

## Low-Temperature Resistivity of Dilute Magnetic Impurities in the Presence of Internal Fields

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The variation of the low-temperature resistivity in the presence of internal fields is examined for dilute concentrations of magnetic impurities in a nonmagnetic metal host. The relaxation times are calculated in the second Born approximation for two different internal fields: one arising in a system in which long-range order exists, and another in which the magnetic impurities interact via a Ruderman-Kittel-Kasuya-Yosida interaction. In the latter case the internal field  $H$  is a random variable whose probability distribution  $P(H)$  can, in principle, be obtained. Using an Ising-like probability distribution, it is predicted that the change in the very low-temperature resistivity  $\Delta\rho(T)$  is, except for a small  $\ln T$  term, linear in  $T$ . This is in agreement with experiment for Au-0.1% Fe, where the experiment was performed at sufficiently low temperatures. More generally we find that  $\Delta\rho(T)$  is approximately proportional to that part of the low-temperature specific heat which arises from the magnetic disordering of the impurities in their internal fields. The "width" of the probability distribution function obtained from the low-temperature specific-heat measurements gives the slope  $m$  of  $\Delta\rho(T)$  in rather good agreement with experiment. This is additional evidence that the excess specific heat in these alloys arises from a magnetic disordering of the impurities. The slope of the resistivity is, from our theory, approximately independent of the impurity concentration and the exchange interaction  $J$  at sufficiently low temperatures. For higher temperatures we obtain a resistivity maximum at a temperature proportional to the impurity concentration. This maximum arises from the suppression of the Kondo  $\ln T$  term by the presence of internal fields. For concentrations of the order of 1%, the maximum as well as the minimum disappears, and the resistivity decreases monotonically as the temperature is lowered. The behavior of the resistivity as a function of the impurity concentration, the strength and the sign of the  $s$ - $d$  interaction, the impurity spin, and the temperature is discussed. It is proposed that low-temperature resistivity measurements be used to probe the behavior of the probability distribution  $P(H)$  of the fields near  $H=0$ . The present results apply only to temperatures much greater than the Suhl-Abrikosov resonance temperature.

### I. INTRODUCTION

VERY dilute concentrations of magnetic impurities distributed in a nonmagnetic host metal exhibit remarkable low-temperature resistive anomalies.<sup>1</sup> As the temperature  $T$  is decreased from very high  $T$ , these systems exhibit a minimum in the resistivity at  $T=T_{\min}$ , where  $T_{\min}$  is approximately proportional to  $c^{1/5}$  and  $c$  is the fractional impurity concentration. As the temperature is further lowered, at least some of the alloys show a maximum in the resistivity. The temperature  $T_{\max}$  at which the maximum occurs decreases with the impurity concentration. For relatively high concentrations (for Cu-Mn, for example, of the order of 1% or higher), the resistivity does not show a minimum but a continuous decrease with decreasing temperature.

Recently, Kondo<sup>2</sup> has explained the temperature and concentration dependence of the low-temperature resistivity minimum. He found that if one calculates the scattering cross section for the electrons scattered by the isolated impurities in the second Born approxima-

tion, using a phenomenological  $s$ - $d$  interaction with negative exchange constant  $J$ , a negative  $\ln T$  term enters into the expression for the resistivity. This  $\ln T$  term, combined with the  $T^5$  term arising from the phonon contribution to the resistivity, gives the minimum in good agreement with experiment. As the temperature is further lowered, the Kondo  $\ln T$  term increases without limit, showing a divergence in the resistivity and hence indicating that the perturbation expansion used must fail at sufficiently low temperatures. Methods of avoiding the difficulties of perturbation theory at the very low temperatures have been the subject of much recent work.<sup>3-6</sup> For example it has been shown by Abrikosov<sup>4</sup> that as the temperature is sufficiently lowered the Kondo logarithmic divergence is replaced

<sup>3</sup> H. Suhl, *Physics* **2**, 39 (1965); *Phys. Rev.* **138**, A515 (1965); Y. Nagaoka, *ibid.* **138**, A1112 (1965).

<sup>4</sup> A. A. Abrikosov, *Physics* **2**, 5 (1965).

<sup>5</sup> K. Yosida and A. Okiji, *Progr. Theoret. Phys. (Kyoto)* **34**, 505 (1965); S. Doniach, *Phys. Rev.* **144**, 382 (1966).

<sup>6</sup> Apart from obtaining a nonperturbative solution to the model Hamiltonian with the assumed form of exchange interaction, the more recent work is also strongly concerned with the more basic problem of explaining the impurity state itself, i.e., the local moment problem. Both these problems are beyond the scope of our present work: With regard to the former we restrict our attention to the region where we expect perturbation results to be valid; with regard to the latter it is acknowledged that our conclusions depend on the validity of the model Hamiltonian and any modification of this must affect our results accordingly.

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<sup>1</sup> For a review of the experimental and some of the theoretical aspects of these resistive anomalies, see G. J. van den Berg, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Company, Amsterdam, 1964), Vol. 4, p. 104.

<sup>2</sup> J. Kondo, *Progr. Theoret. Phys. (Kyoto)* **32**, 37 (1964).

by a resonance scattering of the conduction electrons at the impurity site. The resonance behavior becomes important at or below the resonance temperature<sup>4</sup>  $T_r$ , where

$$T_r \approx (\epsilon_F/k_B) \exp(2\epsilon_F)/(3Jz), \quad (1.1)$$

where  $\epsilon_F$  is the Fermi energy,  $k_B$  the Boltzmann constant,  $J$  the  $s$ - $d$  exchange interaction, and  $z$  the number of conduction electrons per atom. For alloys exhibiting a resistivity minimum  $J$  is negative<sup>2</sup> and is of the order of 0.2 eV,  $z \approx 1$ . For copper alloys,  $\epsilon_F \approx 7$  eV, thus  $T_r$  is of the order of  $10^{-5}$  °K.

So far, the discussion is restricted to the case when the magnetic impurities experience no internal fields. The variation of the resistivity in the presence of internal fields has been considered by Yosida,<sup>7</sup> Mikoshiba and Yoshihiro,<sup>8</sup> Van Peski-Tinbergen and Dekker,<sup>9</sup> Liu,<sup>10</sup> Silverstein,<sup>11</sup> Harrison and Klein,<sup>12</sup> and Abrikosov.<sup>13</sup> Abrikosov<sup>13</sup> finds that the presence of internal fields suppresses the increase of the resistivity at low temperatures for the case when long-range magnetic order is present in the solid. Silverstein<sup>11</sup> and Harrison and Klein<sup>12</sup> discussed recently the variation of the resistivity maximum with impurity concentration and temperature.

The purpose of this paper is to examine the variation of the low-temperature resistivity of dilute concentrations of magnetic impurities dissolved in nonmagnetic metals in the presence of internal (or external) fields. This internal field is assumed to arise from the indirect exchange interaction via the conduction electrons between the magnetic impurities and has been derived by Ruderman and Kittel,<sup>14</sup> Kasuya,<sup>15</sup> and Yosida<sup>16</sup> and will be denoted as the RKKY interaction. The presence of internal fields will quench the  $\ln T$  dependence of the resistivity at very low temperatures as was recently discussed by Abrikosov.<sup>13</sup> Our treatment differs from that of Abrikosov in two important ways: (1) Since we derive the relaxation time in the second Born approximation only, our treatment is more straightforward and transparent and thus serves as a useful comparison of the results obtained by more complicated methods. (The two should give the same result for terms up to the third power of the  $s$ - $d$  exchange interaction  $J$ .) (2) Abrikosov<sup>13</sup> considers the case when there is long-

range magnetic order in the solid which results in a fixed internal field, whereas we believe that only short-range magnetic order exists in these systems. Because of the random distribution of the magnetic impurities in the nonmagnetic host, the internal field, we believe, is a random variable with a well-defined probability distribution. The concentration dependence of the probability distribution of the internal field in an Ising model was previously discussed by Marshall<sup>17</sup> and Klein and Brout.<sup>18,19</sup> Our treatment and that of Abrikosov's<sup>13</sup> predict markedly different behavior of the resistivity as the temperature departs from  $T=0$ . Abrikosov's model yields an increase in the resistivity  $\rho(T)$  which has zero slope as we depart from  $T=0$ ,<sup>20</sup> whereas our model, using an Ising distribution of internal fields, gives a linear increase of the resistivity with  $T$ . Experimental evidence exists which confirms this linear dependence for 0.1% Au-Fe. An Ising distribution also gives a slope of  $\rho(T)$  versus  $T$  near  $T=0$ <sup>20</sup> which is only weakly dependent upon the impurity concentration and the  $s$ - $d$  exchange interaction and depends upon the Fermi energy of the host and a geometrical factor. Thus a precise measurement of the low-temperature resistivity may be useful in obtaining information on the probability distribution of internal fields near zero fields. This information relates in an important way to the question of what causes the large excess low-temperature specific heat in dilute alloys and whether long-range order exists in these materials. There are so far at least three models presented to explain the low-temperature specific heat.<sup>17-19,21-23</sup> Should the experimentally measured low-temperature resistivity agree with that obtained from the probability distribution of internal fields presented in this paper, this would be additional evidence for the validity of the specific-heat calculation presented in Refs. 17, 18, 19, and 23. In the analysis of our calculations, we address ourselves to the following:

(a) Does the existence of the internal field modify the low-temperature resistance minimum? If so, what is the effect of the concentration on this?

