

The plot clearly indicates no evidence for the presence of harmonics of the lens oscillations or for the presence of the short-period Grenier oscillations.

It has been demonstrated that submillimeter diameter cylinders may be prepared by spark-trepanning without significant reduction in the bulk mean free path. This is evidenced by the fact that the observed oscillations are of approximately the same amplitude

as those observed in Sondheimer samples of equivalent thickness. The use of cylindrical samples in conjunction with field-modulation detection of the oscillations will allow probing of the curvature of Fermi surfaces by rotation of the magnetic field without the complication of loss of sample symmetry. Further, the order of magnitude increase in practical current density results in an equivalent increase in signal.

## Scattering-Center Effects in the Magnetoresistance of Nickel

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The magnetoresistance of many polycrystalline samples of nickel alloyed with small concentrations of iron, cobalt, manganese, chromium, and carbon has been measured. An analysis of the data gave the result that all those specimens in which the conduction-electron scattering was dominated by one particular scattering center were found to obey Kohler's rule. However, the details of the magnetoresistance were different in each case, with the exception of the iron, cobalt, and manganese impurities, which formed one group. In addition, the thermal-scattering magnetoresistance and that due to deformation were different again. These observations confirm previous conclusions concerning the validity of Kohler's rule. Extremely large magnetoresistance was found for the iron, cobalt, and manganese group of impurities as scattering centers, which apparently is much larger than observed in any other system. This can be correlated with other electronic properties of the impurities, such as the resistivity and magnetic moment per unit concentration. In particular, the high magnetoresistance appears associated with the presence of large local moments. The zero-field anisotropy in the resistance is also correlated with these same properties. In spite of this correlation, we have no clear interpretation of the extreme magnetoresistance.

### I. INTRODUCTION

NOW that properties of the Fermi surface have become relatively well understood, the role of electron scattering processes has become of importance in understanding some of the electronic transport properties of metals. We note recent work by Dugdale and Basinski<sup>1</sup> relating breakdowns in Matthiessen's rule in noble metals to anisotropies in the relaxation times and work by Bailyn and Dugdale<sup>2</sup> on effects in the electronic contribution to the thermoelectric power, again in the noble-metal alloys. Attention to this aspect of transport properties was first drawn by Coles<sup>3</sup> in connection with Hall effect studies. Since then, further work has been contributed by Cooper and Raimes,<sup>4</sup> Ziman,<sup>5</sup> Hurd,<sup>6</sup> and Heine.<sup>7</sup> In particular, Ziman has

given a discussion of transport properties in noble-metal alloy systems based on well-known Fermi-surface properties and shown that in some cases considerable anisotropy in the relaxation times could be concluded from the experimental results. Deaton and Gavenda<sup>8</sup> have made direct measurements of these anisotropies in copper. Most of the work to date has been carried out on noble-metal alloys. In this paper, we are concerned with magnetoresistance measurements on very dilute alloys of nickel in which effects thought to be due to electron scattering effects are identified. Rather large breakdowns in Matthiessen's rule in more concentrated alloys of the same type have recently been identified by Fert and Campbell<sup>9</sup> and by Farrell and Greig<sup>10</sup> and identified by them as due to spin mixing (see Campbell, Fert and Pomeroy,<sup>11</sup> and Bourquard, Daniel and Fert<sup>12</sup>).

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<sup>1</sup> J. S. Dugdale and Z. S. Basinski, *Phys. Rev.* **157**, 552 (1967).

<sup>2</sup> M. Bailyn, *Phys. Rev.* **157**, 480 (1967); J. S. Dugdale and M. Bailyn, *ibid.* **157**, 485 (1967).

<sup>3</sup> B. R. Coles, *Phys. Rev.* **101**, 1254 (1956).

<sup>4</sup> J. R. A. Cooper and S. Raimes, *Phil. Mag.* **4**, 145 (1959).

<sup>5</sup> J. M. Ziman, *Phys. Rev.* **121**, 1320 (1961).

<sup>6</sup> C. M. Hurd, *Phil. Mag.* **12**, 47 (1965).

<sup>7</sup> V. Heine, *Phil. Mag.* **12**, 53 (1965).

<sup>8</sup> B. C. Deaton and J. D. Gavenda, *Phys. Rev.* **129**, 1990 (1963).

<sup>9</sup> A. Fert and I. A. Campbell, *Phys. Rev. Letters* **21**, 1190 (1968).

<sup>10</sup> T. Farrell and D. Greig, *J. Phys.* **C1**, 1359 (1968).

<sup>11</sup> I. A. Campbell, A. Fert, and A. R. Pomeroy, *Phil. Mag.* **15**, 977 (1967).

<sup>12</sup> A. Bourquard, E. Daniel, and A. Fert, *Phys. Letters* **26A**, 260 (1968).

The intimate connection between the electron scattering process and the magnetoresistive behavior of a metal has recently been emphasized by Pippard.<sup>13,14</sup> Thus, if the scattering contains a significant amount of small-angle scattering, a diffusion model for motion of electrons over the Fermi surface becomes appropriate and this may have marked effects on the magnetoresistance<sup>13</sup> (see also Klemens and Jackson<sup>15</sup>). This conclusion was illustrated by calculations on copper and there is evidence that one section of the Fermi surface of nickel has a similar topology to that of copper.<sup>16</sup> Alternatively, it may be appropriate to make the relaxation time approximation and assume that the relaxation time is anisotropic, i.e., a function of the electron wave vector  $k$ . In that case, the coefficient of  $B^2$  in the low-field expansion for the resistivity  $\rho(B)$  is related to averages of the relaxation time over the Fermi surface<sup>17</sup> and the saturation value of the longitudinal magnetoresistance (current parallel to  $B$ ) depends on anisotropy in the mean free path<sup>18</sup>  $l[\tau(k)]V|V|$ <sup>19</sup> where  $\tau(k)$  is the anisotropic relaxation time and  $V$  is the Fermi velocity.

A first step in the analysis of magnetoresistive behavior is often to see if the data are governed by Kohler's rule<sup>20</sup>  $\Delta\rho/\rho_0 = F(B/\rho_0)$ , i.e., the fractional increase in resistance  $\Delta\rho/\rho_0$  in a magnetic induction  $B$  of a specimen of resistivity  $\rho_0$  at zero induction is a function only of the parameter  $B/\rho_0$ . A discussion of Kohler's rule has been given by Chambers<sup>21</sup> who suggests that the rule is valid provided that a relaxation time approximation can indeed be made, that changes in  $\rho_0$  for a given set of samples give rise to negligibly small changes in electron band-structure details and, finally, that changes in  $\rho_0$  result only in changes in the magnitude of the relaxation time  $\tau(k)$  and not in the form of the dependence on  $k$ . We, therefore, expect that the  $\tau(k)$  corresponding to two differing scattering centers may give rise to two different forms of the function  $F$  in Kohler's rule. Thus, an exploration of the validity of Kohler's rule may identify for us similarities and dissimilarities in the  $\tau(k)$ , provided, of course, that Chambers's other two criteria are satisfied. It is not entirely clear what the status of Kohler's rule is in situations for which the relaxation time approximation is not valid, such as the small-angle

scattering situation discussed by Pippard.<sup>13,14</sup> We shall later outline arguments why it still may be valid under appropriate conditions and we suspect that under these conditions the rule may be relatively general. Experimental breakdowns of Kohler's rule in copper have been reported by de Launay, Dolecek, and Webber<sup>22</sup> and by Jongenburger<sup>23</sup> in circumstances for which it is plausible that changes of anisotropy are occurring, and Tsui and Stark<sup>24</sup> attribute similar breakdowns in this rule in cadmium to anisotropies in the scattering probabilities.

In the following sections, we will describe data for nickel doped with varying amounts of Fe, Co, Mn, Cr, and C which we relate to differing anisotropies of the relaxation time associated with each impurity. The concentrations in each case are small enough that we believe the band structure to be essentially unaffected. One difficulty associated with ferromagnetic metals is the occurrence of the magnetic induction  $B$  and not the applied field  $H$  in Kohler's rule. For nickel, the difference  $4\pi M_s$  is 6.4 kG and changes the value of  $\rho_0$  considerably for a pure sample. We have discussed this problem elsewhere in connection with the determination of the temperature dependence of the "ideal" resistivity<sup>25</sup> and also in connection with the problem of determining the residual resistivity of nickel specimens and the question of the validity of Kohler's rule.<sup>26</sup> In this paper, we lay more emphasis on the magnetoresistance corresponding to well-characterized systems so that comparisons between the Kohler's function  $F$  relating to different scattering centers can be made. We also compare the data with that representing ideal resistivity and with that from dislocation-dominated samples. The observations reinforce the conclusions of our earlier work<sup>25,26</sup> concerning the conditions of validity of Kohler's rule and suggest that breakdowns in Kohler's rule may well yield useful information in studies of the role of the electron scattering process in transport properties.

A final note should be made about the choice of some of the systems for these experiments, i.e., as given above, alloys of Ni with Fe, Co, Mn, and Cr. Experimentally, a distinction can be drawn between Fe, Co, and Mn, on one hand, and Cr on the other, since, according to neutron diffraction, Fe, Co, and Mn are characterized by a local moment which is different from that of the nickel matrix, whereas Cr is not.<sup>27</sup> Corresponding differences are also noted in the changes in magnetic moment/unit concentration (*NiCr* alloys deviate from the Slater-Pauling curve),<sup>27</sup> and in the resistivity/unit concentration (Cr impurities give a much larger re-

<sup>13</sup> A. B. Pippard, Proc. Roy. Soc. (London) **A282**, 464 (1964).

<sup>14</sup> A. B. Pippard, Proc. Roy. Soc. (London) **A305**, 991 (1968).

