Equilibrium current vortices in rare-earth–doped simple metals

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Dilute alloys of rare earths have played a vital role in understanding magnetic phenomena. Here, we model the ground state of dilute 4f rare-earth impurities in light metals. When the 4f subshells are open (but not half-filled), the spin-orbit coupling imprints a rotational charge current of conduction electrons around rare-earth atoms. The sign and amplitude of the current oscillate similar to the RKKY spin polarization. We compute the observable effect, namely the Ørsted field generated by the current vortices and the Knight shift.

Introduction.— The conversion of spin currents into excitations of the charge, phonon, photon, or magnetization degrees of freedom and vice versa often involves spin-orbit interactions (SOI)\(^2\). Examples are the spin-orbit torques\(^3\), charge pumping\(^4\), magnetoelastic interactions\(^5\), and electric-field-induced magnetization dynamics\(^6,7\). The large intraatomic SOI that governs the local moments of lanthanides with partially filled 4f subshells causes novel spin charge coupling\(^8\) and affects device parameters such as the magnetic damping\(^9,10\). Rare-earth (RE) ions with local magnetic moments can partially or entirely substitute the non-magnetic yttrium in the ferrimagnetic insulator yttrium iron garnet Y\(_3\)Fe\(_5\)O\(_{12}\) (YIG)\(^11\). The different magnetic sublattices of RE-IG strongly modify the magnetic properties\(^12,14\), causing, for example, different compensation points for the magnetic and total angular moments\(^16\). A more complex phenomenon is a double radius and shielding by the more extended and fully occupied 5s and 5p orbitals. This does not exclude a significant exchange interaction: the conduction electrons of Pt contacts activate the Gd moments in gadolinium gallium garnet (GGG)\(^31\). The exchange interaction between a local spin with conduction electrons of a metal host generates RKKY spin-density oscillations. Tri-positive lanthanides have electronic configuration [Xe] 4f\(^n\), where the number of 4f electrons \(n\) goes from \(n = 0\) for La\(^{3+}\) to...

Fig. 1. Schematic representation of the rotational velocity field \(v(r)\) around a rare-earth ion with orbital moment \(\mathbf{L}\) embedded in a free electron gas.
n = 14 for Lu$^{3+}$. Except for n = 0 (La$^{3+}$), n = 7 (Gd$^{3+}$), and n = 14 (Lu$^{3+}$), the intra-atomic spin-orbit interaction critically affects the magnetic properties. Here we address a spin-orbit proximity effect of such a magnetic moment embedded in a Fermi sea.

A partially occupied 4f subshell is characterized by a spin $S$, an orbital moment $L$, and a total angular moment $J = L + S$ [23][32]. For the basis $|\Psi\rangle = |S, L, J, J_z\rangle$, $S^2|\Psi\rangle = \hbar^2S(S+1)|\Psi\rangle$, $L^2|\Psi\rangle = \hbar^2L(L+1)|\Psi\rangle$, $J^2|\Psi\rangle = \hbar^2J(J+1)|\Psi\rangle$, $J_z|\Psi\rangle = \hbar J_z|\Psi\rangle$, where $\hbar$ is the reduced Planck constant. Hund’s rules specify the quantum numbers $S$, $L$, and $J$ of the ground state manifold, while $J_z$ depends on the applied magnetic and electric fields. Within a manifold of constant $S$, $L$, and $J$, the Wigner-Eckart theorem ensures collinearity of all angular moment vectors: $|\Psi\rangle = (g_J-1)J, L = (2-g_J)J$, and $L + 2S = g_JJ$, where $g_J = 3/2 + |S(S+1) - L(L+1)|/[2J(J+1)]$ is the Landé $g$-factor.

We model the system as a single RE local moment embedded into a free electron gas, which is appropriate for most dilute alloys. Conduction electrons interact with the rare-earth spin and orbital moment via the Kondo Hamiltonian [25]. Here, we address equilibrium properties that are affected by the spin-independent skew scattering but disregard external current-induced phenomena such as the spin-Hall effect. We operate in a regime of weak coupling, disregarding higher-order terms in the exchange coupling $J_{ex}$ that cause, e.g., the Kondo effect. The strongly localized 4f orbital radius governs the spatial extent of the coupling. When the 4f orbital radius is much smaller than the typical wavelength of the conduction electrons, the moment couples to free electrons by a contact interaction. We treat $J$, $S$, and $L$ as classical vectors.

The $s$-$f$ exchange interaction in the Kondo Hamiltonian is similar to the $s$-$d$ Hamiltonian for 3d-transition metals [28][30][33][34]. It reads

$$H_{sf} = -\frac{J_{ex}}{\hbar^2}\delta^{4f}(\mathbf{r})\mathbf{S} \cdot \frac{\hbar\sigma}{2},$$

where $\sigma$ is the vector of Pauli matrices and $\delta^{4f}(\mathbf{r})$ is a Dirac delta representing the localized 4f subshell. In a free electron gas with Fermi wave number $k_F$, the exchange constant [28] is $J_{ex} = 2e^2A_0/(7e_0k_F^2)$, where the radial integral [28]

