Effect of Ordinary Scattering on Exchange Scattering from Magnetic Impurity in Metals

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We investigate the ordinary and exchange scattering of conduction electrons by magnetic impurities. We find that for thermodynamic properties the effect of ordinary scattering is *entirely* absorbed into an effective exchange interaction $J\cos^2\eta$, where η is the phase shift due to the ordinary scattering alone. The resistivity is calculated by using the Nagaoka-Hamann scheme. The nth-order leading logarithmic term ($\sim J^{n+2}\log^n T$ in the absence of ordinary scattering) is multiplied by $(\cos\eta)^{2n+4}\cos 2\eta$, in agreement with Fischer, who obtained this result for $n=1$. We discuss the alloy Rh-Fe, where the resistivity was interpreted in terms of a positive exchange interaction, whereas the susceptibility data indicated a negative one. We point out that these results are not inconsistent with each other in the light of the present theory.

1. INTRODUCTION

ANOMALOUS scattering[1] of conduction electrons from a magnetic impurity in metals has been the subject of a considerable number of recent works.[2] Most of the authors are concerned with the low-temperature anomaly which is associated with a negative *s-d* exchange interaction. Recently, Fischer[3] has considered the effect of ordinary scattering on the exchange scattering and found that the first ln T term in the resistivity changes its sign when the ordinary scattering is sufficiently large. Since the well-known low-temperature anomaly is associated with the divergence of a series in $2J\rho\ln(T/D)$ in the case of no ordinary scattering, a question immediately arises as to what the series looks like and when the anomaly occurs in the presence of ordinary scattering. We obtained the leading logarithmic terms and found that the series is in $2J\rho\cos^2\eta\ln(T/D)$ so that the low-temperature anomaly occurs at the temperature $D\exp(1/2J\rho\cos^2\eta)$ when J is negative. We also investigated the effect of ordinary scattering on thermodynamic properties and found that it can *entirely* be absorbed into an effective exchange interaction $J\cos^2\eta$. This is in conformity with the result on resistivity.

We also discuss the alloy Rh-Fe where the resistivity was interpreted in terms of a positive exchange interaction, whereas the susceptibility data indicated a negative one. We point out that these results are not inconsistent with each other in the light of the present theory.

2. THERMODYNAMIC PROPERTIES

We consider the free conduction electrons and a single magnetic impurity which is located at the origin of the coordinates and interacts with the conduction electrons via an exchange interaction. We take account of the ordinary potential arising from the impurity as well. Thus our Hamiltonian consists of two terms, H_0 and H', which are expressed by

$$H_0=\sum_{\mathbf{k}s}\epsilon_{\mathbf{k}}a_{\mathbf{k}s}{}^{\dagger}a_{\mathbf{k}s}+(V/N)\sum_{\mathbf{k}\mathbf{k}'s}a_{\mathbf{k}s}{}^{\dagger}a_{\mathbf{k}'s},\tag{1}$$

$$H'=-(J/N)\sum_{\mathbf{k}\mathbf{k}'}\big[(a_{\mathbf{k}\uparrow}{}^{\dagger}a_{\mathbf{k}'\uparrow}-a_{\mathbf{k}\downarrow}{}^{\dagger}a_{\mathbf{k}'\downarrow})S_z +a_{\mathbf{k}\uparrow}{}^{\dagger}a_{\mathbf{k}'\downarrow}S_-+a_{\mathbf{k}\downarrow}{}^{\dagger}a_{\mathbf{k}'\uparrow}S_+\big],\tag{2}$$

where $a_{\mathbf{k}s}{}^{\dagger}$ and $a_{\mathbf{k}s}$ denote the creation and the annihilation operator for the electron $\mathbf{k}s$, S is the spin operator of the impurity, and N is the total number of the unit cells. V and J represent the strength of the ordinary and exchange interaction. We neglect their dependence on the wave vectors. In this section we discuss the thermodynamic properties of the system represented by the Hamiltonian H_0+H'. In the next section we discuss the resistivity using an approach different from that of this section.