<sup>15</sup> P. G. Klemens and J. L. Jackson, Physica **30**, 2031 (1964); **31**, 1421 (1965).

<sup>16</sup> D. C. Tsui and R. W. Stark, Phys. Rev. Letters **17**, 871 (1966); L. Hodges, D. R. Stone, and A. V. Gold, *ibid.* **19**, 655 (1967).

<sup>17</sup> See, for example, J. P. Jan, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1957), p. 26.

<sup>18</sup> A. B. Pippard, *The Dynamics of Conduction Electrons* (Gordon and Breach, Science Publishers, Inc., New York, 1965), p. 99.

<sup>19</sup> The more general case of anisotropic-scattering probabilities has been considered by M. C. Jones and E. H. Sondheimer, Phys. Rev. **155**, 561 (1967).

<sup>20</sup> M. Kohler, Ann. Physik **32**, 211 (1938).

<sup>21</sup> R. G. Chambers, Proc. Roy. Soc. (London) **A238**, 344 (1956).

<sup>22</sup> J. de Launay, F. L. Dolecek, and R. J. Webber, J. Phys. Chem. Solids **11**, 37 (1959).

<sup>23</sup> P. Jongenburger, Acta Met. **9**, 985 (1961).

<sup>24</sup> D. C. Tsui and R. W. Stark, Phys. Rev. Letters **19**, 1317 (1967).

<sup>25</sup> F. C. Schwerer and J. Silcox, Phys. Rev. Letters **20**, 101 (1968).

<sup>26</sup> F. C. Schwerer and J. Silcox, J. Appl. Phys. **39**, 2047 (1968).

<sup>27</sup> G. G. E. Low and M. F. Collins, J. Appl. Phys. **34**, 1195 (1963).

sistivity increment).<sup>28</sup> The first theoretical discussion of these differences was given by Friedel<sup>29</sup> in terms of the relationship of the impurity  $d$  level to the Fermi level. A more recent discussion has been given by Campbell and Gomes.<sup>30</sup> Thus, there are good reasons for expecting qualitatively different impurity electronic structures in the two cases leading to differences in the anisotropy of the electron scattering.

## II. EXPERIMENTAL PROCEDURE

Specimens were cut 5.5 cm long and 3 mm wide from Johnson Matthey and Co. nickel foil (0.003 in, nominal purity 99.997%). For potential probes, nickel wires (0.04 mm diam.) were spot welded along the long axis of the sample at a gauge length of 3.0 cm. With suitable annealing treatments,<sup>26</sup> these foils were found to have resistivity ratios  $R_{273}/R_{4.2}$  in excess of 2500. The solute material (Cr, Mn, Fe, Co, or C) was evaporated onto both large surfaces of the sample either in an electron-beam evaporator or from hot tungsten filaments. Specimens prepared in each way gave identical results. Alloying was accomplished by letting the solute diffuse into the foil to a uniform concentration. For this, the sample was placed in a vacuum chamber which was evacuated to  $10^{-7}$  Torr. The sample was surrounded with a liquid-nitrogen trap and a high-purity atmosphere was flushed continually through the chamber while the sample was heated by passing a dc current through the foil. Annealing conditions were chosen to minimize the diffusion time  $t^0$ , reevaporation of the solute, and reactions with the atmosphere. The samples were annealed for periods in excess of  $t^0 = \delta^2/4D$ , where  $\delta$  is the half-thickness of the foil and  $D$  is the diffusion coefficient for the solute in nickel at the annealing temperature. For iron, cobalt, and chromium solutes, the samples were annealed for 5 h at 1250°C in a nitrogen atmosphere at  $10^{-3}$  Torr. For manganese solute, the samples were annealed for 1.5 h at 1200°C in a nitrogen atmosphere at 0.05 Torr. For carbon solute, anneals were 10 min at 1050°C in a carbon dioxide atmosphere. After annealing, the grain size was typically of order 10  $\mu\text{m}$ . In this manner, alloys of nickel with iron (10–10 000 ppm), cobalt (20–1210 ppm), manganese (12–60 ppm), chromium (5–1700 ppm),<sup>28</sup> and carbon<sup>31</sup> (10–4400 ppm) were prepared. The homogeneity of the alloys was checked by reannealing a few samples from each series for another full period  $t^0$ . The magnetoresistivity of these double annealed samples was entirely compatible with that of the singly annealed samples, a test, which the results presented below will suggest, is quite sensitive.

Some samples prepared in this fashion, particularly those for the highly magnetoresistive series, initially showed large deviations from the appropriate Kohler plot. These “bad” samples could not be related to the evaporation procedure, annealing time, or solute concentration, but rather were ascribed to contamination during the anneals. The remarkable feature, however, is that by annealing these samples in either an argon-oxygen or nitrogen atmosphere (see Schwerer and Silcox<sup>26</sup> for details), the samples could be cleaned up and then fitted the appropriate Kohler plot. This contamination accounted for only a fraction (generally less than one-half) of the total resistivity. Consequently, Kohler’s rule provides a sensitive test for adventitious effects of this sort.

At the time the experiments were done, the geometrical factor (gauge length/cross-section area) for each sample was deduced from Matthiessen’s rule and the measured resistances at 0°C and 4.2°K; White and Woods<sup>32</sup> value of 6.2  $\mu\Omega\text{cm}$  was used as the ideal resistivity. The work by Farrell and Greig<sup>10</sup> and Fert and Campbell<sup>9</sup> on comparable alloys has identified breakdowns in Matthiessen’s rule, and it has proved necessary to reevaluate this procedure. For dilute alloys of the type discussed here, the correction due to Matthiessen’s-rule breakdown has proved relatively small and has not changed the Kohler plots significantly. The main effect has been to change our estimates of the impurity concentration present, since these were determined from the residual resistivity. All estimates of impurity concentration have been corrected for this effect.

Magnetoresistance measurements were made at 4.2°K in applied fields up to 18 kOe. The resistance at zero applied field was measured to  $\pm 0.02 \mu\Omega$  by a four-probe technique using a Minneapolis-Honeywell six-dial potentiometer. The difference of the voltage across the specimen from the zero-field setting of the potentiometer was amplified and plotted on an  $X$ - $Y$  recorder as a function of applied magnetic field for fields parallel and perpendicular to the current. Changes in resistance were measured with a relative accuracy of better than 1%, and nonlinearities due to the measuring circuit were less than 0.5%.

In addition to facilitating the dilute alloying, the sample shape minimized demagnetizing fields for the relevant orientations. Demagnetizing fields in the plane of the foil, due to the saturation moment, were estimated using Osborn’s<sup>33</sup> expression for the general ellipsoid as  $H_{11}^D = 1.7$  Oe for the field parallel to the long axis of the sample and current (longitudinal orientation) and as  $H_{11}^D = 150$  Oe for the field perpendicular to the long axis (transverse orientation). Where necessary, all data has been corrected for this effect.

<sup>28</sup> Estimated from the residual resistivity and the data of C. W. Chen, Phys. Letters 7, 16 (1963).

<sup>29</sup> J. Friedel, Nuovo Cimento 7, Suppl. 2, 287 (1958).

<sup>30</sup> I. A. Campbell and A. A. Gomes, Proc. Phys. Soc. (London) 91, 319 (1967).

<sup>31</sup> Estimated from the residual resistivity and the data of F. C. Schwerer, J. Appl. Phys. 40, 2705 (1969).

<sup>32</sup> G. K. White and S. B. Woods, Phil. Trans. Roy. Soc. London A251, 273 (1959).

<sup>33</sup> J. A. Osborn, Phys. Rev. 67, 351 (1945).

The low-field portion of the transverse resistivity curves (that portion showing effects associated primarily with technical magnetization) was sensitive to a misalignment of  $0.5^\circ$ , whereas a misalignment of  $5.0^\circ$  did not produce a detectable change in the high-field transverse magnetoresistivity data. The longitudinal resistivity was much less sensitive to a misalignment of the sample. All specimens were aligned to within  $0.5^\circ$  of the desired orientation.

### III. ANALYSIS OF DATA

#### A. Background

The change in resistivity  $\Delta\rho(B)$  of a metal in a magnetic field  $B$  due to Lorentz forces on the conduction electrons depends in a complex fashion upon the electronic character of the material and upon the orientation of the field and the current. The magnetoresistivity  $\Delta\rho(B)/\rho_0$  always saturates for fields applied parallel to the current  $\rho_{||}(B)$  (longitudinal magnetoresistivity) and, depending on the topology of the Fermi surface, either saturates or continues to increase for field applied perpendicular to the current  $\rho_{\perp}(B)$  (transverse magnetoresistivity).<sup>34</sup> In general, longitudinal magnetoresistivity is the weaker effect. Kohler's rule, used to describe this behavior, can be deduced from the solution of the relevant Boltzmann equation in a relaxation time approximation. The tube integral expression for the magnetoresistive behavior  $F$  (involving electron velocities and scattering processes over the Fermi surface) contains the magnetic field only in the combination  $B\tau(\mathbf{k})$  where  $\tau(\mathbf{k})$  is the relaxation time as a function of electronic wave vector. If changes in the resistivity  $\rho_{0i}$  result solely from changes in magnitude but not in the functional form of the scattering process [e.g.,  $\tau(\mathbf{k}) \rightarrow (1/c)\tau(\mathbf{k})$ ], then  $F$  is a function of  $B\tau(k)/c$ , i.e., a function of  $B/c$ . Since  $\rho_{0i}$  is a measure of the strength of the scattering processes  $c$ , this implies that  $F$  is a function only of  $B/\rho_{0i}$ . Such considerations led Chambers<sup>21</sup> to the criteria mentioned in the Introduction for the validity of Kohler's rule, namely, (i) collisions can be described in terms of a relaxation time, (ii) the filled region in  $\mathbf{k}$  space does not change appreciably in size over the range of temperature or purity considered, and (iii) changes in temperature or purity simply alter all  $\tau(\mathbf{k})$  by the same factor. Appropriate modifications of these arguments are discussed in Sec. V.