(b) Does the assumption of the internal-field distribution obtained from<sup>19</sup> an RKKY interaction give the experimentally observed concentration and temperature dependence of the resistance maximum and the disappearance of the minimum for more highly concentrated alloys?

<sup>7</sup> K. Yosida, Phys. Rev. **107**, 396 (1957).

<sup>8</sup> N. Mikoshiba and K. Yoshihiro, J. Phys. Soc. Japan **19**, 2346 (1964).

<sup>9</sup> Tineke van Peski-Tinbergen and A. J. Dekker, Physica **29**, 917 (1963).

<sup>10</sup> S. H. Liu, Phys. Rev. **137**, 1209 (1965).

<sup>11</sup> S. D. Silverstein, Phys. Rev. Letters **16**, 466 (1966); Bull. Am. Phys. Soc. **11**, 237 (1966).

<sup>12</sup> R. J. Harrison and M. W. Klein, Bull. Am. Phys. Soc. **11**, 237 (1966). This represents a preliminary report of the present paper.

<sup>13</sup> A. A. Abrikosov, Physics **2**, 61 (1965).

<sup>14</sup> M. A. Ruderman and C. Kittel, Phys. Rev. **96**, 99 (1954).

<sup>15</sup> T. Kasuya, Progr. Theoret. Phys. (Kyoto) **16**, 45 (1956).

<sup>16</sup> K. Yosida, Phys. Rev. **106**, 893 (1957).

<sup>17</sup> W. Marshall, Phys. Rev. **118**, 1520 (1960).

<sup>18</sup> M. W. Klein and R. Brout, Phys. Rev. **132**, 2412 (1963).

<sup>19</sup> M. W. Klein, Phys. Rev. **136**, 1156 (1964).

<sup>20</sup> Throughout this paper whenever we refer to  $T$  near zero, we have in mind temperatures which are quite low compared to that of the resistivity maximum but still much greater than the temperature  $T_r$  given in Eq. (1.1). That is, we assume the temperature remains sufficiently high that perturbation theory is valid.

<sup>21</sup> J. Kondo, Progr. Theoret. Phys. (Kyoto) **33**, 575 (1965).

<sup>22</sup> A. W. Overhauser, Phys. Rev. Letters **3**, 414 (1959).

<sup>23</sup> M. W. Klein, Phys. Rev. Letters **16**, 127 (1966).

(c) Is the Kondo  $\ln T$  term still observable in principle below the temperature of the resistivity maximum?

(d) What information can one obtain about the probability distribution of the internal field from the measurement of the resistivity as a function of concentration and temperature?

## II. MATHEMATICAL DEVELOPMENTS

Let the phenomenological Hamiltonian  $\mathcal{H}$  for a system of  $N$  magnetic impurities randomly distributed in the solid be<sup>24</sup>

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}', \quad (2.1)$$

where

$$\mathcal{H}_0 = \sum_{\mathbf{k}, s} \epsilon_{\mathbf{k}s} a_{\mathbf{k}s}^\dagger a_{\mathbf{k}s} - \sum_n \mu_B \gamma_n \mathbf{H}_n \cdot \mathbf{S}_n. \quad (2.2)$$

Thus, we assume an ensemble of noninteracting electrons in the conduction band, characterized by eigenvectors  $\mathbf{k}$  and spin quantum numbers  $s$ , together with a group of independent localized spins interacting with a set of effective fields  $H_n$  in a purely Zeeman-like classical fashion. Thus,  $S_n$  is the spins operator at site  $n$  whose expectation value in the direction of the field  $H_n$  is  $M_n$ ,  $\mu_B$  and  $\gamma_n$  are the Bohr magneton and the Lande  $g$  factor, respectively, and serve to give  $H_n$  the proper dimensions of magnetic field. Although occasionally one may want to include an external magnetic field as part of  $H_n$ , we shall consider the values of  $H_n$  to correspond to a statistically derived random variable resulting from the random distribution of the magnetic impurities. The distribution function we shall use was originally derived<sup>17,18</sup> by a semiclassical argument, using an Ising model, and involves the assumption of an RKKY interaction.<sup>24</sup> The Zeeman-like interaction with the conduction electron spins is implicitly included in Eq. (2.2) by means of the spin dependence of the one-electron energy terms  $\epsilon_{\mathbf{k}s}$ . The quantities  $a_{\mathbf{k}s}^\dagger$  and  $a_{\mathbf{k}s}$  are, respectively, the creation and annihilation operators for the state  $\mathbf{k}, s$ .

The perturbation  $\mathcal{H}'$  is considered to arise from two parts; one a perturbing potential  $V_n$  at site  $n$ , and the other a spin-spin interaction involving both the electron and localized impurity spin operators. Both of these interactions are assumed to be strictly localized in the sense of involving a single Wannier function localized

at site  $n$ . Thus, we write

$$\begin{aligned} \mathcal{H}' = & N^{-1} \sum_{n, \mathbf{k}, \mathbf{k}'} \exp\{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}_n\} \\ & \times [V(a_{\mathbf{k}+\mathbf{i}}^\dagger a_{\mathbf{k}+\mathbf{i}'} + a_{\mathbf{k}-\mathbf{i}}^\dagger a_{\mathbf{k}-\mathbf{i}'}) \\ & - J\{(a_{\mathbf{k}+\mathbf{i}}^\dagger a_{\mathbf{k}+\mathbf{i}'} - a_{\mathbf{k}-\mathbf{i}}^\dagger a_{\mathbf{k}-\mathbf{i}'}) S_{nz} \\ & + a_{\mathbf{k}+\mathbf{i}}^\dagger a_{\mathbf{k}-\mathbf{i}'} S_n^- + a_{\mathbf{k}-\mathbf{i}}^\dagger a_{\mathbf{k}+\mathbf{i}'} S_n^+\}]. \quad (2.3) \end{aligned}$$

The constants  $V$  and  $J$  represent the localized potential and exchange constant assumed to be the same at all sites, thus dropping the subscript  $n$  on these constants. The spin operator for the conduction electrons has been replaced by its second quantized form.  $S_z, S^\pm$  refer to the localized impurity spins.

The transition probability per unit time,  $W(a \rightarrow b)$ , from an initial state  $a$  to a final state  $b$  in the second Born approximation is given by<sup>2</sup>

$$W(a \rightarrow b) = \frac{2\pi}{\hbar} \sum_b \left\{ [\mathcal{H}'_{ab} \mathcal{H}'_{ba} + \sum_{c \neq a} \{\mathcal{H}'_{ac} \mathcal{H}'_{cb} \mathcal{H}'_{ca} + \text{c.c.}\}] / (\epsilon_a - \epsilon_c) \right\} \delta(\epsilon_a - \epsilon_b), \quad (2.4)$$

where  $\mathcal{H}'$  is given in Eq. (2.3) and  $\epsilon_a, \epsilon_b$ , and  $\epsilon_c$  are the initial, final, and intermediate energies, respectively, and c.c. designates the complex conjugate.

The transition probabilities per unit time for  $N_i$  localized and independent spins from states  $\mathbf{k}$  to  $\mathbf{k}'$  without change of spin state (nonflip scattering) are given by

$$\begin{aligned} W(\mathbf{k} \pm \rightarrow \mathbf{k}' \pm) &= (2\pi N_i / \hbar N^2) \{ (V \mp JM)^2 - 2(V \mp JM)^3 h^\pm(\epsilon_{\mathbf{k} \pm}) \\ &\mp 4MJ^2 (V \mp JM) g^\mp(\epsilon_{\mathbf{k} \pm} \mp \gamma \mu_B H) \\ &- 2(V \mp JM) J^2 (S \pm M + 1) (S \mp M) \\ &\times h^\pm(\epsilon_{\mathbf{k} \pm} \mp (\gamma \mu_B H)) \} \delta(\epsilon_{\mathbf{k} \pm} - \epsilon_{\mathbf{k}' \pm}). \quad (2.5) \end{aligned}$$

The term  $(V \mp JM)^2$  represents the contribution to scattering arising in the first Born approximation.  $M$  is the quantum-mechanical expectation value of the operator  $S_z$ . The functions  $g^\pm(y)$  and  $h^\pm(y)$  are given by

$$g^\pm(y) = N^{-1} \sum_{\mathbf{q}} f_{\mathbf{q} \pm}^0 / (\epsilon_{\mathbf{q} \pm} - y), \quad (2.6a)$$

$$h^\pm(y) = N^{-1} \sum_{\mathbf{q}} (\epsilon_{\mathbf{q} \pm} - y)^{-1}. \quad (2.6b)$$