$$A_0(n) = \int_0^\infty dx_1 x_1^2 \int_0^\infty dx_2 x_2^2 \int_0^\infty dx_3 x_3^2 j_0(x_1)j_0(x_2)x_3^{n+1} R(r_1) R(r_2) R(r_3),$$

with $x_1 = k_FR_1$, $x_2 = k_FR_2$, $x_3 = \min(x_1,x_2)$, and $x_3 = \max(x_1,x_2)$. $A_0(n)$ can be evaluated numerically using a Slater-type orbital for the radial part of the 4f wave function $R(r) \sim r^3e^{-r/\alpha}$ normalized over a large volume, $\int_0^\infty dr r^2 R^2(r) = 1$. The constant $\alpha$ is related to the 4f radius by $r = \int_0^\infty dr r^3 R^2(r) = 9\alpha/2$. With $k_F = 1.75$ Å$^{-1}$ for Al and $r = 0.6$ Å [23], $A_0(0) = 0.33$ and $J_{ex} = 5.6$ eV Å$^3$.

The spin-independent so-called skew scattering generates an anomalous Hall effect in metals with RE impurities [24][28][35], but as shown below, it also affects the ground state. Its Hamiltonian reads [25]

$$H_{skew} = \mathbf{L} \cdot (|\nabla \eta(\mathbf{r})| \times \frac{1}{i} \mathbf{L}) \mathbb{I}_{2\times2},$$

where $\mathbb{I}_{2\times2}$ is the identity matrix in Pauli spin space, and $\eta(\mathbf{r}) = \eta_0\delta^{4f}(\mathbf{r})$ with $\eta_0 = 9e^2|A_2(1) - (5/9)A_4(1)|/(140e_0\hbar k_F^2)$. For the parameters introduced above, $A_2(1) \approx 0.0855$, $A_4(1) = 0.056$, and $\eta_0 k_F^2 = 0.21$ eV Å$^3$. Both exchange and skew-scattering interactions are active in a volume $V_{4f} \lesssim 10$ Å$^3$. The energy scales $\langle H_{skew}\rangle / \eta_0 k_F^2 = O(10$ meV) and $\langle H_{sf}\rangle \sim J_{ex}/V_{4f} = O(100$ meV) are consistent with published values extracted from experiments, such as the Knight shift [36], electron spin resonance [37], and magnetoresistance [38][40]. $H_{skew}$ deflects free electrons via an effective local force caused by the 4f subshell with orbital angular momentum $\mathbf{L}$. Equation (2) does not contain an essential SOI parameter because we operate in the limit of large 4f spin-orbit interaction, which is essential to generate a finite $|\mathbf{L}|$.

The 4f RE impurities in noble metals hybridize with 5d virtual-bound states of the conduction electrons [41], which can be parameterized in terms of phase shifts of angular momentum scattering channels [42]. The enhancement of the magnetic moments of pure RE metals [43][45] and in RE-doped Ag and Au [27][46] has been attributed to those 5d virtual bound states. Here we focus on the spin and orbital polarization induced by the Kondo Hamiltonian on conduction electrons that we describe by plane waves without truncating an expansion into spherical harmonics. To leading order in the contact interaction, we may discard hybridization and orthogonalization corrections.

Next, we discuss the RKKY spin polarization due to the $H_{sf}$ and the response induced by $H_{skew}$. We focus on ions with partially filled 4f shells. Gd$^{3+}$ ($L = 0$) can create a RKKY spin polarization, but its $H_{skew}$ vanishes. We do not address Eu$^{3+}$ since its spin and orbital moment cancel in its ground ($J = 0$) but not in excited states.

**RKKY spin-density oscillations.** In the mean-field, local-density approximation, Eq. (1) for an RE moment at the origin $r = 0$ becomes

$$H_{sf} = -\frac{J_{ex}}{\hbar^2}\mathbf{s}(r = 0) \cdot \mathbf{S},$$

where $\mathbf{s}(\mathbf{r}) = \langle \Psi^+_N(\mathbf{r}) | \mathbf{S} | \Psi_N(\mathbf{r}) \rangle$ is the spin density of the conduction-electron wave function $\Psi_N$. For a static moment and to leading order in $J_{ex}$, we recover the RKKY spin density oscillations

$$\langle \mathbf{s}(\mathbf{r}) \rangle = \frac{J_{ex}}{\hbar^2} \chi(\mathbf{r}) \mathbf{S},$$

$$\chi(\mathbf{r}) = \frac{D_J \hbar^2}{16\pi^3} \left[ \sin(2k_FR) - \cos(2k_FR) \right],$$

where $D_J$ is the exchange constant [28].
where $\chi(r)$ is the susceptibility and $D_e = m_e k_F (\pi \hbar)^{-2}$ the density of states of the host metal at the Fermi energy. Figure 4(a) illustrates the characteristic RKKY oscillations in $r\chi(r)$ that contribute to the total spin magnetic moment $m_S$

$$m_S = -\gamma_0 g_S \int d^3r [S\delta(r) + (s)(r)] = -\gamma_0 g_S (1 + G_i^S) S,$$

where the bare $g$-factor is $g_S = 2$, $\gamma_0 = e/(2m_e)$ is the modulus of the gyromagnetic ratio, $-e$ is the electron charge, and the constant $G_i^S = J_\perp D_e/4$. For example, in Al, $G_i^S \approx 0.13$. The polarization cloud enhances the total spin magnetic moment and $g$-factor by $G_i^S$, which can be observed via the imaginary part of the spin-mixing conductance (effective field) at ferromagnet-normal metal interfaces or spin-dependent interfacial phase shifts at ferromagnet-superconductor interfaces.