#### B. Method of Analysis

Because  $B$  is the relevant field, a problem arises which is illustrated by Fig. 1. The measurements give  $\Delta\rho/\rho_{0i}$  over a window only in the argument  $9kG/\rho_{0i} < B/\rho_{0i} < 24kG/\rho_{0i}$ , where the lower limit is given by the value of the induction corresponding to the field needed to remove the ferromagnetic domain structure

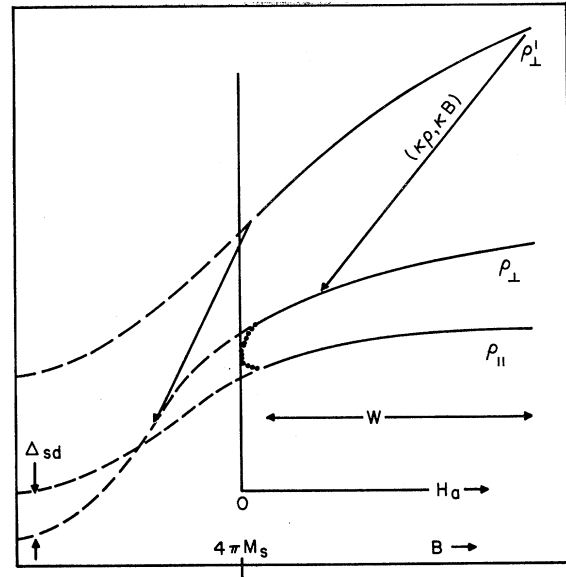


FIG. 1. Schematic representation of the longitudinal  $\rho_{||}$  and transverse  $\rho_{\perp}$  magnetoresistance of a ferromagnetic metal. The solid curves represent the experimentally measurable portions and the arrows indicate the result of reducing the data over the range  $W$  under Kohler's rule.

and the upper limit is instrumental. The procedure which we have followed previously<sup>25,26</sup> and review here is essentially to vary  $\rho_{0i}$  in a controlled fashion, taking care to alter only the number of scattering centers while keeping the type of scattering center the same. It is, thus, possible to map out the function  $F$  over a wide range of the argument. We note that  $\rho_{0i}$ , being the resistivity at zero induction, is experimentally unobtainable and that, accordingly, Kohler's rule has to be approached in an oblique fashion. We write it in the form

$$\kappa_i(c)\rho_i(B) = \rho^* \{1 + F[\kappa_i(c)B/\rho^*]\}, \quad (1)$$

where  $\kappa_i(c) = \rho^*/\rho_{0i}$  is the ratio of a standard resistivity  $\rho^*$  to  $\rho_{0i}$ . Analysis of the data in this form involves plotting  $[\rho(B), B]$  for a low-concentration sample. A value of  $\kappa(c)$  is then chosen for a sample with a slightly larger solute concentration ( $c$ ) such that a data point reduced as  $[\kappa(c)\rho(B), \kappa(c)B]$  falls on the curve for the original sample. If Kohler's rule is obeyed, then not only do the values of the magnetoresistivity match at this point but so will the slopes of the curves and the two sets of data become continuous. It was observed that the data from samples with the same solute did, indeed, reduce in this fashion to a unique curve; however, each solute series described a distinct curve and, naturally, the longitudinal and transverse data reduced to different curves. By a judicious choice of solute concentrations, the data from different samples will overlap sufficiently to ensure that the curve will be smoothly continued and that the total curve will span a large enough range of  $\kappa B$  to facilitate extrapolation to zero induction. The

<sup>34</sup> A. B. Pippard, *The Dynamics of Conduction Electrons* (Gordon and Breach, Science Publishers, Inc., New York, 1965), p. 98.

extrapolated value of the resistivity  $\rho^*$  represents the scattering process resistivity at zero induction for the sample with  $\kappa(c)=1$ . Indeed, the plot represents the full magnetoresistive curve of this  $\kappa(c)=1$  sample including values of  $B$  which are experimentally inaccessible. From  $\rho^*$  and the  $\kappa(c)$  values, the scattering process resistivity may be inferred for each sample in the series and meaningful Kohler plots can be made.

We note that it is not necessary that the transverse and longitudinal Kohler plots reduce to the same value of zero-induction resistivity.<sup>35</sup> Indeed, experimentally, in many cases, they do not. We denote such anisotropy with the notation  $\Delta_{sd}=\rho_{11}-\rho_{\perp}$ , and data on values of  $\Delta_{sd}$  will also be given in this paper.

These features are illustrated in Fig. 1, in which schematic  $[\rho(B), B]$  plots have been made for the longitudinal ( $\rho_{11}$ ) and transverse ( $\rho_{\perp}$ ) magnetoresistivity. The solid portion of the curves is that which is experimentally measurable. A small region associated with magnetic domain effects (technical magnetization) has been included. The curves illustrate the uncertainty in extrapolating the data of a low-resistivity sample from the saturated region to  $B=0$ . The anisotropy in the scattering process resistivity  $\Delta_{sd}$  is shown with the normally observed sign ( $\rho_{011} > \rho_{0\perp}$ ). To illustrate the mapping obtained by reducing data from other samples as  $(\kappa\rho, \kappa B)$ , a "measurable" transverse resistivity curve for a sample with a larger scattering process resistivity is plotted and arrows indicate the result of reducing the data.

That this procedure is necessary is shown by plots of the same data using the applied field  $H_a$  instead of  $B (=H_a + 4\pi M_s)$  [see Fig. 2 in comparison with Fig. 4(a)]. In these cases, data from samples in a given series which reduced appropriately for  $B$  do not reduce to a single plot either as  $(\kappa\rho, \kappa H)$  or as  $(\Delta\rho/\rho_0, H_a/\rho_0)$ .<sup>26</sup> The requirement that the data reduce under Kohler's rule and the experimental conditions of this study determined the field additional to the applied field to be  $4\pi M_s = 6.4$  kG to within 10%. From the saturating form of the magnetoresistivity and the large value of  $4\pi M_s$  relative to the applied fields, it is apparent that an accurate graphical extrapolation of the data to  $B=0$  for a single sample would be very difficult.

### C. Discussion of Errors

The results presented in this paper as well as those of our previous communications on this subject depend critically on the data being described by Kohler's rule. It is difficult to present the agreement with this rule convincingly as a Kohler plot since  $\Delta\rho/\rho_0$  and  $B/\rho_0$  each vary over three or four orders of magnitude for a given series. In turn, each series is composed of typically 15–25 samples with 15 reduced data points per sample. To enhance the credibility of these plots, we will discuss in some detail the limits on the reduction of our data

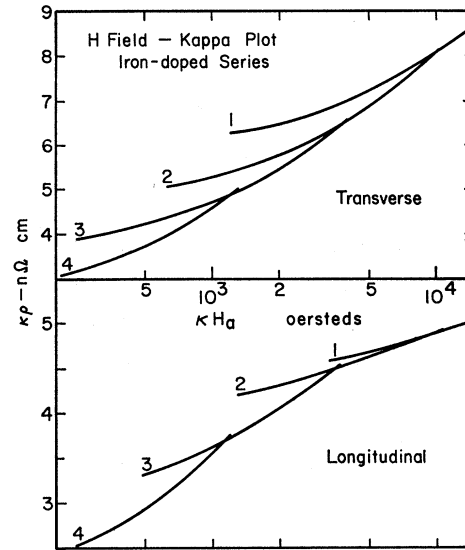


FIG. 2. Magnetoresistance of NiFe reduced as  $\kappa\rho, \kappa H_a$  for samples (1) 10-ppm Fe; (2) 20-ppm Fe; (3) 60-ppm Fe; (4) 275-ppm Fe.

under Kohler's rule. This discussion will be in the context of the graphical analysis technique described above; more sophisticated discussions would be appropriate to analytical, computer-assisted analyses. This discussion will relate to the reduction of the data as  $(\Delta\rho/\rho_0, B/\rho_0)$  rather than as  $(\kappa\rho, \kappa B)$ . Although these are physically identical, the former is a more sensitive test,  $\Delta\rho/\rho_0$  being a difference of two similar quantities.

Four considerations are relevant to a discussion of the degree to which Kohler's rule describes the data from several samples.

1. *Experimental errors.* Such errors always tend to obscure agreement with theoretical descriptions. With the methods described above, the change in resistivity was measured with uncertainties of the order of 1% of the total change, i.e.,  $\delta(\Delta\rho_i) \sim 0.01\rho_0 F$ . At very low values of  $F$ , the measuring system imposed a limit on the resolution of  $(\Delta\rho/\rho_0)$  of order  $2 \times 10^{-4}$ . However, over most of the range the resolution was dominated by errors inherent in the graphical techniques of order  $\delta(\Delta\rho/\rho_0) \sim 0.02F$ . Experimental errors in the determination of  $B$  were negligible and errors in  $B/\rho_{0i}$  arose principally during the assignment of values to  $\rho_{0i}$ . It is to be noted that the geometrical factor for each sample only enters in the variable  $B/\rho_{0i}$ , not in the more sensitive  $(\Delta\rho/\rho_{0i})$ . Thus,  $0.02F$  (or  $2 \times 10^{-4}$ ) essentially defines the uncertainty in the placement of data on a Kohler plot.