In the evaluation of  $g^\pm(y)$  in Eq. (2.6) we have replaced the Fermi function of the intermediate state by its thermal equilibrium value  $f_{\mathbf{q} \pm}^0$  of the argument  $(\epsilon_{\mathbf{q} \pm} - \epsilon_F)$ , i.e.,

$$f_{\mathbf{q} \pm}^0 = 1 / \{1 + \exp[(\epsilon_{\mathbf{q} \pm} - \epsilon_F) / k_B T]\}, \quad (2.7)$$

where

$$\epsilon_{\mathbf{q} \pm} = \epsilon_{\mathbf{q}} \pm \zeta, \quad (2.8)$$

where  $\zeta$  is the Zeeman displacement of the conduction-electron spins in the local field. It is to be noted that  $\zeta$

<sup>24</sup> Here we introduce the RKKY interaction phenomenologically. The Hamiltonian, Eq. (2.2), may be obtained in a more rigorous fashion by performing a unitary transformation on the perturbed Hamiltonian as was done by Kondo (Ref. 21). This transformation is more clearly exhibited by J. D. Kim, Phys. Rev. **149**, 434 (1966). Kim has shown how one can separate the Hamiltonian into two parts, one describing the motion of the conduction electrons and another that of the localized spins. The part that describes the localized spins has the form  $-\gamma \mu_B \mathbf{H}_n \cdot \mathbf{S}_n$ . An alternative point of view on justifying Eq. (2.2) is in a perturbation-theory approach with expansion parameter  $J$ . The term in  $J^2$  gives the RKKY potential and the first Born approximation. The term in  $J^3$  gives the Kondo effect and a modification of the RKKY potential. The latter we have neglected. However, one should realize that Eq. (2.2) has not been rigorously justified.

does not enter into the final result, provided that  $\zeta/\epsilon_F \ll 1$ . The function  $h^\pm(y)$  contributes only to potential scattering, and in this paper we neglect all potential scattering terms arising from higher than the first Born approximation. These would not be expected to contribute appreciably to the very low-temperature resistivity. Rewriting Eq. (2.6a) gives

$$\begin{aligned} g^\pm(y) &= N^{-1} \sum_{\mathbf{q}} f_{\mathbf{q}\pm}^0 / (\epsilon_{\mathbf{q}\pm} - y) \\ &= (\Omega/8N\pi^3) \int d\mathbf{q}_{\pm} f_{\mathbf{q}\pm}^0 / (\epsilon_{\mathbf{q}\pm} - y) \\ &\approx (3z)/(2\epsilon_F) \left[ 2 + \int_{-\infty}^{\infty} d\epsilon_{\mathbf{q}\pm} (-\partial f_{\mathbf{q}\pm}^0 / \partial \epsilon_{\mathbf{q}\pm}) \right. \\ &\quad \left. \times \ln |\epsilon_{\mathbf{q}\pm} - y| / 4\epsilon_F \right], \end{aligned} \quad (2.9)$$

where  $\Omega$  is the volume of the solid and  $z$  is the number of conduction electrons per atom.

It follows from the limits on the integral in Eq. (2.9) that  $g^+ = g^-$ ; we henceforth drop the  $\pm$  signs from the  $g$ 's. With this notation, and neglecting the function  $h(y)$  for reasons described above, the transition probabilities  $W(\mathbf{k}\pm \rightarrow \mathbf{k}'\pm)$  become

$$\begin{aligned} W(\mathbf{k}\pm \rightarrow \mathbf{k}'\pm) &= (2\pi N_i / \hbar N^2) \{ (V \mp JM)^2 \mp 4MJ^2 (V \mp JM) \\ &\quad \times g(\epsilon_{\mathbf{k}\pm} \mp \gamma\mu_B H) \} \delta(\epsilon_{\mathbf{k}\pm} - \epsilon_{\mathbf{k}'\pm}). \end{aligned} \quad (2.10)$$

$$\begin{aligned} \frac{1}{\tau(\mathbf{k}\pm)} &= km\Omega N_i / (\pi \hbar^3 N^2) \left[ \sum_{M=-S}^S P_M \{ (V \mp JM)^2 \mp 4MJ^2 (V \mp JM) g(\epsilon_{\mathbf{k}\pm} \mp \gamma\mu_B H) \} \right. \\ &\quad \left. + \sum_{M=-S}^S P_M \left\{ \frac{J^2 (S \mp M) (S \pm M + 1) \{ 1 + 2Jg(\epsilon_{\mathbf{k}\pm}) + 2Jg(\epsilon_{\mathbf{k}\pm} \mp \gamma\mu_B H) \} e^{\pm x/2}}{(1 - f_{\mathbf{k}\pm}) \exp \pm x/2 + f_{\mathbf{k}\pm} \exp \mp x/2} \right\} \right]. \end{aligned} \quad (2.13)$$

In Eq. (2.13) we replaced  $[2m(\epsilon_{\mathbf{k}\pm} \pm \gamma\mu_B H)]^{1/2} / \hbar$  by  $k$ , except in the expression for  $g$ , since in our final result the error due to this will be of the order of  $\gamma\mu_B H / \epsilon_F$ , which we neglect. The first curly bracket in Eq. (2.13) arises from the elastic scattering, whereas the second from the inelastic scattering between initial and final states.  $P_M$  is the probability of finding the ion in state  $M$  at temperature  $T$ , i.e.,

$$P_M = \exp Mx / \sum_{M'=-S}^S \exp M'x,$$

where  $x = \gamma\mu_B H / k_B T$ .

The following identity will be useful in evaluating Eq. (2.13):

$$\begin{aligned} \langle (S-M)(S+M+1) \rangle_{av} e^{x/2} &= \langle (S+M)(S-M+1) \rangle_{av} e^{-x/2} \\ &= (\sinh \frac{1}{2}x) [ (S^2 - \langle M^2 \rangle_{av}) \coth \frac{1}{2}x \\ &\quad + (S \coth \frac{1}{2}x - \bar{M}) ] \equiv \Lambda_S(x) \\ &= \bar{M} \operatorname{csch} \frac{1}{2}x, \end{aligned} \quad (2.14)$$

With similar approximations the spin-flip transition probabilities  $W(\mathbf{k}\pm \rightarrow \mathbf{k}'\mp)$  with the  $z$  component of the ion spin changing from  $M$  to  $M \pm 1$  are

$$\begin{aligned} W(\mathbf{k}\pm \rightarrow \mathbf{k}'\mp) &= (2\pi N_i / \hbar N^2) J^2 (S \mp 1) (S \pm M + 1) \{ 1 + 2Jg(\epsilon_{\mathbf{k}\pm}) \\ &\quad + 2Jg(\epsilon_{\mathbf{k}\pm} \mp \gamma\mu_B H) \} \delta(\epsilon_{\mathbf{k}\pm} - \epsilon_{\mathbf{k}'\mp} \pm \gamma\mu_B H). \end{aligned} \quad (2.11)$$

The first term in curly brackets again represents the contribution from the first Born approximation.

The transition probabilities are to be introduced into the collision term of the Boltzmann equation. The relaxation times  $\tau(k\pm)$  are given by Van Peski-Tinbergen and Dekker<sup>9</sup> in terms of transition probabilities as

$$\begin{aligned} 1/\tau(\mathbf{k}\pm) &= \sum_{\substack{a \\ k'}} W_a(\mathbf{k}\pm \rightarrow \mathbf{k}'\pm) + \sum_{\substack{b \\ k'}} W_b(\mathbf{k}\pm \rightarrow \mathbf{k}'\mp) / \\ &\quad \{ 1 - f_{\mathbf{k}\pm} [ 1 - \exp(\epsilon_{\mathbf{k}\pm} - \epsilon_{\mathbf{k}'\mp}) / k_B T ] \}. \end{aligned} \quad (2.12)$$

The result of replacing the summation over  $k'$  in Eq. (2.12) by an integral

$$\sum_{k'} \rightarrow \frac{\Omega}{(2\pi)^3} \int 4\pi k'^2 dk' \rightarrow \frac{m\Omega}{2\pi^2 \hbar^2} \int k' dk'$$

and using Eqs. (2.10) and (2.11) in Eq. (2.12) and integrating is

where

$$\bar{M} = \sum P_M M \quad \text{and} \quad \langle M^2 \rangle_{av} = \sum M^2 P_M.$$

Note that  $\Lambda_S(x)$  is an even function of  $x$ . For large  $|x|$ , it is proportional to  $e^{-|x|/2}$ . This enters as a factor of the inelastic scattering term in Eq. (2.13). The denominator of the same term gives another factor  $e^{-|x|/2}$  for large  $x$ . Thus, the inelastic scattering term for large fields is asymptotically proportional to  $e^{-|x|}$  [apart from the slow dependence of  $g(\epsilon_{\mathbf{k}\pm} \mp \gamma\mu_B H)$  on  $x$ ].

