Crystal field effects on spin pumping

Adam B. Cahaya, 1 Alejandro O. Leon, 1 and Gerrit E. W. Bauer 1,2,3
1 Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
2 WPI-AMR & CSRN, Tohoku University, Sendai 980-8577, Japan
3 Zernike Institute for Advanced Materials, Groningen University, The Netherlands

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“Spin pumping” is the injection of spin angular momentum by a time-dependent magnetization into an adjacent normal metal proportional to the spin mixing conductance. We study the role of electrostatic interactions in the form of crystal fields on the pumped spin currents generated by insulators with exchange-coupled local moments at the interface to a metal. The crystal field is shown to render the spin currents anisotropic, which implies that the spin mixing conductance of insulator | normal metal bilayers depends on crystal cut and orientation. We interpret the interface “effective field” (imaginary part of the spin mixing conductance) in terms of the coherent motion of the equilibrium spin density induced by proximity in the normal metal.

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I. INTRODUCTION

The interaction between the magnetization and currents in small structures and devices has attracted much attention in the last two decades. The generation of a spin current by magnetization dynamics is referred to as spin pumping [1,2]: A time-dependent magnetization “pumps” a spin current with magnitude and polarization \( \mathbf{J} = g_\text{β} \mathbf{m} \times \mathbf{m} - g_\text{β} \mathbf{m} \times \mathbf{m} \) into a normal metal, where \( \mathbf{m} \) is the unit magnetization vector, \( \mathbf{m} \) its time derivative, and \( g_\text{β} = g_\text{β} + ig_\text{β} \) is the complex interfacial spin mixing conductance. The spin pumping enhances the magnetization damping and can be interpreted as the Onsager reciprocal effect to the current-induced spin transfer torque, both being governed by the same spin mixing conductance [3]. The mixing conductance of the magnetic insulator yttrium iron garnet (YIG) was predicted to be of the same order of magnitude as that of magnetic metals [4], which was subsequently confirmed by experiments [5,6]. A dependence of the spin mixing conductance on the interface cut and orientation to the normal metal has also been predicted [4] and confirmed [7,8]. This anisotropy could partly be explained by the density of the local Fe magnetic moment directly at the interface. The rotational symmetry of magnetic atoms can be broken by the electric fields generated by neighboring atoms, i.e., the so-called crystal field. The relationship between the spin pumping and the local symmetry of magnetic moments at the interface has, to the best of our knowledge, not been studied yet. We therefore focus here on noncubic crystal fields of 3d transition metal ions with partially (not fully or half-) filled shells. These are predicted to cause effects that are much stronger than those generated by a cubic crystal field or when acting on 4f moments. Also, in the former case the spin orbit interaction is much weaker than the spin orbit interaction that we, hence, disregard here.

Under crystal fields, the angular part of the single 3d electron is described by the real valued doubly degenerate \( e_g \) and triply degenerate \( t_{2g} \) orbitals [9,10]. For transition metal ions on sites with octahedral symmetry, the energy level order is \( E_{e_g} > E_{t_{2g}} \) while in tetrahedral environment \( E_{t_{2g}} > E_{e_g} \) [9,10]. The total orbital angular momentum in this basis is quenched, \( \langle L_z \rangle = 0 \) [9,10]. The magnetism is then predominantly caused by the electron (Pauli) spins. When the spin orbit interaction is not negligible but competes with the crystal fields, the eigenstates are complex combinations of the sets \( e_g \) and \( t_{2g} \).

The orbital moment is then not completely quenched \( \langle L_z \rangle \neq 0 \) and the energy depends on the direction of the magnetization relative to the crystal axes (magnetic anisotropy) [11]. The effects of the spin-orbit interaction is discussed in a forthcoming paper with emphasis on partially filled 4f shells [12].

Here we study the role of crystal fields on the spin mixing conductance that governs spin pumping and other properties of interfaces. The paper is organized as follows. In Sec. II, we review the static and dynamics of 3d transition metal magnetic moments, disregarding their weak spin-orbit interactions. In the presence of crystal fields, the ground state electronic density of individual ions is nonspherical. By the exchange interaction such local moment induces in a metal an oscillating proximity spin density and associated Ruderman-Kittel-Kasuya-Yosida interaction (RKKY) [13–15] that are also anisotropic. This has, for example, been confirmed by first principles calculations on metallic surfaces [16]. The effects of such anisotropies on the spin dynamics are discussed in Sec. III, where we find that the spin current emitted by a dynamic magnetization is enhanced in certain directions. We discuss how the anisotropy influences local magnetization dynamic in term of enhanced damping in Sec. IV. In Sec. V, we extend our analysis to magnetic insulators in which the local moments at the interface are exposed to normal metal contacts. In Sec. VI we conclude that the crystal fields induce differences in the spin pumping for different crystal growth directions, which might help to explain some experiments.