**Rotational currents.** We now show that the Kondo Hamiltonian generates equilibrium charge-current vortices around the impurity. In Fourier representation with linear momentum $\mathbf{b}q$ and unperturbed wave function $\langle \mathbf{r} | q \rangle = e^{i\mathbf{r} \cdot \mathbf{q}} / \sqrt{\Omega}$, the matrix elements of the skew-type interaction read

$$\langle \mathbf{q} + \mathbf{k} | H_{skew} | \mathbf{q} \rangle = i\eta_0 \Omega^{-1} e^{-2k^2 a^2} (\mathbf{k} \times \mathbf{q}) \cdot \mathbf{L},$$

where $e^{-2k^2 a^2}$ cuts off an ultraviolet divergence of a delta-function potential. To leading order in $\eta_0$, we find a spin-independent velocity field of conduction electrons [19]:

$$\langle \mathbf{v}(\mathbf{r}) \rangle = \frac{\eta_0}{2\pi^3 \hbar} \frac{F(r)}{r} \mathbf{L} \times \hat{\mathbf{r}},$$

where

$$F(r) = \frac{1}{a r \sqrt{2\pi}} \int_0^\infty \frac{d r'}{r} e^{-r'^2/4a^2} f_1(r, r') f(r'),$$

$$f_1(r, r') = r' r \cos \left( \frac{r' r}{4a^2} \right) - 4a^2 \sinh \left( \frac{r' r}{4a^2} \right),$$

$$f(x/k_F) = \frac{2x (9 - 2x^2) \cos (2x) + (9 - 14x^2) \sin (2x)}{8(x/k_F)^6},$$

and $x = k_F r$. In the delta-function $a \to 0$, the response function $f$ should used in Equation (8) instead of $F$. Outside the 4f subshell, $F$ and $f$ are similar, see Fig. 4(b). $F$, $f$, and $\chi$ oscillate with wavenumber $2k_F$ and are approximated by $\cos (2k_F r) / r^3$ far from the atom. The integrated modulus of the velocity,

$$\bar{v}_s = \int |\langle \mathbf{v} \rangle| d^3r = 0.01 v_F \left( \frac{2 - g_f}{4} \right) \sqrt{J(J+1)},$$

is of the order of a percent of the Fermi velocity $v_F = \hbar k_F / m_e \sim 2 \times 10^8$ m/s and $\bar{v}_s \sim 10$ km/s for Al.

**FIG. 2.** Distribution of the spin and orbital polarizations close to a RE local moment in the free electron gas. (a) RKKY spin density oscillations $r\chi(r) / F_0$ normalized by $F_0 = \lim_{r \to 0} r\chi(r)$. (b) Distribution of the induced orbital moment obtained with the regularized and non-regularized response functions, $rF(r) / F_1$ and $rf(r) / F_2$, respectively, normalized by $F_1 = \lim_{r \to 0} rF(r)$ and $F_2 = \lim_{r \to 0} rf(r)$. The response is dominantly paramagnetic but oscillates with diamagnetic contributions. Far from the atom, the spin and orbital responses share an oscillating algebraic decay $\cos (2k_F r) / r^3$. As this figure illustrates, the distributions have a phase difference for small radius.

The radial density of the orbital angular momentum

$$\langle \mathbf{L} \rangle = \int \frac{d\phi d\theta \sin \theta}{4\pi} m_e \mathbf{r} \times \langle \mathbf{v}_s \rangle = \frac{m_e \eta_0 F(r)}{3\pi^3 \hbar} \mathbf{L},$$

is at equilibrium always collinear with $\mathbf{L}$ and parallel to it near the origin. Both the orbital density of the rotational current and the RKKY spin polarization decay algebraically and oscillate with increasing distance from the origin [see Fig. 4(b)]. Therefore, the current response has paramagnetic as well as diamagnetic contributions. Note that the spin and orbital radial distributions have a phase difference for small radius, as shown in Fig. 4(b).

The orbital magnetic moment is described by the electron gas

$$\mathbf{m}_L = -\gamma_0 g_L \int d^3r \left[ J(\mathbf{r}) + \langle \mathbf{L} \rangle \right] = -\gamma_0 g_L (1 + G_i^L) \mathbf{L},$$

where the bare orbital $g$-factor $g_L = 1$, and $G_i^L = 2m_e \eta_0 k_F / (3\pi^2 \hbar)$. For the present parameters we find $G_i^L \approx 3.2 \times 10^{-3}$. 