The electrical conductivity  $\sigma_I^\pm$  due to impurity scattering is given by standard theory:<sup>9</sup>

$$\sigma_I^\pm = -e^2 / (6\pi^2 m) \int \tau(k\pm) k_\pm^3 (\partial f_{k\pm} / \partial \epsilon_{k\pm}) d\epsilon_{k\pm}. \quad (2.15)$$

The electrical resistivity  $\rho_I$  arising from impurity scattering is

$$\rho_I = (\sigma_I^+ + \sigma_I^-)^{-1}. \quad (2.16)$$

We assume  $|V| \gg |J|$  and expand the resistivity keeping only the lowest order terms in  $(J/V)^2$ . Using Eq. (2.13) in combination with Eqs. (2.14), (2.15), and (2.16) gives

$$\begin{aligned} \rho_I = & (3\pi mc\Omega)/(2e^2 N \epsilon_F \hbar) \left[ V^2 + J^2 \langle M^2 \rangle_{av} \left( 1 + 2J \int (-\partial f/\partial \epsilon) [g(\epsilon - \gamma \mu_B H) + g(\epsilon + \gamma \mu_B H)] d\epsilon \right. \right. \\ & \left. \left. + (J^2/2) \bar{M} \operatorname{csch} x/2 \left\{ \int (-\partial f^0/\partial \epsilon) d\epsilon [1 + 2Jg(\epsilon) + 2Jg(\epsilon - \gamma \mu_B H)]/D^+ \right. \right. \right. \\ & \left. \left. \left. + \int (-\partial f^0/\partial \epsilon) d\epsilon [1 + 2Jg(\epsilon) + 2Jg(\epsilon + \gamma \mu_B H)]/D^- \right\} \right], \quad (2.17) \end{aligned}$$

where

$$D^\pm = (1-f)e^{\pm x/2} + f e^{\mp x/2}, \quad (2.18)$$

where in Eq. (2.17) we have assumed isotropic scattering and factors of  $k$  and  $k^2$  have been replaced by their values at the Fermi surface. The error introduced because of this is of the order of  $(\gamma \mu_B H)/\epsilon_F$  and is neglected. Also  $(\partial f_{k\pm}/\partial \epsilon_{k\pm}) \equiv (\partial f_{k^0}/\partial \epsilon_k)$ , where the  $\pm$  subscripts have been dropped since the final result in Eq. (2.17) is independent of  $\epsilon_{\pm}$ .

To evaluate Eq. (2.17) we define the following integrals:

$$I_0^\pm(x) = \int d\epsilon (\partial f^0/\partial \epsilon)/D^\pm = (x/2) \operatorname{csch} x/2, \quad (2.19)$$

$$I_1^\pm(x) = \iint d\epsilon d\epsilon' (\partial f/\partial \epsilon) (\partial f'/\partial \epsilon') \ln |v \mp x/2|, \quad (2.20)$$

$$I_2^\pm(x) = (2/x) (\sinh \frac{1}{2} x) \iint d\epsilon d\epsilon' (\partial f/\partial \epsilon) (\partial f'/\partial \epsilon') \times (\ln |v|)/D^\pm, \quad (2.21)$$

$$I_3^\pm(x) = (2/x) (\sinh \frac{1}{2} x) \iint d\epsilon d\epsilon' (\partial f/\partial \epsilon) (\partial f'/\partial \epsilon') \times (\ln |v \mp x/2|)/D^\pm, \quad (2.22a)$$

where

$$v = (\epsilon - \epsilon')/2k_B T.$$

By making the substitution  $u = (\epsilon + \epsilon' - 2\epsilon_F)/2k_B T$ , the integral over the variable  $u$  may be carried out analytically, leading to

$$I_1^\pm(x) = \int_{-\infty}^{\infty} F(v, 0) \ln |v \mp x/2| dv, \quad (2.22b)$$

$$I_2^\pm(x) = \int_{-\infty}^{\infty} F(v, \pm x) \ln |v| dv, \quad (2.22c)$$

$$I_3^\pm(x) = \int_{-\infty}^{\infty} F(v, \pm x) \ln |v \mp x/2| dv. \quad (2.22d)$$

The function  $F(v, x)$  is given by

$$F(v, x) = (\partial/\partial v) \{ [\phi(v) - \phi(v - x/2)]/x \}, \quad (2.23)$$

where

$$\phi(v) = v \coth v,$$

$$\begin{aligned} F(v, 0) &= \frac{1}{2} (2v \coth v - 2) \operatorname{csch}^2 v \\ &= \frac{1}{2} (d^2/dv^2) [\phi(v)]. \quad (2.24) \end{aligned}$$

Using Eq. (2.23) it is easily verified that

$$\begin{aligned} F(v, x) &= F(-v, -x), \\ F(v, -x) &= F(-v, x), \\ F(x/2 - v, x) &= F(v - x/2, -x) = F(v, x). \end{aligned} \quad (2.25)$$

Using Eq. (2.25) in Eqs. (2.19) to (2.22a) gives

$$\begin{aligned} I_1^+(x) &= I_1^-(x) \equiv I_1(x), \\ I_2^+(x) &= I_2^-(x) = I_3^+(x) = I_3^-(x) \equiv I_2(x). \end{aligned} \quad (2.26)$$

Using Eqs. (2.19) to (2.26) in Eq. (2.17) in conjunction with Eq. (2.9) gives

$$\begin{aligned} \rho_I = & \frac{3\pi mc\Omega}{2e^2 N \epsilon_F \hbar} \left( V^2 + J^2 \langle M^2 \rangle_{av} \right) \\ & \times \left\{ 1 + \frac{6Jz}{\epsilon_F} [2 + \ln(k_B T/2\epsilon_F) + I_1(x)] \right\} \\ & + \frac{J^2 x}{2} \bar{M} [\operatorname{csch}^2(\frac{1}{2} x)] \left[ 1 + \frac{6Jz}{\epsilon_F} \left( 2 + \ln \frac{k_B T}{2\epsilon_F} + I_2(x) \right) \right]. \end{aligned} \quad (2.27)$$

From Eqs. (2.22a), (2.23), (2.24), and (2.26) it follows that

$$I_1(x) = \frac{1}{2} \int_{-\infty}^{\infty} \ln |v + x/2| \frac{d^2}{dv^2} [v \coth v] dv, \quad (2.28)$$

$$I_2(x) = \int_{-\infty}^{\infty} \ln |v| \frac{\partial}{\partial v} \{ [\phi(v) - \phi(v - x/2)]/x \} dv.$$

The integrals  $I_1(x)$  and  $I_2(x)$  can not be evaluated in closed form. For small and large values of  $x$  we obtain

$$\begin{aligned} I_1(x) &\approx -0.432 + 0.091x^2; \quad |x| < 2 \\ &\approx \ln \left| \frac{x}{2} \right| - \frac{1.63}{x^2}; \quad |x| > 10 \end{aligned} \quad (2.29)$$

and

$$I_2(x) \approx -0.432 + 0.030x^2; \quad |x| < 2$$

$$\approx \ln \left| \frac{x}{2} \right| - 1 + \frac{3.27}{x^2}; \quad |x| > 10. \quad (2.30)$$

The asymptotic values of  $I_1(x)$  and  $I_2(x)$  will be used later to evaluate the variation of the resistivity near  $T=0$ . The functions  $I_1(x)$  and  $I_2(x)$  are shown in Fig. 1.

### III. CALCULATION OF THE RESISTIVITY

The expression Eq. (2.27) for  $\rho$  contains the variable  $x = \gamma\mu_B H / (k_B T)$  and is therefore dependent upon the internal (or external) field in the system. By measuring the resistivity of dilute magnetic alloys as a function of the temperature and the impurity concentration one hopes to learn something about the distribution of the internal fields in the alloy. The theoretical interpretation of the experimental resistivity measurements must also be examined with respect to consistency with other results, such as the low-temperature specific heat, the magnetic susceptibility, Mössbauer determination of hyperfine fields, etc., in order to distinguish between alternative models proposed for the magnetic state of the alloy system.

It was proposed by Marshall<sup>17</sup> that the very low temperature specific-heat anomaly of dilute copper manganese arises from the distribution of random internal fields resulting from the random positions of the impurities which interact via a RKKY potential. Marshall argued qualitatively that the probability distribution  $P(H, T)$  of the internal fields  $H$  at a temperature  $T$  is a truncated Lorentzian. Later Klein and Brout<sup>18,19</sup> used more rigorous methods to obtain the  $P(H, 0)$ , the distribution function at  $T=0$ , in an Ising model. It was found that spin correlations between the magnetic impurities reduce the probability of obtaining very small (near zero) fields by roughly a factor of 4 from that of an uncorrelated system, and correspondingly increases the probability for large fields. For the purposes of this paper the Ising model  $P(H, 0)$  is approximated sufficiently well by

$$P(\gamma\mu_B H, 0) = (2 \tan^{-1} 4)^{-1} \frac{\Delta}{\Delta^2 + (\gamma\mu_B H)^2} \quad |H| < 4\Delta/\gamma\mu_B$$

$$= 0, \quad |H| > 4\Delta/\gamma\mu_B \quad (3.1)$$

where  $\Delta$  is the spread in the distribution function, again calculated in an Ising model. In most of our calculations we will use the value of  $\Delta$  adjusted so that it gives agreement with the experimental low-temperature specific heat,<sup>19</sup> i.e.,

$$\Delta \approx 750c \propto J^2 c / \epsilon_F, \quad (3.2)$$

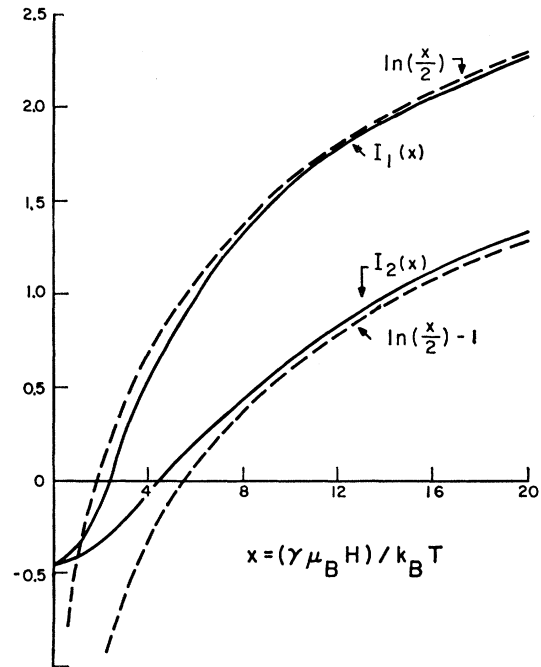


FIG. 1. The function  $I_1(x)$  and  $I_2(x)$ , where  $x = \gamma\mu_B H / k_B T$ . The functions were obtained by numerical integration of Eq. (2.27).