The first question concerns the extent to which experimental errors obscure true deviations from Kohler's rule. That is, data from sample 1 with Kohler function  $F_1$  is compared with either a known Kohler plot  $F_2$  or with data from a sample 2 described by  $F_2$ . If  $\rho_{01}$  and  $\rho_{02}$  are chosen such that the two Kohler plots coincide at  $b$ , and if the range of overlap of the two sets

<sup>35</sup> H. C. Van Elst, *Physica* 25, 708 (1959).

of data is  $\Delta b$ , then the deviation of the two curves at  $b + \Delta b$  will be

$$\Delta F \simeq (F_1' - F_2') \Delta b \simeq (n_1 - n_2) F \Delta b / b,$$

where the second approximation comes from using  $F = Ab^{n_i}$  (a most useful description for log-log Kohler plots). To be detectable, this deviation must be greater than  $\delta(\Delta\rho/\rho_0) \sim 0.02 F$ . In our experiment  $4 > \Delta b/b > 1$  and  $n$  ranges from 2 to 0.5. We, therefore, find a limit of resolution for  $n_1 - n_2$  of several percent.

A similar argument can be used to determine the usefulness of Kohler's rule in determining the field  $B$  governing the magnetoresistive behavior. In particular, we consider  $B = B + \Delta B$  where  $\Delta B$  is the deviation from the true value of the induction. The data from two samples described by the same Kohler function  $F(B/\rho_0)$  are assumed to be fitted at the high limit of  $B$  with appropriate values for  $\rho_{01}$  and  $\rho_{02}$ . Equating the deviation at the other limit of their range of overlap to the experimental limit of detection ( $0.02F$ ), we find a limit of resolution given by  $(\Delta B/B) = (\Delta F/F)/n(\Delta\rho/\rho)$ , where  $\Delta\rho = \rho_{01} - \rho_{02}$ . Inserting values of  $n = 1$ ,  $\Delta\rho/\rho = 0.5$  and  $\Delta F/F \simeq 0.02$ , values met in the course of our work, we find that  $\Delta B \simeq 0.1 (4\pi M_s)$ , i.e.,  $B$  has to be  $H + 4\pi M_s$  to within 10% of  $4\pi M_s$ .

If the  $\rho_{0i}$  are determined for a series by fitting  $\rho_i(B)$  to a curve of the form  $\rho_{0i}[1 + F(B/\rho_{0i})]$ , then the errors in  $\rho_{0i}$  will be of order

$$(\delta\rho_{0i}/\rho_{0i}) = [\delta\rho_i(B)/\rho_i(B)] + (\delta F/1 + F).$$

The first term  $\delta\rho_i(B)/\rho_i(B)$  is generally negligible and, as discussed above, we may put  $\delta F = 0.02F$ . Hence, for common situations with  $F$  less than unity, we should expect errors in the relative values of  $\rho_{0i}$  of less than 1%. However, in dilute highly magnetoresistive alloys,  $F$  can be as high as 10 and the limiting accuracy of 2% applied in these cases.

Note that, in order to minimize errors, measurements should be taken over as wide a range in  $H$  as possible.

2. *Systematic errors.* Certain systematic errors in analysis of the data are compatible with Kohler's rule. For example, if  $\rho_{0i} = \rho_{0i}^t(1 + \delta)$  is used to reduce the data where  $\rho_{0i}^t$  is the "true" zero-induction resistivity, then Kohler's rule gives

$$\frac{\rho(B) - \rho_{0i}}{\rho_{0i}} = \frac{-\delta_i}{(1 + \delta_i)} + \frac{1}{(1 + \delta_i)} F \left( \frac{B}{\rho_{0i}} (1 + \delta_i) \right).$$

Consequently, if  $\delta_i$  is constant for the series,  $[\rho(B) - \rho_{0i}]/\rho_{0i}$  remains a function only of  $B/\rho_{0i}$  and a Kohler-like plot will be obtained. If the resistivities  $\rho_{0i}$  are deduced from an erroneous extrapolated value  $\rho^*$  of the  $(\kappa_i\rho, \kappa_i B)$  plot as  $\rho_{0i} = \rho^*/\kappa_i(c)$ , then exactly this type of error ( $\delta_i$  constant) will result. However, the resultant Kohler-like plot will not extrapolate to zero as  $B/\rho_0 \rightarrow 0$ , may become negative, and will not approach a quadratic low-field dependence. Such criteria were used to reduce

these systematic errors such that the absolute accuracy of  $\rho_{0i}$  (and the placement of the Kohler plot) was limited by the random relative errors described above.

This type of systematic error gives a measured anisotropy in the scattering process resistivity ( $\Delta_{sd} = \rho_{011} - \rho_{01}$ ) proportional to  $\rho_{0i}$  with a proportionality constant equal to the difference between  $\delta_1$  and  $\delta_{11}$ , the fractional errors for the transverse and longitudinal resistivities. Since we wish to analyze  $\Delta_{sd}$  for a linear dependence on concentration ( $\rho_0^t$ ), these errors could be particularly troublesome. From the above consideration, we expect these systematic errors to limit our resolution of the coefficient ( $\Delta_{sd}/\rho_0^t$ ) to  $\sim \pm 0.01$ .

A similar troublesome systematic error is the one involved by assuming the validity of Matthiessen's rule and deriving the geometrical factor (Sec. II) in preference to direct measurement.

3. *Field dependences of the resistivity.* There exist other field dependences of the resistivity which are not due to Lorentz forces and are not described by Kohler's rule. These dependences may be of sufficient magnitude to give apparent deviations from a Kohler description of the resistivity. In this study, the fields used (the values of  $B/\rho_0$ ) were not large enough to necessitate considerations of magnetic breakdown and the field dependence of the spontaneous magnetization  $M_s$  (once the sample is technically saturated) is expected to be well below our limit of resolution. However, there remain scattering process resistivities which are field dependent, such as those associated with spin-disorder scattering or localized magnetic moments. If the scattering process resistivity is described by  $\rho_0(H) = \rho_0[1 + r(H)]$  and the Kohler description is

$$[\rho(B) - \rho_0(H)]/\rho_0(H) = F(B/\rho_0(H)),$$

then data reduced with a fixed value  $\rho_0$  will deviate from the Kohler plot by  $r(H)[1 + (1 - n)F]$ , where again we have used  $F = A(B/\rho_0)^n$ . At low fields ( $n > 1$ ),  $F$  is small and the deviations are of order  $r(H)$ . These deviations should be detectable only if the total change  $\int r(H) dH$  is larger than  $0.02F$ . At high fields  $n \rightarrow 0$ ,  $F$  may be as large as 10 or 20 and a total change greater than  $\sim 0.02$  should be detectable.

4. *Chamber's criteria.* Finally, there are deviations from Kohler's rule due to violations of one or more of Chambers's criteria. Having eliminated or put bounds on other sources of deviations and apparent agreements, we are now prepared to discuss observed deviations in terms of violations of these criteria.

## IV. OBSERVATIONS

### A. Magnetoresistivity

Magnetoresistivity data obtained with the carbon-, chromium-, and iron-doped samples in the transverse orientation are shown in the reduced form  $\kappa\rho, \kappa B$  in Fig. 3, together with data for a temperature dependent

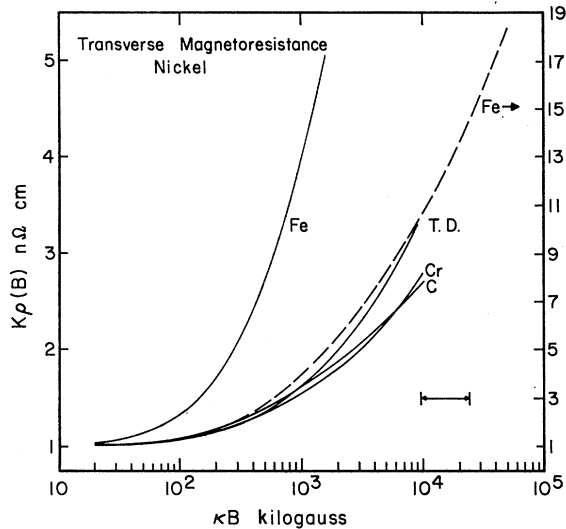


FIG. 3. Transverse magnetoresistance of nickel reduced as  $(\kappa\rho, \kappa B)$  for several scattering centers. The bar represents the range of  $\kappa B$  for a single sample. The scale for the dashed curve is to the right.

series.<sup>25</sup> Measurements were made at 14 field values between 2.4 and 18 kOe on 29 iron-doped samples, 15 chromium-doped samples, and 13 carbon-doped samples. The wide ranges of  $\kappa\rho$  and  $\kappa B$  covered and the high accuracy and large number of points involved preclude a meaningful reproduction of the actual data points. Rather, the data is presented as smooth curves where the width of the curve approximates the errors involved. A representative region of the iron-doped specimen data is shown expanded in Fig. 4(a) to indicate the excellent reduction of the data under Kohler's rule. A comparable region for chromium-doped data is shown in Fig. 4(b). This data has been normalized to a value  $\rho^* = 1 \text{ n}\Omega\text{cm}$  for all series. This same data failed to reduce appropriately when the applied field  $H_a$  was used, as shown in Fig. 2 (see Fig. 4, Schwerer and Silcox,<sup>26</sup> for plots of  $\kappa\rho$ , and  $\kappa H_a$  for the chromium-doped series). The data from each series was distinctive in the sense that it did not reduce compatibly with the data from any other series. In addition, series prepared by recovering cold-worked material<sup>26</sup> and by varying the temperature of a high-purity sample<sup>25</sup> also gave distinctive plots. However, data from the cobalt-doped series was indistinguishable from the iron-doped series and the manganese-doped series very closely approximated it. Over the concentrations studied, the cobalt and manganese solutes behaved similarly to the iron solutes, but doping was not achieved over as wide a range in these samples. Kohler plots for the iron-doped series are shown in Fig. 5 and the carbon-doped series in Fig. 6. A comparison of Kohler plots for all the scattering centers so far studied is given in Fig. 7 and illustrates the dramatic difference between the iron-, cobalt-, and manganese-doped series and the others. We emphasize

again that each Kohler plot apart from the iron, cobalt, and manganese series seems to be distinctive.

