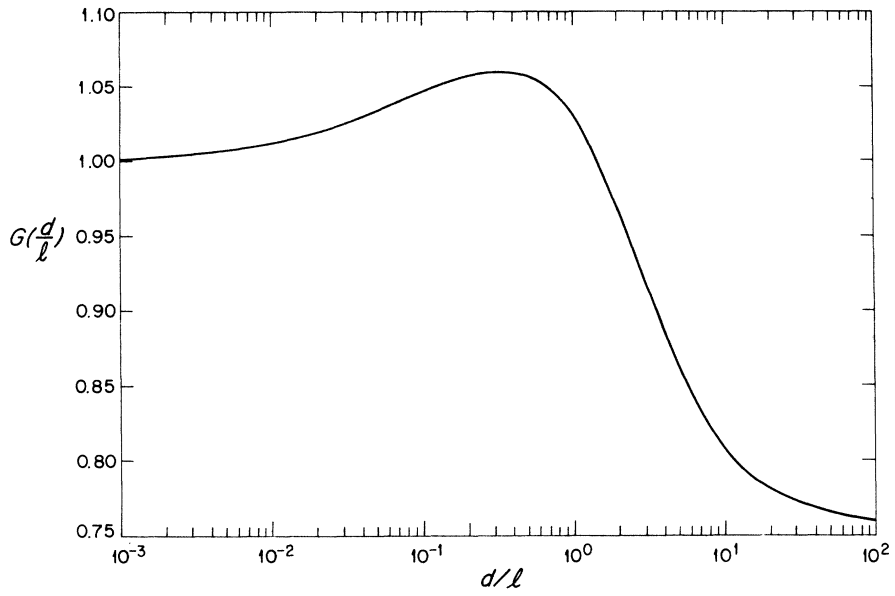

FIG. 4. Function $G(d/l)$ vs d/l according to the size-effect theory of Dingle.

temperature dependence of Δ_2' is seen to consist of the incremental variation of $G(d/l)$ as l varies from $l(0)$ to some smaller value $l(T)$. Using the value of 8.17×10^{-12} ohm cm^2 for ρl, as obtained by Cochran and Yaqub[24] for b-axis gallium, Δ_2' will contribute an amount to the resistivity equal to ~0.1 ΔGnΩ cm for specimens of the size used in this investigation. In the pure case, the mean free path varies by at least an order of magnitude in going from 0 to 4.2 K and so ΔG is approximately 0.1. This results in a total contribution from Δ_2' of ~0.01 nΩ cm, or about 20% of the entire temperature-dependent resistivity of the pure specimen at 4.2 K. Although Dingle's model is much too oversimplified for application to a metal with such a complicated band structure, it is nevertheless interesting that the temperature dependence of Δ_2' predicted for a specimen where l varies from a value greater than d at $T=0$ K to a value somewhat less than d as the temperature is increased exhibits an inflection similar to that found in the data for the pure specimens if it is assumed that $1/l \propto T^3$ or some higher power of T. Furthermore, as the impurity concentration is increased, the starting value of $l(0)$ approaches d, which means that the inflection moves to lower temperatures as the residual resistivity is increased. Eventually the starting value of $l(0)$ becomes smaller than d, and the inflection disappears as the total contribution to $\Delta\rho$ becomes negative. At the same time, the range of variation in l between 0 K and some fixed temperature T decreases, thereby decreasing the magnitude of this contribution.

In view of the fact that the exact temperature dependence of l is not known and the model used by Dingle is not sufficiently realistic, it is not worthwhile to attempt a more detailed comparison with the experimental data. This matter will be treated in a future paper on the deviations from MR due to boundary scattering. It nevertheless is clear that it is possible to associate the observed inflections in the data with the predicted dependence of Δ_2' on ρ_0 and T. It follows that if one wishes to gain information about the impurity deviation function Δ_1', care must be exercised to ensure that Δ_2' makes a negligible contribution. In this temperature range, it appears that this assumption can safely be made for all specimens which exhibit a residual resistivity at least one order of magnitude greater than that of the pure specimen.