By using a unitary transformation

$$a_{\mathbf{k}s}{}^{\dagger}=\sum_n\langle n\,|\,\mathbf{k}\rangle a_{ns}{}^{\dagger},\tag{3}$$

we diagonalize H_0 as

$$H_0=\sum_n\epsilon_n a_{ns}{}^{\dagger}a_{ns}.\tag{4}$$

The secular equation for this transformation is expressed by

$$\epsilon_{\mathbf{k}}\langle\mathbf{k}\,|\,n\rangle+(V/N)\sum_{\mathbf{k}'}\langle\mathbf{k}'\,|\,n\rangle=\epsilon_n\langle\mathbf{k}\,|\,n\rangle.\tag{5}$$

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We define a Green's function[4] which takes full account of the ordinary scattering by

$$G_{\mathbf{k}'\mathbf{k}}{}^{0}(\omega)=\sum_{n}\langle\mathbf{k}'\,|\,n\rangle(\omega-\epsilon_n)^{-1}\langle n\,|\,\mathbf{k}\rangle. \qquad (6)$$

The equation for the Green's function is obtained as

$$G_{\mathbf{k}'\mathbf{k}}{}^{0}(\omega)=\delta_{\mathbf{k}\mathbf{k}'}(\omega-\epsilon_{\mathbf{k}})^{-1}+(V/N)(\omega-\epsilon_{\mathbf{k}})^{-1}\sum_{\mathbf{k}''}G_{\mathbf{k}''\mathbf{k}'}{}^{0}(\omega), \qquad (7)$$

which is solved as

$$G_{\mathbf{k}'\mathbf{k}}{}^{0}(\omega)=\delta_{\mathbf{k}\mathbf{k}'}(\omega-\epsilon_{\mathbf{k}})^{-1}+[t^{0}(\omega)/N](\omega-\epsilon_{\mathbf{k}})^{-1}(\omega-\epsilon_{\mathbf{k}'})^{-1}, \qquad (8)$$

where

$$t^{0}(\omega)=V[1-VF(\omega)]^{-1}, \qquad (9)$$

$$F(\omega)=N^{-1}\sum_{\mathbf{k}}(\omega-\epsilon_{\mathbf{k}})^{-1}=\int\rho(\omega')[\omega-\omega']^{-1}d\omega'. \qquad (10)$$

$\rho(\omega)$ denotes the density of states per atom per spin.

We now apply the transformation (3) to H' and obtain

$$H'=-(J/N)\sum_{nn'}c_n{}^{*}c_{n'}[(a_{n\uparrow}{}^{\dagger}a_{n'\uparrow}-a_{n\downarrow}{}^{\dagger}a_{n'\downarrow})S_z+a_{n\uparrow}{}^{\dagger}a_{n'\downarrow}S_{-}+a_{n\downarrow}{}^{\dagger}a_{n'\uparrow}S_{+}], \qquad (11)$$

where

$$c_n=\sum_{\mathbf{k}}\langle\mathbf{k}\,|\,n\rangle. \qquad (12)$$

From (4) and (11) we see that our problem reduces to an *s-d* problem with a factorizable exchange interaction $Jc_n{}^{*}c_{n'}$ but without an ordinary potential. We note from (11) that with each annihilation (creation) operator a_n ($a_n{}^{\dagger}$) is associated a factor c_n ($c_n{}^{*}$). Let us consider the perturbation expansion of the partition function considering (11) as a perturbation. We note that with each factor H' we create an electron and destroy another in such a way that an electron once created is eventually destroyed or vice versa. This is because we are interested in thermal averages. This means that the factor c_n appears in the form $|\,c_n\,|^2$ in the partition function. Thus if the rth-order term of the partition function in the absence of the ordinary scattering is given by

$$J^{r}\sum_{\mathbf{k}_1\cdots\mathbf{k}_r}H(\epsilon_{\mathbf{k}_1},\cdots,\epsilon_{\mathbf{k}_r}), \qquad (13)$$

the same quantity in the presence of it will be expressed by

$$J^{r}\sum_{n_1\cdots n_r}|\,c_{n_1}|^{2}\cdots|\,c_{n_r}|^{2}\,H(\epsilon_{n_1},\cdots,\epsilon_{n_r}). \qquad (14)$$