One feature of the data deserves comment. In terms of concentration dependence, the extensive iron-doped series divided into three regions. In the middle region of intermediate concentrations, estimated to be 600–2400 ppm, the data reduced to the Kohler plot only for fields greater than 10 kOe. For samples in the other two concentration regions, the data reduced to the Kohler plot for fields greater than 2.5 kOe (the field needed to remove the magnetic domain structure). Within experimental error, the same Kohler plot applied in all three ranges. The behavior in the intermediate range samples is suggestive of an impurity spin-ordering process. We return to this point later.

### B. Form of Longitudinal Magnetoresistance

The differences in the magnetoresistive behavior could be described quantitatively when it was observed that the longitudinal Kohler plots  $[\ln \Delta\rho(B)/\rho_0, \ln B/\rho_0]$  had identical shapes for all the series [i.e., iron, manganese, cobalt, and chromium doped, temperature-dependent,<sup>25</sup> recovery (i.e., dislocation scattering<sup>26</sup>), and some specimens doped by gas chemistry<sup>26</sup>] with the exception of the carbon-doped series. The common shapes implied that two parameters ( $\alpha^{-1}, \beta^{-1}$ ) could be found for each series such that the longitudinal data from all the series fell on a unique curve for the variables  $[\alpha^{-1}(\Delta\rho(B)/\rho_0); \beta^{-1}(B/\rho_0)]$ . Values for  $\alpha, \beta$  for each series were taken as those values which adjust the data to a master curve of the same shape which approaches a saturation value of  $\Delta\rho/\rho_0 = 1.0$  and which reaches half-saturation  $\Delta\rho/\rho_0 = \frac{1}{2}$  at  $B/\rho_0 = 10^8 \text{ kG}/\mu\Omega \text{ cm}$ . Discussion of the physical significance of  $\alpha, \beta$  will be given later. At this point, we note that  $\alpha$  becomes the saturation value of the longitudinal magnetoresistance for each series which, in the normal nearly free-electron picture, has been shown by Pippard<sup>24</sup> to be related to the anisotropy of the electron velocity and relaxation time over the Fermi surface. We relate  $\beta$  in some sense to the Hall constant  $A_H$  by remarking that in the tube integral formula for  $\sigma_{ij}$ , the appropriate argument for  $F$  is  $\omega_c \tau$ , i.e.,  $A_H' B/\rho_0'$ , where the prime denotes values for a specific orbit in  $k$  space. Since  $\beta$  comes into the argument as  $\beta^{-1} B/\rho_0$ , we may expect  $\beta^{-1}$  to be some measure of the Hall constant. The relevant data are given in Table I for the series discussed in this paper and by Schwerer and Silcox.<sup>25, 26</sup> Similar parameters for the carbon-doped series which did not reduce to this common curve were found from the criteria that  $\alpha$  is the saturation value of  $\Delta\rho/\rho_0$  and that  $\Delta\rho/\rho_0 = \frac{1}{2}\alpha$  at  $\omega_c \tau = 1$ . For comparison, the master curve and the carbon-doped series reduced by these parameters are shown in Fig. 8.

### C. Residual Resistivity Anisotropy

The anisotropy in the scattering process resistivity  $\Delta_{sd} = \rho_{011} - \rho_{01}$  (see Fig. 1) was calculated from the

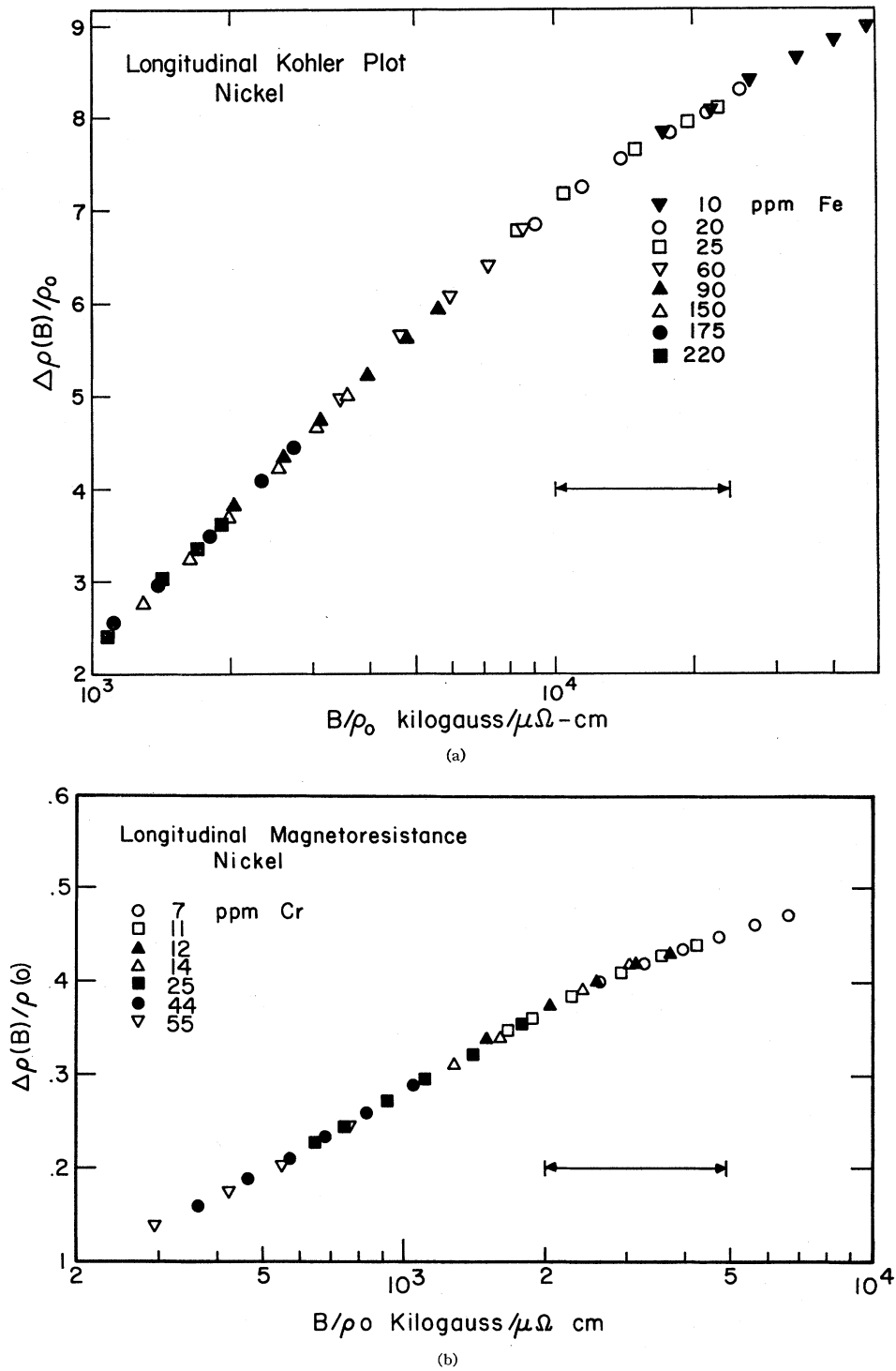


FIG. 4. (a) Longitudinal magnetoresistance of several samples from the *NiFe* series reduced as  $[\rho(B) - \rho_0]/\rho_0; B/\rho_0$ . The bar represents the range of  $B/\rho_0$  for a single sample. Compare this plot with the same data given in Fig. 2. (b) A comparable plot for a chromium-doped sample. Compare with Fig. 4 of Schwerer and Silcox (Ref. 26).

scattering process resistivities  $\rho_0$  used to form the Kohler plots. For the chromium-doped series  $\Delta_{sd}/\rho_{0\perp} = +(0.9 \pm 0.8)\%$  with no systematic deviations of the

values about the mean. The anisotropy at zero applied field  $\Delta(H_a=0)$  includes an additional anisotropy  $\Delta_{MR}$  from the magnetoresistance which also may be calcu-



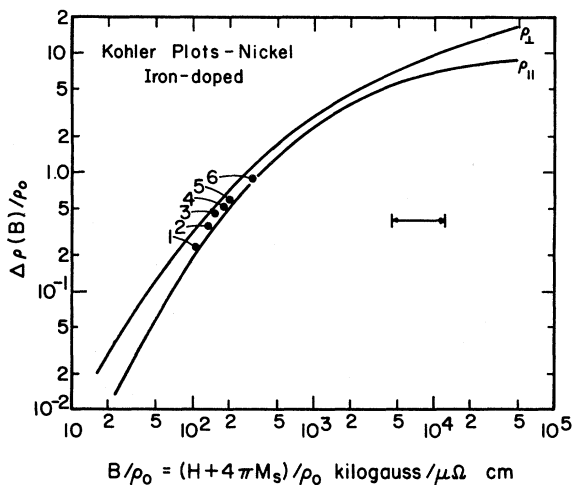


FIG. 5. Kohler plots of the transverse and longitudinal magnetoresistance of NiFe. The bar represents the range of  $B/\rho_0$  for a single sample. The numbered points were measured at an applied longitudinal field of 3.6 kOe for samples with (1) 1800-ppm Fe; (2) 1500 ppm; (3) 1360 ppm; (4) 1160 ppm; (5) 1060 ppm; (6) 680 ppm.

lated from the Kohler plots and is, in general, negative. If  $|\Delta_{MR}| > |\Delta_{sd}|$ , then the resistivity anisotropy at zero applied field  $\Delta(H_a=0)$  will be negative. For the chromium-doped series,  $\Delta(H_a=0)$  was always negative, as is seen in Fig. 9. For this series, we conclude that  $\Delta_{sd}$  is very small and, considering the experimental error, we are entitled to doubt whether any such anisotropy exists.