B. Comparison with theory

As is apparent from Fig. 3, the temperature dependence of $\Delta\rho$ for those specimens in which the deviations from MR are impurity dominated is quite accurately described by a cubic power law in the range 1.2 to 4.2 K. Even the presence of the deviations produced by boundary scattering only slightly alters the accuracy of the above statement. For this reason, we have chosen to fit the data for all of the specimens investigated to the form $\rho(T)=\rho_0+\mathfrak{B}T^3$. The results of a least-squares fit of all the alloy data obtained at 4.2 K and below are presented in Table II. Although no clear-cut indication of the existence of saturation of $\Delta\rho$ in the limit of large ρ_0 is exhibited by the results, if we may assume that the highest $\Delta\rho$ values measured must be fairly close to the limiting value, then several conclusions emerge upon comparison

TABLE II. Results of a least-squares fit of the data taken between 1.2 and 4.2 K to the form $\rho(T)=\rho_0+\mathfrak{G}T^3$. The quantity $\Delta\mathfrak{G}$ is the standard deviation of the slope.

Specimen no.	ρ_0 ($n\Omega$ cm)	$\mathfrak{G}$ ($n\Omega$ cm/K^3)	$\Delta\mathfrak{G}/\mathfrak{G}$ (%)
1	0.0521	7.81×10^{-4}	4.0
2	0.0540	8.00	3.6
3	0.0873	9.01	2.6
4	0.0865	8.58	3.0
5	0.0962	9.10	2.1
6	0.1256	9.36	3.1
7	0.1401	8.96	1.1
8	0.1697	8.56	1.1
9	0.1732	9.87	1.7
10	0.1941	10.6	1.5
11	0.3809	11.9	2.3
12	0.4072	11.1	2.7
13	0.5568	10.9	1.8
14	0.7116	13.3	1.3
15	2.700	12.0	1.5
16	11.47	15.1	2.0
17	11.40	14.0	2.0
18	51.80	13.6	4.6
19	57.05	16.1	5.5

with the various theoretical models considered previously.

According to the Dosdale-Morgan[8] model, the dependence of $\Delta\rho$ on $\log\rho_0$ should resemble a "flattened" step. Such a functional variation certainly could be used to fit the data for gallium to within the scatter. The observed variation of $\Delta\rho$ by approximately a factor of 2 over variation of several decades in ρ_0 is also consistent with the predictions of this as well as a number of other isotropization models.[7] In the Dosdale-Morgan model the temperature dependence of $\Delta(T)$ in the dirty limit provides information about the temperature dependence of the umklapp contribution to electron-phonon scattering. If we take the dependence of $\Delta(T)$ to be proportional to T^3, then the umklapp processes would contribute a T^2 term to the scattering frequency according to Eq. (2). It is interesting to note that a similar conclusion was drawn by the above authors in the case of aluminum. Although no explicit statement is made to this effect in the Dosdale-Morgan paper, one would expect that as ρ_0 is decreased from the dirty limit region, the temperature dependence of $\Delta\rho$ will exhibit a changing temperature dependence to reflect the increasing importance of normal phonon scattering. It is clear that, apart from the contribution from boundary scattering, the experimentally determined temperature dependence of $\Delta\rho$ remains cubic to a very good approximation, so apparently the expected changeover in the temperature dependence does not occur in this metal. The only consistent explanation would include the highly unlikely condition that normal phonon scattering also contributes the same kind of cubic dependence.

The striking similarities[14] between the gallium data and those obtained on other metals tend to point to an explanation for the deviations which is not as strongly sensitive to the specific properties of a given metal as is the Dosdale-Morgan model, or for that matter, most of the theoretical descriptions in the isotropization category. On the other hand, the explanation proposed by Campbell, Caplin, and Rizzuto[15] to explain the data on aluminum can, if valid, be expected to apply to most metals in a very general way. The idea that the observed deviations from MR at low temperatures are the result of a breakdown of the adiabatic approximation leads to a quite general prediction concerning the temperature dependence of $\Delta\rho$ in the dirty limit. It is assumed that all phonon scattering events affect the resistivity equally in this limit so that $\Delta\rho$ should be proportional to the available phonon density which depends on T^3 at low temperatures. This dependence is of course what is observed experimentally. Using the expression obtained by Campbell *et al.* in Eq. (3), the dirty limit value of $\Delta\rho$ (4.2 K) predicted for b-axis gallium is ~0.15 nΩ cm. The experimental results indicate a value close to 0.12 nΩ cm which is in fair agreement with this prediction. A key bit of evidence favoring the model of Campbell *et al.* over the numerous alternative isotropization explanations is the fact that neglecting size effects, the experimentally observed temperature dependence of $\Delta\rho$ is seen to conform to a T^3 law over the entire range of variation in the residual resistivity. Caplin and Rizzuto[9] have suggested that a dependence given by $\Delta\rho(T)=CT^3\log\rho_0$ provides a reasonable fit for aluminum. Clearly the b-axis data for gallium can be represented by a similar functional dependence. The best fit to a function of this form yields for the coefficient C a value of 2.4×10^{-4} nΩ cm/K^3.

