Self-Consistent Treatment of the Kondo Effect*

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(Received 19 December 1966)

We investigate the scattering of conduction electrons by magnetic impurities for arbitrary exchange and normal interaction using Nagaoka's self-consistent-field approach. The corresponding self-consistent equations are solved by iteration in the limit of high and low temperatures. One finds for an extended exchange interaction a series of Suhl-Abrikosov resonances corresponding to different partial waves, which determine the scattering at low temperatures. A part of these resonances is affected by the normal interaction. In the special case of a δ-function exchange interaction corresponding to a single resonance, the exchange component of the resistivity goes to zero as $T\rightarrow 0$. For temperatures high compared to the temperature where the first resonance becomes possible, we find generalizations for Kondo's expressions for the resistivity and thermopower. However, the exchange and normal interactions no longer give independent contributions to the resistivity. One finds for a sufficiently large normal interaction a resistivity minimum for ferromagnetic exchange coupling.

I. INTRODUCTION

THE first consistent explanation for the anomalous temperature dependence of the resistivity $\rho(T)$ and for the giant thermoelectric power $S(T)$ observed in some metals containing a small concentration of magnetic impurities at low temperatures has been given by Kondo.[1,2] He showed that the spin-flip or virtual spin-flip scattering of a conduction-electron spin by a localized impurity spin, calculated in third order in the exchange coupling, gives rise to a term proportional to $\ln T$ in the resistivity. He also showed that one obtains the correct order of magnitude for the thermopower if one includes terms of the order VJ^3 in the perturbation series, where V and J correspond to the normal and exchange interaction, respectively.

In the meantime the scattering of the conduction electrons by a small concentration of magnetic impurities at low temperatures has been investigated by several authors.[3–11] Abrikosov[3] used the field-theoretic perturbation technique of summing up the most divergent terms within each order of the (contact) exchange interaction J. This summation results for antiferromagnetic coupling ($J<0$) in the replacement of Kondo's singularity in the transition probability by a scattering resonance corresponding to a maximum in the exchange part $\rho_{\text{ex}}(T)$ of the resistivity at a temperature T_c. The resistivity $\rho_{\text{ex}}(T)$ goes to zero as $(\ln T)^{-2}$ as $T\rightarrow 0$ for both signs of the exchange interaction J. Very similar results have been obtained by Suhl,[4] using nonperturbative scattering theory and solving the corresponding nonlinear integral equations for one-particle intermediate states. However, there appeared for $T<T_c$ and $J<0$ inadmissible complex poles in the spin-flip scattering amplitude. By making a proper analytic continuation in temperature, Suhl and Wong[5] found a solution free from such difficulties, leading to a finite exchange resistivity $\rho_{\text{ex}}(T)$ as $T\rightarrow 0$ without any resonance behavior. They also found, in contradiction to Abrikosov, that the resistivities due to the exchange interaction J and normal interaction V do not simply add. The coupling between exchange and nonexchange scattering turned out to be essential for the removal of the spurious poles.

These results do not seem to be in agreement with results obtained by Nagaoka,[6] who used a truncated set of self-consistent Green's-function equations. Nagaoka suggested for $T<T_c$ and $J<0$ a quasibound state around the impurity spin and found a maximum of $\rho_{\text{ex}}(T)$ at $T=0$. In this paper we reinvestigate his self-consistent equations. We generalize them for arbitrary exchange and normal interaction, and solve them by iteration for both signs of the exchange interaction in the limit of high and low temperatures. We find in contrast to Nagaoka, for a contact exchange interaction $J<0$ and for a weak normal interaction (corresponding to the resistivity ρ_n), Abrikosov's resonance and $\rho=\rho_n+\rho_{\text{ex}}(T)$ with $\rho_{\text{ex}}(T)\rightarrow 0$ as $(\ln T)^{-2}$ as $T\rightarrow 0$. The same asymptotic behavior holds for ferromagnetic exchange coupling ($J>0$).

In the case of an extended exchange interaction $J_{kk'}<0$ (which is assumed to depend only on the wavevector difference $|\mathbf{k}-\mathbf{k}'|$) a series of resonances appear below critical temperatures $T_{c,l}$, defined by

$$J_l g(0,T_{c,l})=1\,, \tag{1}$$

where J_l is the l component in the expansion of $J_{kk'}$ into Legendre polynomials, and

$$g(\omega,T)\equiv N^{-1}\sum_q \frac{f_q}{\epsilon_q-\omega}=\int_0^\infty \rho(\epsilon_q)P\frac{f_q}{\epsilon_q-\omega}d\epsilon_q\,. \tag{2}$$

* Work supported in part by the U. S. Air Force Office of Scientific Research under Grant Nos. AF 328 and AF 803 and the Advanced Research Projects Agency, Contract No. SD-131.

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Here, N is the number of atoms in the crystal, f_q the Fermi distribution function, ϵ_q the one-electron energy, and P means principle part. All these resonances contribute at sufficiently low temperatures to $\rho_{\text{ex}}(T)$, since Eq. (1) with $g(0,T)\propto \ln T$ can be fulfilled for arbitrary small components J_l. The behavior of the resistivity $\rho_{\text{ex}}(T)$ at low temperatures is then determined by the l dependence of J_l or by the range and form of the exchange interaction. However, we should like to emphasize that for $T\ll T_{c,l}$ we could solve the self-consistent equations only for energies $\omega\ll\omega_{c,l}$ and $\omega\gg\omega_{c,l}$, where $\omega_{c,l}$ corresponds to $T_{c,l}$, and no simple solution seems to exist near $\omega_{c,l}$. Therefore, we cannot exclude the possible existence of inadmissible poles in the self energy of the electron Green's function near $\omega_{c,l}$, similar to those found by Suhl.[4]

We include in our calculations the normal interaction V between conduction electrons and magnetic impurities to arbitrary order. This is important since many systems exhibiting a resistivity minimum show also a resonance in the normal scattering explained first by Friedel.[12] In agreement with Ref. 5, it turns out that the resistivities ρ_n and ρ_{ex} for strong normal scattering do not simply add, as suggested by Abrikosov.[3]

In Sec. II the self-consistent equations are derived for arbitrary exchange and normal interactions. The basic approximation involved is the factorization of terms in the second Green's-function equation of motion, and we make no attempt to justify it. In Secs. III and IV we solve the self-consistent equations by iteration in the limits $T\gg T_{c,l}$ and $T\ll T_{c,l}$, and derive expressions for the resistivity and for the thermoelectric power.

II. SELF-CONSISTENT EQUATIONS

A. Derivation

We consider a system consisting of independent conduction electrons scattered by a small concentration c of noninteracting impurities with a localized magnetic moment. The spin-dependent interaction is described by the usual exchange term which is proportional to the product $\mathbf{S}\cdot\mathbf{S}_e$ of the impurity-spin $\mathbf{S}$ and of the conduction-electron-spin $\mathbf{S}_e$.

We can restrict ourselves in the limit $c\to 0$ to the scattering by a single impurity. The corresponding Hamiltonian is[13]

$$H=\sum_{ks}\epsilon_k c_{ks}^\dagger c_{ks}+N^{-1}\sum_{kk's}V_{kk'}c_{ks}^\dagger c_{k's}-(2N)^{-1}\sum_{kk'}J_{kk'}[(c_{k\uparrow}^\dagger c_{k'\uparrow}-c_{k\downarrow}^\dagger c_{k'\downarrow})S_z+c_{k\uparrow}^\dagger c_{k'\downarrow}S_-+c_{k\downarrow}^\dagger c_{k'\uparrow}S_+], \quad (3)$$

where $V_{kk'}$ and $J_{kk'}$ are the matrix elements for normal and exchange interactions, respectively, c_{ks} and $c_{ks}^\dagger$ are annihilation and creation operators for a conduction electron with wave vector k and z component of spin s, and S_z, $S_\pm$ are components of the impurity spin operator.