where  $c$  is the fractional impurity concentration.  $\Delta$  in Eq. (3.2) is expressed in degrees Kelvin. We remark that the distribution function, Eq. (3.1), is truncated at a value proportional to the impurity concentration, unlike the truncation used by Marshall.<sup>17</sup> In the more correct probability distribution function calculated in Ref. 19, there is a finite probability that  $|H| > 4\Delta/\gamma\mu_B$ , but the qualitative (or semiquantitative) results of the resistivity calculation will not be appreciably affected by considering these large fields because of the relatively small probability associated with them. Throughout this paper we use the  $T=0$  probability distribution of the fields and then later discuss qualitatively the errors arising from this. Therefore, from now on we suppress the index 0 in  $P(\gamma\mu_B H, 0)$ , i.e.,  $P(\gamma\mu_B H, 0) \equiv P(\gamma\mu_B H)$ .

The existence of another magnetic state for these alloys was proposed by Abrikosov and Gorkov.<sup>4,25</sup> According to this, long-range magnetic order with a well-defined transition temperature<sup>26</sup> exists in dilute magnetic alloys interacting via an  $s-d$  exchange interaction. Part of the purpose of this paper is to suggest experiments which may differentiate between the

<sup>25</sup> A. A. Abrikosov and L. P. Gorkov, Zh. Eksperim. i Teor. Fiz. 43, 2230 (1962) [English transl.: Soviet Phys.—JETP 16, 1575 (1962)].

<sup>26</sup> A third model to explain the low-temperature specific heat of the dilute alloys was proposed by A. W. Overhauser, Phys. Rev. Letters 3, 414 (1959). We have not calculated the predictions for the resistivity arising from this model.

Marshall-Brout-Klein (MBK) and Abrikosov models. In particular, the change in the very low-temperature resistivity of dilute magnetic alloys obtained from the MBK model is linear in  $T$ , with slope proportional to the  $T \approx 0$  specific heat and to the magnetic susceptibility (that these two quantities are related has been discussed before).<sup>27</sup> In a long-range-ordered system the change in  $\rho(T)$  has zero slope near  $T=0$ . The resistivity as a function of temperature is

$$\rho(T) = \int_{-\infty}^{\infty} P(\gamma\mu_B H) \rho_I(x, T) d(\gamma\mu_B H) + aT^5. \quad (3.3a)$$

$$\rho_I(0) = (3\pi mc\Omega) / (2e^2 N \epsilon_F \hbar z)$$

$$\times \left\{ V^2 + J^2 S^2 \left[ 1 + (6J/\epsilon_F)(2 + \log(k_B/4\epsilon_F)) + \int_{-4\Delta}^{4\Delta} P(\gamma\mu_B H) \log(\gamma\mu_B H/k_B) d(\gamma\mu_B H) \right] \right\}. \quad (3.4)$$

The value of the integral in Eq. (3.4) is  $-0.443 + \log(\Delta/k_B)$ . We define the change in the resistivity from its  $T=0$  value by  $\Delta\rho(T)$ , then

$$\Delta\rho(T) = \rho(T) - \rho(0). \quad (3.5)$$

#### A. The Resistivity near $T=0$

We next show that for the short-range-ordered system,  $\Delta\rho(T)$  near<sup>20</sup>  $T=0$  is nearly linear in  $T$  and is only weakly dependent upon the impurity concentration and the strength of the  $s$ - $d$  exchange interaction  $J$ . Also for a fixed  $\Delta$ ,  $\Delta\rho(T)$  is proportional to the impurity spin  $S$ .

Let  $m$  be the slope of the resistivity as a function of temperature near  $T \rightarrow 0$ .<sup>20</sup> Then the coefficient  $m$  of the linear term in  $T$  is

$$m = \lim_{T \rightarrow 0} d\rho/dT. \quad (3.6)$$

As  $T \rightarrow 0$ ,  $x \rightarrow \infty$ , and we use the asymptotic form of  $I_1(x)$  and  $I_2(x)$  as given by Eqs. (2.29) and (2.30) to obtain

$$m \approx AJ^2 c \lim_{T \rightarrow 0} \int_0^{4\Delta/T} \frac{\Delta}{\Delta^2 + x^2 T^2} x \times \left[ 1 + \frac{6J}{\epsilon_F} \left\{ 2 + \ln \left( \frac{xT}{4\epsilon_F} \right) \right\} \right] \frac{dF_3(x)}{dx} dx, \quad (3.7)$$

where

$$A = (3\pi m\Omega/2e^2 N \epsilon_F \hbar) \approx 2.3 \times 10^{-8} [\epsilon_F(\text{copper})/\epsilon_F]^{1/2} \epsilon_F^{-2} \Omega \text{ cm}/\text{eV}^2.$$

$A$  is conveniently written so that the interaction potentials in the equation for the resistivity are expressed in units of the Fermi energy of the host, and

$$-F_3(x) = S(S+1) + \bar{M} \left[ \frac{1}{2} x \operatorname{csch}^2 \left( \frac{1}{2} x \right) - \coth \frac{1}{2} x \right]. \quad (3.8)$$

<sup>27</sup> M. W. Klein, Phys. Rev. **141**, 489 (1966).

For the Ising-like distribution of fields this becomes

$$\rho(T) = \Gamma \int_0^{4\Delta/T} \frac{(\Delta/T)}{(\Delta/T)^2 + x^2} \rho_I(x, T) dx + aT^5, \quad (3.3b)$$

where  $a$  is assumed to be  $2.6 \times 10^{-16} \Omega \text{ cm}/\text{deg}^5$ , as in Kondo's discussion.<sup>2</sup> Here  $\rho(x, T)$  is given by Eq. (2.26) and  $\Gamma = (\tan^{-1} 4)^{-1}$ . In order to compare results with experiment it is convenient to subtract out the zero-temperature limiting value of the resistivity  $\rho(0)$  from Eq. (3.3b).  $\rho(0)$  can be evaluated by using the asymptotic forms of  $I_1(x)$  and  $I_2(x)$  as given in Eqs. (2.28) and (2.29). We find

Using Eq. (3.8) in Eq. (3.7) gives

$$m \approx AS c J^2 Q P(0) [1 + (6Jz/\epsilon_F)(2 + \ln(k_B/4\epsilon_F))] \times (1 - \alpha \ln T), \quad (3.9a)$$

where  $P(0)$  is the value of the probability distribution function at  $H=0$ ,  $P(0) = \Gamma/\Delta$ , and  $\alpha = -(6Jz/\epsilon_F)/[1 + (6Jz/\epsilon_F)(2 + \ln(k_B/4\epsilon_F))]$ .  $Q$  is a number of the order of unity and is independent of  $J$  and  $c$  and only weakly dependent upon  $S$ . The low-temperature specific heat is also proportional<sup>17-19</sup> to  $P(0)$ . Using the expression Eq. (3.2) for  $\Delta$  gives for the slope of the resistivity  $m$ :

$$m \propto S \epsilon_F^{-3/2} (1 - \alpha \ln T) \times \{1 + (6Jz/\epsilon_F)[2 + \ln(k_B/4\epsilon_F)]\}. \quad (3.9b)$$

