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Effect of crystal fields on the Hall effect in Kondo-type systems

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We calculate the effect of crystal fields on the skew scattering and anomalous velocity contributions to the Hall effect of Kondo systems. Above the Kondo temperature T_K the crystal field reduces the extraordinary Hall effect in the same proportion as the product of the magnetic resistivity ρ and susceptibility χ. Below T_K the effect of the crystal field cannot be accounted for by only the reduction of $\rho\chi$, and the Hall effect can be either enhanced or suppressed.

I. INTRODUCTION

The Hall effect has been a particularly effective probe of conduction-electron scattering in Kondo-type compounds.[1] In particular the extraordinary Hall effect (EHE) probes the scattering of conduction electrons by the *orbital* angular momentum of localized electrons.[2] As such, it is sensitive to crystalline electric fields, because they reduce the orbital degrees of freedom of localized electrons. Here, we show how to include the effects of crystal fields on the EHE. We determine these effects for single crystals and powdered samples.

In the conventional Hall geometry one applies a magnetic field **H** perpendicular to the current which is parallel to the applied electric field $\mathcal{E}$, and measures a Hall voltage along the direction $\hat{\mathcal{E}}\times\hat{\mathbf{h}}$. The presence of a crystal field introduces a *second* set of axes, i.e., the set of local axes about which one defines symmetry operations of the crystalline environment, in which the crystal-field Hamiltonian assumes its simplest form. In general these two sets of axes are not coincident, and it is necessary to rotate one set into the other. The Hamiltonian for the Kondo ions in a metallic system contains, among other things, two terms which depend on the relative orientation of the two sets of axes, viz., the crystal field

$$V_{\mathrm{cr}}=\sum_{lm} B_{lm}(\hat{\mathbf{z}})O_m^l(\mathbf{J},\hat{\mathbf{z}}) , \tag{1.1}$$

and the Zeeman interaction

$$V_z=g_J\mu_B\mathbf{H}\cdot\mathbf{J}=g_J\mu_B H O_0^1(\mathbf{J},\hat{\mathbf{h}}) , \tag{1.2}$$

where the angular momentum tensor operators $O_m^l(J)$ are defined by Smith and Thornley,[3] e.g., $O_0^1(\mathbf{J},\hat{\mathbf{h}})=\mathbf{J}\cdot\hat{\mathbf{h}}=J_h$. The additional variable $\hat{\mathbf{z}}$ or $\hat{\mathbf{h}}$ on these operators denotes the direction of spatial quantization. If we choose to rotate the crystal-field Hamiltonian to the set of field axes $[\hat{\mathcal{E}},-(\hat{\mathcal{E}}\times\hat{\mathbf{h}}),\hat{\mathbf{h}}]$ we use

$$|m(\hat{\mathbf{z}})\rangle=\mathcal{R}|m(\hat{\mathbf{h}})\rangle=\sum_{m'}|m'(h)\rangle\langle m'(h)|R|m(h)\rangle , \tag{1.3}$$

and find

$$\mathcal{H}=\mathcal{H}_0+\sum_{lm'}\bar{B}_{lm'}(\hat{\mathbf{h}})O_{m'}^l(\mathbf{J},\hat{\mathbf{h}})+g_J\mu_B H O_0^1(\mathbf{J},\hat{\mathbf{h}}) , \tag{1.4}$$

where

$$\bar{B}_{lm'}(\hat{\mathbf{h}})\equiv\sum_m B_{lm}(\hat{\mathbf{z}})\mathcal{D}_{m'm}^{(l)}(\mathcal{R}) ,$$

and $\mathcal{D}_{m'm}^{(l)}$ is a matrix element of the rotation from the field $\hat{\mathbf{h}}$ to local $\hat{\mathbf{z}}$ axes,[4]

$$\mathcal{D}_{m'm}^{(l)}(\mathcal{R})=\langle lm'|R|lm\rangle . \tag{1.5}$$

On the other hand, if we choose to rotate the Zeeman interaction, we find

$$\mathcal{H}=\mathcal{H}_0+\sum_{lm}B_{lm}(\hat{\mathbf{z}})O_m^l(\mathbf{J},\hat{\mathbf{z}})+g_J\mu_B H\sum_m\mathcal{D}_{0m}^{(1)*}O_m^1(\mathbf{J},\hat{\mathbf{z}}) . \tag{1.6}$$

In either case the eigenstates of $\mathcal{H}$ depend on the relative orientation of the two sets of axes, and on the field H, because the commutator $[V_{\mathrm{cr}},V_z]$ is nonzero. Thus for a single crystal one cannot express the eigenstates of $\mathcal{H}$ in a simple form because they vary with H. For a polycrystalline sample the different orientations of local axes relative to the field axes further complicate the situation. One could diagonalize the Hamiltonian numerically and obtain solutions for arbitrary field strengths. However, that would not be very enlightening; we are more interested in trends. Therefore, we limit ourselves to effects *linear* in the magnetic field so that we obtain analytic results.

II. HALL EFFECT

The Hall constant due to the linear extraordinary Hall effect (LEHE) is

$$R_H=\frac{\rho_H}{H}=\frac{\sigma_H/H}{\sigma_N^2} . \tag{2.1}$$

When we consider only the spin-independent contribution to the LEHE, the Hall conductivity σ_H is given as

$$\sigma_H=\tfrac{2}{3}e^2\int n(\varepsilon)d\varepsilon\left[-\frac{\partial f^0}{\partial\varepsilon}\right]\times[v^2(\varepsilon)\tau_0^2(\varepsilon)B(\varepsilon)+2v(\varepsilon)\tau_0(\varepsilon)\omega_a(\varepsilon)]\ , \tag{2.2}$$

where the first term in square brackets comes from skew scattering,[5] and the second from anomalous velocity contributions[6] to the EHE. The normal conductivity σ_N is

$$\sigma_N=\tfrac{2}{3}e^2\int n(\varepsilon)d\varepsilon\left[-\frac{\partial f^0}{\partial\varepsilon}\right]v^2(\varepsilon)\tau_0(\varepsilon)\ . \tag{2.3}$$