Following Nagaoka (henceforth referred to as N) we define a retarded time-dependent single-particle Green's function[14] for $s=s'=\uparrow$ (note that our notation differs slightly from N),

$$G_{kk'}(t)=-i\langle[c_{k\uparrow}(t),c_{k'\uparrow}^\dagger(0)]_+\rangle\Theta(t). \quad (4)$$

The average is taken over a grand-canonical ensemble and $\Theta(t)$ is the step function. The equation of motion for the Fourier transform

$$G_{kk'}(\omega)=\int_{-\infty}^{\infty}e^{i\omega t}G_{kk'}(t)dt \quad (5)$$

is given by (we set $\hbar=1$)

$$(\omega-\epsilon_k)G_{kk'}(\omega)-N^{-1}\sum_q V_{kq}G_{qk'}(\omega)+(2N)^{-1}\sum J_{kq}\Gamma_{qk'}(\omega)=\delta_{kk'}. \quad (6)$$

Here we introduced the Fourier transform of a second Green's function:

$$\Gamma_{kk'}(t)=-i\times\langle[(c_{k\uparrow}(t)S_z(t)+c_{k\downarrow}(t)S_-(t)),c_{k'\uparrow}^\dagger(0)]_+\rangle\Theta(t), \quad (7)$$

which obeys the equation of motion

$$\begin{aligned}(\omega-\epsilon_k)\Gamma_{kk'}(\omega)&-N^{-1}\sum_q(V_{kq}-\tfrac{1}{2}J_{kq})\Gamma_{qk'}(\omega)\\&+(2N)^{-1}S(S+1)\sum_q J_{kq}G_{qk'}(\omega)+(2N)^{-1}\sum_{qq'}J_{qq'}\\&\times(-\langle c_{k\uparrow}c_{q\uparrow}^\dagger c_{q'\downarrow}S_-|c_{k'\uparrow}^\dagger\rangle+\langle c_{k\uparrow}c_{q\downarrow}^\dagger c_{q'\uparrow}S_+|c_{k'\uparrow}^\dagger\rangle\\&+\langle c_{k\downarrow}c_{q\uparrow}^\dagger c_{q'\uparrow}S_-|c_{k'\uparrow}^\dagger\rangle-\langle c_{k\downarrow}c_{q\downarrow}^\dagger c_{q'\downarrow}S_-|c_{k'\uparrow}^\dagger\rangle\\&-2\langle c_{k\downarrow}c_{q\downarrow}^\dagger c_{q'\uparrow}S_z|c_{k'\uparrow}^\dagger\rangle)=0. \quad (8)\end{aligned}$$

We used the notation $\langle A|B\rangle$ for the Fourier transform of the quantity

$$-i\langle[A(t),B(0)]_+\rangle\Theta(t)$$

and we used the spin commutators

$$[S_z,S_\pm]_-=\pm S_\pm,\quad [S_+,S_-]_-=2S_z. \quad (9)$$

Following N we decompose now the higher-order Green's functions into products of one-particle Green's functions and the thermal average of the remaining operators, taking into account particle conservation and conservation of total spin; for instance

$$\langle c_{k\uparrow}c_{q\uparrow}^\dagger c_{q'\downarrow}S_-|c_{k'\uparrow}^\dagger\rangle=\langle c_{k\uparrow}c_{q\uparrow}^\dagger\rangle\langle c_{q'\downarrow}S_-|c_{k'\uparrow}^\dagger\rangle+\langle c_{q\uparrow}^\dagger c_{q'\downarrow}S_-\rangle\langle c_{k\uparrow}|c_{k'\uparrow}^\dagger\rangle. \quad (10)$$

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In the absence of a magnetic field all thermal averages are independent of the direction of the impurity spin in space:

$$\langle S_z\rangle=\langle S_+\rangle=\langle S_-\rangle=0\,, \tag{11}$$

$$\langle c_{k\uparrow}{}^\dagger c_{k'\uparrow}\rangle=\langle c_{k\downarrow}{}^\dagger c_{k'\downarrow}\rangle\,, \tag{12}$$

$$\langle c_{k\uparrow}{}^\dagger c_{k'\downarrow}S_-\rangle=\langle c_{k\downarrow}{}^\dagger c_{k'\uparrow}S_+\rangle =2\langle c_{k\uparrow}{}^\dagger c_{k'\uparrow}S_z\rangle=-2\langle c_{k\downarrow}{}^\dagger c_{k'\downarrow}S_z\rangle\,. \tag{13}$$

From Eqs. (9) to (13) follows the second equation of motion:

$$(\omega-\epsilon_k)\Gamma_{kk'}(\omega)-N^{-1}\sum_q V_{kq}\Gamma_{qk'}(\omega) +(2N)^{-1}\sum_{qq'}(2n_{kq}-\delta_{kq})J_{qq'}\Gamma_{q'k'}(\omega) -(2N)^{-1}\sum_{qq'}[m_{kq}-S(S+1)\delta_{kq}]J_{qq'}G_{q'k'}(\omega)=0, \tag{14}$$

with

$$n_{kk'}=\langle c_{k'\uparrow}{}^\dagger c_{k\uparrow}\rangle\,, \tag{15}$$

$$m_{kk'}=3\langle c_{k'\uparrow}{}^\dagger c_{k\downarrow}S_-\rangle\,. \tag{16}$$

The thermal average $\langle BA\rangle$ of two arbitrary Fermi operators is connected with the corresponding Green's function $\langle A\,|\,B\rangle$ by[14] (μ is the chemical potential, k_B the Boltzmann constant)

$$\langle B\,|\,A\rangle=-\pi^{-1}\int_{-\infty}^{\infty}d\omega\, f(\omega)\,\mathrm{Im}\langle A\,|\,B\rangle\,,$$

$$f(\omega)=(e^{\beta(\omega-\mu)}+1)^{-1},\quad \beta=(k_BT)^{-1}, \tag{17}$$

and therefore

$$n_{kk'}=-\pi^{-1}\int_{-\infty}^{\infty}d\omega f(\omega)\,\mathrm{Im}G_{kk'}(\omega)\,, \tag{18}$$

$$m_{kk'}=-2\pi^{-1}\int_{-\infty}^{\infty}d\omega f(\omega)\,\mathrm{Im}\Gamma_{kk'}(\omega)\,. \tag{19}$$

Equations (6), (14), (18), and (19) form a complete set of self-consistent equations. They are linear besides Eq. (14) which is nonlinear due to the truncation of the hierarchy of Green's functions.

B. Effective Exchange Interaction

We can simplify Eqs. (6) and (14) by introducing the t matrix for normal scattering. The matrix elements $t_{kk'}$ can be expressed in terms of the phase shifts for the normal scattering which we assume to be known. We write

$$G_{kk'}(\omega)=G_{kk'}{}^{(0)}(\omega)+G_{kk'}{}^{(1)}(\omega)\,, \tag{20}$$

where $G_{kk'}{}^{(0)}(\omega)$ satisfies the equation for $J_{kk'}\equiv 0$.

$$G_{kk'}{}^{(0)}(\omega)-\frac{1}{N}G_k{}^{(0)}(\omega)\sum_q V_{kq}G_{qk'}{}^{(0)}(\omega) =G_k{}^{(0)}(\omega)\delta_{kk'}\,, \tag{21}$$

with the retarded Green's function for free electrons

$$G_k{}^{(0)}(\omega)=(\omega-\epsilon_k+i\eta)^{-1},\quad \eta=0_+\,. \tag{22}$$

The t matrix for the scattering by a single impurity is defined by[15]

$$t_{k'k}=\langle k'\,|\,V\,|\,k^{(+)}\rangle\,, \tag{23}$$

where $|k^{(+)}\rangle$ satisfies the Schrödinger equation for a single-electron

$$|k^{(+)}\rangle=|k\rangle+(\omega-h+i\eta)^{-1}V\,|\,k^{(+)}\rangle\,, \tag{24}$$

(with $|k\rangle$ the incoming plane wave, h the single-particle Hamiltonian). Using Eq. (23) and the completeness of the functions $|k\rangle$, we find

$$t_{kk'}=V_{kk'}+N^{-1}\sum_q V_{kq}G_q{}^{(0)}(\omega)t_{qk'}\,. \tag{25}$$

Equation (21) may now be written as

$$G_{kk'}{}^{(0)}(\omega)=G_k{}^{(0)}(\omega)\delta_{kk'}+N^{-1}G_k{}^{(0)}(\omega)t_{kk'}G_{k'}{}^{(0)}(\omega)\,. \tag{26}$$

We insert now Eq. (20) into (6) and make use of Eqs. (21) and (22):

$$G_{kk'}{}^{(1)}(\omega)-N^{-1}G_k{}^{(0)}\sum_q V_{kq}G_{qk'}{}^{(1)}(\omega) =-(2N)^{-1}G_k{}^{(0)}\sum_q J_{kq}\Gamma_{qk'}(\omega)\,. \tag{27}$$