The value of  $\alpha$  is approximately 0.06 for copper and gold if  $J$  is taken as  $-0.15 \text{ eV}$ . We find, therefore, that except for the relatively weak dependence on  $J$ , the initial slope from one material to another is, in our model, proportional to the impurity spin and is proportional to  $\epsilon_F^{-3/2}$ . The larger the impurity concentration the greater is the range in which the resistivity is linear in  $T$ . The behavior of  $\Delta\rho(T)$  versus  $T$  for low temperatures and for several concentrations is shown in Fig. 2. The solid lines are the results obtained when using the probability distribution of the internal field given in Eq. (3.1) with the impurity spin  $S=1$ . The results were obtained by numerical integration of Eq. (3.3). The dashed lines in Fig. 2 show the calculated results obtained when one assumes long-range magnetic order to exist in the system. For this case we assume for  $T \ll T_0$

$$P(\gamma\mu_B H) = \delta(\gamma\mu_B H - k_B T_0), \quad (3.10)$$

where  $T_0$  is the temperature at which the magnetic susceptibility is a maximum, i.e.,  $\gamma\mu_B H \approx 750c$ . The long-range-ordered model of the form presented by Abrikosov

and Gorkov<sup>25</sup> gives, in the molecular-field approximation, a change in  $\rho(T)$  near  $T \rightarrow 0$  which has zero slope, in contrast to the linear behavior predicted by the short-range order of Klein and Brout.<sup>18,19</sup> The very low-temperature resistivity of Au-Fe and Au-Mn was measured by MacDonald *et al.*<sup>28</sup> For the Au-Fe alloy with a nominal concentration of 0.1 at. % Fe the linear dependence of the resistivity with  $T$  is clearly exhibited<sup>28</sup> in Fig. 11(b). Their data on the 0.04% Au-Fe alloy also suggests linearity. The linear behavior of  $\Delta\rho(T)$  for copper alloys ( $\epsilon_F = 7$  eV,  $J = -0.15$  eV) is shown in Fig. 2. The slope is about  $5 \times 10^{-9}$   $\Omega$  cm/deg. For gold-iron the value of  $\Delta$  obtained from the low-temperature specific-heat data<sup>19,29</sup> is about the same as that for copper alloys. Using Eq. (3.9b) we find that the slope has to be multiplied by the ratio of the Fermi energy of copper to that of gold to the three-halves power. Taking the iron spin to be unity and  $\epsilon_F$  for gold to be 5 eV gives for the slope  $\Delta\rho(T)$  about  $0.8 \times 10^{-8}$   $\Omega$  cm/deg. The measurements of MacDonald *et al.* give  $10^{-8}$   $\Omega$  cm/deg. (It is our opinion that the very good agreement between theory and experiment is accidental. For our purposes a disagreement of only a factor of 2 between them is still considered good.)

We consider the qualitative and relatively good quantitative agreement between the low-temperature specific heat<sup>29</sup> and the low-temperature resistivity as additional evidence that, at least for the 0.1% Au-Fe for which the very low-temperature resistivity has been measured, the probability for zero internal fields is well described by an Ising-like probability distribution function of the form given by Eq. (3.1). Our model predicts this approximately linear behavior at sufficiently low temperatures for all dilute magnetic alloys in which the excess specific heat is linear in  $T$  and  $J$  is negative. We should also remark that the temperature of the maximum in  $\rho(T)$  for the 0.1% Au-Fe is  $\approx 2.5^\circ\text{K}$  from experiment and  $1.8^\circ\text{K}$  from theory.

Another difference between the two models is predicted in the resistivity at high temperatures. On the basis of a long-range-ordered model, with  $T_0$  as the ordering temperature, as  $T$  is decreased from high temperatures  $\rho(T)$  would reach its minimum and then increase again, provided that  $T_{\min}$  is greater than  $T_0$ . Slightly above  $T_0$ ,  $\rho(T)$  may possibly decrease somewhat due to the onset of short-range order, but as the temperature is further lowered to  $T_0$  there would be an abrupt, almost discontinuous decrease of the resistivity due to the onset of long-range ordering. The

<sup>28</sup> D. K. C. MacDonald, W. B. Pearson, and I. M. Templeton, Proc. Roy. Soc. (London) **A266**, 161 (1962).

<sup>29</sup> B. Dreyfus, S. Souletie, R. Tournier, and L. Weil, Compt. Rend. **259**, 4266 (1964). The linearity of the low-temperature specific heat of Au-Fe with temperature is exhibited in this reference. Here also, the excess low-temperature specific heat is approximately independent of the impurity concentration. The fact that the excess specific heat changes by about a factor of 2 as the concentration changes by a factor of 40 is presently still unexplained.

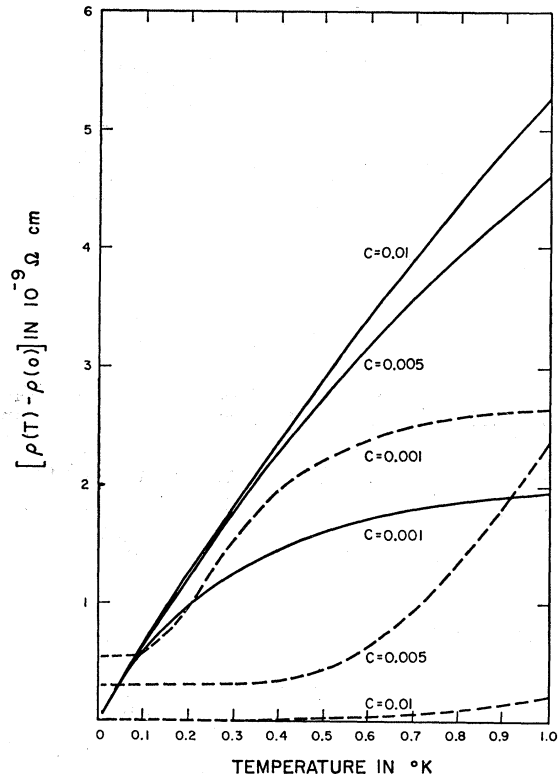


FIG. 2. The change in resistivity  $\Delta\rho(T)$  with temperature for three different concentrations. The solid lines were obtained by using an Ising-like distribution of internal fields, whereas the dashed lines are the results obtained by using Eq. (3.10) for the field distribution. Both sets of curves were obtained by numerical integration.

short-range-ordered model, on the other hand, predicts a continuous change of the resistivity with temperature.

### B. The Resistivity Maximum

To obtain the condition for an extremum in the resistivity we differentiate Eq. (3.3) with respect to  $T$  and set the derivative equal to zero. Let  $\delta = \Delta/T$  and let

$$Y_1(\delta) = \Gamma \int_0^{4\delta} \langle M^2(x) \rangle_{av} I_1(x) \frac{\delta}{\delta^2 + x^2} dx, \quad (3.11a)$$

$$Y_2(\delta) = \Gamma \int_0^{4\delta} \bar{M}(x)(x/2) \operatorname{csch}^2(\frac{1}{2}x) \frac{\delta}{\delta^2 + x^2} dx, \quad (3.11b)$$

$$Y_3(\delta) = \Gamma \int_0^{4\delta} \{S(S+1) + \bar{M}(x)[\frac{1}{2}x \operatorname{csch}^2(\frac{1}{2}x) - \coth \frac{1}{2}x]\} \times \frac{\delta}{\delta^2 + x^2} dx, \quad (3.11c)$$

where in Eq. (3.11) we used the relationship  $\langle M^2 \rangle_{av}$



$$= S(S+1) - \bar{M} \coth \frac{1}{2} x. \text{ Then}$$

$$d\rho/dT \propto Y_3(\delta) - \delta \{ [\epsilon_F / (6J) + 2 + \ln(k_B T / 2\epsilon_F)] \\ \times Y_3'(\delta) + Y_1'(\delta) + Y_2'(\delta) \} \\ + (5aT^5 \epsilon_F) / (6AcJ^3) = 0, \quad (3.12)$$

where the primes in Eq. (3.12) are derivatives with respect to  $\delta$ . It is convenient to rewrite Eq. (3.12) in the form

$$\ln T_{\text{ext}} = -\ln(k_B / 2\epsilon_F) - \epsilon_F / (6J) - 2 \\ + \{ Y_3(\delta) / \delta - Y_1'(\delta) - Y_2'(\delta) \} \\ + (5aT_{\text{ext}}^5 \epsilon_F) / (6AcJ^3 \delta) / Y_3'(\delta), \quad (3.13)$$

where  $T_{\text{ext}}$  is the temperature of the extremum. Equation (3.11) will be used to discuss the two extrema, namely  $T_{\text{max}}$  the temperature at which the resistivity is a maximum, and  $T_{\text{min}}$  the temperature of the resistivity minimum. As long as these temperatures are reasonably well separated and  $T_{\text{max}} < T_{\text{min}}$ , the value of  $T_{\text{max}}$  may be determined by neglecting the  $T^5$  term in Eq. (3.13). We obtain

$$\ln T_{\text{max}} = G(\delta) = G(\Delta / T_{\text{max}}) \approx G(750c / T_{\text{max}}). \quad (3.14)$$

The left-hand side of Eq. (3.14) varies slowly with  $T_{\text{max}}$ , whereas the right-hand side of Eq. (3.14) is found to be, using numerical integrations, rapidly varying with  $\delta$ . Thus, the resistivity maximum is approximately determined by the value of  $\delta_{\text{max}}$ . Therefore, a change in the concentration by a factor of 2 requires a change in  $T_{\text{max}}$  also by a factor of 2 in order to keep  $\delta_{\text{max}}$  a constant. We therefore find that the temperature at which the resistivity is a maximum is proportional to the

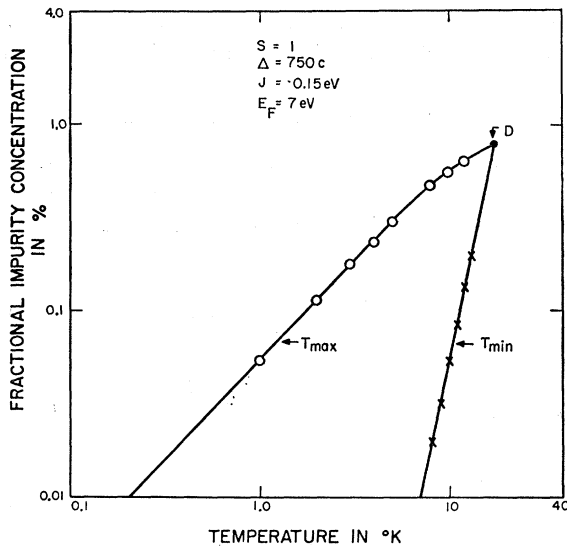


FIG. 3. The temperature of the resistivity maximum and minimum as a function of the impurity concentration. The results are obtained by numerical solution of Eq. (3.13). The point  $D$  gives the concentration where the maximum and minimum temperatures coincide. At this concentration the maximum as well as the minimum disappears.

impurity concentration. We have solved Eq. (3.13) as a function of the impurity concentration and the results are shown in Fig. 3. The circles in the figure represent the solution obtained numerically. It is seen from Fig. 3 that  $T_{\text{max}}$  is proportional to the impurity concentration up to a temperature relatively close to the resistivity minimum.