The relaxation rate τ_0^{-1} is[5,6]

$$\tau_0^{-1}(\varepsilon)=-c_i(|V_k|^2/\hbar)\mathrm{Im}G_0(\varepsilon)\ , \tag{2.4}$$

the skew scattering coefficient B is

$$B(\varepsilon)=-\tfrac{24}{49}c_i(|V_k|^2/\hbar)\sin\eta_2(\varepsilon)\mathrm{Im}[e^{-i\eta_2(\varepsilon)}G_1(\varepsilon)]\ , \tag{2.5}$$

and when we neglect terms proportional to $\partial_k\eta_2(k)$, which is quite small, the anomalous velocity ω_a is[6]

$$\omega_a=-\tfrac{12}{49}c_i(|V_k|^2/\hbar)\sin\eta_2(\varepsilon)\times\mathrm{Re}[e^{-i\eta_2(\varepsilon)}(\partial_k+6/k)G_1(\varepsilon)]\ . \tag{2.6}$$

Here c_i is the concentration of Kondo ions, V_k is the Anderson mixing parameter, and η_2 the phase shift due to potential scattering in the nonresonant $l=2$ channel. The sums over the $4f$ electron Green's functions are defined as[6]

$$G_0=\sum_{m_j=-j}^{j}G_{m_j}^{4f}=\sum_{m_j}G_{m_j}^{4f}(\varepsilon,H=0)+O(H^2)\ , \tag{2.7a}$$

$$\begin{aligned}G_1&=\sum_{m_j}m_jG_{m_j}^{4f}\\&=g_J\mu_BH\Big(\sum_{m_j}m_j^2\Big)\frac{\partial G_{m_j}^{4f}}{\partial\varepsilon_h}\bigg|_{H=0}+O(H^3)\ ,\end{aligned} \tag{2.7b}$$

and

$$\begin{aligned}\partial_kG_1(\omega=\varepsilon_k)&=\frac{\partial\varepsilon_k}{\partial k}\frac{\partial G_1}{\partial\omega}\\&=\frac{\partial\varepsilon_k}{\partial k}\sum_{m_j}m_j\frac{\partial G_{m_j}^{4f}}{\partial\omega}\\&=\frac{\partial\varepsilon_k}{\partial k}g_J\mu_BH\Big(\sum_{m_j}m_j^2\Big)\frac{\partial^2G_{m_j}^{4f}}{\partial\omega\partial\varepsilon_h}\bigg|_{H=0}\\&\quad+O(H^3)\ .\end{aligned} \tag{2.7c}$$

We have expanded the field dependence of the Green's function as

$$G(\varepsilon,H)=G(\varepsilon,H=0)+\frac{\partial G}{\partial\varepsilon_h}\bigg|_{H=0}\varepsilon_h+O(H^2)\ , \tag{2.8}$$

where $\varepsilon_h\equiv g_J\mu_BHm_j$. It can be expanded in this form when the Zeeman interaction commutes with the unperturbed Hamiltonian as it does for a free $4f$ electron.

In general, in the presence of crystal fields the commutator is nonzero and the expansion Eq. (2.8) is more complicated. However, when we restrict our attention to *one* degenerate crystal-field level

$$\langle\Gamma\alpha|V_{\mathrm{cr}}|\Gamma\beta\rangle=V_\Gamma\delta_{\alpha\beta}\ , \tag{2.9}$$

so that

$$\langle\Gamma\alpha|[V_{\mathrm{cr}},V_z]|\Gamma\beta\rangle=0\ ,$$

where $|\Gamma\alpha\rangle$ is an eigenstate of the crystal-field Hamiltonian

$$(\mathcal{H}_0+V_{\mathrm{cr}})|\Gamma\alpha\rangle=E_\Gamma|\Gamma\alpha\rangle\ , \tag{2.10}$$

and α denotes the degenerate partners of the manifold Γ. Under these conditions the magnetic field dependence of the crystal-field Green's function can be expanded as shown in Eq. (2.8). The sums over the $4f$ Green's functions Eqs. (2.7) can be rewritten in operator form as

$$\begin{aligned}G_0&=\mathrm{Tr}G^{4f}(\varepsilon)\\&=\mathrm{Tr}G^{4f}(\varepsilon,H=0)+O(H^2)\ ,\end{aligned} \tag{2.11a}$$

$$G_1=g_J\mu_BH\ \mathrm{Tr}J_h^2\frac{\partial G^{4f}}{\partial\varepsilon_h}\bigg|_{H=0}+O(H^3)\ , \tag{2.11b}$$

and

$$\partial_kG_1(\omega=\varepsilon_k)=\frac{\partial\varepsilon_k}{\partial k}g_J\mu_BH\ \mathrm{Tr}J_h\frac{\partial^2G^{4f}}{\partial\omega\partial\varepsilon_h}\bigg|_{H=0}J_h+O(H^3), \tag{2.11c}$$

where $J_h=\mathbf{J}\cdot\hat{\mathbf{h}}=O_0^1(\hat{\mathbf{h}})$.

To proceed we consider the Zeeman interaction is small compared to the crystal field, and keep only the leading order in H terms. The Green's function G^{4f} $(\varepsilon,H=0)$ is diagonal in the crystal-field eigenstates $|\Gamma\alpha\rangle$, therefore

$$\langle\Gamma\alpha|G^{4f}(\varepsilon,H=0)|\Gamma'\beta\rangle\equiv G_\Gamma\delta_{\Gamma\Gamma'}\delta_{\alpha\beta}\ . \tag{2.12}$$