This can be simplified by iteration with the right-hand side of the equation as the inhomogeneous term. We obtain a sum of terms which may be expressed by the t-matrix Eq. (25):

$$G_{kk'}{}^{(1)}(\omega)=-(2N)^{-1}G_k{}^{(0)}(\omega) \times\sum_q(J_{kq}+N^{-1}\sum_{q'}t_{kq'}G_{q'}{}^{(0)}(\omega)J_{q'q})\Gamma_{qk'}(\omega)\,, \tag{28}$$

or, introducing the effective exchange interaction

$$J_{k'}{}^{\mathrm{eff}}=J_{kk'}+N^{-1}\sum_q t_{kq}G_q{}^{(0)}(\omega)J_{qk'}\,, \tag{29}$$

$$G_{kk'}{}^{(1)}(\omega)=-(2N)^{-1}G_k{}^{(0)}(\omega)\sum_q J_{kq}{}^{\mathrm{eff}}\Gamma_{qk'}(\omega)\,. \tag{30}$$

The effective exchange interaction $J_{kk'}{}^{\mathrm{eff}}$ is complex even in Born approximation for the normal scattering ($t_{kk'}=V_{kk'}$ real). We will show that it is the imaginary part of $J_{kk'}{}^{\mathrm{eff}}$ which causes part of the giant thermopower. In this sense both explanations of Kondo for the giant thermopower (compare the second footnote in Ref. 2, p. 373) are equivalent. However, the Green's function $G_{kk'}{}^{(1)}(\omega)$ Eq. (30) is coupled to $\Gamma_{kk'}{}^{(1)}(\omega)$, which also depends on the normal scattering. We will see later that it is impossible to absorb the normal scattering entirely into an effective exchange interaction.

[15] V. Ambegaokar, in *Astrophysics and the Many-Body Problem*, edited by E. N. Parker, J. S. Goldstein, A. A. Maradudin, and V. Ambegaokar (W. A. Benjamin, Inc., New York, 1963), pp. 321, 381.

Equation (14) for the Green's function $\Gamma_{kk'}(\omega)$ can be iterated in a similar way. We write it in the form

$$\Gamma_{kk'}(\omega)-N^{-1}G_k^{(0)}(\omega)\sum_q V_{kq}\Gamma_{qk'}(\omega)=G_k^{(0)}(\omega)\alpha_{kk'}(\omega)\,, \quad (31)$$

$$\alpha_{kk'}(\omega)=-(2N)^{-1}\sum_{qq'}(2n_{kq}-\delta_{kq})J_{qq'}\Gamma_{q'k'}(\omega)+(2N)^{-1}\sum_{qq'}(m_{kq}-S(S+1)\delta_{kq})J_{qq'}G_{q'k'}(\omega)\,, \quad (32)$$

and obtain

$$\Gamma_{kk'}(\omega)=G_k^{(0)}(\omega)\times[\alpha_{kk'}(\omega)+N^{-1}\sum_q t_{kq}G_q^{(0)}(\omega)\alpha_{qk'}(\omega)]. \quad (33)$$

C. Partial Waves

The scattering of conduction electrons by a single impurity depends only on the product $\mathbf{k}\cdot\mathbf{k}'$ of the wave vectors of the incoming and scattered waves. We expand, therefore, all quantities into Legendre polynomials $P_l(\cos\Theta_{kk'})$:

$$G_{kk'}(\omega)=G(\mathbf{k}\cdot\mathbf{k}',\omega)=\sum_{l=0}^{\infty}(2l+1)G_l(\epsilon_k,\epsilon_{k'},\omega)P_l(\cos\Theta_{kk'})\,, \quad (34a)$$

$$J_{kk'}=\sum_{l=0}^{\infty}(2l+1)J_l(\epsilon_k,\epsilon_{k'})P_l(\cos\Theta_{kk'})\,, \quad (34b)$$

and similar expressions for $\Gamma_{kk'}(\omega)$, $t_{kk'}$, $J_{kk'}^{\text{eff}}$, $n_{kk'}$, $m_{kk'}$. Replacing the sums by integrals

$$N^{-1}\sum_k\rightarrow(4\pi)^{-1}\int d\Omega_k\int_0^{\infty}\rho(\epsilon_k)d\epsilon_k\,, \quad (35)$$

and using the relation

$$(2l+1)P_l(\cos\Theta_{kk'})=4\pi\sum_m Y_{lm}(\Theta_k,\varphi_k)Y_{lm}^*(\Theta_{k'},\varphi_{k'})$$

between the spherical harmonics $Y_{lm}(\Theta,\varphi)$ and the Legendre polynomials, we can easily perform all angle integrations in Eqs. (29), (30), (32), and (33). All integrals over the energy ϵ_q contain free-electron Green's functions $G_q^{(0)}(\omega)$ which have a sharp peak at $\epsilon_q=\omega$. We therefore replace $t_l(\epsilon_k,\epsilon_q)$, $J_l^{\text{eff}}(\epsilon_k,\epsilon_q)$, and $J_l(\epsilon_k,\epsilon_q)$ by $t_l(\epsilon_k,\omega)$, etc. Since $\epsilon_k=\epsilon_{k'}$ and since the Green's functions $G_{kk'}(\omega)$ and $\Gamma_{kk'}(\omega)$ contain also free electron Green's functions $G_k^{(0)}(\omega)$, we replace $t_l(\epsilon_k,\omega)$ by $t_l(\omega)$ etc. and finally drop the argument ω.

We obtain from Eqs. (20), (29), and (30)

$$J_l^{\text{eff}}=J_l[1+F(\omega)t_l]\,, \quad (36)$$

$$G_l(\epsilon_k,\epsilon_{k'},\omega)=G_l^{(0)}(\epsilon_k,\epsilon_{k'},\omega)-(2N)^{-1}G_k^{(0)}(\omega)J_l^{\text{eff}}\bar{\Gamma}_l(\epsilon_{k'},\omega)\,, \quad (37)$$

with

$$F(\omega)\equiv\int_0^{\infty}\rho(\epsilon_k)G_k^{(0)}(\omega)d\epsilon_k\,, \quad (38)$$

$$\bar{\Gamma}_l(\epsilon_k,\omega)\equiv N\int_0^{\infty}\rho(\epsilon)\Gamma_l(\epsilon,\epsilon_k,\omega)d\epsilon. \quad (39)$$

We define in a similar way

$$\bar{G}_l(\epsilon_k,\omega)\equiv N\int_0^{\infty}\rho(\epsilon)G_l(\epsilon,\epsilon_k,\omega)d\epsilon\,, \quad (40)$$

$$\tilde{n}_l(\epsilon_k)\equiv N\int_0^{\infty}\rho(\epsilon)n_l(\epsilon_k,\epsilon)d\epsilon\,, \quad (41)$$

$$\tilde{m}_l(\epsilon_k)\equiv N\int_0^{\infty}\rho(\epsilon)m_l(\epsilon_k,\epsilon)d\epsilon\,, \quad (42)$$

$$g_l(\omega)\equiv\int_0^{\infty}\rho(\epsilon_q)G_q^{(0)}(\omega)(\tilde{n}_l(\epsilon_q)-\tfrac{1}{2})d\epsilon_q\,, \quad (43)$$

$$\gamma_l(\omega)\equiv\int_0^{\infty}\rho(\epsilon_q)G_q^{(0)}(\omega)[\tilde{m}_l(\epsilon_q)-S(S+1)]d\epsilon_q. \quad (44)$$

[Note that n_{kq} and m_{kq} are integrated over the second variable, whereas $\Gamma_{qk'}(\omega)$ and $G_{qk'}(\omega)$ are integrated over the first ones.] We obtain with these definitions for the coefficients of $\Gamma_{kk'}(\omega)$:

$$\Gamma_l(\epsilon_k,\epsilon_{k'},\omega)=(2N)^{-1}G_k^{(0)}(\omega)\times J_l\{[\tilde{m}_l(\epsilon_k)-S(S+1)+t_l\gamma_l(\omega)]\bar{G}_l(\epsilon_{k'},\omega)-[2\tilde{n}_l(\epsilon_k)-1+2t_lg_l(\omega)]\bar{\Gamma}_l(\epsilon_{k'},\omega)\}. \quad (45)$$

For $V_{kk'}=0$, $J_l^{\text{eff}}=J_l=J_0\delta_{l0}$, and $S=\frac{1}{2}$, Eqs. (37) and (45) correspond to Eqs. (2.8) and (2.14) of Nagaoka. For free electrons

$$n_{kk'}^{(0)}=f_k\delta_{kk'} \quad (46)$$

and with $P_l(0)=1$ for all l

$$n_l^{(0)}(\epsilon_k,\epsilon_{k'})=(4\pi)^{-1}\int d\Omega n_{kk'}^{(0)}P_l(\cos\Theta_{kk'})\,, \quad (47)$$

$$\tilde{n}_l^{(0)}(\epsilon_k)=\sum_{k'}n_{kk'}^{(0)}P_l(\cos\Theta_{kk'})=f_k\,, \quad (48)$$

and similarly for the Green's function for normal scattering only

$$\bar{G}_l^{(0)}(\epsilon_k,\omega)=G_k^{(0)}(\omega)[1+F(\omega)t_l]. \quad (49)$$