Let us consider the sum

$$\sum_{n}|\,c_n\,|^{2}\,H(\epsilon_n), \qquad (15)$$

where $H(\epsilon_n)$ is any function of ϵ_n. We note that $|\,c_n\,|^2=|\,\psi_n(0)\,|^2/|\,\psi_{\mathbf{k}}(0)\,|^2$, where $\psi_n=\sum_{\mathbf{k}}\langle\mathbf{k}\,|\,n\rangle\psi_{\mathbf{k}}$ and $\psi_{\mathbf{k}}$ is the plane-wave function. We define the density of states at the origin by

$$\begin{aligned}\sum_{\omega\le\epsilon_n\le\omega+d\omega}|\,c_n\,|^{2}&=-\pi^{-1}\mathrm{Im}\sum_{\mathbf{k}\mathbf{k}'}G_{\mathbf{k}\mathbf{k}'}{}^{0}(\omega+i\delta)\,d\omega\\&=-\pi^{-1}\mathrm{Im}\{NF(\omega+i\delta)/[1-V\\&\qquad\times F(\omega+i\delta)]\}\,d\omega\\&=N\rho(\omega)\,|\,1-VF(\omega+i\delta)\,|^{-2}d\omega,\end{aligned}$$

where use has been made of (8), (9), and (10). Then (15) is expressed as

$$N\int\rho(\omega)\,|\,1-VF(\omega+i\delta)\,|^{-2}\,H(\omega)\,d\omega. \qquad (16)$$

We see that the effect of the ordinary scattering is wholly incorporated into the factor $|\,1-VF(\omega+i\delta)\,|^{-2}$. We are interested in the case where this factor does not have a strong dependence on ω. Since the characteristic energy range of the low-temperature anomaly will be of order kT_K, this factor as well as $\rho(\omega)$ may be taken out of the integral sign with their values at the Fermi surface. Thus in order to translate the results on thermodynamic properties obtained in the absence of the ordinary scattering to those in its presence, we just replace $J\rho(\epsilon_F)$ by $J\rho(\epsilon_F)\,|\,1-VF(\epsilon_F+i\delta)\,|^{-2}$. There is no change of the sign of the exchange integral.

In the following, we use a widely adopted approximation

$$F(\omega+i\delta)=-i\pi\rho=\text{const.} \qquad (17)$$

Then we have

$$[1-VF(\omega+i\delta)]^{-1}=\cos\eta\;e^{i\eta}, \qquad (18)$$

where η is defined by $\tan\eta=-\pi V\rho$. We define the effective exchange integral $\tilde{J}$ by

$$\tilde{J}=J\cos^{2}\eta. \qquad (19)$$

This is the real part of what Fischer defines as the effective exchange interaction.

3. RESISTIVITY

We now discuss the resistivity, where the argument of the previous section is no longer valid. We use a different approach, namely, that which has been used by Hamann[5] and Falk and Fowler[6] recently. We use Nagaoka's truncation scheme for the electron Green's functions, convert the self-consistency equation into an integral equation for the t matrix, and solve it within an approximation which is valid for the leading logarithmic terms. We could pursue this program only with the approximation (17). Furthermore, we could do so to a lesser extent than Hamann could. However, the result obtained is sufficiently general to answer the question raised in the Introduction.

We go back to (1) and (2) and look at Green's

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[4] P. W. Anderson, Phys. Rev. **124**, 41 (1961).

[5] D. R. Hamann, Phys. Rev. **158**, 570 (1967).