In addition, we assume only *one* crystal-field level Γ is occupied for *all* temperatures. If this is not the case the problem is more complicated because the crystal field becomes inextricably intertwined with the Kondo effect itself. With this approximation only one Green's function exits. We evaluate the traces in Eqs. (2.11) by inserting the crystal-field eigenstates of the ground-state manifold, and find

$$G_0^\Gamma=N_\Gamma G_\Gamma(\varepsilon,H=0)+O(H^2)\ , \tag{2.13a}$$

$$G_1^\Gamma=g_J\mu_BH\frac{\partial G_\Gamma}{\partial\varepsilon_h}\bigg|_{H=0}R_\Gamma+O(H^3)\ , \tag{2.13b}$$

and

$$\partial_kG_1^\Gamma(\omega=\varepsilon_k)=g_J\mu_BH\frac{\partial\varepsilon_k}{\partial k}\frac{\partial^2G_\Gamma}{\partial\omega\partial\varepsilon_h}\bigg|_{H=0}R_\Gamma+O(H^3)\ , \tag{2.13c}$$

where

$$R_\Gamma(\hat{\mathbf{h}}) \equiv \sum_{\alpha\beta} |\langle \Gamma\alpha | O_0^1(\hat{\mathbf{h}}) | \Gamma_\beta \rangle|^2 \tag{2.14}$$

is an orbital factor, and N_Γ is the degeneracy of the level Γ. For a free $4f$ electron in a $j=\frac{5}{2}$ state, viz., for cerium, we have[6]

$$G_0^{4f} = (zj+1) G^{4f}(\varepsilon, H=0) + O(H^2) \ , \tag{2.15a}$$

$$G_1^{4f} = g_J \mu_B H \left. \frac{\partial G_m^{4f}}{\partial \varepsilon_h} \right|_{H=0} R_0 + O(H^3) \ , \tag{2.15b}$$

and

$$\partial_k G_1^{4f}(\omega=\varepsilon_k) = \frac{\partial \varepsilon_k}{\partial k} g_J \mu_B H \left. \frac{\partial^2 G_m^{4f}}{\partial\omega \partial\varepsilon_h} \right|_{H=0} R_0 + O(H^3), \tag{2.15c}$$

where

$$R_0 \equiv \sum_{m_j} m_j^2 = \tfrac{1}{3} j(j+1)(2j+1) \ . \tag{2.16}$$

On comparing Eqs. (2.13) and (2.15) we note that the crystal field affects the quantities entering the Hall constant in two ways. First it replaces R_0 by $R_\Gamma(\hat{\mathbf{h}})$; we will call this an orbital reduction effect. As the crystal-field states are referred to the local crystal symmetry axes and O_0^1 is quantized along $\hat{\mathbf{h}}$, to evaluate the matrix element in R_Γ we write $O_0^1(\hat{\mathbf{h}})$ in terms of tensor operators referred to the local axes,

$$O_0^1(\hat{\mathbf{h}}) = \mathcal{R}' O_0^1(\hat{\mathbf{z}}) = \sum_m O_m^1(\hat{\mathbf{z}}) \langle 1m | R' | 10 \rangle \ , \tag{2.17}$$

where $\mathcal{R}'$ is the *inverse* of the rotation $\mathcal{R}$ defined in Eq. (1.3). We find $\mathcal{R}' = \mathcal{R}^{-1} = \mathcal{R}^+$, so that

$$O_0^1(\hat{\mathbf{h}}) = \sum \mathcal{D}_{0m}^{(1)*}(\hat{\mathbf{h}}, \hat{\mathbf{z}}) O_m^1(\hat{\mathbf{z}}) \tag{2.17a}$$

and

$$\langle \Gamma\alpha | O_0^1(\hat{\mathbf{h}}) | \Gamma\beta \rangle = \sum_m \mathcal{D}_{0m}^{(1)*}(\hat{\mathbf{h}}, \hat{\mathbf{z}}) \langle \Gamma\alpha | O_m^1 | \Gamma\beta \rangle \ . \tag{2.18}$$

This result is for single crystals. For polycrystalline samples we perform a powder average. In the present context this is done by averaging $R_\Gamma(\hat{\mathbf{h}})$ over all orientations of the local axes relative to the fixed field axes $[\hat{\mathscr{E}}, -(\hat{\mathscr{E}} \times \hat{\mathbf{h}}), \hat{\mathbf{h}}]$,

$$\begin{aligned} \bar{R}_\Gamma &= \frac{1}{4\pi} \int d\Omega_z |\langle \Gamma\alpha(\hat{\mathbf{z}}) | O_0^1(\hat{\mathbf{h}}) | \Gamma\beta(\hat{\mathbf{z}}) \rangle|^2 \\ &= \sum_{mm'} \langle \Gamma\alpha | O_m^1 | \Gamma\beta \rangle \langle \Gamma\beta | (O_{m'}^1)^* | \Gamma\alpha \rangle \\ &\quad \times \frac{1}{4\pi} \int d\Omega_h \mathcal{D}_{0m}^{(1)*}(\hat{\mathbf{h}}) \mathcal{D}_{0m'}^{(1)}(\hat{\mathbf{h}}) \ , \end{aligned} \tag{2.19}$$

where we average over directions of the field instead of local axes because it is irrelevant to the present application whether we rotate crystal-field states or the field direction. The average over the rotation matrix yields[4]

$$\frac{1}{4\pi} \int d\Omega_h \mathcal{D}_{0m}^{(1)*}(\hat{\mathbf{h}}) \mathcal{D}_{0m'}^{(1)}(\hat{\mathbf{h}}) = \tfrac{1}{3} \delta_{mm'} \ , \tag{2.20}$$

and the powder-averaged orbital factor Eq. (2.19) is

$$\bar{R}_\Gamma = \tfrac{1}{3} \sum_{\alpha\beta m} |\langle \Gamma\alpha | O_m^1 | \Gamma\beta \rangle|^2 \ . \tag{2.21}$$

This is to be compared with the single-crystal expression, see Eqs. (2.14) and (2.18),

$$R_\Gamma(\hat{\mathbf{h}}) = \sum_{\alpha\beta} \left| \sum_m \mathcal{D}_{0m}^{(1)*}(\hat{\mathbf{h}}, \hat{\mathbf{z}}) \langle \Gamma\alpha | O_m^1 | \Gamma\beta \rangle \right|^2 \ . \tag{2.22}$$

The second effect of crystal fields is to replace the free $4f$ electron Green's function G^{4f} by the crystal-field Green's function $G_\Gamma(H=0)$. There is no need for the index α, because in zero field the levels in the Γ manifold are not split and have the same Green's function, see Eq. (2.12). In the approximation within which we are working, viz., of considering only one crystal-field manifold, the Green's function $G_\Gamma(H=0)$ is identical to that for a state of fictitious anger momentum $\tilde{j}$, where $\tilde{j}$ is defined by the degeneracy of the manifold Γ,

$$N_\Gamma \equiv 2\tilde{j} + 1 \ , \tag{2.23}$$

and whose energy is E_Γ. So long as the crystal-field splitting Δ_{cr} is small so that $|E_\Gamma - E_F| \cong |E_f - E_F|$ where E_F is the Fermi energy, the sole effect of the crystal field on the Green's function is to replace $(2j+1)$ by N_Γ.