The current vortex induces an Ørsted field

$$\mathbf{B}_0(\mathbf{r}) = \frac{\mu_0}{4\pi} \int d^3r' \langle \mathbf{v}(\mathbf{r}') \rangle \times \frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3},$$

(15)

where $\mu_0 = 4\pi \times 10^{-7} \text{J/(mA}^2\text{)}$ is the magnetic permeability of free space. Far from the RE ion, $R \gg \langle r \rangle$,

$$\mathbf{B}_0(\mathbf{R}) = \frac{\mu_0}{4\pi} 3 (\mathbf{m}_{rc} \cdot \mathbf{R}) \mathbf{R} - R^2 \mathbf{m}_{rc}. \tag{16}$$

is the field generated by the magnetic dipole $\mathbf{m}_{rc} = -\gamma_0 G^L_z \mathbf{L}$. At the origin and for $k_F = 1.75/\AA$

$$\mathbf{B}_0(0) = -3.5 \times 10^{-7} e\gamma_0 \mu_0 a^{-6} (\mathbf{L}/\hbar),$$

$$= 0.06 \text{ T} \left( \frac{(2 - g_J) \sqrt{J(J + 1)}}{4} \right).$$

Figure 3 shows the $z$-component of this field as a function of distance $\mathbf{r} = x\mathbf{e}_x$, where the unit vectors $\mathbf{e}_x$ and $\mathbf{e}_z$ pointing along the $x$- and $z$-Cartesian axis, respectively. The field is negative close to the origin but turns positive and decays to zero in an oscillatory fashion.

The field at the origin, $\mathbf{B}_0(0)$, couples to the local nuclear spin by the Zeeman interaction, causing an additional Knight shift of the nuclear magnetic resonance. In an NMR experiment, a constant magnetic field $\mathbf{B}_0 \mathbf{e}_z$ polarizes the nuclear moments as well as the $4f$ moments along the $z$-direction. The Knight shift $K$ produced by the current and parameterized by the ratio of the internal and applied magnetic fields, at low temperatures ($T \lesssim 1 \text{ K}$) is

$$K \equiv \frac{|\mathbf{B}_0(0)|}{\mathbf{B}_0} = -0.3\% \left( \frac{(2 - g_J) \sqrt{J(J + 1)}}{4} \right) \left( \frac{20 \text{ T}}{B_0} \right). \tag{17}$$

where we assume full polarization of the magnetic moment of the $4f$ subshell. The nuclear magnetic resonance frequency is typically in the 100 MHz regime for applied (constant) fields of $B_0 \sim 10 \text{ T}$. For a given rf frequency we therefore predict different resonance magnetic fields for rare earth impurities in an insulating and metallic host. We hope that our results stimulate experiments that can identify the ground state current vortices.

Orbital contributions to the Knight shift have been predicted before [27,54], holding virtual bound states of conduction electrons at RE impurities in a metal host responsible [27]. These theories are not compatible with our model since they predict effects for half-filled shells without orbital moment (Gd).

**Conclusions.**— We predict that RE local moments interact with the conduction electrons of a metallic host differently from transition metal moments by generating both an oscillating spin density and charge current around it. The skew-scattering spin-orbit interaction causes the latter. The radial distribution of the induced velocity field oscillates with the same period as the RKKY spin polarization induced by the local exchange interaction. A finite $4f$ orbital moment $\mathbf{L}$ is necessary to form the current in the electron gas. Therefore, the predicted magnetic field and Knight shift depend linearly with $|\mathbf{L}| \propto (2 - g_J) |\mathbf{J}|$ and vanish for RE-doped insulators. Furthermore, the induced magnetic field depends on the atomic number via the Landé $g$-factor ground-state quantum numbers. Several approximations are crude, but we are confident about the predicted trends. Relativistic first-principles calculations of open $4f$ subshells in a metal host should improve the accuracy of the predictions.

The Ørsted fields generated by RE impurities at the surface of a metal with sufficiently large Fermi wavelength can be measured by scanning magnetometers based on nano-scale superconducting quantum interference devices (nanoSQUID) [50] or optically read-out nitrogen-vacancy (NV)-centers [51,52]. RE impurities adsorbed at a surface 2D electron gas, or graphene monolayer are promising candidate systems to image the predicted equilibrium current vortices.

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Rotational currents

Here we derive the linear response of a simple metal to a rare-earth (RE) magnetic impurity characterized by the classical vectors $\mathbf{S}$, $\mathbf{L}$, and $\mathbf{J}$, i.e. the spin, orbital, and total angular momenta, respectively. The conduction-electron orbital angular momentum density $\mathbf{l} = m_e \mathbf{r} \times \mathbf{v}(\mathbf{r})$ relative to the local moment at the origin, where $m_e \mathbf{v}(\mathbf{r})$ is the linear momentum density, reads in second quantization (see also Sec. of this SM)

$$\mathbf{l}(\mathbf{r}) = \hbar \mathbf{r} \times \frac{1}{\Omega} \sum_{pq\gamma} e^{i(q-p)\cdot \mathbf{r}} \frac{q^2 + q^2}{2} a^\dagger_{p\gamma} a_{q\gamma}. \tag{18}$$