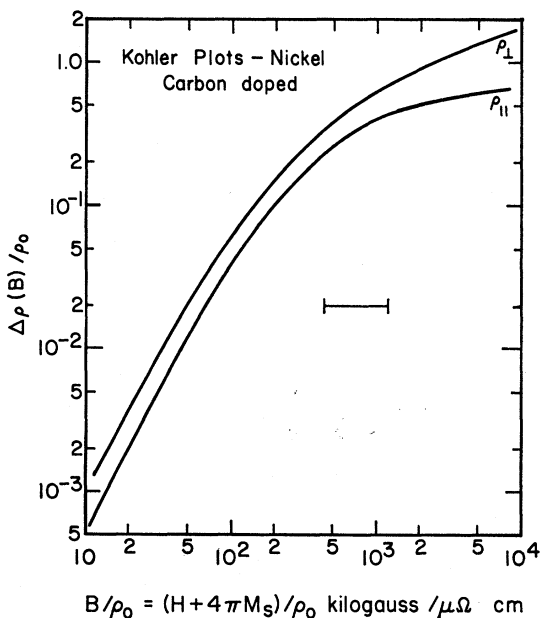


FIG. 6. Kohler plots of the transverse and longitudinal magnetoresistance of NiC. The bar represents the range of  $B/\rho_0$  for a single sample.

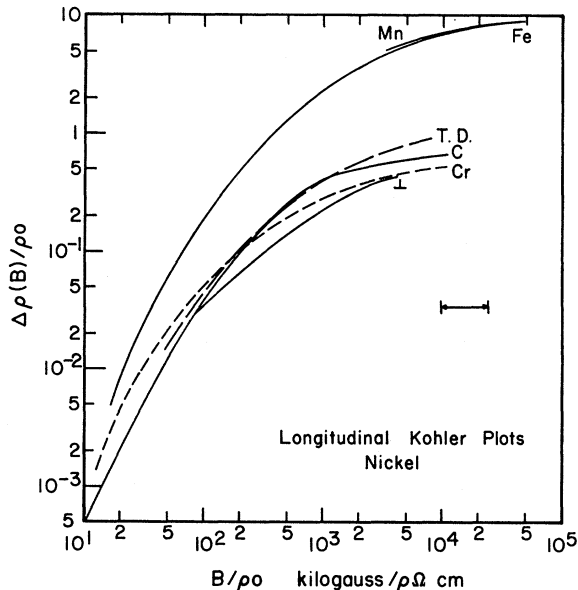


FIG. 7. Longitudinal Kohler plots of nickel with various scattering centers. The bar denotes the range of  $B/\rho_0$  for a single sample.

In contrast, the carbon-doped series showed a definite increase in anisotropy. At high concentrations,  $\Delta(H_a=0)$  is clearly positive (see Fig. 9) implying that  $\Delta_{sd}$  is positive and of sufficient magnitude to outweigh the negative  $\Delta_{MR}$ . From the zero-flux resistivities obtained from the Kohler plots,  $(\Delta_{sd}/\rho_0) \sim 2\%$  with probable errors less than 0.5%. Neither of these two series showed any field dependence in the scattering-process resistivity in the form of deviations from Kohler's rule above  $H_a = 2.5$  kOe.

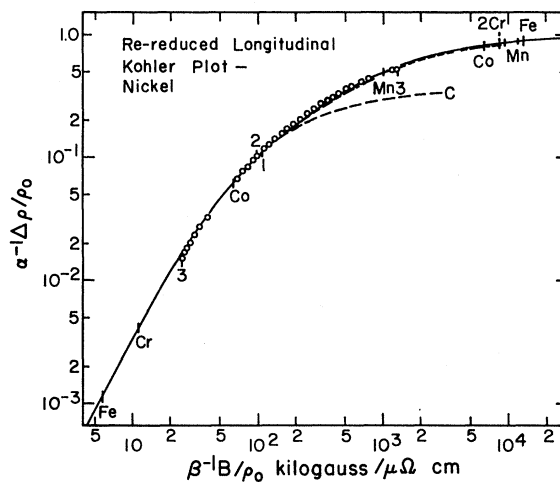


FIG. 8. Longitudinal Kohler plots reduced as  $(\Delta \rho / \rho_0) \rightarrow \alpha^{-1} F(\beta^{-1} B / \rho_0)$

using  $\alpha, \beta$  values given in Table I. Symbols denote the extent of the data for each series. Several plots are taken from Ref. 26, and include (1) reduction series; (2) CN series; (3) recovery (dislocation) series circles.

TABLE I. Magnetic resistivity and magnetoresistivity parameters for scattering resulting from Cr, Mn, Fe, Co, and C impurities, dislocations ( $\perp$ ) and temperature-dependent effects (TD).

	Cr	Mn	Fe	Co	TD	$\perp$	C
Bohr magneton change <sup>a</sup> $d\mu/dc$	-4.5	1.8	2.2	1.2	...	...	...
Moment at impurity site <sup>b</sup>	0.6	2.4	2.8	1.8	0.6	...	...
Resistivity per defect <sup>c</sup> $\frac{d\rho/dc}{\mu\Omega\text{cm}}$	500	50	50	50	...	...	350 <sup>d</sup>
Anisotropy $\Delta/\rho_0\%$ <i>atomic conc</i>	0	...	+14	>0	+3.5	+3.3	+1.8
Magnetoresistivity parameters							
$\alpha$	0.6	10.2	10	10	1.27	0.76	0.68
$\beta$	0.885	0.290	0.272	0.272	0.410	0.310	0.835
Matthiessen's rule parameters							
$\rho_{\uparrow}/\rho_{\downarrow}^e$	0.38	16	...	30	2.3	...	...
$\rho_{\uparrow}/\rho_{\downarrow}^f$	0.21	5.4	7.35	13.2	1	...	...

<sup>a</sup> Reference 1. <sup>e</sup> Reference 28. <sup>g</sup> Reference 9.  
<sup>b</sup> Reference 27. <sup>d</sup> Reference 31. <sup>f</sup> Reference 10.

As mentioned above, the iron-doped series show three distinct regions of behavior depending on solute concentration in terms of deviations from Kohler's rule. As shown in Fig. 10, this division is also reflected, more markedly, in the anisotropy behavior. The zero applied-field anisotropy changed sign on going from light to heavy solute concentrations, confirming the positive increase in the scattering process anisotropy  $\Delta_{sd}$ . While it is difficult to make very definitive statements about this data, it is consistent with the following pattern. The anisotropy per solute as determined from the reducing resistivities  $\rho_0$  was  $(\Delta_{sd}/\rho_{0\pm}) \sim -14\%$  for concentrations  $< 600$ -ppm Fe and  $(\Delta_{sd}/\rho_{0\pm}) \sim +14\%$  for concentrations  $> 2400$  ppm (although for most of the low-concentration samples the anisotropy was as close to zero as the experimental error). Between these limits shown in Fig. 10, the anisotropy demonstrated a smooth change from one value to the other. A preliminary temperature study of this effect was made by cooling the

sample to 1.7°K. This had the result of increasing the anisotropy of a sample (1360 ppm) from  $-2.0$  to  $+0.5$  n $\Omega$  cm and decreasing the deviation from the Kohler plot at  $H_a = 2.4$  kOe by about 10%. From its effect on the anisotropy, this temperature change is roughly equivalent to an increase in concentration of 20 ppm. Over the ranges examined, cobalt- and manganese-doped samples behaved in the same fashion.

## V. DISCUSSION

It is apparent that the work reported here lends further support to the position that Kohler's rule is valid in nickel in the restricted form that the resistivity be varied by altering only one kind of scattering center (see also Schwerer and Silcox<sup>25,26</sup>). Indeed, so valid does the rule appear under this condition that it seems reasonable that the relaxation time approximation may not be necessary. The following argument suggests that this is, indeed, the case.

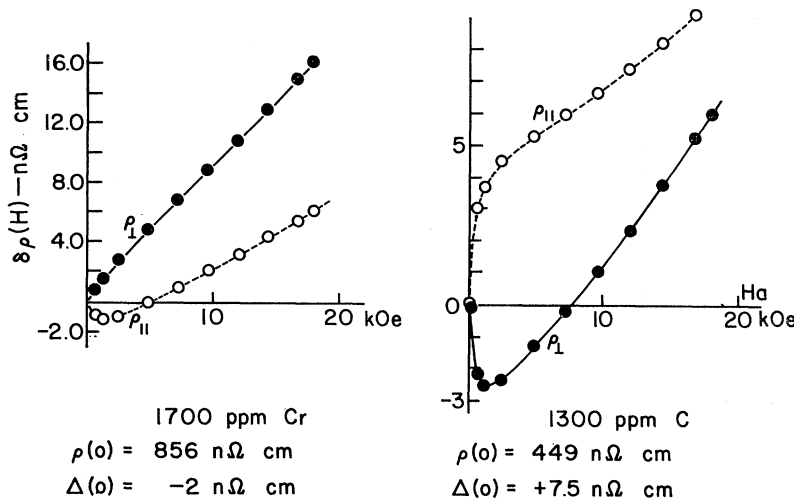
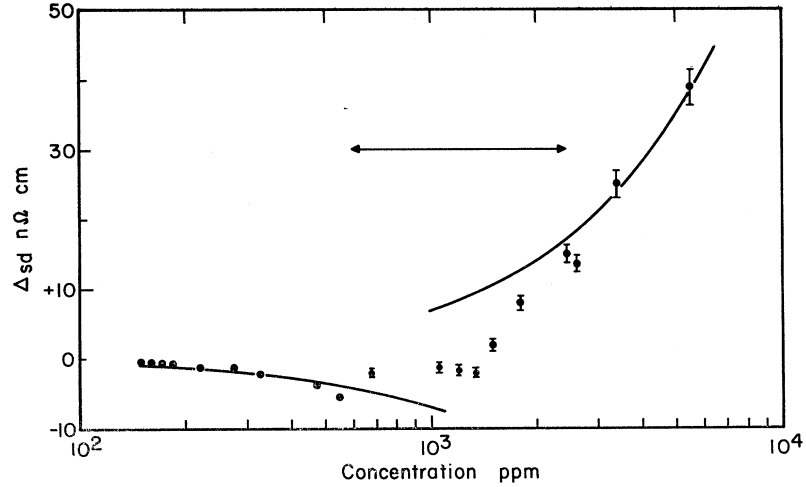


FIG. 9. Typical longitudinal (open circles, dashed lines) and transverse magnetoresistance data illustrating negative zero-field anisotropy for NiCr and positive anisotropy for NiC at high solute concentrations.