III. WEAK-COUPLING LIMIT

With the approximations made above we can obtain reasonable estimates of the conductivities σ_H and σ_N [Eqs. (2.2) and (2.3)] by replacing $(-\partial f^0/\partial\varepsilon)$ by $\delta(\varepsilon)$, so that the integrands are evaluated at $\varepsilon=0$, the Fermi level. In the limits of weak coupling ($T \gg T_K$, the Kondo temperature), and strong coupling ($T=0$ K) we have explicit forms for the Green's functions entering σ_H and σ_N. For intermediate cases only numerical results are possible and we do not consider them here.

In the limit of weak coupling the Green's functions for a free $4f$ electron [Eqs. (2.15)] are[6,7]

$$G_0^{4f}(\varepsilon \sim 0) = \frac{1}{|E_f|} \left[1 - i(2j+1) \frac{\Gamma}{|E_f|} \right] , \tag{3.1a}$$

$$\begin{aligned} G_1^{4f}(\varepsilon \sim 0) = &- \frac{\frac{1}{3} j(j+1)}{|E_f|} \frac{g_J \mu_B H}{k_B T} \\ &\times \left[1 - (i/2)(2j+1) \frac{\Gamma}{|E_f|} \right] , \end{aligned} \tag{3.1b}$$

and

$$\partial_k G_1^{4f}(\varepsilon \sim 0) = \frac{2}{k_F} \frac{E_F}{|E_f|} \left[\frac{1}{3} j(j+1) \frac{g_J \mu_B H}{k_B T} \right] G_0^{4f}(\varepsilon \sim 0) \ , \tag{3.1c}$$

where Γ is a measure of the strength of the Anderson mixing parameter, $\Gamma \equiv \pi N(0) |V_k|^2$, $|E_f|$ is the energy of the $4f$ electron relative to the Fermi level, E_F is the Fermi energy, and k_F is the Fermi momentum. To obtain the crystal-field Green's functions [Eqs. (2.13)], we replace $2j+1$ by N_Γ, $|E_f|$ by $|E_\Gamma|$, and $\frac{1}{3}j(j+1)$, which is just $R_0/(2j+1)$, by R_Γ/N_Γ for a single crystal or $\bar{R}_\Gamma/N_\Gamma$ for a polycrystalline sample. We find

$$G_0^\Gamma(\varepsilon\sim 0)=\frac{1}{|E_\Gamma|}\left[1-iN_\Gamma\frac{\Gamma}{|E_\Gamma|}\right], \tag{3.2a}$$

$$G_1^\Gamma(\varepsilon\sim 0)=-\frac{R_\Gamma}{N_\Gamma|E_\Gamma|}\frac{g_J\mu_B H}{k_B T}\left[1-\frac{i}{2}N_\Gamma\frac{\Gamma}{|E_\Gamma|}\right], \tag{3.2b}$$

and

$$\partial_k G_1^\Gamma(\varepsilon\sim 0)=\frac{2}{k_F}\frac{E_F}{|E_\Gamma|}\frac{R_\Gamma}{N_\Gamma}\frac{g_J\mu_B H}{k_B T}G_0^\Gamma(\varepsilon\sim 0)\ . \tag{3.2c}$$

By placing these results in the conductivities, Eqs. (2.2) and (2.3), and by setting $v(0)/k_F=\hbar/m$, and $mv^2(0)=2E_F$ we find the Hall conductivity in weak coupling is

$$\sigma_H^\Gamma=-\frac{16}{49}\frac{n(0)e^2\hbar}{m}\frac{g_J\mu_B H}{k_B T}\frac{|E_\Gamma|}{\Delta_\Gamma}\frac{R_\Gamma}{N_\Gamma}\sin\eta_2$$
$$\times\left\{\frac{3\pi z N_\Gamma}{4c_i}\frac{|E_\Gamma|}{\Delta_\Gamma}\left[\cos\eta_2+2\frac{|E_\Gamma|}{\Delta_\Gamma}\sin\eta_2\right]\right.$$
$$-\left[6-2\frac{E_F}{|E_\Gamma|}\right]\cos\eta_2$$
$$\left.+\left[3-2\frac{E_F}{|E_\Gamma|}\right]\frac{\Delta_\Gamma}{|E_\Gamma|}\sin\eta_2\right\}\ . \tag{3.3}$$

We set $N(0)E_F=3z/4$ by using the free electron density of states where z is the number of conduction electrons per Kondo ion, and $\Delta_\Gamma\equiv N_\Gamma\Gamma$. The normal conductivity Eq. (2.3) reduces to

$$\sigma_N^\Gamma=2N_\Gamma\frac{\pi N(0)ne^2\hbar}{mc_i}\frac{|E_\Gamma|^2}{\Delta_\Gamma^2}\ . \tag{3.4}$$

By placing these conductivities in Eq. (2.1) we find the Hall constant in the limit of weak coupling is

$$R_H^\Gamma=-\frac{6}{49}\frac{mc_i/N_\Gamma}{\pi N(0)ne^2\hbar}\left[\frac{\Delta_\Gamma}{|E_\Gamma|}\right]^2\frac{R_\Gamma}{N_\Gamma}\frac{g_J\mu_B}{k_B T}\sin\eta_2$$
$$\times\left\{\left[\cos\eta_2+2\frac{|E_\Gamma|}{\Delta_\Gamma}\sin\eta_2\right]-\frac{4c_i}{3\pi z N_\Gamma}\frac{\Delta_\Gamma}{|E_\Gamma|}\left[\left[6-2\frac{E_F}{|E_\Gamma|}\right]\cos\eta_2-\left[3-2\frac{E_F}{|E_\Gamma|}\right]\frac{\Delta_\Gamma}{|E_\Gamma|}\sin\eta_2\right]\right\}\ . \tag{3.5}$$