We define the expectation values $\langle \mathbf{v} \rangle = \text{Tr}[\rho \mathbf{v}]$, where $\text{Tr}$ stands for the trace, and $\rho$ is the density matrix of the full Hamiltonian. With time-evolution operator in the interaction picture, $U(t) \equiv \exp \left[-(i/\hbar) \int_0^t H_{\text{skew}}(t')dt' \right]$, the total density matrix $\rho$ can be written in terms of the ground state density matrix $\rho_0$ of the unperturbed free electron gas with Hamiltonian $H_0$ and the regularized skew scattering Hamiltonian

$$H_{\text{skew}} = \frac{i\hbar}{\Omega} \sum_{kq\gamma \gamma'} e^{-2k^2a^2} a^\dagger_{q+k, \gamma} a_{q, \gamma'}(\mathbf{k} \times \mathbf{q}') \cdot \mathbf{L}, \tag{19}$$

where the constant $a = 2\langle r \rangle /9$, related to the 4f subshell radius $\langle r \rangle \approx 0.6\text{Å}$, accounts for the finite spatial extension of the 4f Slater-type orbital $R(r) \propto r^3 e^{-r/a}$. The exponential $e^{-2k^2a^2}$ cuts off an ultraviolet divergence that would
arrive for a delta-function perturbation. For an Al host metal, \( \hbar \nu_0 k_F^2 = 0.21 \text{ eV } \text{Å}^3 \). Then,

\[
\langle \mathbf{v}(r) \rangle = \text{Tr} \left[ \rho_0 \hat{U}^{-1}(t) \mathbf{v} \hat{U}(t) \right],
\]

\[
\approx \text{Tr} \left[ \rho_0 \left( 1 + \frac{i}{\hbar} \int_{-\infty}^{t} \hat{H}_{\text{skew}}(t') dt' \right) \mathbf{v} \left( 1 - \frac{i}{\hbar} \int_{-\infty}^{t} \hat{H}_{\text{skew}}(t') dt' \right) \right],
\]

\[
= \frac{i}{\hbar} \left\langle \int_{-\infty}^{t} dt' \left[ \hat{H}_{\text{skew}}(t'), \mathbf{v}(r) \right] \right\rangle_0,
\]

where \( \langle A \rangle_0 = \text{Tr}[\rho_0 A] \). This leads to

\[
\langle \mathbf{v}(r) \rangle = -\frac{i}{m_e \Omega} \int_{-\infty}^{t} dt' \sum_{\mathbf{p} \mathbf{q} \gamma} e^{i(\mathbf{q}-\mathbf{p}) \cdot \mathbf{r}} \frac{\mathbf{p} + \mathbf{q}}{2} \left\langle \left[ a_{\mathbf{p} \gamma}(t) a_{\mathbf{q} \gamma}(t), \hat{H}_{\text{skew}}(t') \right] \right\rangle_0
\]

\[
= \frac{\eta_0}{m_e \Omega^2} \sum_{\mathbf{k} \mathbf{q} \gamma'} \sum_{\mathbf{p} \mathbf{q} \gamma} e^{-2k^2 a^2} \sum_{\mathbf{p} \mathbf{q} \gamma} e^{i(\mathbf{q}-\mathbf{p}) \cdot \mathbf{r}} \frac{\mathbf{p} + \mathbf{q}}{2} \int_{-\infty}^{t} dt' \left\langle \left[ a_{\mathbf{p} \gamma}(t) a_{\mathbf{q} \gamma}(t), a_{\mathbf{q}' + \mathbf{k} \gamma'}(t') a_{\mathbf{q}' \gamma'}(t') \right] \right\rangle_0 \mathbf{(k \times q')} \cdot \mathbf{L}.
\]

The susceptibility is

\[
\chi(t - t') = \Theta(t - t') \sum_{\gamma \gamma'} \left\langle \left[ a_{\mathbf{p} \gamma}^\dagger(t) a_{\mathbf{q} \gamma}(t), a_{\mathbf{q}' + \mathbf{k} \gamma'}(t') a_{\mathbf{q}' \gamma'}(t') \right] \right\rangle_0,
\]

where \( \Theta \) is the Heaviside step function with time derivative

\[
\partial_t \chi(t - t') = \delta(t - t') \sum_{\gamma \gamma'} \left\langle \left[ a_{\mathbf{p} \gamma}^\dagger(t) a_{\mathbf{q} \gamma}(t), a_{\mathbf{q}' + \mathbf{k} \gamma'}(t') a_{\mathbf{q}' \gamma'}(t') \right] \right\rangle_0
\]

\[
+ \Theta(t - t') \sum_{\gamma \gamma'} \left\langle \left[ \partial_t \left( a_{\mathbf{p} \gamma}^\dagger(t) a_{\mathbf{q} \gamma}(t) \right), a_{\mathbf{q}' + \mathbf{k} \gamma'}(t') a_{\mathbf{q}' \gamma'}(t') \right] \right\rangle_0.
\]

\[
\partial_t \left[ a_{\mathbf{p} \gamma}^\dagger(t) a_{\mathbf{q} \gamma}(t) \right] \text{ can be calculated by the Heisenberg equation for the electron gas } H_0 = \sum_{\mathbf{k} \gamma} \epsilon_\mathbf{k} a_{\mathbf{k} \gamma}^\dagger a_{\mathbf{k} \gamma} \text{ with parabolic dispersion relation } \epsilon_\mathbf{k} = \hbar^2 \mathbf{k}^2 / (2m_e),
\]