Morelli *et al.*[14] have noted that all the metals in the III-B column of the periodic table (Al, In, Ga) exhibit low-temperature deviations of this form. In fact, when the $\Delta\rho(T)$ values are normalized by the quantity $\Delta\rho'=\rho_\Theta(T/\Theta_D)^3$, the slope of the normalized deviation $\Delta\rho(T)/\Delta\rho'\log\rho_0$ is very nearly the same for all three metals over the entire temperature range in each case where $\Delta\rho$ is found to be proportional to T^3. This result is strongly suggestive of the presence of a single scattering mechanism which exhibits increasing strength as ρ_0 is increased. Caplin *et al.*[22] have also considered the implications of the model for the residual resistivity value necessary to induce the onset of the

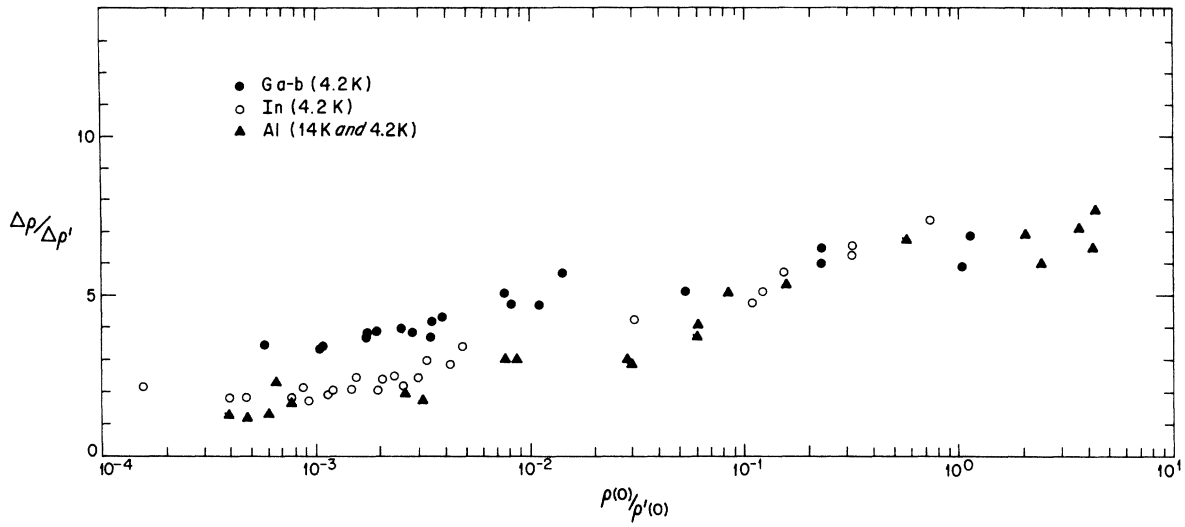

FIG. 5. Normalized temperature-dependent part of the resistivity vs logarithm of the residual resistivity normalized by the onset value ρ_0' for the metals Al, In, and Ga (b axis).

deviations from MR. In the case of b-axis gallium, Eq. (5) yields a value for ρ_0' of 5×10^{-8} Ω cm at 4.2 K. Clearly this value overestimates the onset resistivity by several orders of magnitude. Keeping in mind that the argument of the above authors is essentially of a dimensional nature, we can however attempt to test the result more qualitatively by normalizing ρ_0 by ρ_0'. In the case of the three