In the limit of weak coupling we can express the Hall constant in terms of the normal resistivity

$$\rho^\Gamma=(\sigma_N^\Gamma)^{-1}$$
$$=\frac{N_\Gamma}{2}\frac{mc_i}{\pi N(0)ne^2\hbar}\left[\frac{\Gamma}{|E_\Gamma|}\right]^2, \tag{3.6}$$

and the magnetic susceptibility

$$\chi^\Gamma=\frac{(g_J\mu_B)^2}{k_B T}\frac{R_\Gamma}{N_\Gamma}\ . \tag{3.7}$$

In terms of these quantities the dimensionless ratio

$$f_\Gamma\equiv g_J\mu_B\frac{R_H^\Gamma}{\rho^\Gamma\chi^\Gamma} \tag{3.8a}$$

is given as

$$f_\Gamma=f_\Gamma^{\rm sk}+f_\Gamma^{\rm av}\ , \tag{3.8b}$$

where

$$f_\Gamma^{\rm sk}=-\tfrac{12}{49}\sin\eta_2\left[\cos\eta_2+\frac{2|E_\Gamma|}{\Delta_\Gamma}\sin\eta_2\right] \tag{3.8c}$$

$$\cong-\tfrac{12}{49}\sin\eta_2\cos\eta_2 \tag{3.8d}$$

is the skew scattering contribution, and

$$f_\Gamma^{\rm av}=\frac{16c_i}{49\pi z N_\Gamma}\sin\eta_2\frac{\Delta_\Gamma}{|E_\Gamma|}$$
$$\times\left[\left[6-2\frac{E_F}{|E_\Gamma|}\right]\cos\eta_2\right.$$
$$\left.-\frac{\Delta_\Gamma}{|E_\Gamma|}\left[3-2\frac{E_F}{|E_\Gamma|}\right]\sin\eta_2\right] \tag{3.8e}$$

is the anomalous velocity contribution.

We note for $T\gg T_K$ the ratio f_Γ is relatively insensitive to the effects of crystal fields, at least under the conditions that $|\eta_2|\ll 1$ and $\Delta_\Gamma/|E_\Gamma|\ll 1$ which make the skew scattering term Eq. (3.8c) the dominant one.[6] Therefore we conclude that the main effects of crystal fields on the extraordinary Hall effect are accounted for by the changes of the resistivity (3.6), and of the susceptibility, Eq. (3.7).

IV. STRONG COUPLING

Now, let us see how these results are modified when we go to the limit of strong coupling, specifically $T=0$ K. The Green's functions Eq. (2.13) take the form[6]

$$G_0^\Gamma=-\frac{N_\Gamma}{\Gamma}e^{i\eta_\Gamma}\sin\eta_\Gamma+O(H^2)\ , \tag{4.1a}$$

$$G_1^\Gamma = e^{i2\eta_\Gamma}\sin^2\eta_\Gamma \frac{R_\Gamma}{\Gamma}\frac{g_J\mu_B H}{\Gamma^*} + O(H^3)\ , \tag{4.1b}$$

and

$$\partial_k G_1^\Gamma = \frac{2}{k_F}\left[\frac{E_F}{\Gamma^*}\right](2e^{i\eta_\Gamma}\sin\eta_\Gamma - A\pi/N_\Gamma)G_1^\Gamma + O(H^3)\ , \tag{4.1c}$$

where we used $(\partial\varepsilon_k/\partial k)|_{E_F} = 2E_F/k_F$, A is a constant of order one, and $\Gamma^* = \Gamma Z$ is the magnetic Kondo resonance scale where Z is a renormalization constant.[6,8] At $T=0$ K the phase shift at the Fermi surface $\eta_\Gamma(0)$ is given by the Friedel-Langreth rule[8]

$$\eta_\Gamma(0) = \frac{\pi n_f(0)}{N_\Gamma}\ , \tag{4.2}$$

where $n_f(0)$ is the number of electrons in the $4f$ state at $T=0$ K. This is one in the absence of mixing. In the Kondo regime $n_f(0)$ is less than but close to one. By setting $v(0)/k_F = \hbar/m$ and $v^2(0) = 2E_F/m$ we find the Hall conductivity Eq. (2.2) at $T=0$ K is

$$\sigma_H^\Gamma = -\frac{32}{49}\frac{n(0)e^2\hbar}{m}\frac{g_J\mu_B H}{\Gamma^*}\frac{R_\Gamma}{N_\Gamma}\sin\eta_2 \times\left\{\frac{3\pi z}{4c_i N_\Gamma}\frac{\sin[2\eta_\Gamma(0)-\eta_2]}{\sin^2\eta_\Gamma(0)} + \frac{E_F}{\Gamma^*}\left[2\sin\eta_\Gamma(0)\cos[3\eta_\Gamma(0)-\eta_2] - \frac{A\pi}{N_\Gamma}\cos[2\eta_\Gamma(0)-\eta_2]\right]\right\}\ , \tag{4.3}$$

where we have neglected the term $6G_1/k_F$ in Eq. (2.6) because the dominant term, $\partial_k G_1 \sim (E_F/\Gamma^*)G_1/k_F$ where $E_F/\Gamma^* \gg 1$, is much larger. The normal conductivity Eq. (2.3) at $T=0$ K is

$$\sigma_N^\Gamma = \frac{2}{N_\Gamma}\frac{\pi N(0)ne^2\hbar}{mc_i}\frac{1}{\sin^2\eta_\Gamma(0)} \tag{4.4}$$

so that the resistivity is

$$\rho^\Gamma = \frac{N_\Gamma}{2}\frac{mc_i}{\pi N(0)ne^2\hbar}\sin^2\eta_\Gamma(0)\ . \tag{4.5}$$