\[
\partial_t \left( a_{\mathbf{p} \gamma}^\dagger(t) a_{\mathbf{q} \gamma}(t) \right) = \frac{1}{i \hbar} \left[ a_{\mathbf{p} \gamma}^\dagger(t) a_{\mathbf{q} \gamma}(t), H_0 \right] = \frac{1}{i \hbar} \left[ a_{\mathbf{p} \gamma}^\dagger(t) a_{\mathbf{q} \gamma}(t), \sum_{\mathbf{k} \gamma'} \epsilon_\mathbf{k} a_{\mathbf{k} \gamma'}^\dagger a_{\mathbf{k} \gamma'} \right] = -\frac{i}{\hbar} (\epsilon_\mathbf{q} - \epsilon_\mathbf{p}) a_{\mathbf{p} \gamma}^\dagger a_{\mathbf{q} \gamma},
\]

and

\[
\sum_{\gamma \gamma'} \left[ a_{\mathbf{p} \gamma}^\dagger a_{\mathbf{q} \gamma}, a_{\mathbf{q}' + \mathbf{k} \gamma'}^\dagger a_{\mathbf{q}' \gamma'} \right] = \sum_{\gamma} \left( \delta_{\mathbf{q} \mathbf{q}' + \mathbf{k} \gamma} a_{\mathbf{q} \mathbf{k} \gamma} - \delta_{\mathbf{p} \mathbf{q} \mathbf{p}' \mathbf{k} \gamma} a_{\mathbf{p} \mathbf{q} \mathbf{p}' \mathbf{k} \gamma} \right).
\]

\( \chi \) then satisfies the equation of motion

\[
\left( \partial_t + \frac{i}{\hbar} (\epsilon_\mathbf{q} - \epsilon_\mathbf{p}) \right) \chi(t - t') = \delta(t - t') \left\langle \sum_{\gamma} \left( \delta_{\mathbf{q} \mathbf{q}' + \mathbf{k} \gamma} a_{\mathbf{q} \mathbf{k} \gamma} - \delta_{\mathbf{p} \mathbf{q} \mathbf{p}' \mathbf{k} \gamma} a_{\mathbf{p} \mathbf{q} \mathbf{p}' \mathbf{k} \gamma} \right) \right\rangle_0.
\]

In the frequency domain, with \( \chi(t) = (2\pi)^{-1} \int d\omega \chi(\omega) e^{-i\omega t} \)

\[
\chi(\omega) = i\hbar \sum_{\gamma} \left\langle \delta_{\mathbf{q} \mathbf{q}' + \mathbf{k} \gamma} a_{\mathbf{q} \mathbf{k} \gamma} - \delta_{\mathbf{p} \mathbf{q} \mathbf{p}' \mathbf{k} \gamma} a_{\mathbf{p} \mathbf{q} \mathbf{p}' \mathbf{k} \gamma} \right\rangle_0 = 2i\hbar \delta_{\mathbf{q} \mathbf{q}' + \mathbf{k} \gamma} \delta_{\mathbf{p} \mathbf{q} \mathbf{p}' \mathbf{k} \gamma} / (\epsilon_\mathbf{p} - \epsilon_\mathbf{q} + \hbar \omega + i0^+),
\]

\[
\epsilon_\mathbf{p} - \epsilon_\mathbf{q} + \hbar \omega + i0^+.
\]
where $f_p$ is the (spin-degenerate) Fermi-Dirac distribution. Substituting $\chi$ into Eq. (21) after transformation into the frequency domain and in the steady state ($\omega \to 0$)

$$
\langle \mathbf{v}(\mathbf{r}) \rangle = \eta_0 \frac{1}{m_0 \Omega^2} \sum_{kq} e^{-2k^2a^2} \sum_{pq} e^{i(q-p)\cdot r} \langle k \cdot q \rangle \left( \langle k \times q \rangle \cdot \mathbf{L} \right) \epsilon_p \left( \delta_{q,q} + k \delta_{p,q} - \epsilon_p, \epsilon_q + i0^+ \right)
$$

(28)

$$
= i\eta_0 \frac{\hbar}{m_0 \Omega^2} \sum_{pq} e^{-2a^2|p-q|^2} e^{i(q-p)\cdot r} \langle p + q \rangle \left( \mathbf{L} \cdot \frac{-p \times q f_p - q \times p f_q}{\epsilon_p - \epsilon_q + i0^+} \right)
$$

(29)

$$
= i\eta_0 \frac{\hbar}{m_0 \Omega^2} \int \frac{d^3p}{(2\pi)^3} \int \frac{d^3q}{(2\pi)^3} e^{-2a^2|p-q|^2} e^{i(q-p)\cdot r} \langle p + q \rangle \left( \mathbf{L} \cdot \frac{-p \times q f_p - q \times p f_q}{\epsilon_p - \epsilon_q + i0^+} \right)
$$

(30)

$$
= i\eta_0 \frac{\hbar}{m_0 \Omega^2} \int \frac{d^3p}{(2\pi)^3} \int \frac{d^3q}{(2\pi)^3} e^{-2a^2|p-q|^2} e^{i(q-p)\cdot r} \langle p + q \rangle \left( \mathbf{L} \cdot \frac{-p \times q f_p - q \times p f_q}{\epsilon_p - \epsilon_q + i0^+} \right) + c.c.,
$$

(31)

where c.c. stands for the complex conjugate of the other terms.