[6] D. S. Falk and M. Fowler, Phys. Rev. **158**, 567 (1967).

functions:

$$G_{\mathrm{k'k}}(\omega)=\langle a_{\mathrm{k'}\uparrow}\mid a_{\mathrm{k}\uparrow}{}^{\dagger}\rangle, \tag{20}$$

$$\Gamma_{\mathrm{k'k}}(\omega)=\langle a_{\mathrm{k'}\uparrow}S_z+a_{\mathrm{k'}\downarrow}S_-\mid a_{\mathrm{k}\uparrow}{}^{\dagger}\rangle. \tag{21}$$

The equations of motion are written, after Nagaoka's truncation, as

$$(\omega-\epsilon_{\mathrm{k'}})G_{\mathrm{k'k}}+(J/N)\sum_{\lambda}\Gamma_{\lambda\mathrm{k}}=\delta_{\mathrm{kk'}}+(V/N)\sum_{\lambda}G_{\lambda\mathrm{k}}, \tag{22}$$

$$(\omega-\epsilon_{\mathrm{k}}{}')\Gamma_{\mathrm{k'k}}+(J/N)(2n_{\mathrm{k'}}-1)\sum_{\lambda}\Gamma_{\lambda\mathrm{k}} +(J/N)[S(S+1)-m_{\mathrm{k'}}]\sum_{\lambda}G_{\lambda\mathrm{k}}=(V/N)\sum_{\lambda}\Gamma_{\lambda\mathrm{k}}, \tag{23}$$

where

$$n_{\mathrm{k}}=\sum_{\lambda}\langle a_{\lambda\uparrow}{}^{\dagger}a_{\mathrm{k}\uparrow}\rangle, \tag{24}$$

$$m_{\mathrm{k}}=3\sum_{\lambda}\langle a_{\lambda\uparrow}{}^{\dagger}a_{\mathrm{k}\downarrow}S_-\rangle. \tag{25}$$

Equations (22) and (23) are solved as

$$G_{\mathrm{k'k}}(\omega)=\delta_{\mathrm{kk'}}(\omega-\epsilon_{\mathrm{k}})^{-1}+[t(\omega)/N](\omega-\epsilon_{\mathrm{k}})^{-1}(\omega-\epsilon_{\mathrm{k'}})^{-1}, \tag{26}$$

where

$$t(\omega)=t^0(\omega)-[1-VF(\omega)]^{-1} \times\frac{J_{\mathrm{eff}}{}^2(\omega)\Gamma(\omega)}{1+2J_{\mathrm{eff}}(\omega)G(\omega)+J_{\mathrm{eff}}{}^2(\omega)F(\omega)\Gamma(\omega)}, \tag{27}$$

$$J_{\mathrm{eff}}(\omega)=J[1-VF(\omega)]^{-1}, \tag{28}$$

$$G(\omega)=N^{-1}\sum_{\mathrm{k}}(n_{\mathrm{k}}-\tfrac{1}{2})/(\omega-\epsilon_{\mathrm{k}}), \tag{29}$$

$$\Gamma(\omega)=N^{-1}\sum_{\mathrm{k}}[m_{\mathrm{k}}-S(S+1)]/(\omega-\epsilon_{\mathrm{k}}). \tag{30}$$

Following Hamann, we express $G(\omega)$ and $\Gamma(\omega)$ in terms of $t(\omega)$. The results are

$$G(\omega)=G^0(\omega)+\mathfrak{F}_{\omega'}\left[\frac{F(\omega')-F(\omega)}{\omega-\omega'}F(\omega')t(\omega')\right], \tag{31}$$

$$\Gamma(\omega)=-2\mathfrak{F}_{\omega'}\left[\frac{F(\omega')-F(\omega)}{\omega-\omega'}\frac{\Delta t(\omega')}{J_{\mathrm{eff}}(\omega')}\right] -S(S+1)F(\omega), \tag{32}$$

where

$$G^0(\omega)=N^{-1}\sum_{\mathrm{k}}[f(\epsilon_{\mathrm{k}})-\tfrac{1}{2}]/(\omega-\epsilon_{\mathrm{k}}), \tag{33}$$

$$\Delta t=t-t^0. \tag{34}$$

f denotes the Fermi distribution function. The functional notation $\mathfrak{F}$ is defined by

$$\mathfrak{F}_{\omega'}[G_{AB}(\omega')]=(i/2\pi)\int_{-\infty}^{\infty}d\omega'\quad f(\omega') \times[G_{AB}(\omega'+i\delta)-G_{AB}(\omega'-i\delta)]. \tag{35}$$