By placing these conductivities in Eq. (2.1) we find the Hall constant at $T=0$ K is

$$R_H^\Gamma(0) = -\tfrac{18}{49}R_\Gamma\frac{g_J\mu_B}{\Gamma^*}\frac{m}{\pi N(0)ne^2\hbar}\sin\eta_2\sin^2\eta_\Gamma(0) \times\left\{c_i\sin[2\eta_\Gamma(0)-\eta_2] + \frac{4N_\Gamma c_i^2}{3\pi z}\sin^2\eta_\Gamma(0)\frac{E_F}{\Gamma^*}\left[2\sin\eta_\Gamma(0)\cos[3\eta_\Gamma(0)-\eta_2] - \frac{A\pi}{N_\Gamma}\cos[2\eta_\Gamma(0)-\eta_2]\right]\right\}\ . \tag{4.6}$$

At $T=0$ K the magnetic susceptibility is

$$\chi_\Gamma(0) = \frac{(g_J\mu_B)^2\sin^2\eta_\Gamma(0)}{\pi\Gamma^*}R_\Gamma \tag{4.7}$$

so that the dimensionless ratio f_Γ, see Eq. (3.8a), at $T=0$ K is

$$f_\Gamma(0) = f_\Gamma^{\rm sk}(0) + f_\Gamma^{\rm av}(0)\ , \tag{4.8a}$$

where

$$f_\Gamma^{\rm sk}(0) = -\tfrac{24}{49}\pi\sin\eta_2\frac{\sin[2\eta_\Gamma(0)-\eta_2]}{N_\Gamma\sin^2\eta_\Gamma(0)} \tag{4.8b}$$

and

$$f_\Gamma^{\rm av}(0) = -\frac{32}{49}\frac{c_i}{z}\frac{E_F}{\Gamma^*}\left[2\sin\eta_\Gamma(0)\cos[3\eta_\Gamma(0)-\eta_2] - \frac{A\pi}{N_\Gamma}\cos[2\eta_\Gamma(0)-\eta_2]\right] \tag{4.8c}$$

are the skew scattering and anomalous velocity contributions, respectively.

In contrast to the results for $T \gg T_K$ the ratio $f_\Gamma(0)$ is sensitive to the effects of crystal fields as reflected by $\eta_\Gamma(0)$ and N_Γ. That is to say, with respect to the case without crystal field for which $\eta_j = \pi n_f(0)/(2j+1)$ this is now replaced by $\eta_\Gamma(0)$, and $2j+1=6$ (for cerium) is replaced by N_Γ. We conclude that for $T \ll T_K$ the changes in the resistivity ρ and susceptibility χ Eqs. (4.5) and (4.7) are unable to completely account for the effects of crystal fields on the Hall constant. As we will see in the next section, while $\rho\chi$ is always reduced by the crystal field, the Hall constant can be enhanced by the increase of $f_\Gamma^{\rm av}(0)$. On the other hand, it must be pointed out that $f_\Gamma(0)$ is independent of the orientation of the magnetic field with respect to the crystal axes. When the orientation is changed (suppose the current remains fixed in one direction) the anisotropy of the Hall constant R_H is fully accounted for by the anisotropy of the susceptibility, i.e., $R_H^\Gamma(\hat{\mathbf{h}})$ depends on $\hat{\mathbf{h}}$ in the same way as $\chi_\Gamma(\hat{\mathbf{h}})$ does. Similarly, if one compares the Hall constant of a polycrystalline sample $\bar{R}_H^\Gamma$ with that of a single crystal $R_H^\Gamma(\hat{\mathbf{h}})$ one expects

$$\frac{R_H^\Gamma(\hat{\mathbf{h}})}{\bar{R}_H^\Gamma} = \frac{\chi_\Gamma(\hat{\mathbf{h}})}{\bar{\chi}_\Gamma}\ . \tag{4.9}$$

V. CRYSTAL-FIELD DOUBLET

To obtain an idea of the magnitude of these crystal-field effects, we consider a crystal field with rhombic symmetry; the 4f $j=\frac{5}{2}$ state of cerium, for example, is split into three doublets. We focus on the ground doublet which is written as

$$|\Gamma^{\pm}\rangle = a|\pm\tfrac{5}{2}\rangle + b|\mp\tfrac{3}{2}\rangle + c|\pm\tfrac{1}{2}\rangle \ , \tag{5.1}$$

where $a^2+b^2+c^2=1$. To simplify the calculation for single crystals we assume the magnetic field is parallel to the local symmetry $\hat{\mathbf{z}}$ axis. In this case there is no need to rotate the field operator $O_0^1(\hat{\mathbf{h}})$, see Eq. (2.17a), because it acts on states [Eq. (5.1)] written in the same axis of quantization, i.e., $\mathcal{D}_{0m}^{(1)}(\hat{\mathbf{h}},\hat{\mathbf{z}})=\delta_{m0}$ in Eq. (2.22) and $O_0^1(\hat{\mathbf{h}})=J_h=J_z$. We first calculate the orbital factor R_Γ which fixes the reduction of the product $\rho\chi$. From Eq. (2.14) we write R_Γ as

$$R_\Gamma = \sum_{\alpha\beta} |\langle \Gamma\alpha|J_z|\Gamma\beta\rangle|^2 \ . \tag{5.2}$$

For the crystal-field doublet, Eq. (5.1), we find

$$\langle \Gamma\alpha|J_z|\Gamma\beta\rangle = \sum_m m|\langle m|\Gamma\alpha\rangle|^2\delta_{\alpha\beta} \ , \tag{5.3}$$

so that

$$R_\Gamma = \sum_{\alpha=\pm}\left[\sum_m m|\langle m|\Gamma\alpha\rangle|^2\right]^2 = 2(\tfrac{5}{2}a^2-\tfrac{3}{2}b^2+\tfrac{1}{2}c^2)^2 \ . \tag{5.4}$$

For a polycrystalline sample the orbital factor is given by Eq. (2.21). This can be written as

$$\bar{R}_\Gamma = \tfrac{1}{3}\sum_{\alpha\beta}(|\langle\Gamma\alpha|J_z|\Gamma\beta\rangle|^2 + |\langle\Gamma\alpha|J_+|\Gamma\beta\rangle|^2) \ , \tag{5.5}$$

where we used[3] $O_{\pm1}^1=\mp(1/\sqrt{2})J_\pm$. For the doublet wave functions [Eq. (5.1)] we find

$$\bar{R}_\Gamma = \tfrac{1}{3}[2(\tfrac{5}{2}a^2-\tfrac{3}{2}b^2+\tfrac{1}{2}c^2)^2 + (2\sqrt{5}ab+3c^2)^2+32b^2c^2] \ . \tag{5.6}$$