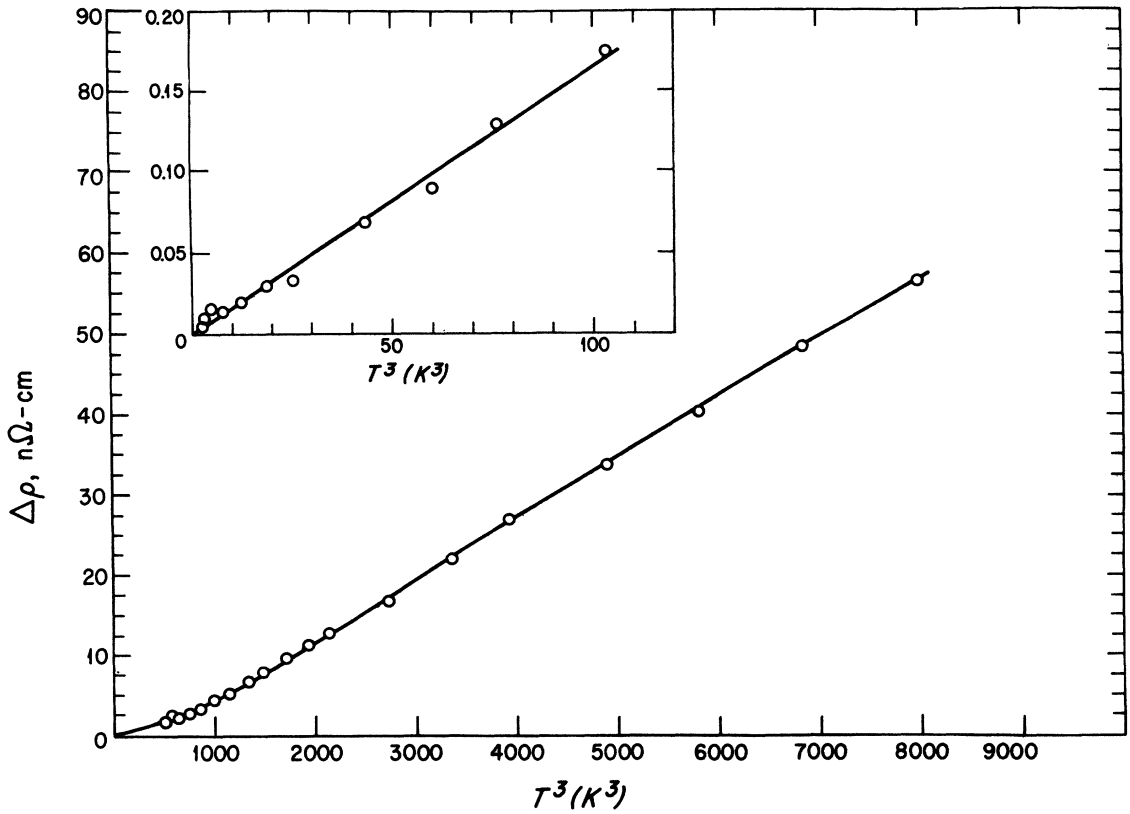

FIG. 6. Temperature-dependent part of the resistivity $\Delta\rho(T)$ vs T^3 for the most impure specimen measured over the range 1.2–20 K.

III-B metals, a sufficient amount of data exists where the deviations are proportional to T^3 to consider a plot of the normalized deviation $\Delta\rho/\Delta\rho'$ vs $\log(\rho_0/\rho_0')$. The result is presented in Fig. 5 where data on indium and gallium (b axis) at 4.2 K and aluminum at 14 K are plotted. The values used for the various parameters which appear in the expression for ρ_0' were the following. Al: $\Theta_D = 428$ K, $\rho l = 5.5\ \text{p}\Omega\ \text{cm}^2$; In: $\Theta_D = 111$ K, $\rho l = 12.5\ \text{p}\Omega\ \text{cm}^2$; Ga($b$ axis): $\Theta_D = 325$ K, $\rho l = 8.2\ \text{p}\Omega\ \text{cm}^2$. The normalized data for both aluminum and indium are in apparently quite good agreement over the entire range of more than four decades of variation of ρ_0/ρ_0'. The data for gallium b axis appear to lie above the curve determined by the other two metals in the region where ρ_0/ρ_0' is small. It is possible that the discrepancy arises from the contribution to $\Delta\rho$ from boundary scattering which was estimated to account for ~20% of the total temperature dependence for the purest samples. In the dirty limit region however, where the complication due to boundary scattering does not arise, the data for all three metals exhibit a close correspondence. It is clear that this normalization provides additional evidence that there indeed appears to be a universal behavior with the III-B metals. Taken at face value, the momentum nonconservation model appears to be the only theory presently available which can be used to predict such close agreement. Whether or not the choice of parameters which combine to give ρ_0' is fortuitous remains to be seen after enough data for other metals become available. Based on rather meager amounts of data for other metals, Cimberle *et al.*[13] have reached a similar conclusion concerning this aspect of the momentum nonconservation model, by considering the temperature dependence of ρ_0'.