Multiplying Eqs. (37) and (45) by $N\rho(\epsilon_k)$ and integrating over ϵ_k we obtain

$$\bar{G}_l(\epsilon_k,\omega)=G_k^{(0)}(\omega)[1+F(\omega)t_l]-\tfrac{1}{2}J_l^{\text{eff}}F(\omega)\bar{\Gamma}_l(\epsilon_k,\omega). \quad (50)$$

$$\bar{\Gamma}_l(\epsilon_k,\omega)=J_l^{\text{eff}}[\tfrac{1}{2}\gamma_l(\omega)\bar{G}_l(\epsilon_k,\omega)-g_l(\omega)\bar{\Gamma}_l(\epsilon_k,\omega)]. \quad (51)$$

We solve Eqs. (50) and (51) for $\tilde{G}_l(\epsilon_k,\omega)$ and $\tilde{\Gamma}_l(\epsilon_k,\omega)$ and insert these into (37) and (45):

$$G_{kk'}(\omega)=G_{kk'}^{(0)}(\omega)-(4N)^{-1}G_k^{(0)}(\omega)G_{k'}^{(0)}(\omega)\times\sum_{l=0}^{\infty}(2l+1)P_l(\cos\Theta_{kk'})\times(J_l^{\text{eff}})^2[1+F(\omega)t_l]\gamma_l(\omega)\times[1+J_l^{\text{eff}}g_l(\omega)+\tfrac{1}{4}(J_l^{\text{eff}})^2F(\omega)\gamma_l(\omega)]^{-1}, \quad (52)$$

$$\Gamma_{kk'}(\omega)=(2N)^{-1}G_k^{(0)}(\omega)G_{k'}^{(0)}(\omega)\times\sum_{l=0}^{\infty}(2l+1)P_l(\cos\Theta_{kk'})J_l^{\text{eff}}\times\{[\tilde{m}_l(\epsilon_k)-S(S+1)+t_l\gamma_l(\omega)](1+J_l^{\text{eff}}g_l(\omega))-[\tilde{n}_l(\epsilon_k)-\tfrac{1}{2}+t_lg_l(\omega)]J_l^{\text{eff}}\gamma_l(\omega)\}\times[1+J_l^{\text{eff}}g_l(\omega)+\tfrac{1}{4}(J_l^{\text{eff}})^2F(\omega)\gamma_l(\omega)]^{-1}. \quad (53)$$

The functions $\tilde{n}_l(\epsilon_k)$ and $\tilde{m}_l(\epsilon_k)$ and the corresponding Green's functions $\tilde{G}_l(\epsilon_k,\omega)$ and $\tilde{\Gamma}_l(\epsilon_k,\omega)$ have to be determined self-consistently. We find from Eqs. (52) and (53)

$$\tilde{G}_l(\epsilon_k,\omega)\equiv N\int_0^{\infty}\rho(\epsilon)G_l(\epsilon_k,\epsilon,\omega)d\epsilon=\tilde{G}_l(\epsilon_k,\omega)=G_k^{(0)}(\omega)[1+F(\omega)t_l][1+J_l^{\text{eff}}g_l(\omega)]\times[1+J_l^{\text{eff}}g_l(\omega)+\tfrac{1}{4}(J_l^{\text{eff}})^2F(\omega)\gamma_l(\omega)]^{-1} \quad (54)$$

and

$$\tilde{\Gamma}_l(\epsilon_k,\omega)\equiv N\int_0^{\infty}\rho(\epsilon)\Gamma_l(\epsilon_k,\epsilon,\omega)d\epsilon=\tfrac{1}{2}G_k^{(0)}(\omega)J_l^{\text{eff}}F(\omega)\times[1+J_l^{\text{eff}}g_l(\omega)+\tfrac{1}{4}(J_l^{\text{eff}})^2F(\omega)\gamma_l(\omega)]^{-1}\times\{[\tilde{m}_l(\epsilon_k)-S(S+1)+t_l\gamma_l(\omega)]\times[1+J_l^{\text{eff}}g_l(\omega)]-[\tilde{n}_l(\epsilon_k)-\tfrac{1}{2}+t_lg_l(\omega)]J_l^{\text{eff}}\gamma_l(\omega)\} \quad (55)$$

or

$$\tilde{\Gamma}_l(\epsilon_k,\omega)=\tfrac{1}{2}\tilde{G}_l(\epsilon_k,\omega)J_lF(\omega)\{\tilde{m}_l(\epsilon_k)-S(S+1)+t_l\gamma_l(\omega)-[\tilde{n}_l(\epsilon_k)-\tfrac{1}{2}+t_lg_l(\omega)]\times[1+J_l^{\text{eff}}g_l(\omega)]^{-1}J_l^{\text{eff}}\gamma_l(\omega)\}. \quad (56)$$

The self-consistency conditions corresponding to Eqs. (18) and (19) are

$$\tilde{n}_l(\epsilon)=-\pi^{-1}\int_{-\infty}^{\infty}d\omega f(\omega)\,\text{Im}\tilde{G}_l(\epsilon,\omega), \quad (57)$$

$$\tilde{m}_l(\epsilon)=-2\pi^{-1}\int_{-\infty}^{\infty}d\omega f(\omega)\,\text{Im}\tilde{\Gamma}_l(\epsilon,\omega). \quad (58)$$

Equations (54) and (55), (57) and (58) generalize Eqs. (2.24) to (2.27) of N. They are completely decoupled in the partial waves l. We expect to find a series of "critical" temperatures $T_{c,l}$ defined by Eq. (1) which may be affected by the normal interaction. The normal scattering cannot entirely be absorbed into the effective exchange interaction in Eqs. (54) and (55).

The scattering by the normal potential may be expressed in terms of phase shifts η_l. From the definition (23) follows ($f_{kk'}$ is the scattering amplitude, Ω_{At} the atom volume, m the electron mass)

$$t_{kk'}=-(2m)^{-1}4\pi(\Omega_{At})^{-1}(2ik)^{-1}\times\sum_{l=0}^{\infty}(2l+1)(e^{2i\eta_l}-1)P_l(\cos\Theta_{kk'}). \quad (59)$$

We restrict ourselves in the following to energies ω near the Fermi energy ϵ_F and assume [see Eq. (38)]

$$F(\omega)=-i\pi\rho, \quad (60)$$

with $\rho\equiv\rho(\omega)\approx\rho(\epsilon_F)$. This corresponds to the assumptions that (a) the Fermi surface is in the middle of the conduction band; (b) the density of states $\rho(\epsilon)$ is symmetric around the Fermi surface; and (c) the density of states changes little near ϵ_F. We find in this approximation with $(\Omega_{At})^{-1}=k_F^3/3\pi^2$ and $\rho=\frac{3}{4}\epsilon_F^{-1}$ for monovalent metals

$$F(\omega)t_l\equiv\tilde{t}_l=\tfrac{1}{2}(e^{2i\eta_l}-1). \quad (61)$$

D. Relaxation Time

(a) *Normal Scattering*

The relaxation time for normal scattering of conduction electrons by N_i statistically distributed impurities may be calculated either from the imaginary part of the self-energy $\sum^{(0)}(\epsilon_k,\omega)$ of the Green's function $G_{kk}^{(0)}(\omega)$, or from the t matrix[15]:

$$\begin{aligned}(2\tau_n)^{-1}&=-\text{Im}\sum^{(0)}(\epsilon_k,\omega)\\&\equiv N_i\,\text{Im}\{[G_{kk}^{(0)}(\omega)]^{-1}-G_k^{(0)}(\omega)^{-1}\}\\&=-c\,\text{Im}t_{kk}\\&=-N_i\,\text{Im}[G_k^{(0)}(\omega)^{-2}G_{kk}^{(0)}(\omega)-G_k^{(0)}(\omega)^{-1}],\end{aligned} \quad (62)$$

where we used Eq. (26). Both expressions are identical if higher-order terms in the impurity concentration $c=N_i/N$ are neglected. This may be seen by expressing $[G_{kk}^{(0)}(\omega)]^{-1}-G_k^{(0)}(\omega)^{-1}$ in terms of t_{kk} and $G_k^{(0)}(\omega)$. Using the optical theorem,[15] one obtains the usual result

$$(2\tau_n)^{-1}=c\pi\sum_{k'}|t_{kk'}|^2\delta(\epsilon_k-\epsilon_{k'})=c(\pi\rho)^{-1}\sum_l(2l+1)\sin^2\eta_l. \quad (63)$$