To compare the single-crystal and powder-averaged orbital factors we consider axial crystal fields where only a, b, or c is nonzero. For a or b nonzero the powder-averaged $\bar{R}_\Gamma$ is $\frac{1}{3}$ that of the single crystal, while for $c\neq 0$, $\bar{R}_\Gamma$ is *enhanced* by $\frac{19}{3}$ or nearly sixfold due to powder averaging. The enhancement comes from the matrix element $\langle\frac{1}{2}|J_+|-\frac{1}{2}\rangle$ that is not present in the single-crystal orbital factor, see Eqs. (5.2) and (5.5). Even though there is a large enhancement $\bar{R}_\Gamma(a=b=0,c=1)=\frac{19}{6}$, the orbital factor is still small compared to the free 4f $j=\frac{5}{2}$ electron value $R_0=\frac{35}{2}$. That is, the overall reduction for a powder-averaged $|\pm\frac{1}{2}\rangle$ crystal-field doublet is $\frac{19}{105}$.

For the crystal-field doublet Eq. (5.1) and for $T\gg T_K$ we find from Eqs. (3.6) and (3.7) that, with respect to the case without crystal field, the magnetic resistivity ρ, the susceptibility for $\hat{\mathbf{h}}$ along $\hat{\mathbf{z}}$, $\chi(\hat{\mathbf{z}})$, and the susceptibility of a polycrystalline sample $\bar{\chi}$ are, respectively, multiplied by

$$r_\rho = \tfrac{1}{3} \ , \tag{5.7a}$$

$$r_{\chi(\hat{\mathbf{z}})} = \tfrac{12}{35}(\tfrac{5}{2}a^2-\tfrac{3}{2}b^2+\tfrac{1}{2}c^2)^2 \ , \tag{5.7b}$$

and

$$r_{\bar{\chi}} = \tfrac{4}{35}[(\tfrac{5}{2}a^2-\tfrac{3}{2}b^2+\tfrac{1}{2}c^2)^2 + \tfrac{1}{2}(2\sqrt{5}ab+3c^2)^2+16b^2c^2] \ , \tag{5.7c}$$

where we set $N_\Gamma=2$, and $j=\frac{5}{2}$. At $T=0$ K we find from Eqs. (4.5) and (4.7) that with respect to the case without crystal field ρ, $\chi(\hat{\mathbf{z}})$ and $\bar{\chi}$ are, respectively, multiplied by

$$r_\rho(0) = r_\rho\frac{\sin^2\eta_\Gamma(0)}{\sin^2 n_j(0)} \ , \tag{5.8a}$$

$$r_{\chi(\hat{\mathbf{z}})}(0) = \tfrac{1}{3}r_{\chi(\hat{\mathbf{z}})}\frac{\sin^2\eta_\Gamma(0)}{\sin^2\eta_j(0)} \ , \tag{5.8b}$$

and

$$r_{\bar{\chi}}(0) = \tfrac{1}{3}r_{\bar{\chi}}\frac{\sin^2\eta_\Gamma(0)}{\sin\eta_j(0)} \ , \tag{5.8c}$$

where $n_\Gamma(0)=\pi n_f(0)/2$ and $\eta_j(0)=\pi n_f(0)/6$. The reduction factor for the product $\rho\chi$ is derived from the above results in a straightforward manner.

In addition we have calculated the factor f_Γ [Eq. (3.8a)] for a crystal-field doublet. For $T\gg T_K$, as stated in Sec. III, f_Γ is insensitive to the effects of crystal fields, and approximately takes the same value as in the case without a crystal field. For $T\ll T_K$, $f_\Gamma(0)$ depends on the degeneracy of the crystal-field ground doublet. For a doublet, $f_\Gamma(0)$ is obtained from Eq. (4.8) with $N_\Gamma=2$ and $\eta_\Gamma(0)=\pi n_f(0)/2$, whereas the corresponding ratio $f_j(0)$ in the case without crystal field is obtained from Eq. (4.8) with

$$N_j=2j+1=6 \text{ and } \eta_j(0)=\pi n_f(0)/6 \ .$$

We have also calculated the ratios $f_\Gamma^{\text{sk}}(0)/f_j^{\text{sk}}(0)$ and $f_\Gamma^{\text{av}}(0)/f_j^{\text{av}}(0)$ as a function of $n_f(0)$ in the range $0.8\le n_f(0)\le 1$ and for $\eta_2=-0.1$ rad, which is typical of the order of magnitude of the phase shifts η_2 derived from experimental data.[2]

In Fig. 1(a) we display the results of our calculation for the skew scattering contribution to the Hall constant. For the majority of the range of $n_f(0)$ studied, i.e., $n_f(0)<0.97$, we find $f_\Gamma^{\text{sk}}(0)/f_j^{\text{sk}}(0)$ is less than one, i.e., the crystal field *reduces* the skew scattering contribution to f for $T\ll T_K$. In a very narrow range of $n_f(0)$, $0.97<n_f(0)\le 1$, we find the crystal field changes the sign of $f^{\text{sk}}(0)$, i.e., the skew scattering contribution is negative at low temperatures [the value $n_f(0)=0.97$ is for $\eta_2=-0.1$ and slightly different values would be obtained for other values of η_2].