We process the functional operations in the same way as Hamann did to the point that G and Γ are now expressed by

$$G(\omega)=G^0(\omega)+i\pi\rho^2\int t^*(\omega'+i\delta) \times[f(\omega')-\tfrac{1}{2}]/(\omega-\omega'+i\delta)d\omega', \tag{36}$$

$$\Gamma(\omega)=-S(S+1)F(\omega)-(2\rho/J_{\mathrm{eff}}{}^*)\int\Delta t^*(\omega'+i\delta) \times[f(\omega')-\tfrac{1}{2}]/(\omega-\omega'+i\delta)d\omega', \tag{37}$$

where we assumed $J_{\mathrm{eff}}(\omega)$ is a constant.

Using these results in (27) and using (17), (18), (19), (33), and (34), we obtain the integral equation for Δt:

$$\Delta t(\omega+i\delta)=\frac{-i\pi J^2\rho S(S+1)\cos\eta e^{3i\eta}+2J\rho e^{4i\eta}\displaystyle\int\frac{\Delta t^*(\omega'+i\delta)}{\omega-\omega'+i\delta}[f(\omega')-\tfrac{1}{2}]d\omega'}{\cos^{-2}\eta+\pi^2J^2\rho^2S(S+1)e^{2i\eta}+2J\rho\displaystyle\int[1+2\pi i\rho e^{2i\eta}\Delta t^*(\omega'+i\delta)]\frac{f(\omega')-\frac{1}{2}}{\omega-\omega'+i\delta}d\omega'}. \tag{38}$$

This becomes identical to Hamann's (2.29) when $\eta=0$. At this point it is no longer possible to pursue the scheme further in the same way as Hamann did. The next step which he took can be carried through only when the symmetry relation

$$\Delta t(\omega+i\delta)=-\Delta t^*(-\omega+i\delta) \tag{39}$$

is satisfied [see Hamann's (2.32)]. In our case, (39) no longer holds because of the phase factors of η in (38). Since (39) would imply that the imaginary part of Δt is an even function of ω and since giant thermoelectric power is obtained only when it has an odd part, the fact that (39) no longer holds for $\eta\neq0$ is nothing but another expression of our previous result[7] that giant thermoelectric power is a result of a cooperation of V and J.

We can solve (38) if we restrict ourselves to the leading logarithmic terms. The leading logarithmic terms of Δt will turn out to be $\sum_n C_nJ^{n+2}\ln^n\omega$. The part of the

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[7] J. Kondo, Progr. Theoret. Phys. (Kyoto) **34**, 372 (1965).

integral in the denominator which contains Δt^* will then be at most $\sum_n d_n J^{n+3}\ln^{n+1}\omega$, which will contribute $\sum_n b_n J^{n+4}\ln^n\omega$ to Δt. Thus within the approximation of keeping the leading logarithmic terms, the mentioned part as well as the second term of the denominator may be neglected. This is consistent with the later approximation (45) which is also valid for the leading logarithmic terms.

Making a transformation

$$\Delta t=\phi e^{2i\eta}-i\pi J^2\rho S(S+1)\cos^3\eta e^{3i\eta}, \qquad (40)$$

we obtain the equation for ϕ:

$$\phi(\omega+i\delta)=\left\{2\tilde{J}\rho\int\frac{\phi^*(\omega'+i\delta)}{\omega-\omega'+i\delta}\left[f(\omega')-\tfrac{1}{2}\right]d\omega'+4\pi i\tilde{J}^3\rho^2S(S+1)\int\frac{f(\omega')-\frac{1}{2}}{\omega-\omega'+i\delta}d\omega'\right\}\Bigg/\left\{1+2\tilde{J}\rho\int\frac{f(\omega')-\frac{1}{2}}{\omega-\omega'+i\delta}d\omega'\right\}. \qquad (41)$$

It is now possible to show

$$\phi(\omega+i\delta)=-\phi^*(-\omega+i\delta) \qquad (42)$$

from (41). We change the variable from ω to y by

$$y=-\int\frac{f(\omega')-\frac{1}{2}}{\omega-\omega'+i\delta}d\omega'. \qquad (43)$$