II. SINGLE-ION MODEL

Consider a single localized magnetic moment generated by a partially filled 3d shell with spin density \( S_j(r,t) \) that depends adiabatically on time. In terms of the single electron wave functions \( \psi_j(r) \) with orbital index \( j \), the ground state spin density reads

\[
S_j(r,t) = S(t)n_d(r),
\]

where the electron density distribution of unpaired electrons

\[
n_d(r) = \sum_j S_j |\psi_j(r)|^2,
\]

\[
S_j = \frac{\int \mathbf{f}_{j} \cdot \mathbf{f}_{j} \, d^3r}{\int \mathbf{f}_{k} \cdot \mathbf{f}_{k} \, d^3r}
\]
depends on the occupation numbers \( f_{j,m} \) of orbital \( j \) and spin label \( m = \{ \uparrow, \downarrow \} \) and is normalized, \( \int d\mathbf{r} n_{j}(\mathbf{r}) = 1 \). The occupation numbers \( f_{j,m} \) are governed by the aufbau principle when the thermal energy is much smaller than the crystal field splitting \( (\Delta) \), i.e., \( k_B T/|\Delta| \ll 1 \), where \( k_B \) is the Boltzmann constant and \( T \) the temperature. Since spin orbit interaction is disregarded, the time dependence is encoded exclusively in the unit vector of the total spin \( \mathbf{S}(t) \).

In the presence of crystal fields, \( n_d(\mathbf{r}) \) has the point symmetry of the crystal site (or higher) that is characterized by a multipolar expansion. Here we focus on the common case of uniaxial deformation along the \( z \) direction, which allows parameterizing of the anisotropy in the spin density by its quadrupole moment

\[
Q_2 = \int d\mathbf{r} r^2 \left( \frac{3z^2}{r^2} - 1 \right) n_d(\mathbf{r}),
\]

where \( z = r \cos \theta \) is the coordinate along the symmetry axis of \( n_d(\mathbf{r}) \). \( Q_2 > 0(0 < 0) \) describes a prolate (oblate) ellipsoidal distribution. Decomposing the orbitals in the radial and angular functions, \( \psi_j(\mathbf{r}) = R_{j}(r)Y_j(\Omega) \), the quadrupole reads

\[
Q_2 = \langle r^2 \rangle \sum_j \int \left( 3 \cos^2 \theta - 1 \right) |Y_j(\Omega)|^2 d\Omega,
\]

where \( r = |\mathbf{r}| \), \( \langle r^2 \rangle = \int r^2 d\mathbf{r} R_{j}^2(r) \), \( \Omega = r \theta / d \Omega \theta \) and \( d\Omega = d\theta d\phi \). The radial function \( R_{j}(r) \) can be approximated by Slater-type orbitals [17,18], while the angular function are linear combinations of spherical harmonics (see Appendix A). \( Q_2 \) is calculated using the occupation numbers and the coefficients \( \langle \Omega_j \rangle = \int \left( 3 \cos^2 \theta - 1 \right) |Y_j(\Omega)|^2 d\Omega \), listed in Table 1.

Crystal fields can be parameterized by a point charge model of the local environment. The Hamiltonian close to the center of an octahedron made from point charges \( q \) is

\[
H_{cf}(\mathbf{r}) = \sum_n \frac{-qe^2}{4\pi\varepsilon_0 |\mathbf{r} - \mathbf{r}_n|} \approx \frac{-3qe^2}{2\pi\varepsilon_0 R_0} + \Delta_{\text{oct}}\left( x^4 + y^4 + z^4 - 3(x^2y^2 + x^2z^2 + y^2z^2) \right),
\]