Most cerium Kondo systems appear to correspond to the range $n_f(0)<0.97$ for which the skew scattering contribution remains positive. For these systems the reduction of $f^{\text{sk}}(0)$ by the crystal field is necessary to quantitatively account for the experimental results. As an example we consider CeAl$_3$ for which a good fit with experi-

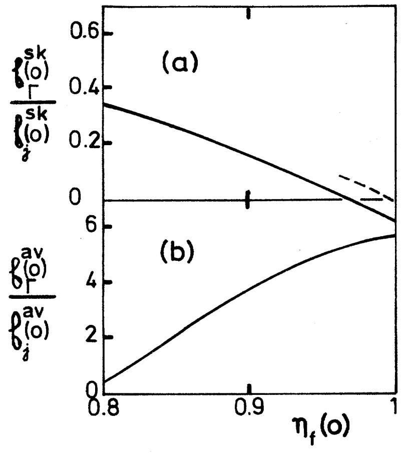

FIG. 1. Crystal-field effects on the contributions from skew scattering and anomalous velocity to the coefficient f_Γ, Eq. (3.8a), for $T \ll T_K$ and for a doublet ground state. (a) $f_\Gamma^{\mathrm{sk}}(0)/f_j^{\mathrm{sk}}(0)$ vs the number of f electrons $n_f(0)$. (b) $f_\Gamma^{\mathrm{av}}(0)/f_j^{\mathrm{av}}(0)$ vs $n_f(0)$. The calculations were based on Eq. (4.8) with $N_\Gamma=2$, $N_j=6$, and $\eta_2=-0.1$. The results are weakly dependent on the value of η_2, e.g., the dashed line indicates the shift of the curve in (a) when η_2 is reduced by a factor of 10.

mental data is obtained with the values[2] $\gamma_1=0.075$ K/T for $T \gg T_K$ and $\gamma_2=0.097$ K/T for $T < T_K$ where the coefficient γ is defined as

$$\gamma \equiv R_H/\rho\chi = \frac{g_J \mu_B j(j+1) f}{3 k_B T} \; . \tag{5.9}$$

The value of γ_1 is accounted for by Eq. (48) of Ref. 2, or equivalently our Eq. (3.8c), with $\eta_2=-0.078$ rad, independent of any assumption about the crystal field. The ratio γ_2/γ_1 and thereby γ_2 is accounted for by using our results for a doublet ground state,[9] i.e., from Eqs. (4.8b) and (3.8c),

$$\frac{\gamma_2}{\gamma_1} \equiv \frac{f_\Gamma^{\mathrm{sk}}(0)}{f_\Gamma^{\mathrm{sk}}(T \gg T_k)} = \pi \frac{\sin[2\eta_\Gamma(0)-\eta_2]}{\sin^2 \eta_\Gamma(0)} \tag{5.10}$$

with $\eta_\Gamma(0)=1.33$ rad. This corresponds to $n_f(0)=0.85$, which is a plausible value for the number of f electrons. In contrast the experimental data for $\mathrm{CeAl_3}$ cannot be accounted for by the expression for γ_2/γ_1 in the absence of crystal-field effects:

$$\frac{\gamma_2}{\gamma_1} \equiv \frac{f_j^{\mathrm{sk}}(0)}{f_j^{\mathrm{sk}}(T \gg T_k)} = \pi \frac{\sin[2\eta_j(0)-\eta_2]}{3\sin^2 \eta_j(0)} \; . \tag{5.11}$$

For realistic values of $n_f(0)$ this expression predicts values of γ_2/γ_1 that are too large. In the same way the experimental results for CePtSi (Ref. 10) yield $\gamma_1 \simeq 0.028$ K/T and $\gamma_2/\gamma_1 \simeq 1.2$. They can be accounted for only by a doublet ground state with $\eta_2 \simeq 0.029$ and $n_f(0) \simeq 0.87$.

To summarize, in the absence of crystal fields one would expect a significant increase for f or γ below T_K, e.g., see Fig. 1 of Ref. 5. With the crystal field we can understand why the values of f or γ are not significantly different above and below T_K. Therefore, in general, it is necessary to include the effects of the crystal field for a quantitative interpretation of the data on the Hall constant.

The case of $\mathrm{CeCu_6}$ (Refs. 1 and 11) is less clear. Here the Hall effect exhibits a positive maximum at 10 K, becomes negative below 3 K, and presents a negative minimum at about 0.1 K. One could posit that the change of sign is that expected for f below T_K in the case where $n_f(0)$ is very close to one. However, the change of sign is no longer observed when La is substituted for Ce, and the effects of coherence at low temperatures are suppressed. Thus we are inclined to ascribe the change of sign to the development of coherence. If the behavior of R_H only at relatively high temperatures is interpreted in our single impurity model of skew scattering we find[2]

$$\gamma_1 \simeq 0.082 \text{ K/T} \text{ and } \gamma_2/\gamma_1 \simeq 0.82 \; ,$$

which is consistent with a doublet ground state with $\eta_2=-0.09$ and $n_f(0) \simeq 0.87$.

Finally with $\eta_2 \simeq -0.1$ and for the range $0.8 \le n_f(0) \le 1$ the anomalous velocity contribution to the Hall constant is negative without crystal fields and positive for a crystal-field doublet. Data on the anomalous velocity contribution exist only for $\mathrm{CeCu_{5-x}Al_x}$ alloys;[12] this indicates a positive sign in agreement with out results for a crystal-field doublet. The absolute value of the anomalous velocity contribution, which is significant only for $T < T_K$,[6] is *enhanced* at low temperatures by the crystal field as shown in Fig. 1(b).

VI. SUMMARY

We have calculated the effects of the crystal field on the skew scattering and anomalous velocity contributions to the Hall effect for *incoherent* Kondo systems. Our results are summarized as follows.

(1) For $T \gg T_K$ the Hall constant is affected by the crystal field in the same way as the product of the magnetic resistivity ρ and the susceptibility χ. In other words, the coefficient f_Γ [Eq. (3.8a)] is independent of the crystal-field splitting.

(2) For $T \ll T_K$ not only are ρ and χ, but also $f_\Gamma(0)$ is affected by the crystal field. If the crystal-field ground state is a doublet the skew scattering contribution to f is generally reduced by the crystal field, while the anomalous velocity contribution is enhanced. It is worth noting that f depends only on the degeneracy of the ground state, so that if the applied field is rotated the anisotropy of the Hall constant R_H is fixed by the anisotropy of the susceptibility χ.[13]

(3) It is necessary to take into account crystal-field effects in order to obtain a *quantitative* interpretation of the Hall effect data in Kondo compounds such as $\mathrm{CeAl_3}$, CePtSi, and $\mathrm{CeCu_6}$.

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