We define a function $\Phi(y)$ of y by

$$\Phi(y)=\int\frac{\phi^*(\omega'+i\delta)}{\omega-\omega'+i\delta}\left[f(\omega')-\tfrac{1}{2}\right]d\omega'. \qquad (44)$$

Correct up to the leading logarithmic terms we have

$$d\Phi(y)/dy=\phi(y), \qquad (45)$$

where use has been made of (42) [see Hamann's (3.14)]. Solving (41) for Φ and differentiating both sides with respect to y, we obtain

$$d\phi/[\phi-\pi i\tilde{J}^2\rho S(S+1)]=4\tilde{J}\rho dy/(1-2\tilde{J}\rho y). \qquad (46)$$

This differential equation is easily integrated as

$$\phi=\pi i\tilde{J}^2\rho S(S+1)[1-(1-2\tilde{J}\rho y)^{-2}], \qquad (47)$$

which satisfies the boundary condition that the first term of the expansion should be $-4\pi i\tilde{J}^3\rho^2S(S+1)y$, as easily seen from (41). Using (40) and (34) we finally obtain

$$t=V[1-VF(\omega)]^{-1}-\pi iJ^2\rho S(S+1)\cos^3\eta e^{3i\eta}$$
$$+\pi i\tilde{J}^2\rho S(S+1)e^{2i\eta}[1-(1-2\tilde{J}\rho y)^{-2}]. \qquad (48)$$

The relaxation time of the conduction electron is expressed by

$$[2\tau(\omega)]^{-1}=-cN\ \mathrm{Im}t(\omega+i\delta),$$

where c is the fraction of the impurities. The resistivity is obtained as

$$R=\frac{3c}{\pi e^2v_F{}^2\rho^2\hbar}\frac{\Omega}{N}\{\sin^2\eta-2\pi^2J^2\rho^2S(S+1)\cos^4\eta\sin^2\eta$$
$$+\pi^2J^2\rho^2S(S+1)\cos^4\eta\cos2\eta[1-2\tilde{J}\rho\ln(T/D)]^{-2}\}, \qquad (49)$$

where Ω is the volume of the metal and D is a cut-off energy. The first two terms of the expansion of (49) (J^2 and J^3 terms) have been obtained by Fisher. The effect of the ordinary potential on the nth-order logarithmic term is seen to replace J^{n+2} by $\tilde{J}^{n+2}$ and multiply by $\cos2\eta$. The temperature dependence of (49) is such that the resistivity increases (decreases) with decreasing temperature when $J\cos2\eta<0$ (>0). For negative J, Eq. (49) is no longer valid in the region $T\lesssim T_K\equiv De^{1/2\tilde{J}\rho}$, where we will have to look for another solution.

4. DISCUSSION

We have seen that the effect of the ordinary scattering on the exchange scattering is not to change the sign of the effective exchange integral. Thus the low-temperature singularity will always be associated with a negative exchange integral.

However, the slope of the resistivity-versus-T curve can change its sign when η is greater than $\frac{1}{4}\pi$, as first noted by Fischer. This result seems to be particularly important in view of the recent experimental results on Rh-Fe alloys. The resistivity of this alloy decreases with decreasing temperature.[8] This result was interpreted as arising from a positive exchange interaction.[1] However the recent measurement on the susceptibility made by Knapp[9] shows that the magnetic moment of the iron atom is considerably smaller than what is expected for a free spin. This is a feature generally accepted as arising from Nagaoka-type spin compensation associated with a negative exchange interaction. This apparent contradiction may be reconciled by the results obtained in this paper, because the above experimental results[10] are consistent with a negative exchange interaction and η greater than $\frac{1}{4}\pi$.

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[8] R. B. Coles, Phys. Letters **8**, 243 (1964).

[9] G. Knapp, J. Appl. Phys. **38**, 1267 (1967).

[10] Recently G. Knapp [Phys. Letters **25A**, 114 (1967)] has proposed an alternative explanation of these results.