where the subscript \( n \) labels the point charges at \((±R_0,0,0),(0,±R_0,0),(0,0,±R_0)\), \( \mathbf{r} = x\hat{x} + y\hat{y} + z\hat{z} \), and the Cartesian axes \( \{x,y,z\} \) are oriented along the crystallographic directions \((100),(010),(001)\), respectively. \( R_0 \) is the nearest-neighbor distance, \( \varepsilon_0 \) is the vacuum permittivity, \(-e \) is the electron charge, and \( q_e \) is the electric charge of neighboring ions. In metals, ion cores are positively charged, i.e., \( q > 0 \), while in transition metal oxides the oxygen anions dominate and \( q < 0 \). The crystal field parameter is \( \Delta_{\text{oct}} = -7qe^2(R_0^3 - 32\pi\varepsilon_0)^{-1} \) and can be estimated as \( \Delta \sim 2 \) eV for \( q = -2, (r^4)^{1/4} = 1.5 \AA, R_0 = 3 \AA \). In the tetrahedral site, on the other hand, the magnetic atom sits in the center of a cube defined by \((±R_0,±R_0,±R_0)/\sqrt{3} \). Both octahedral and tetrahedral sites are described by the same Hamiltonian [10] but \( \Delta_{\text{oct}} = -4\Delta_{\text{tet}}/9 \).

Half-filled shells, such as Mn\(^{2+}\) and Fe\(^{3+}\) are isotropic (spherical) and their \( Q_2 \) vanishes in any crystal field. The quadrupole moments vanish as well for octahedral and tetrahedral crystal fields, because the half-filled \( g \) and \( t_{2g} \) shells are still nearly spherical:

\[
e_g : \int d\Omega (3\cos^2 \theta - 1)|Y_{2z^2-r^2}|^2 = 0,
\]

\[
t_{2g} : \int d\Omega (3\cos^2 \theta - 1)|Y_{y^2-z^2}|^2 + |Y_{x^2-y^2}|^2 = 0.
\]

The quadrupole in the presence of compressive and tensile uniaxial strains depends on the occupation numbers as

\[
Q_2 = \frac{2}{5}\left[ S_{x^2} + S_{y^2} + 2( S_{y^2} - S_{x^2} - 3S_{xy} ) \right].
\]

where \( S_{xy} \) is given by Eq. (3). We note that even in distorted octahedral sites, some ions such as V\(^{2+}\), Cr\(^{3+}\), Ni\(^{2+}\), and Cu\(^{3+}\) have \( Q_2 = 0 \) because the \( g \) and \( t_{2g} \) are half filled.

**Interaction between a magnetic ion and conduction electrons**

The interaction between localized magnetic moments and conduction electrons with spin density \( s_{c}(\mathbf{r},t) \) is described by the \( s-d \) exchange Hamiltonian [22,23]. In the local-density approximation:

\[
H_{s-d} = -\frac{J}{\hbar^2} \int S_{c}(\mathbf{r},t) \cdot s_{c}(\mathbf{r},t) d\mathbf{r},
\]

where in the static and strong screening limit of the Coulomb interaction the exchange constant \( J = \frac{g_e^2}{4\pi\varepsilon_0} \) is the reciprocal conduction electron density of states \( g_e \) of the host metal.
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and \( \hbar \) is Planck’s constant divided by 2\( \pi \). For free electrons, \( g_e = m_e k_F/(\hbar^2) = 3n_r/(2E_F) \), in terms of the electron density \( n_e \), Fermi energy \( E_F \), and the effective electron mass \( m_r \). In the ground state, a static magnetic moment induces spin density oscillations. \( H_{\text{static}} \) also communicates the time dependence of the magnetic moment \( S \neq 0 \) to the conduction electrons, which can be formulated by extending the RKKY perturbation theory into the time domain [2]. Magnetization dynamics can be excited by magnetic or spin resonance, but also by spin transfer torques due to voltage and temperature gradients, lattice vibrations, etc. [24,25].

For sufficiently weak coupling, the response of the conduction electrons to a time dependent local moment \( S_d(r,t) \) reads

\[
\chi_s(r,t) = \frac{J}{\hbar^2} \int dr' dt' \chi(r-r',t-t') S_d(r',t'),
\]

where \( \chi(r,t) \) is the (scalar) dynamic spin susceptibility of the homogeneous host metal. In frequency and momentum space

\[
\chi_s(q,\omega) = \frac{J}{\hbar^2} \chi(q,\omega) S_d(q,\omega),
\]

where

\[
f(q,\omega) = \int \frac{dr}{(2\pi)^3} \int dt \chi(r,t) e^{-iqr} e^{i\omega t},
\]

\[
f(r,t) = \int \frac{dq}{(2\pi)^3} \int \frac{d\omega}{2\pi} f(q,\omega) e^{iqr} e^{-i\omega t}.
\]