C. Temperature dependence in the dirty limit

In Fig. 6 the temperature-dependent part of the resistivity for the sample with the highest residual resistivity measured in this study is plotted against T^3 over the temperature range from 1.2 to 20 K. Clearly, the fit to a T^3 dependence is strictly valid only below ~3.5 K. Above this temperature the temperature dependence becomes stronger than T^3. In order to determine the nature of the dependence in this range, a plot of $\log\Delta\rho$ vs $\log T$ is shown in Fig. 7. There are several regions which can be delineated according to the assumed fit to a power law. However, in no case does the power approach the value of 4.5 which has been determined by Olsen-Bär and Powell[33] and Weisberg and Josephs[34] to fit the data for pure gallium. This case also contrasts with that of aluminum where the changeover from the "ideal" T^5 law to a T^3 dependence takes place over a comparatively smaller temperature span. The details of the nature of the scattering processes in gallium are obviously rather complex. In this context, it is therefore even more significant that in the limit of low temperatures the same cubic temperature dependence appears in the dirty limit for gallium as it does in

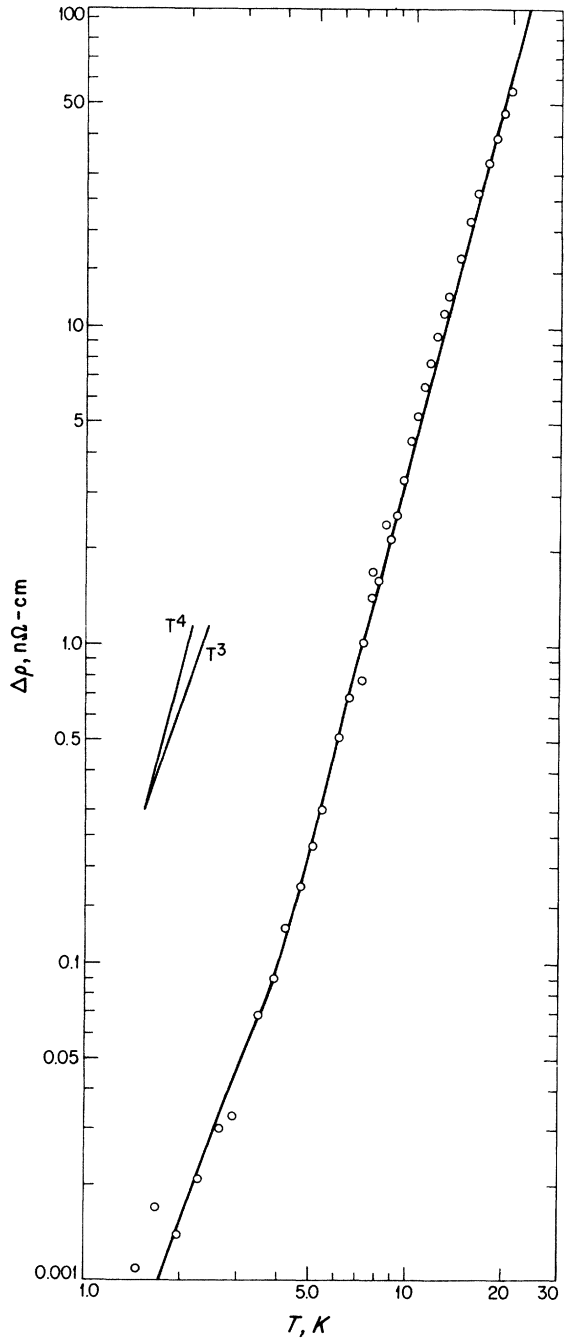

FIG. 7. Log-log plot of $\Delta\rho(T)$ vs T for the most impure sample to determine power-law fit.