At this point we should note that the result that  $T_{\text{max}} \propto c$  was obtained without a moment expansion of the effective internal field. In fact this result is independent upon the details of the probability distribution used and would be valid for a cutoff Lorentzian as well as a Gaussian. The change in the concentration dependence of  $T_{\text{max}}$  from a linear dependence for a Gaussian to a  $c^{1/2}$  dependence for a cutoff Lorentzian as found by Silverstein<sup>11</sup> arises only because one uses a moment expansion.

The low-temperature resistivity maximum occurs with negative values of  $J$  only. However, when  $J$  is positive an increase in the internal field tends to reduce the increased resistance arising from the  $\ln T$  term. Therefore, for positive  $J$ , as the temperature departs from  $T=0$  the initial slope of the resistivity will be negative from our model.<sup>30</sup> This behavior is exhibited in Fig. 4 where it is found that with positive  $J$  value the resistivity has a low-temperature minimum. The minimum was found, by our numerical calculations, to be approximately proportional to the impurity concentration for low concentrations. In Fig. 4 we also show the variation of the resistivity with the value of the impurity spin  $S$  keeping  $\Delta$  constant. Note that  $T_{\text{max}}$  is somewhat spin-dependent. We find that the value of  $\delta_{\text{max}}$  which satisfies Eq. (3.14) is  $\delta_{\text{max}} = 0.395$  for  $S = \frac{1}{2}$ ,  $\delta_{\text{max}} = 0.415$  for  $S = 1$ , and  $\delta_{\text{max}} = 0.55$  for  $S = \frac{5}{2}$ . The initial slope of the  $S = 1$  curve is that shown in Fig. 2.

The value of  $\Delta\rho(T)$  at  $T = T_{\text{max}}$  increases with the impurity concentration and the value of  $J$ . An approximate relation for the value of  $\Delta\rho$  at its maximum,  $\Delta\rho(T_{\text{max}})$ , is

$$\Delta\rho(T_{\text{max}}) \propto cJ^2 \{ 1 + (6Jz / \epsilon_F) (2 + \ln(k_B \Delta / \epsilon_F)) \}. \quad (3.15)$$

The second term in Eq. (3.15) is not very accurate and should only be considered as a guide to obtain approximate values. We have performed computations in which we varied  $J$  [including the value of  $J$  occurring in Eq. (3.2)] for a 0.1% alloy and found Eq. (3.15) correct to within 20% as  $J$  was varied from 0.075 to 0.225 eV.

### C. The Resistivity Minimum

For low concentration of magnetic impurities (of the order of 0.1% or less) the resistivity minimum in the presence of internal fields is not drastically changed from the case when the field is zero. Thus, the Kondo

<sup>30</sup> It is not clear whether this can be taken seriously in view of the Suhl-Abrikosov resonance temperature, which is not small for positive  $J$ .

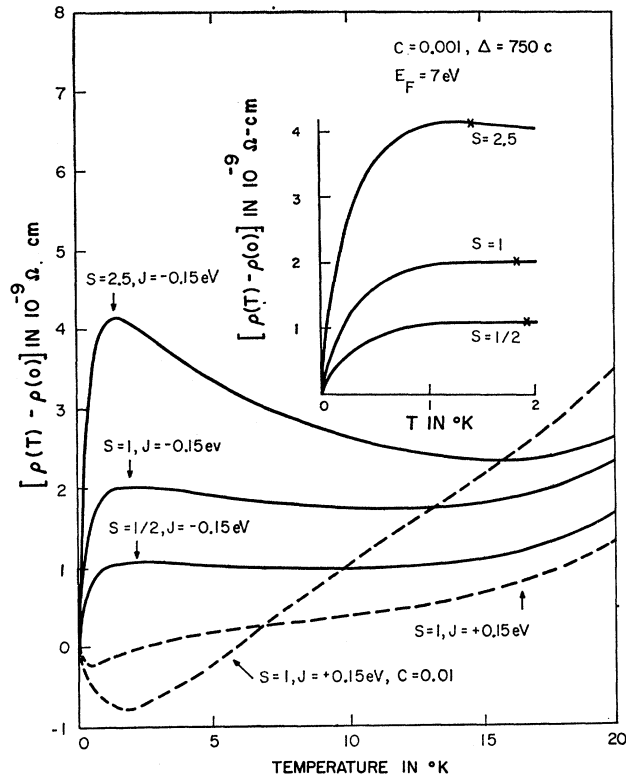


FIG. 4. The change in resistivity  $\Delta\rho(T)$  for several impurity spins  $S$ , and two values of  $J$ . The crosses in the upper figure show  $T_{\max}$ . The impurity concentration is 0.1% unless otherwise indicated. The solid lines represent the results of numerical integration assuming negative  $J$ . Positive  $J$  is assumed in the results indicated by dashed lines.

treatment<sup>2</sup> gives the value of  $T_{\min}$  quite well. From our analysis we find that at  $T=T_{\min}$  there are still some impurities which experience large internal fields and quench the Kondo effect completely, others experience small internal fields and quench the Kondo effect only partially. For this reason the resistivity near  $T=T_{\min}$  will be somewhat lower than that given by the Kondo treatment.<sup>2</sup> But since it is difficult to estimate the variation of the probability distribution of the internal field as a function of the temperature we can not give a good quantitative estimation of how much the resistivity is suppressed around  $T=T_{\min}$  for concentration of the order of 0.1%. However, we can describe qualitatively, and semiquantitatively by machine calculation, the resistivity behavior as the concentration is increased. We recall that the temperature at which the resistivity maximum occurs is proportional to the impurity concentration. Therefore, as the concentration is increased the maximum moves to higher and higher temperatures. As this happens the whole resistivity curve as a function of temperature flattens, until the maximum and the minimum "coalesce" and both disappear, resulting in a monotonic decrease in the resistivity as the temperature is lowered. An approximate expression for the

disappearance of the maximum and the minimum can be simply derived by requiring that  $T_{\min} > T_{\max}$ .  $T_{\min}$  is of the order of  $10^\circ\text{K}$  and is proportional to  $c^{1/5}$ , whereas  $T_{\max} \approx 1800c$  for  $S=1$ . We therefore obtain the condition

$$1800c > 10(c/c_0)^{1/5}, \quad (3.16)$$

where  $c_0$  is 0.01% for copper manganese.<sup>31</sup> This rough calculation gives that the minimum disappears at a concentration of about 1%. We shall discuss later that for Cu-Mn the experimental value of  $T_{\max}$  is about twice that which one obtains using  $\Delta=750c$  and  $S=\frac{5}{2}$ , and therefore the minimum may be expected to disappear at about 0.5% concentration. To obtain a more accurate description of the concentration where the minimum disappears one has to solve Eq. (3.13) with the  $T^5$  term included. This we have done for an impurity spin  $S=1$ , and the calculated results obtained are shown by the crosses in Fig. 3. We note that the  $c^{1/5}$  dependence of  $T_{\min}$  is retained in the presence of internal fields as seen from the slope of the  $T_{\min}$  curve as a function of the impurity concentration. We find that for the parameters chosen the maximum and the minimum temperature coalesce at a concentration slightly below 0.8%. For concentrations greater than 0.8% the resistivity is monotonically decreasing with decreasing temperatures for negative values of  $J$ .

The variation of the resistivity as a function of the temperature and for several values of the impurity concentration is shown in Fig. 5. Note how the characteristic curves change as the concentration is increased. For the 0.1% concentrated alloy there is well defined maximum and minimum, whereas for the 0.5% alloy the curve is flat over a wide range of temperatures but the minimum is still retained. The 1% alloy shows neither a minimum nor a maximum but the resistivity decreases monotonically with decreasing temperature. The slopes for each of these curves near  $T=0$  is given in Fig. 2. They are approximately independent of the impurity concentration and their value is given in Eq. (3.9). The dashed line in Fig. 5 shows the resistivity arising from a long-range-ordered system with a fixed internal field. Its slope near  $T=0$  is essentially zero.