Using

$$
e^{-2a^2|p-q|^2} = \frac{\sqrt{2}}{32\pi^{3/2}a^3} \int d^3r' e^{i(q-p)\cdot r'} e^{-r'^2/(8a^2)},
$$

we can write the regularized velocity, $\langle \mathbf{v}(\mathbf{r}) \rangle$, as the integral of the non-regularized one, $\langle \mathbf{v}_\infty(\mathbf{r}) \rangle$. The later has a divergence in the origin due to the delta nature of the skew scattering when $a \to 0$, as shown later.

$$
\langle \mathbf{v}(\mathbf{r}) \rangle = \frac{\sqrt{2}}{32\pi^{3/2}a^3} \int d^3r' e^{-r'^2/(8a^2)} \langle \mathbf{v}_\infty(\mathbf{r} + \mathbf{r}') \rangle,
$$

(32)

$$
\langle \mathbf{v}_\infty(\mathbf{r}) \rangle = i\eta_0 \frac{\hbar}{m_0} \int \frac{d^3p}{(2\pi)^3} \int \frac{d^3q}{(2\pi)^3} e^{i(q-p)\cdot r} \langle p + q \rangle \left( \mathbf{L} \cdot \frac{-p \times q f_p - q \times p f_q}{\epsilon_p - \epsilon_q + i0^+} \right) + c.c.,
$$

(33)

The angular part of the integral over $q = q \sin \theta_q \cos \phi_q \hat{x} + q \sin \theta_q \sin \phi_q \hat{y} + q \cos \theta_q \hat{z}$ reads

$$
I_1 = i \int_0^\pi d\theta_q \sin \theta_q \int_0^{2\pi} d\phi_q e^{iq \cdot r} \langle q + p \rangle \left( \mathbf{L} \cdot \langle q \times p \rangle \right) = \frac{4\pi i}{q r^3} \left[ q r \cos (qr) - \sin (qr) \right] \left[ \mathbf{L} \times p + i (r \cdot \mathbf{J} \times p) p \right]
$$

$$
- \frac{4\pi i}{q r^3} \left[ 3 q r \cos (qr) - 3 q^2 r^2 \sin (qr) \right] \left( \hat{r} \cdot \mathbf{L} \times \hat{p} \right),
$$

(34)

such that the angular integral over $p = p \sin \theta_p \cos \phi_p \hat{x} + p \sin \theta_p \sin \phi_p \hat{y} + p \cos \theta_p \hat{z}$ of the previous expression is

$$
\int_0^\pi d\theta_p \sin \theta_p \int_0^{2\pi} d\phi_p e^{-ip \cdot \mathbf{r}} I_1 = - \frac{32\pi^2}{pq r^5} \left[ q r \cos (qr) - \sin (qr) \right] \left[ \mathbf{L} \times \hat{r} \right],
$$

(35)

which reveals the rotational (i.e., $\propto \mathbf{L} \times \hat{r}$) character of the current.

$$
\langle \mathbf{v}_\infty \rangle = - \frac{\eta_0}{\pi^2 \hbar^2} \mathbf{L} \times \hat{r} \int_0^{k_F} dpp \left[ p r \cos (pr) - \sin (pr) \right] \int_0^{\infty} dq \left[ q r \cos (qr) - \sin (qr) \right] + c.c.
$$

(36)

Using $\cos (qr) = (e^{iqr} + e^{-iqr}) / 2$ and $\sin (qr) = (e^{iqr} - e^{-iqr}) / (2i)$

$$
\int_0^{\infty} dq \left[ q r \cos (qr) - \sin (qr) \right] \frac{q^2}{p^2 - q^2 + i0^+} = - \frac{1}{4} \int_{-\infty}^{\infty} dq q \left[ e^{iqr} (q r + i) + e^{-iqr} (q r - i) \right] q^2 - (p + i0^+)^2
$$

(37)

the integral over $q$ can be carried out by a contour integral in the complex plane. For $r > 0$ only the poles with a positive (negative) imaginary part contribute for integrands containing $e^{iqr}$ ($e^{-iqr}$),

$$
\int_0^{\infty} dq \left[ q r \cos (qr) - \sin (qr) \right] \frac{q^2}{p^2 - q^2 + i0^+} = - \frac{\pi i}{2} \left( pr + i \right) e^{ipr}.
$$

(38)

and

$$
\langle \mathbf{v}_\infty \rangle = \frac{\eta_0}{2\pi^2 \hbar^2} \mathbf{L} \times \hat{r} \int_0^{k_F} dpp \left[ p r \cos (pr) - \sin (pr) \right] (ipr - 1) e^{ipr} + c.c.
$$

$$
= - \frac{\eta_0}{\pi^2 \hbar^2} \mathbf{L} \times \hat{r} \int_0^{k_F} dpp \left[ p r \cos (pr) - \sin (pr) \right] \left[ \cos (pr) + pr \sin (pr) \right]
$$

(39)
where in the second step we used \( \eta = 0 \) for an Al host metal. The constant \( G_i^L \) plays the role of a g-factor, and then the rotational current contributes by about 0.3% to the total 4f orbital moment.
Ørsted field generated by the equilibrium currents

According to Maxwell’s equations the equilibrium charge current vortex around the rare-earth moment generates a magnetic field

\[
B_0(R) = -\frac{e\mu_0}{4\pi} \int d^3r' \langle v(r') \rangle \times \frac{R - r'}{|R - r'|^3}.
\]  
(47)

The derivation of an analytic expression for general \( R \) is tedious. However, far from the RE ion, \( R = |R| \gg (r) \) the Taylor expansion of \(|R\tilde{R} - r|^{-3} \) gives

\[
B_0(R) = \frac{\mu_0}{4\pi} \frac{3}{3} \left( m_{rc} \cdot R \right) R - R^2 m_{rc},
\]

where \( m_{rc} = -\gamma_0 G^L \). The above expression is the expected result of a field generated by the magnetic moment \( m_{rc} \) of the current vortex.