(b) *Exchange and Normal Scattering*

One finds from the self-energy of the Green's function $G_{kk}(\omega)$, to order c,

$$\tau^{-1}(\omega)=-2N_i\,\mathrm{Im}[G_k^{(0)}(\omega)^{-2}G_{kk}(\omega)-G_k^{(0)}(\omega)^{-1}]$$
$$=\tau_n^{-1}+\tfrac{1}{2}c\sum_{l=0}^{\infty}(2l+1)\,\mathrm{Im}\{(J_l^{\mathrm{eff}})^2(1+\bar{t}_l)\gamma_l(\omega)$$
$$\times[1+J_l^{\mathrm{eff}}g_l(\omega)+\tfrac{1}{4}(J_l^{\mathrm{eff}})^2F(\omega)\gamma_l(\omega)]^{-1}\}\,. \quad (64)$$

We could not find a general expression for the transport relaxation time τ_{tr}, which is actually needed for transport properties. In order to calculate τ_{tr} one has to know the matrix $T_{kk'}$ for the total scattering. For the relaxation time τ one needs to know only $\mathrm{Im}T_{kk}$. However, τ_{tr} can be found in special cases from τ, using

$$\tau_{\mathrm{tr}}^{-1}\propto\int|T_{kk'}|^2(1-\cos\Theta_{kk'})d\Omega$$
$$\propto\sum_l[(2l+1)|T_l|^2$$
$$-(l+1)(T_lT_{l+1}^*+T_{l+1}T_l^*)]\,, \quad (65)$$

where we expanded $T_{kk'}$ into Legendre polynomials. Since the first term is proportional to τ^{-1}, and since the second term has to be symmetric in l and $l+1$, one can guess the second term in simple cases.

III. ITERATION AT HIGH TEMPERATURES

We expect that the self-consistent Eqs. (54) to (58) with the definitions (43) and (44) can be iterated at sufficiently high temperatures, starting from free electrons, since the expansion parameter $|J_l^{\mathrm{eff}}|\epsilon_F^{-1}$ is of the order of 10^{-1} or smaller. It turns out that there exists a second parameter, $g(\omega,T)\,\mathrm{Re}J_l^{\mathrm{eff}}$, with $g(\omega,T)$ defined by Eq. (2), which has to be small enough to allow an iteration. This limits the temperature range to $T<T_c$, where T_c is the highest critical temperature $T_{c,l}$ and may depend on the normal interaction. The following expressions hold for arbitrary normal scattering.

(a) *Born Approximation*

To lowest order in J_l^{eff} one has $\tilde{m}_l(\epsilon_k)=0$ and therefore from Eqs. (44) and (60)

$$\gamma_l=i\pi\rho S(S+1)\,. \quad (66)$$

We insert Eq. (66) into (64) and neglect all terms of higher order than $(J_l^{\mathrm{eff}})^2$. We find in Born approximation in the exchange scattering

$$\tau_B^{-1}=\tau_n^{-1}+\tfrac{1}{2}c\pi\rho S(S+1)\sum_l(2l+1)J_l^2\,\mathrm{Re}(1+\bar{t}_l)^3$$
$$=\tau_n^{-1}+\tau_m^{-1}J_0^{-2}\sum_l(2l+1)J_l^2\cos^4\eta_l$$
$$\times(1-4\sin^2\eta_l)\,, \quad (67)$$
$$\tau_m^{-1}\equiv\tfrac{1}{2}c\pi\rho S(S+1)J_0^2\,. \quad (68)$$

Equation (67) reduces for $\eta_l\ll 1$ to the result obtained from usual perturbation theory. The lowest-order mixed term of normal and exchange interactions is proportional to $(J_lV_l)^2$. The second term in Eq. (67) becomes negative for sufficiently strong normal scattering. However, the total relaxation time will always be positive.

We find the transport relaxation time by identifying $|t_l|^2+\frac{1}{4}S(S+1)J_l^2\,\mathrm{Re}(1+\bar{t}_l)^3$ with $|T_l|^2$. It follows with Eq. (65):

$$\tau_{\mathrm{tr},B}^{-1}=\tau_{\mathrm{tr},n}^{-1}+\tau_m^{-1}J_0^{-2}$$
$$\sum_l\{(2l+1)J_l^2\cos^4\eta_l(1-4\sin^2\eta_l)$$
$$-(l+1)J_lJ_{l+1}[\cos^2\eta_l\cos^2\eta_{l+1}(\cos2\eta_l+\cos2\eta_{l+1})$$
$$-\tfrac{1}{2}(\cos^2\eta_l+\cos^2\eta_{l+1})\sin2\eta_l\sin2\eta_{l+1}]\}\,. \quad (69)$$

(b) *Kondo's Approximation*

In order to find $\tau(\omega)^{-1}$ to order $|J_l^{\mathrm{eff}}|^3$, we calculate $\gamma_l(\omega)$ to order J_l^{eff}, and $g_l(\omega)$ without exchange interaction. From Eq. (54) follows

$$\tilde{G}_l(\epsilon_k,\omega)=G_k^{(0)}(\omega)(1+\bar{t}_l)\,, \quad (70)$$

and from Eq. (57) with $\bar{t}_l=\bar{t}_l'+i\bar{t}_l''$

$$\tilde{n}_l(\epsilon_k)=f_k(1+\bar{t}_l')-g_k(\pi\rho)^{-1}\bar{t}_l''\,, \quad (71)$$

$$g_k=\rho\int_0^{\epsilon_m}P\frac{f(\omega)}{\omega-\epsilon_k}d\omega\,, \quad (72)$$

where we cut off the integral g_k at the band limits $\omega=0$ and $\omega=\epsilon_m$. The integral g_k reduces to $g(\epsilon_k,T)$ given by Eq. (2) only if $\rho(\epsilon)=\mathrm{const}$ for $0<\epsilon<\epsilon_m$ and $\rho(\epsilon)=0$ otherwise. We use this approximation in the following for all integrals containing the Green's function $G_k^{(0)}(\omega)$ with $\omega\approx\epsilon_F$. We find in the limits $\epsilon_k=\epsilon_F$, $T\to 0$, and $\epsilon_k\to\epsilon_F$, $T=0$,

$$g(\epsilon_F,T)=\rho\ln k_BT/\epsilon_F\,,$$
$$g(\epsilon_k,0)=\rho\ln|\epsilon_k-\epsilon_F|/\epsilon_F\,, \quad (73a,b)$$

respectively. From Eq. (43) follows

$$g_l(\omega)=-g(\omega)(1+\bar{t}_l^*)-i\pi\rho[f(\omega)(1+\bar{t}_l')-\tfrac{1}{2}]\,. \quad (74)$$

We neglected for $\omega\approx\epsilon_F$ the integral

$$I_1(\omega)\equiv\int_0^{\infty}P\frac{g_k}{\omega-\epsilon_k}\rho(\epsilon_k)d\epsilon_k\approx\rho\int_0^{\epsilon_m}P\frac{g_k}{\omega-\epsilon_k}d\epsilon_k\,, \quad (75)$$

since $I_1(\omega-\epsilon_F)=-I_1(\epsilon_F-\omega)$, or since

$$I_1(\omega)=\rho^2\int_0^{\epsilon_m}\int_0^{\epsilon_m}f(\epsilon')P\frac{d\epsilon d\epsilon'}{(\omega-\epsilon)(\epsilon'-\epsilon)}$$
$$=-\rho^2\int_0^{\epsilon_m}\int_0^{\epsilon_m}f(\epsilon')P\frac{1}{\omega-\epsilon'}\left(\frac{1}{\omega-\epsilon}+\frac{1}{\epsilon-\epsilon'}\right)d\epsilon d\epsilon'$$

is zero if one extends the limits of the ϵ integration to $\pm\infty$.