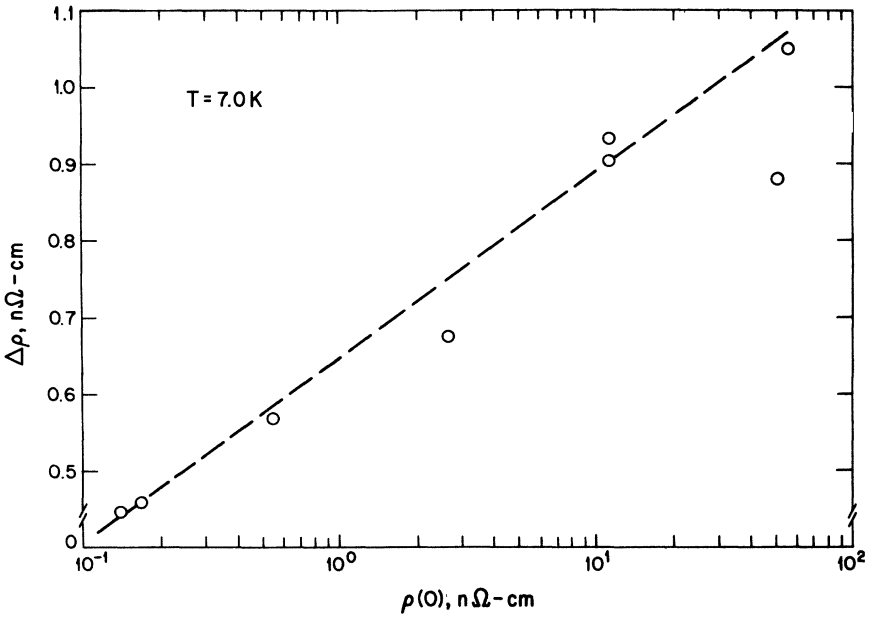

FIG. 8. Temperature-dependent part of the resistivity $\Delta\rho$ at $T=7.0$ K vs $\log(\rho_0)$ for gallium alloys with zinc impurity.

the much simpler case of aluminum. This is not to say that the deviations from MR disappear or become lessened as soon as the T^3 dependence is surpassed. On the contrary, the dependence of $\Delta\rho$ on ρ_0 continues to increase as the temperature is increased. In Fig. 8 the temperature-dependent part of the resistivity at 7.0 K, $\Delta\rho(7\text{ K})$ is plotted versus $\log\rho_0$ for the zinc alloys, to reveal increased contribution to the deviations at this temperature. It is noteworthy that although the absolute value of $\Delta\rho$ for each specimen has of course increased, the fractional increase in $\Delta\rho$ between the most pure and the most impure specimens has remained essentially the same as it is at and below 4.2 K. This result suggests that the deviations from MR scale in proportion to the ideal resistivity over the entire temperature range covered in this investigation. The limiting T^3 behavior at low temperatures thus appears to represent a "saturation" in the effectiveness of the mechanism responsible for the deviations. It is felt that this conclusion also argues in favor of the existence of an independent and universal scattering process in these metals which is not simply the result of isotropization of the electron distribution for phonon scattering.

V. CONCLUSIONS

The data on the temperature-dependent part of the resistivity for the b-axis orientation of gallium can be represented as a function of temperature and of residual resistivity by $\Delta\rho = CT^3\log\rho_0$. It has been demonstrated that this form is similar to that suggested by Caplin and Rizzuto to represent their results for aluminum. The present results have been compared qualitatively with the isotropization model of Dosdale and Morgan and could readily be represented by such a theory with the appropriate choice of adjustable parameters. We observe, however, that such a choice would lead to a rather implausible assumption concerning the nature of Normal electron-phonon scattering in gallium.

On the other hand, the present results, when taken together with data on two other metals in the III-B group display striking similarities. We conclude that, apart from some accidental coincidence, such behavior must be characteristic of the presence of some quite general scattering mechanism with effects which are independent of details of band structure, etc.

The momentum nonconservation model put forward by Campbell *et al.*[15] provides a consistent framework within which the low-temperature data for the III-B metals are well represented. However, it is not possible at present to reach any final conclusion concerning its validity. In our view, several avenues of investigation must be pursued in order to settle the question. First and most importantly, it is necessary that a detailed theory of the effect be completed, so that direct comparison can be made with the data in the intermediate region between onset and the dirty limit. Second, further experimental investigation should be undertaken on transport effects in which the anisotropy of the electron-phonon scattering distribution is expected to be markedly different than

in the case of the electrical resistivity in the presence of impurity scattering. For example, the contribution due to anisotropy in the electron distribution for phonon scattering is appreciably different[21] in the case of heat transport and so measurement of the deviations from MR in the thermal resistivity would provide a key indication of the presence of any universal mechanism such as the momentum nonconservation ideal of Campbell *et al.*[15]

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