On the other hand, the magnetic field at the origin \( R = 0 \) reads

\[
B_0(0) = \frac{\mu_0}{4\pi} \int_0^\infty dr \int_0^\pi d\theta \sin \theta \int_0^{2\pi} d\phi \langle v(r) \rangle \times \hat{r},
\]

\[
= \frac{\gamma_0}{2\pi I_0} \frac{\mu_0}{4\pi} \int_0^\infty dr \frac{F(r)}{r} \int_0^\pi d\theta \sin \theta \int_0^{2\pi} d\phi (L \times \hat{r}) \times \hat{r}
\]

\[
= -3.5 \times 10^{-7} \frac{\gamma_0 \mu_0}{Ia^6} L
\]  
(48)

Orbital angular momentum density in second quantization

The Pauli equation for an electron wave function \( \psi(r) \) with energy \( E \) in a homogeneous magnetic field \( B \) reads

\[
E \psi(r) = \left[ \frac{1}{2m_e} (-i\hbar \nabla + eA)^2 + \frac{e\hbar}{2m_e} \sigma \cdot B \right] \psi(r).
\]  
(49)

In the symmetric gauge \( A = \frac{1}{2} B \times r \)

\[
E = \int d^3r \psi^\dagger(r) \left[ \frac{1}{2m_e} (-i\hbar \nabla + eB \times r)^2 + \frac{e\hbar}{2m_e} \sigma \cdot B \right] \psi(r)
\]

\[
= \int d^3r \psi^\dagger(r) \left[ -\frac{\hbar^2 \nabla^2}{2m_e} - ie\hbar (B \times r) \cdot \nabla + \frac{\nabla \cdot (B \times r)}{4m_e} + \frac{e\hbar}{2m_e} \sigma \cdot B + O(B^2) \right] \psi(r).
\]  
(50)

Then, the energy of the Zeeman coupling \( E_Z \) is

\[
E_Z = \frac{e}{2m_e} (l_T + 2s_T) \cdot B,
\]  
(52)

where the factor 2 is the single-electron orbital \( g \)-factor. In terms of the total spin \( s_T \) and orbital \( l_T \) angular momenta

\[
s_T = \frac{\hbar}{2} \int d^3r \psi^\dagger(r) \sigma \psi(r),
\]  
(53)

\[
l_T = \frac{i\hbar}{2} \int d^3r \psi^\dagger(r) (\nabla \times r - r \times \nabla) \psi(r).
\]  
(54)

Substituting

\[
\psi(r) = \frac{1}{\sqrt{\Omega}} \sum_{p,\alpha} e^{ipr} \chi_\alpha a_{p\alpha},
\]  
(55)

\[
\psi^\dagger(r) = \frac{1}{\sqrt{\Omega}} \sum_{q,\beta} e^{-iqr} \chi_\beta a_{q\beta}^\dagger,
\]  
(56)
where the spinors $\chi_\uparrow$ and $\chi_\downarrow$ are the basis of $\sigma_z$. The second quantized version of $s(r)$ and $l(r)$, the local densities of spin and orbital momentum (relative to the origin), respectively:

\[
\begin{align*}
s_T &= \int s(r) d^3r, \\
s(r) &= \frac{\hbar}{2\Omega} \sum_{pq\alpha\beta} e^{i(p-q) \cdot r} a_{q\alpha}^\dagger \sigma_{\alpha\beta} a_{p\beta}, \\
\end{align*}
\]

and

\[
\begin{align*}
l_T &= \frac{i\hbar}{2} \int d^3r \psi^\dagger(r) (\nabla \times \mathbf{r} - \mathbf{r} \times \nabla) \psi(r) = \int l(r) d^3r, \\
\end{align*}
\]

with

\[
\begin{align*}
l(r) &= \frac{\hbar}{\Omega} \mathbf{r} \times \sum_{pq\gamma} e^{i(p-q) \cdot \mathbf{r}} p a_{q\gamma}^\dagger a_{p\gamma} = \frac{\hbar}{\Omega} \mathbf{r} \times \sum_{pq\gamma} e^{i(p-q) \cdot \mathbf{r}} \frac{\mathbf{p} + q}{2} a_{q\gamma}^\dagger a_{p\gamma}. \\
\end{align*}
\]

Note that $l_T = m_e \mathbf{r}_{op} \times \mathbf{v}_{op}$, where $\mathbf{r}_{op}$ and $\mathbf{v}_{op}$ are the position and velocity operators, respectively.