The Green's function $\tilde{\Gamma}_l(\epsilon_k,\omega)$ has to be calculated to order J_l^{eff}. We find from Eqs. (56), (58), (70), (66), and (44),

$$\tilde{\Gamma}_l(\epsilon_k,\omega)=\tfrac{1}{2}i\pi\rho S(S+1)J_l(1+\tilde{t}_l)^2G_k^{(0)}(\omega)\,, \tag{76}$$

$$\tilde{m}_l(\epsilon_k)=-S(S+1)J_l[\mathrm{Re}(1+\tilde{t}_l)^2g_k+\pi\rho f_k\,\mathrm{Im}(1+\tilde{t}_l)^2]\,, \tag{77}$$

$$\gamma_l(\omega)=i\pi\rho S(S+1)[1+J_lg(\omega)(1+\tilde{t}_l^*)^2+\pi\rho J_lf(\omega)\,\mathrm{Im}(1+\tilde{t}_l)^2]\,. \tag{78}$$

It follows from Eqs. (64), (60), (74), (78), and (68) to order J_l^3

$$\begin{aligned}\tau_{\text{tr}}^{-1}(\omega)=\tau_{\text{tr},B}^{-1}+\tau_m^{-1}J_0^{-2}\sum_l\{&(2l+1)J_l^3\cos^6\eta_l\\&\times[2g(\omega)\cos2\eta_l-\pi\rho f(\omega)\sin2\eta_l]\\&-(l+1)J_lJ_{l+1}\cos^2\eta_l\cos^2\eta_{l+1}[J_l\cos\eta_l\cos(2\eta_l-\eta_{l+1})\\&+J_{l+1}\cos\eta_{l+1}\cos(2\eta_{l+1}-\eta_l)]\\&\times[2g(\omega)\cos(\eta_l+\eta_{l+1})-\pi\rho f(\omega)\sin(\eta_l+\eta_{l+1})]\}\,.\end{aligned} \tag{79}$$

Equation (79) reduces for $\eta_l=0$, $J_l=2\delta_{l0}J_{\text{Kondo}}$ and $\rho=\frac{3}{4}\epsilon_F^{-1}$ to Kondo's[1] Eq. (17) with the number of conduction electrons per atom $z=1$. The second term in Eq. (79) was found by expressing all quantities in terms of J_l^{eff} and comparing with Eq. (65).

(c) *Resistivity*

The conductivity for independent electrons which are scattered elastically by isotropic defects is given by[16] (n is the electron density)

$$\sigma=-\frac{ne^2}{m}\int_0^\infty\tau_{\text{tr}}(\omega)\frac{df(\omega)}{d\omega}d\omega\,. \tag{80}$$

We assume first that the inverse relaxation time $\tau_{\text{tr},B}^{-1}$ (which includes normal scattering) is large compared to the strongly energy-dependent inverse relaxation time $\tau_{\text{tr},an}^{-1}(\omega)\equiv\tau_{\text{tr}}^{-1}(\omega)-\tau_{\text{tr},B}^{-1}$. We can then neglect higher-order terms in the expansion of $\tau_{\text{tr},B}/\tau_{\text{tr},an}$ and find the total impurity resistivity

$$\rho_i=\rho_B-\frac{m}{ne^2}\int_0^\infty\tau_{\text{tr},an}^{-1}(\omega)\frac{df(\omega)}{d\omega}d\omega\,, \tag{81}$$

where ρ_B is the resistivity in Born approximation for the exchange scattering. The integral with $g(\omega)$ has been calculated by Kondo.[1] The terms proportional to

$$\int f(\omega)(df/d\omega)d\omega=-\tfrac{1}{2} \tag{82}$$

are temperature-independent and will be neglected. If $\tau_{\text{tr},an}$ and $\tau_{\text{tr},B}$ are the same order of magnitude, one obtains correction terms which correspond to deviations from Matthiessen's rule. The first correction term is proportional to

$$\int_0^\infty g^2(\omega)(-df/d\omega)d\omega-\left[\int_0^\infty g(\omega)(-df/d\omega)d\omega\right]^2, \tag{83}$$

and has been estimated to be small.[7] Hence we obtain

$$\begin{aligned}\rho_i=\rho_B+\rho_mJ_0^{-2}\tfrac{3}{2}\epsilon_F^{-1}\ln(k_BT/\epsilon_F)&\\\times\sum_{l=0}^\infty\{&(2l+1)J_l^3\cos^6\eta_l\cos2\eta_l\\&-(l+1)J_lJ_{l+1}\cos^2\eta_l\cos^2\eta_{l+1}\cos(\eta_l+\eta_{l+1})\\&\times[J_l\cos\eta_l\cos(2\eta_l-\eta_{l+1})\\&+J_{l+1}\cos\eta_{l+1}\cos(2\eta_{l+1}-\eta_l)]\}\,,\end{aligned} \tag{84}$$

$$\rho_m=m/ne^2\tau_m\,. \tag{85}$$

The correction to the $\ln T$ term due to normal scattering is in lowest order proportional to $V_l^2J_l^3$. For sufficiently strong normal scattering (approximately $\eta_l>\pi/4$ for the most important scattering phases) the $\ln T$ term changes its sign. One finds in this case a resistivity minimum for *ferromagnetic* exchange interaction ($J_l>0$). In all cases the normal scattering reduces the temperature-dependent resistivity term. A resonance in the d scattering due to the normal interaction influences the resistivity minimum only, if the component J_2 is not too small. The corresponding part of the reciprocal transport relaxation time vanishes if the resonance is at the Fermi surface ($\eta_2=\pi/2$) corresponding to a maximum in the normal scattering. These results are in contrast to Abrikosov's,[3] who found no change in the temperature-dependent resistivity due to normal scattering.

(d) *Thermopower*

The thermopower is given by[16] ($e<0$)

$$S(T)=(eT)^{-1}\frac{\int(\omega-\epsilon_F)\tau_{\text{tr}}(\omega)v^2\rho(df/d\omega)d\omega}{\int\tau_{\text{tr}}(\omega)v^2\rho(df/d\omega)d\omega}\,. \tag{86}$$

We neglect contributions due to the energy dependence of the density of states ρ, electron velocity v, and of J_l and t_l, since they are all small compared to the contribution arising from the term proportional to $f(\omega)$ in $\tau_{\text{tr}}^{-1}(\omega)$. Furthermore, we can neglect in the relaxation time terms proportional to $g(\omega)$, since $g(\omega-\epsilon_F)$ is an even function. Expanding again $\tau_{\text{tr}}^{-1}(\omega)$ in powers of $\tau_{\text{tr},B}/\tau_{\text{tr},an}$ and neglecting all deviations from

[16] N. F. Mott and H. Jones, *The Theory of the Properties of Metals and Alloys* (Dover Publications, Inc., New York, 1958), p. 305.

Matthiessen's rule, we find with

$$\int(\omega-\epsilon_F)f(\omega)(df/d\omega)=\tfrac{1}{2}k_BT\,, \tag{87}$$

$$S(T)=-\frac{k_B}{2e}\frac{\rho_m}{\rho_{\text{tot}}}\pi\rho J_0^{-2}\sum_l\{(2l+1)J_l^3\cos^6\eta_l\sin2\eta_l$$
$$-(l+1)J_lJ_{l+1}\cos^2\eta_l\cos^2\eta_{l+1}\sin(\eta_l+\eta_{l+1})$$
$$\times[J_l\cos\eta_l\cos(2\eta_l-\eta_{l+1})$$
$$+J_{l+1}\cos\eta_{l+1}\cos(2\eta_{l+1}-\eta_l)]\}\,. \tag{88}$$

We include in the total resistivity $\rho_{\text{tot}}=\rho_i+\rho_T$, the term ρ_T due to the lattice vibrations. This is again correct in the approximation in which the resistivities add. The dependence of the thermopower Eq. (88) on the temperature and on the impurity concentration is then of the form

$$S(T)=S_0\rho_i(\rho_i+\rho_T)^{-1}. \tag{89}$$

Equation (89) is within about 30% in agreement with experimental results of MacDonald *et al.*[17] for 0.0022–0.02 at. % Fe in Au and at temperatures where the plateau in the thermopower is already reached (and at higher temperatures), when one fits S_0 and uses $\rho_B=7.3\,\mu\Omega$ cm/at. %. The decrease of $S(T)$ at lower temperatures was explained by Kondo[2] by the interaction of the impurity spin with internal magnetic fields.

In the Born approximation for normal scattering ($\eta_0=-\pi\rho V$) and for $J_l=J_0\delta_{l0}$, Eq. (88) reduces to Kondo's[2] result except for a factor $-\frac{1}{2}$. Kondo erroneously neglected a term in his expansion [the next term in the expansion Eq. (2) of Ref. 2] which contains a product of two δ functions and therefore contributes an additional term of the order VJ_0^3 to the thermopower. This term is of the correct order of magnitude to account for the discrepancy. The thermopower Eq. (88) is negative, if the most important components $J_l<0$, and if the normal interaction is attractive with $\eta_l<\pi/2$. It changes its sign if $\eta_l>\pi/2$ for the most important scattering phases, compared to $\eta_l>\pi/4$ for the change in sign of the resistivity term.