FIG. 10. Resistivity anisotropy at zero induction as a function of iron concentration. The bar represents the region of large deviations from the Kohler plot for data at 3.6 kOe. The solid curves are for  $\Delta_{sd}/\rho_0 = \pm 0.14$ .



The normal procedure (see Ziman<sup>36</sup>) is to write down a Boltzmann equation

$$-e\mathbf{E} \cdot \mathbf{v} \frac{\partial f_{\mathbf{k}}^0}{\partial \epsilon_{\mathbf{k}}} = -\dot{f}_{\mathbf{k}}|_{\text{scatt}} + \frac{e}{ch} (\mathbf{v} \times \mathbf{B}) \cdot \frac{\partial g_{\mathbf{k}}}{\partial \mathbf{k}}, \quad (2)$$

where  $f_{\mathbf{k}}^0$  is the Fermi distribution function,  $g_{\mathbf{k}}$  is the out-of-balance function generated by the electric ( $\mathbf{E}$ ) and magnetic ( $\mathbf{B}$ ) fields,  $\dot{f}_{\mathbf{k}}|_{\text{scatt}}$  is the effect of scattering on the distribution function,  $\mathbf{v}$  is the velocity of the electrons at the Fermi surface, and  $e$ ,  $c$ , and  $h$  have the usual significance. We refer the interested reader to Stinchcombe's discussion<sup>37</sup> of the regions of validity of the Boltzmann equation. The equation is solved for  $g_{\mathbf{k}}(E, B)$  which is inserted into the equation

$$\mathbf{J} = e \int \mathbf{v}_{\mathbf{k}} g_{\mathbf{k}} d\mathbf{k} \quad (3)$$

for the current. The term in the solution of  $g_{\mathbf{k}}$  of interest takes the form  $\sigma_{\mathbf{k}}(B) \cdot \mathbf{E}$  to yield Ohm's law, i.e., after inversion of the conductivity tensor,  $E_i = \rho_{ij}(B) J_j$ . Suppose we now consider a specimen  $s$  in which the strength of the scattering has been increased by a scalar  $\lambda_s$  independent of  $k$ , such as one might expect if the number of scattering centers has been changed. The Boltzmann equation for this specimen will be identical to Eq. (2) except for  $\lambda_s$  which can be regarded as multiplying the scattering term  $\dot{f}_{\mathbf{k}}|_{\text{scatt}}$  or, more profitably, as resulting in reduced fields  $E/\lambda_s$ ,  $B/\lambda_s$ . In the latter case, we then know that  $g_{\mathbf{k}} = g_{\mathbf{k}}(E/\lambda_s, B/\lambda_s)$ , where  $g_{\mathbf{k}}$  is the same function as in the previous situation and that, therefore, Ohm's law takes the form  $E_i/\lambda_s = \rho_{ij}(B/\lambda_s) J_j$ . On comparing this with  $E_i = \rho_{ij}^s(B) J_j$ , with  $\rho_{ij}^s(B)$  being the resistance of specimen  $s$  in a

magnetic field, we find  $\rho_{ij}^s(B) = \lambda_s \rho_{ij}(B/\lambda_s)$  and a little algebra produces Kohler's rule.

Thus, the validity of Kohler's rule does not depend on making the relaxation time approximation. Indeed, a derivative of Eq. (2) for collisions restricted to small angles is the diffusion equation formulated by Pippard<sup>13</sup> which, in turn, is an appropriate form of the Fokker-Planck equation.<sup>38</sup> We, therefore, expect Kohler's rule to be valid in the sense used in this paper for the situation discussed by Pippard<sup>13,14</sup> and Klemens and Jackson.<sup>15</sup> The argument given above does depend on being able to write down the Boltzmann equation. We believe that even this may not be necessary, although we have no rigorous proof of this. The argument is essentially a dimensional one and begins by suggesting that any model set up to describe this type of behavior will result in a steady-state equation opposing the effect of the Lorentz force in pulling the Fermi distribution out of shape to the restoring effect produced by the effect of scattering. If we alter the number of scattering centers but keep the type the same, we may expect the strength of the restoring force to be altered in an amount proportionally dependent on the change in concentration of the center. Following the same argument as above, we may then treat this as a reduced field situation where, by virtue of the Lorentz force form, both the electric and magnetic fields are reduced by an amount proportionally dependent on the change in concentration as above. Finally, because we require an Ohm's law form, we find the same expression as before, namely,  $E_i/\lambda_s = \rho_{ij}(B/\lambda_s) J_j$  giving Kohler's rule. Note that the rule is valid in this form for any number of carriers such as a two-band model, which could represent neck and belly electrons<sup>5</sup> or spin-up and spin-down electrons.<sup>9-11</sup>

<sup>36</sup> J. M. Ziman, *Electrons and Phonons* (Oxford University Press, Oxford, 1960), p. 491.

<sup>37</sup> R. B. Stinchcombe, Proc. Phys. Soc. (London) **A78**, 275 (1961).

<sup>38</sup> See, e.g., R. L. Liboff, *Theory of Kinetic Equations* (John Wiley & Sons, Inc. New York, 1969), p. 252.

It is perhaps worthwhile at this point to reiterate in terms of this discussion what are felt to be the essential conditions of validity for Kohler's rule. First, the resistivity must be altered by changing the number but not the type of scattering center and this center must be the dominant scattering mechanism. Also, the density of scattering centers should be sufficiently dilute so that no sensible interference can take place between them and the Fermi surface properties should not be significantly altered. Finally, as discussed in Ref. 39, the effect of possible anomalous terms in the Hall constant should be small. Under these circumstances, studies of breakdowns in Kohler's rule appear to be a promising avenue for the study of scattering center effects in transport properties (cf. the studies of breakdowns in Matthiessen's rule<sup>1</sup>). It appears to be particularly useful in the situation in which it is necessary to know that one particular scattering process is dominant.

We turn now to considering the magnetoresistance itself, where our experiments have demonstrated that the nature of the scattering center clearly has a controlling effect. The possibly fortuitous similarities in shape discussed in Sec. IV B make it possible to make quantitative comparisons between the longitudinal magnetoresistance data. We have not been able to find similar behavior for the transverse magnetoresistance which, because of the existence of open orbits, does not saturate. Nevertheless, the transverse data shows similar differences according to the nature of the scattering center. Table I points out the correlation between the magnetoresistance parameter  $\alpha$  and  $\beta^{-1}$  and other relevant numbers pertaining to the scattering centers, i.e., the resistivity per defect, the total average magnetic moment change for the appropriate alloy and the moment localized at the impurity site as determined by neutron scattering.

The most striking characteristic is the difference in the saturation value of the longitudinal magnetoresistance  $\alpha$  for the local moment group (Fe, Co, Mn) and the remainder. Thus, there is a factor of roughly 20 between  $\alpha$  for the local-moment group and the nickel-chromium alloy. Clearly, characteristics of the Fermi surface have to enter into any quantitative discussion of these data, but a relevant quick comparison might be made with copper alloys. The Fermi-surface scheme for nickel proposed by Hodges, Ehrenreich, and Lang<sup>40</sup> involves two spin bands for the *s* electrons with the

electrons split approximately equally between the bands. For the majority spins, the Fermi surface is similar to that of the noble metals, i.e., it has necks across the (111) Brillouin-zone boundaries, whereas the minority spin surface is like a distorted free-electron sphere. Since a free-electron sphere introduces little magnetoresistance, it appears reasonable to make the comparison with copper because of the similarity with the majority spin surface. The published data for copper<sup>41,42</sup> suggest values of  $\alpha \approx 0.5-1.5$  which are comparable to the values of  $\alpha$  found in nickel for chromium and carbon impurities and the dislocation and thermal scattering. These saturation values correspond to variations in mean free path of approximately 1.5-5 and are, therefore, consistent with the direct measurements of Deaton and Gavenda<sup>8</sup> and the inferred values of Dugdale and Bazinski<sup>1</sup> in copper. The very strong magnetoresistance exhibited by the local moment group of impurities in nickel appears most unusual. If interpreted as simple relaxation time anisotropies, these values correspond to variations in mean free path of order 30. We will explore the ramification of this a little further.