#### D. The Variation of the Resistivity in the Presence of External Fields

We have calculated the part of the resistivity which arises from the  $s$ - $d$  scattering mechanism in the presence of an external field. In this calculation the  $v \times B$  term in the Boltzmann transport equation leading to the ordinary magnetoresistance of the solvent is neglected. The concentration was chosen to be sufficiently low that the internal field effects are not very large. In fact we set the internal field equal to zero and varied the

<sup>31</sup> D. K. C. MacDonald, *An Introduction to the Principles of Thermoelectricity* (John Wiley & Sons, Inc., New York, 1962), p. 33.

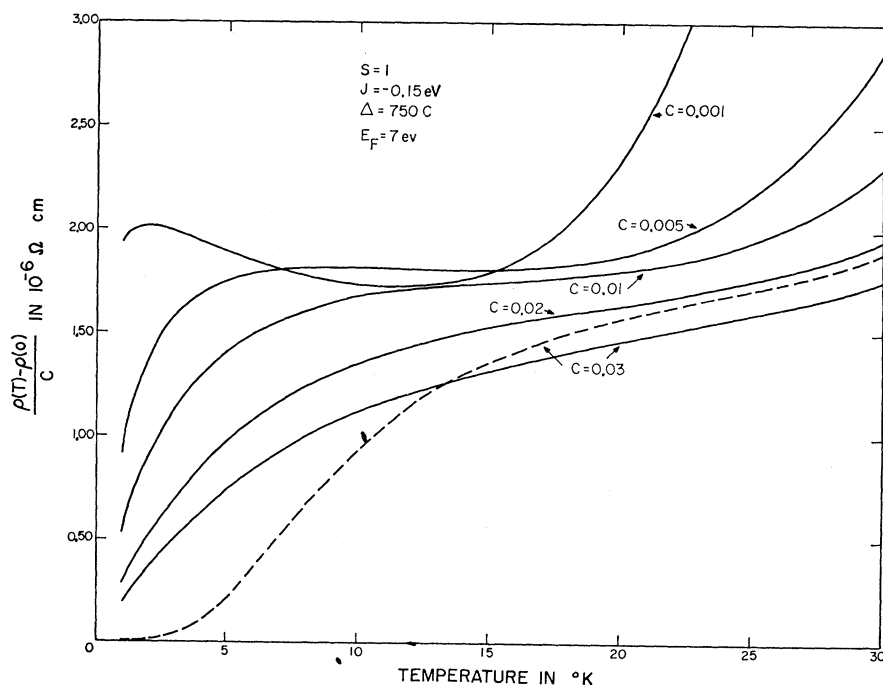


FIG. 5. The change in resistivity  $\Delta\rho(T)$  divided by the impurity concentration as a function of temperature and several concentrations. The solid lines are obtained by using an Ising-like distribution of fields, whereas the dashed line is the result from a long-range-ordered system. The graphs were obtained by numerical integration.

external field. The result of this calculation is  $\rho_I(T) - \rho_I(0)$  as given in Eq. (2.27) and is shown in Fig. 6. It is important to emphasize that in an experiment there will be internal-field effects which will, in general, interfere with the external field. However, in the limit as the concentration goes to zero the internal fields are negligible the external field alone will cause the suppression of the Kondo  $\ln T$  term. The correct way to combine the internal- and external-field effects for higher concentration is a difficult problem and is not treated in this paper.

#### IV. COMPARISON WITH EXPERIMENTS ON Cu-Mn AND Au-Fe

We now briefly compare the theoretical resistivity calculation obtained from the second Born approximation with experiments on Cu-Mn and Au-Fe. The agreement of the initial slope of the resistivity and the temperature at which the resistivity is a maximum was discussed in the previous section. We recall that this agreement was obtained by adjusting  $\Delta$  so that it gives the low-temperature specific heat in agreement with experiment. The value of the experimental  $\Delta\rho(T_{\max})$  for 0.1% Au-Fe is of the order of  $10^{-8} \Omega \text{ cm}$ , whereas the theoretical value for gold iron is about  $4 \times 10^{-9} \Omega \text{ cm}$  for spin  $S=1$  and  $9 \times 10^{-9}$  for spin  $S=\frac{5}{2}$ . Therefore, the theoretical and experimental low-temperature resistivities for Au-Fe are consistent with each other and are also consistent with the low-temperature specific-heat measurements. It should be pointed out, however, that the close numerical agreement of the theoretical and experimental resistivities at  $T=T_{\max}$  may be acci-

dental, for this value depends upon  $J$  which we have arbitrarily chosen to be 0.15 eV. The agreement of the linearity in initial slope of the resistivity we consider to be very significant.

For Cu-Mn we are not aware of an experiment from which the initial slope of  $\Delta\rho(T)$  can be determined. However, the resistivity maximum for a spin  $S=\frac{5}{2}$  occurs at  $T=1.4^\circ\text{K}$  from our calculations and at  $T \approx 4^\circ\text{K}$  from experiments on a 0.1% Cu-Mn alloy.<sup>31</sup> This result indicates that the spread of the distribution function  $\Delta$  which fits the low-temperature specific-heat data has to be increased by a factor of 2 to 3 to fit the experimental resistivity maximum, resulting in a quantitative discrepancy between the two.

#### V. DISCUSSION

There are several difficulties in the approach presented in this paper. One of these is that we have considered the resistivity to second Born approximation only. It was already found previously<sup>3-5</sup> that serious difficulties arise at low enough temperatures if one considers the resistivity to second Born approximation only. However, we believe that this approximation gives the correct qualitative behavior of the resistivity maximum and minimum for  $T \gg T_r$ . This appears to be borne out by comparing with experiments to date, at least for negative values of  $J$ .

We can now calculate the approximate concentration above which the resistivity maximum will be at  $T$  greater than  $T_r$ . For this purpose we set  $\Delta=750C=T_r$ , where  $T_r$  is given in Eq. (1.1). This gives a concentration of the order of  $10^{-6}$  to  $10^{-7}$ . Therefore, one expects

that for concentrations much greater than  $10^{-6}$  there will be a maximum in the resistivity arising from internal field effects at a temperature much greater than  $T_r$ .

There is an error arising in our calculations from the fact that we have used the  $T=0$  probability distribution of the fields for higher temperatures. A preliminary estimate indicates the correct temperature-dependent probability distribution may give a quantitative change in the resistivity, but this change will not be very large. We therefore feel that the qualitative behavior of the resistivity is correctly described by the  $T=0$  probability distribution.

### VI. CONCLUSION

We have calculated the effect of internal fields on the low-temperature resistivity of dilute concentrations of magnetic impurities in a nonmagnetic host using the second Born approximation. We have shown that in an Ising-like distribution of internal fields the low-temperature resistivity varies approximately linearly with temperature. More generally, the variation of the low-temperature resistivity is approximately proportional to the low-temperature specific heat arising from magnetic disordering. Comparison of the experimental low-temperature specific heat with the experimental low-temperature resistivity for a 0.1% concentrated Au-Fe alloy substantiates the proportionality between the above two quantities. The predicted change in the very low-temperature resistivity with temperature is approximately concentration-independent. The theory predicts a maximum in the low-temperature resistivity and  $T_{\max}$  is, for negative  $J$ , proportional to the impurity concentration. At concentration of about 1% the maximum as well as the minimum disappears for iron or manganese in copper or gold. The theory gives qualitative, and in some cases quantitative, agreement with the experimental low-temperature resistivity in dilute magnetic alloys. It is indicated that low-temperature

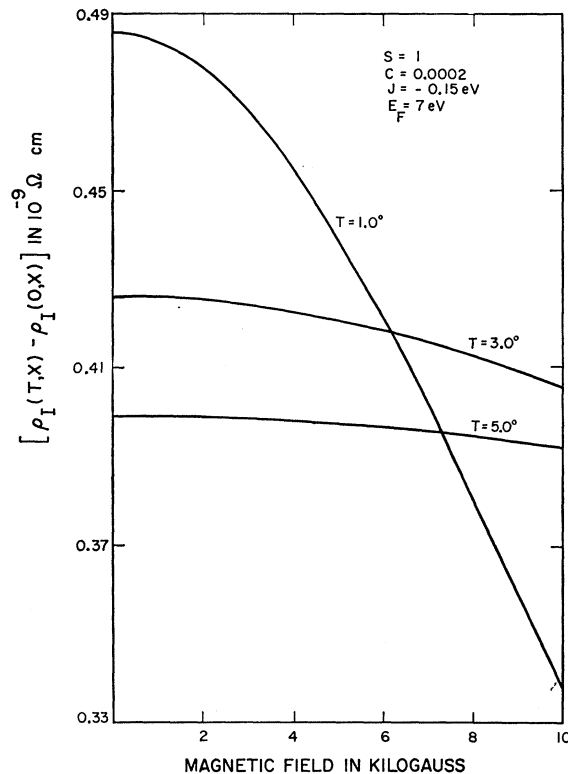


FIG. 6. The change in the resistivity  $\Delta\rho(T)$  as a function of the external field. For these results the internal field was taken to be zero; therefore the curves do not exhibit the interference between the internal and external fields.

resistivity measurements may be used for probing the internal-field distribution of these alloys.

### ACKNOWLEDGMENTS

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