(e) *Terms of Order J_l^4*

We calculate the next iteration step (order J_l^4 in the relaxation time) only for $t_l=0$. The Green's function $\tilde{G}_l(\epsilon_k,\omega)$ can be replaced in order J_l by the free Green's function

$$\tilde{G}_l(\epsilon_k,\omega)=G_k^{(0)}(\omega)\,, \tag{90a}$$

$$\tilde{n}_l(\epsilon_k)=f_k\,, \tag{90b}$$

$$g_l(\omega)=-g(\omega)-i\pi\rho[f(\omega)-\tfrac{1}{2}]. \tag{90c}$$

Using Eqs. (90) we solve Eqs. (56) and (58) with $\gamma_l=\gamma_l'+i\gamma_l''$ self-consistently (the term proportional $\gamma_l'(\omega)$ is of higher order):

$$\tilde{\Gamma}_l(\epsilon_k,\omega)=-\tfrac{1}{2}i\pi\rho J_lG_k^{(0)}(\omega)\{\tilde{m}_l(\epsilon_k)-S(S+1)$$
$$+i\pi\rho J_l(f_k-\tfrac{1}{2})[\tilde{m}_l(\omega)-S(S+1)]\}\,, \tag{91}$$

$$\tilde{m}_l(\epsilon_k)=S(S+1)\{1-[1-J_lg_k$$
$$-(\pi\rho J_l)^2f_k(f_k-\tfrac{1}{2})]^{-1}\}\,, \tag{92}$$

$$\gamma_l''(\omega)=\pi\rho S(S+1)\{1-J_lg(\omega)$$
$$-(\pi\rho J_l)^2f(\omega)[f(\omega)-\tfrac{1}{2}]\}^{-1}. \tag{93}$$

Equations (91) to (93) are correct to order J_l^2. We will prove later that they hold also to order $[J_lg(\omega)]^{-2}$, if $J_lg(\omega,T)\gg1$, corresponding to very low temperatures. The real part $\gamma_l'(\omega)$ is also of order J_l^2, but it enters into the expression for the relaxation time only to order J_l^5. We obtain to order J_l^4

$$\tau^{-1}(\omega)=\tau_m^{-1}J_0^{-2}\sum_l(2l+1)$$
$$\times\frac{J_l^2}{[1-J_lg(\omega)]^2+\frac{1}{4}(\pi\rho J_l)^2[S(S+1)-2f(\omega)+1]}. \tag{94}$$

Equation (94) holds [besides the factor $f(\omega)-\frac{1}{2}$] in all powers of $J_lg(\omega)$ if we keep in each order only the leading terms and neglect (a) all terms proportional $f(\omega)-\frac{1}{2}$, (b) all corrections arising from $\gamma_l'(\omega)$, and use (c) in all orders the free-electron Green's function (90).[18] One obtains in this approximation for $J_l<0$ a series of resonances of the Suhl-Abrikosov type below critical temperatures $T_{c,l}$, defined by Eqs. (1) and (73):

$$k_BT_{c,l}=\epsilon_F\exp(J_l\rho)^{-1}. \tag{95}$$

At least part of the normal interaction can be included by replacing J_l by $\text{Re}J_l^{\text{eff}}=J_l(1+\dot{t}_l')$. This shifts $T_{c,l}$ to the temperature

$$k_BT_{c,l}^*=\epsilon_F\exp(J_l\rho\cos^2\eta_l)^{-1}\,, \tag{96}$$

which is always lower than $T_{c,l}$. The neglected terms, however, are not small near $T_{c,l}^*$, and we expect in this temperature range strong deviations from Eq. (94).

The resistivity calculated from Eq. (94) to order J_l^4 contains terms proportional to $(\ln k_BT/\epsilon_F)^2$. The thermopower S_0 in Eq. (89) remains constant. One finds, however, to order J_l^5 terms proportional to $g(\omega)f(\omega)$ in the relaxation time, which give rise to a temperature dependent thermopower $S_0(T)$.

IV. ITERATION AT LOW TEMPERATURES

We consider now the temperature range $T\ll T_{c,l}^*$ for the partial wave l, corresponding to

$$|\text{Re}J_l^{\text{eff}}g(\omega,T)|\gg1\,, \tag{97}$$

[17] D. K. C. MacDonald, W. B. Pearson, and I. M. Templeton, Proc. Roy. Soc. (London) **A266**, 161 (1962).

[18] Assumption (a) corresponds in the perturbation expansion of $T_{kk'}$ to neglecting all energy conserving processes involving intermediate states. Assumption (c) corresponds to the linearization of the equation of motion (14). The neglected terms to order n are proportional to $\rho J_l[J_lg(\omega)]^{n-1}$ or smaller.

with $\omega \approx \epsilon_F$. The electron Green's function (54) is unaffected by the exchange scattering, if in addition to Eq. (97)

$$|\tfrac{1}{4}(J_l^{\text{eff}})^2 F(\omega)\gamma_l(\omega)| \ll |J_l^{\text{eff}} g_l(\omega)| \tag{98}$$

holds. We will show that Eqs. (97) and (98) are consistent with the full set of self-consistent equations (54) to (58) with (43) and (44). The first set of equations has with Eq. (98) the solution (70) to (72). We insert this into $\tilde{\Gamma}_l(\epsilon_k,\omega)$, Eq. (56). The terms $t_l\gamma_l(\omega)$ cancel in $\tilde{\Gamma}_l(\epsilon_k,\omega)$ in the limit (97). Since $|\gamma_l'(\omega)| \ll |\gamma_l''(\omega)|$ (as will be proved), we find

$$\tilde{\Gamma}_l(\epsilon_k,\omega) = -\tfrac{1}{2} i\pi\rho J_l G_k^{(0)}(\omega)\{\tilde{m}_l(\epsilon_k) - S(S+1) + i g_k \tilde{t}_l''[g(\omega)(1+\tilde{t}_l^*)]^{-1}[\tilde{m}_l(\omega) - S(S+1)]\}. \tag{99}$$

We obtain in order $(J_l g_k)^{-1}$

$$\tilde{m}_l(\epsilon_k) = S(S+1)\{1+[J_l g_k(1+\tilde{t}_l')(1+\beta_l)]^{-1}\}, \tag{100}$$

where approximately

$$\beta_l = -\tilde{t}_l'' \operatorname{Im}(1+\tilde{t}_l)^2(1+\tilde{t}_l')^{-1}|1+\tilde{t}_l|^{-2} = -\tfrac{1}{2}(\sin 2\eta_l)^2 \cos^{-2}\eta_l. \tag{101}$$

This is in contrast to the high-temperature limit where $\tilde{m}_l(\epsilon_k) \to 0$. We find

$$\gamma_l(\omega) = -i\pi\rho S(S+1)[J_l g(\omega)(1+\tilde{t}_l')(1+\beta_l)]^{-1} \times [1+i(\pi\rho)^{-1} I_{-1}(\omega) g(\omega)]. \tag{102}$$

The integral

$$I_{-1}(\omega) \equiv \int_0^\infty P\frac{\rho(\epsilon)d\epsilon}{(\omega-\epsilon)g(\epsilon)} \approx \rho \int_0^{\epsilon_m} P\frac{d\epsilon}{(\omega-\epsilon)g(\epsilon)} \tag{103}$$

vanishes for $\omega = \epsilon_F$ and arbitrary T, since $I_{-1}(\omega-\epsilon_F) = -I_{-1}(\epsilon_F-\omega)$. Therefore Eq. (98) holds for ω sufficiently near to ϵ_F, and the solution is consistent. The l component of the inverse relaxation time is

$$\tau_l^{-1}(\omega) = \tau_{n,l}^{-1} + \tau_m^{-1} J_0^{-2} g(\omega)^{-2} \times [1 - 4\sin^2\eta_l - \tan\eta_l(4\cos^2\eta_l - 1) g(\omega) \times (\pi\rho)^{-1} I_{-1}(\omega)](1+\beta_l)^{-1}. \tag{104}$$

The term proportional to $I_{-1}(\omega)$ vanishes for $t_l = 0$ and does not contribute to the resistivity since

$$\int_0^\infty \frac{df}{d\omega}\frac{I_{-1}(\omega)}{g(\omega)} = 0. \tag{105}$$

We assume for the next iteration step $t_l = 0$ and neglect terms proportional to $I_{-1}(\omega)$. The calculation is then completely analogous to Sec. III(e). We find to order $[J_l g(\omega)]^{-3}$

$$\tau_l^{-1}(\omega) = \tau_m^{-1} J_0^{-2} \frac{J_l^2}{[1-J_l g(\omega)]^2 + \text{const} \times J_l^2}, \tag{106}$$

where the constant is of the order $(\pi\rho)^2 S(S+1)$ and can be calculated exactly only in the next order for the relaxation time.