The discussions of longitudinal magnetoresistance by Pippard<sup>13,14</sup> and Klemens and Jackson<sup>15</sup> point out the possibility of extreme magnetoresistance variations for the combination of a multiply connected Fermi surface and a scattering process dominated by small-angle scattering. An attempt<sup>42</sup> to observe such small-angle scattering effects by a study of phonon scattering in copper was apparently frustrated by impurity effects. Nevertheless, this mechanism is apparently the only one predicting increases in the magnetoresistance of the size observed (i.e.,  $\alpha \sim 10$ ). With this basis, consideration of the scattering centers studied in the present paper leads to the following contradiction. Small-angle scattering is to be expected from long-range potentials, e.g., in the scattering from dislocations and according to the Friedel<sup>29,30</sup> model (see also Hayakawa<sup>43</sup>), chromium, and other impurities with nonlocalized moments. We may also expect small-angle scattering from phonon scattering at low temperatures. On the other hand, isotropic scattering is anticipated from those impurities where the potential is very strongly localized, such as the local moment group. Similarly, any spin-dependent scattering is usually considered to be associated with very localized potential. Hence, we would anticipate weak magnetoresistance from the local moment group and striking magnetoresistance from the remainder in contradiction to observation. Our observations, therefore, suggest that if the scattering potential is, indeed, the source of these phenomena, then the potential associated with the local moment group is likely to have some unusual features.

<sup>39</sup> J. M. Luttinger [Phys. Rev. **112**, 789 (1958)] predicts two terms in the anomalous Hall coefficient for a ferromagnetic material which are linearly proportional to, and independent of  $\sigma_0$ , respectively. The linear term will not give rise to deviations from Kohler's rule but a contribution will come from the term independent of  $\sigma_0$ . These deviations become increasingly important at large values of  $B/\rho_0$ . Using experimental values for the anomalous Hall coefficients reported by Van Elst (Ref. 35) the deviation at our high-field values of  $B/\rho_0 = 10^4$  kG/ $\mu\Omega$  cm would be of order 0.5% and undetectable.

<sup>40</sup> L. Hodges, H. Ehrenreich, and N. D. Lang, Phys. Rev. **152**, 505 (1966).

<sup>41</sup> J. O. Strom-Olsen, Proc. Roy. Soc. (London) **A302**, 83 (1957).

<sup>42</sup> A. L. Clark and R. L. Powell, Phys. Rev. Letters **21**, 802 (1968).

<sup>43</sup> H. Hayakawa, Progr. Theoret. Phys. (Kyoto) **37**, 213 (1967).

We note next that these observations parallel the unusually large deviations from Matthiessen's rule discovered recently in iron<sup>11</sup> and nickel alloys.<sup>9,10</sup> Since in both the present paper and in work on Matthiessen's rule a prime consideration is the anisotropy of the scattering process, it will be appropriate to discuss the spin-mixing model<sup>10,11</sup> proposed to account for the Matthiessen's rule breakdown in nickel. Briefly, each spin band is considered to conduct independently at low temperatures and two relaxation times, one for each band, are postulated. As the temperature is raised, a nonresistive spin-flip mechanism is postulated as becoming important and eventually dominating the scattering process at high temperatures, so that between collisions contributing to resistance, electrons scatter from one spin band to the other many times. An essential feature of the model is that, at low temperatures, the bands conduct in parallel so that that resistivity is dominated by the longest relaxation time. At high temperatures, the bands are mixed and the relaxation times are averaged, resulting naturally in a resistivity increase. The large values of the deviations are accounted for by considerable differences between the resistivities due to each band which are, in turn, related to the Friedel<sup>29</sup> models for the impurity electronic structure. Finally, since the nonresistive spin-flip scattering is proportional to the square of the temperature, the process is tentatively identified as that of electron-electron scattering which results in a spin flip but conserves the total electron momentum.

We note a rough similarity between this type of model and the model of magnetoresistance in the relaxation time approximation. Thus, in zero-magnetic field, electrons at each point on the Fermi surface contribute independently and in parallel to the current. Correspondingly, the resistivity is dominated by those current carriers with the lowest resistance, i.e., the longest relaxation time (taking into account projection factors, etc.). As the field is increased, the current carriers sample larger and larger sections of orbits on the Fermi surface averaging out the relaxation times and, thus, giving more weight than previously to the shorter relaxation times. Naturally, the resistance increases. In this case, the role of the magnetic field is to average the relaxation time and, therefore, the resistance over sections of the orbit, while in the spin-mixing case, the averaging is over bands and is carried out by thermal effects.

Bearing in mind the possibility that spin-mixing effects may enter into the magnetoresistance, then, in general, we are concerned with both spin and momentum characteristics and we need to discuss both. In the present case, the experiments suggest that spin mixing is not the relevant process according to the following argument. Successful Kohler plots have been obtained for these alloys, implying that the scattering is independent of  $B$  and that the primary role of the magnetic field is to make the carriers describe appropri-

ate orbits on the Fermi surface. Thus, either each band has the same magnetoresistance—in which case the spin mixing, however strong, is irrelevant—or alternatively, spin mixing is introduced by the magnetic field in a rather special way to preserve the form of Kohler's rule. The latter appears unlikely. Hence, if spin mixing is the relevant process for the breakdowns in the Matthiessen's rule, our preference is to seek the origin of the magnetoresistance in other processes. Note, however that a two-band model is not inconsistent with Kohler's rule, provided each band separately obeys Kohler's rule and provided the resistivity in each band arises from the same scattering center as in our earlier discussion. Thus, in the range of temperatures with which we are concerned, each spin band can conduct separately in accord with the spin-mixing model.

It may indeed be possible to deduce parameters associated with the spin-mixing model from the magnetoresistance. A tempting first approximation is to assume that all the magnetoresistance is associated with the copperlike majority spin surface and that the nearly spherical minority spin surface contributes no magnetoresistance at all. In these circumstances it is possible that in zero field almost all the current is carried by one band and at the highest fields the current is carried by the other band. The saturation longitudinal magnetoresistance ratio  $\alpha$  is then a direct measure of the ratio of the resistivities associated with each band. The conditions under which this will obtain include very strong magnetoresistance for the one band and a marked difference between the resistivities of the two bands (as obtained in the spin-mixing model). The values obtained in this work and those obtained by Fert and Campbell<sup>9</sup> and Farrell and Greig<sup>10</sup> lead to qualitative agreement but with considerable scatter (see Table I). More detailed discussion of these numbers and the appropriate method of analyzing both types of data is left for future consideration.

We remark now that the other magnetoresistance parameters, i.e.,  $\beta$  and  $\Delta_{sd}$ , also show systematic variations with scattering center. As noted above, a naive connection between  $\beta$  and the reciprocal of the Hall constant can be made through the tube-integral formula. This point needs to be explored further both experimentally and theoretically. Two remarks can be made concerning the zero-field anisotropy  $\Delta_{sd}$ . First,  $\Delta_{sd}$  is proportional to  $\rho_0$ , i.e., the anisotropy depends on the number of scattering centers present. Second,  $\Delta_{sd}$  seems to be associated with the local moment characteristic of the scattering center. Thus, the only center for which  $\Delta_{sd}$  is zero, i.e., the chromium impurity, is also the only center for which the local moment is known<sup>27</sup> to be zero. Dislocations have an associated local moment distributed over a relatively large volume as a result of the magnetoelastic coupling between the strain field and the spin distribution.<sup>44</sup> We almost certainly should

<sup>44</sup> F. Vicena, Czech. J. Phys. 5, 480 (1954).

expect a considerable contribution from the dislocation core. It seems reasonable to expect a similar coupling with the weaker strain field associated with an interstitial impurity such as carbon. The observations are in line with these speculations. Similarly, the largest zero-field anisotropy (with due reservations concerning the concentration dependence) is observed in those samples with the iron group of impurities, i.e., those scattering centers which undoubtedly have the largest local moment. This discussion would suggest that the observations on the temperature-dependent scattering indicate that a magnetic disturbance of this nature is associated with the thermal excitations responsible for scattering in that case. Indeed, the parallel between scattering by a static strain field (i.e., dislocations) and by a dynamic strain field (i.e., phonons) has been drawn elsewhere<sup>45</sup> and we comment that our observations are consistent with a parallel of this nature.

The concentration dependence of  $\Delta_{sd}$  for iron-group solutes is an oddity with the following characteristic features. At the highest and lowest concentrations, the data for fields  $H > 2.5$  kOe follow the same Kohler plot but the value of  $\Delta_{sd}/\rho_0$  differs in sign in the two regions. For intermediate concentrations the data did fit the Kohler plot at high fields ( $H > 10$  kOe) but did not do so in the intermediate field region ( $2.5 \text{ kOe} < H < 10 \text{ kOe}$ ). These observations suggest a two-state ordering process in which one state of order exists at the lowest concentration and the second state exists at the high-concentration end. Since the same Kohler plot applies in each case (although  $\Delta_{sd}$  changes sign), a plausible possibility is that the impurities may be antiferromagnetically aligned with the matrix magnetization at low concentrations and become ferromagnetically aligned at high concentrations. At the intermediate

concentration, the coupling is sufficiently weak that the magnetic field can align the initially random local moments (thus, explaining the breakdown of Kohler's rule in intermediate concentration samples in intermediate fields). Obviously, such an occurrence implies a relatively long-range impurity-impurity interaction, since this occurs at somewhat low concentrations, i.e., at an impurity-impurity spacing of about 10 atomic spacings. Nevertheless, it is difficult to conceive of a preferable explanation of the experimental results.

In conclusion, these experiments have underlined the role played by the nature of the scattering center in this transport process. In particular, the extremely high magnetoresistance shown by impurities of the iron group in nickel has been demonstrated. Finally, breakdowns in Kohler's rule appear to be useful in studies of scattering center effects, just as are breakdowns in Matthiessen's rule.

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<sup>45</sup> Z. S. Basinski, J. S. Dugdale, and A. Howie, *Phil. Mag.* **8**, 199 (1963).