The sum over the inverse relaxation times $\tau_l^{-1}(\omega)$, Eq. (104), is limited for a given temperature by the condition $T \ll T_{c,\bar{l}}$, where $T_{c,\bar{l}}$ corresponds to the smallest component J_l which satisfies Eq. (97). Besides this sum contributions $\bar{\tau}^{-1}$ from resonances with $T_{c,l}$ comparable or smaller than T enter into $\tau^{-1}(\omega)$. The normal potential effects only the first few partial waves, and we will neglect the corresponding change in the resistivity in the following. All components $\tau_l^{-1}(\omega)$ are in the limit (106) independent of J_l and give then the same contributions to the total relaxation time.

We obtain with Eq. (65) the transport relaxation time

$$\tau_{\text{tr}}^{-1}(\omega,T) = \tau_{\text{tr},n}^{-1} + \tau_m^{-1}[J_0 g(\omega,T)]^{-2} \times \left[\sum_{l=0}^{\bar{l}}(2l+1) - 2\sum_{l=0}^{\bar{l}-1}(l+1)\right] + \bar{\tau}^{-1} = \tau_{\text{tr},n}^{-1} + \tau_m^{-1}[J_0 g(\omega,T)]^{-2} \times (\bar{l}+1) + \bar{\tau}^{-1}. \tag{107}$$

The second sum is restricted to $\bar{l}-1$ corresponding to $J_{\bar{l}+1} = 0$. We find the resistivity

$$\rho_i(T) = \rho_n + \rho_m(J_0\rho \ln k_B T/\epsilon_F)^{-2}[\bar{l}(T)+1] + \bar{\rho}(T), \quad T \ll T_{c,\bar{l}}, \tag{108}$$

where $\bar{\rho}$ corresponds to $\bar{\tau}$. Next we calculate the resistivity $\rho(T_{c,\bar{l}+n})$ for the temperatures $T = T_{c,\bar{l}+n}$, $n = 1, 2, 3 \cdots$. Condition (97) reduces for these temperatures with $J_{\bar{l}+n} g(0, T_{c,\bar{l}+n}) = 1$ to

$$J_l/J_{\bar{l}+n} \gg 1, \quad l \leq \bar{l}, \tag{109}$$

where we considered only partial waves unaffected by normal scattering.

We consider only the case $l = \bar{l}$ and $n = 1$, corresponding to a very short-range-exchange interaction. The resistivity $\rho(T)$ then contains only contributions from partial waves $l \geq \bar{l}+1$. Since $J_{\bar{l}+n+1} g(0, T_{c,\bar{l}+1}) \ll 1$, the contributions from resonances $l > \bar{l}+1$ are again small. We obtain

$$\rho_i(T_{c,l}) = \rho_n + \rho_m(J_l/J_0)^2 l + \rho_{i,l}. \tag{110}$$

The second term tends to zero as $l \to \infty$, if J_l vanishes faster than $l^{-1/2}$. This is much less stringent than condition (109). There remains the resistivity $\rho_{i,l}$ at the resonance l. We cannot determine $\rho_{i,l}$, since we cannot solve the self-consistent Eq. (54) to (58) at $T = T_{c,l}$. However, all approximations indicate that the scattering at $T_{c,l}$ is also independent of J_l and therefore $\rho_{i,l}$ is independent of l.

The resistivity $\rho(T)$ oscillates corresponding to the temperatures $T_{c,l}$, where the different resonances become effective. However, for a more extended exchange interaction, the contributions from the different resonances overlap, and the oscillations will be smeared

out. At very low temperatures the resonances lie very near to each other, and one will observe a temperature-independent resistivity in which the normal and exchange parts simply add.

V. CONCLUSIONS

We studied the scattering by a single magnetic impurity using the self-consistent field method first applied to the present problem by Nagaoka.[6] Our basic approximation is the factorization of two-particle Green's functions in the equation of motion for the Green's function $\Gamma_{kk'}(\omega)$, Eq. (8). We also made simplifying assumptions concerning the density of states of the conduction band. We solved the self-consistent equations of motion for ferromagnetic and antiferromagnetic exchange interaction and for $T \gg T_{c,l}$ and $T \ll T_{c,l}$ with $T_{c,l}$ defined by Eq. (1).

Our result at high temperatures [Eqs. (84) and (88)] generalizes Kondo's[1,2] expressions for the resistivity $\rho_i(T)$ and thermopower $S(T)$ for arbitrary normal and exchange interaction. The normal scattering always reduces the anomalous scattering due to the exchange interaction. The resonance in the normal scattering phase η_2 affects only the component $l=2$ of the exchange scattering. We expect therefore a sizeable correlation between the large impurity resistivity caused by the resonance of η_2 and the resistivity minimum only if the component $|J_2|$ is large. A connection between the parameters $J_{kk'}$ and the position and width of the impurity d state, has been discussed recently by Schrieffer and Wolff.[19]

Our result at low temperatures [Eqs. (104), (106), and (108)] is completely different from Nagaoka's[6] solution. We found for both signs of a δ-function exchange interaction J_0 (corresponding to a single partial wave $l=0$) vanishing exchange scattering and $\rho_{\text{ex}}(T) \propto (\ln k_B T/\epsilon_F)^{-2}$ as $T \to 0$, in contradiction to Eqs. (5.8) and (5.10) of N. Nagaoka discusses only the case $J_0<0$. He assumes $\tilde{m}_0(\omega)$ to be singular for $T=0$ and $\omega=\epsilon_F$, and $J_0 \operatorname{Re} g_0(\omega,T)=1$ for $\omega=\epsilon_F$, and $T \leq T_{c,0}$ [Eqs. (4.1) and (4.2) in Ref. 6], and shows this to be consistent for $\omega=\epsilon_F$. However, the energy range in which this solution holds remains unclear. Both conditions disagree with our solution $\tilde{m}_0(\epsilon_k) \sim S(S+1)$ and $\operatorname{Re} g_0(\epsilon_F,T) = -g(\epsilon_F,T) = -\rho \ln(k_B T/\epsilon_F)$ as $T \to 0$ and for $t_l=0$ [Eqs. (90c) and (92)]. The assumption of a singularity in the thermal average $\tilde{m}_0(\epsilon_k)$, which is related to the correlation between the conduction electrons and the impurity spin, seems to us rather unphysical. We could not find any instability in the relaxation time in the considered temperature range, which would suggest a phase transition. In analogy to a normal-scattering resonance the conduction electrons are most disturbed at $T=T_{c,0}$. Sufficiently far off the resonance they can be treated as unperturbed in the self-consistent Eqs. (54) to (58). This corresponds to the linearization of the equation of motion (14) for $\Gamma_{kk'}(\omega)$. The first corrections to the free-electron Green's function enter into the inverse relaxation time Eq. (64) to order $[J_0 g(\omega,T)]^5$ for $T \gg T_{c,0}$, and to order $[J_0 g(\omega,T)]^{-4}$ for $T \ll T_{c,0}$ and $\omega=\epsilon_F$.

In the case of an extended exchange interaction, we obtained a resonance for each partial wave l [Eq. (106)]. The corresponding temperatures $T_{c,l}$ converge to zero very fast for increasing l. The distances between the resonances depend on the l dependence of the components J_l. The resonances overlap only slightly if $J_l/J_{l+1} \gg 1$. The relaxation time is determined in this case mostly by the nearest resonance. The resistivity due to the exchange interaction oscillates as a function of T and tends to a finite value as $T \to 0$. This behavior changes, however, if one takes into account the effect of an internal magnetic field on the scattering. The magnetic field causes Zeeman splitting of the impurity spin levels. The spin flip freezes out, and the resonances of partial waves with large l are suppressed.

ACKNOWLEDGMENTS

I would like to express my sincere thanks to Professor J. Bardeen and to Professor T. Rowland for many valuable discussions during all phases of this work, and to Dr. W. Goetze for a critical reading of the manuscript. A travel grant from the Fulbright Foundation is gratefully acknowledged.

[19] J. R. Schrieffer and P. A. Wolff, Phys. Rev. 149, 491 (1966).