Here the integration domain is a large system volume. In the free electron gas

\[
\chi(q,\omega) = \sum_p \frac{f_p - f_{p+1}}{e\epsilon_p - e\epsilon_0 + i\hbar \omega + i0^+},
\]

where \( f_p = [\exp((\epsilon_p - \mu)/(k_B T)) + 1]^{-1} \) is the Fermi-Dirac distribution, \( \epsilon_p = h^2 q^2 / (2m_e) \), \( \mu \) is chemical potential, and \( 0^+ \) is a positive infinitesimal. The time constants of the conduction electrons in high density metals are governed by the Fermi velocity (fs) and are much smaller than that of the magnetization dynamics (ns), which justifies expansion to leading order in velocity (fs) and are much smaller than that of the magnetization dynamics. Figure 2(a) and 2(c) illustrate that with increasing Fermi velocity the electron gas is polarized, as in the RKKY polarization function (16). The ion anisotropy is parameterized by the quadrupole \( Q_2 \), which is proportional to \( (q^2) \), see Eq. (5); larger ions induce a stronger anisotropy, cf. Figs. 2(c) and 2(d). This can also be seen from Eq. (23) by approximating \( (j_2(qr)) \approx q^2(r^2)/15 \), which leads to \( n_{\text{ani}} \sim (r^2) \). The sign of \( Q_2 \) can enlarge or decrease the total conduction electron spin polarization, as shown in Figs. 2(c) and 2(e).

When the atomic radius is small \( n_{\text{ani}}(r) \rightarrow \delta(r) \), the static spin polarization reduces to the well-known RKKY spatial oscillations

\[
\lim_{n_{\text{ani}}(r) \rightarrow \delta(r)} \rho_i(r) = \chi_i(r),
\]

while \( \rho_i(r) \rightarrow \chi_i(r) \). In this limit all crystal field effects vanish.

III. SPIN CURRENT

Conduction electron spin density and local moments are also related by the spin conservation equation

\[
\partial_t \mathbf{s}_s(r,t) + \nabla \cdot \mathbf{j}(r) = \left( \frac{d\mathbf{S}_s(r,t)}{dt} \right)_{\text{source}},
\]

where \( k_F = (3\pi^2 n_e)^{1/3} \) is the Fermi wave number. Using Eq. (1)

\[
\mathbf{s}_s(q,\omega) = \frac{J}{\hbar^2} \mathbf{S}(\omega) \chi(q,\omega)[n_{\text{iso}}(q) + n_{\text{ani}}(q)].
\]

The Fourier transform of the density distribution \( n_d = n_{\text{iso}}(r) + n_{\text{ani}}(r) \) is the sum of

\[
n_{\text{iso}}(q) = \langle j_0(qr) \rangle
\]

and

\[
n_{\text{ani}}(q) = -\pi \langle j_2(qr) \rangle Y_2 \left( \sin \frac{q}{r} \sqrt{\frac{5}{\pi}} \frac{Q_2}{r^2} \right)
\]

with \( \cos \theta_q = q \cdot \hat{z} \) and \( \langle j_0(qr) \rangle \) is the expectation value of the spherical Bessel function for a radial 3d wave function. Explicit formulas for \( \langle j_0(qr) \rangle \) and \( \langle j_2(qr) \rangle \) are demoted to the Appendix A.1.

Substituting \( \chi(q,\omega) \equiv \chi_i(q) + i\omega \chi_s(q) \) and keeping only linear terms in the frequency \( \omega \) (adiabatic approximation)

\[
\mathbf{s}_s(q,\omega) = \frac{J}{\hbar^2} [\mathbf{S}(\omega) \chi_i(q) + i\omega \mathbf{S}(\omega) \chi_s(q)] n_d(q).
\]

Transforming back into time domain

\[
\mathbf{s}_s(r,t) = \frac{J}{\hbar^2} [\rho_i(r) \mathbf{S}(t) - \rho_i(r) \mathbf{S}(t)].
\]

The densities

\[
\rho_i(r) = \int \frac{d\mathbf{q} e^{i\mathbf{q} \cdot \mathbf{r}}}{(2\pi)^3} \chi_i(q) n_d(q)
\]

and

\[
\rho_i(r) = \int \frac{d\mathbf{q} e^{i\mathbf{q} \cdot \mathbf{r}}}{(2\pi)^3} \chi_i(q) n_d(q)
\]

are plotted in Fig. 2 for several values of \( k_F^2 \) and \( Q_2 \). Figures 2(a) and 2(c) illustrate that with increasing Fermi energy a larger region of the electron gas is polarized, as in the RKKY polarization function (16). The ion anisotropy is parameterized by the quadrupole \( Q_2 \), which is proportional to \( (q^2) \), see Eq. (5); larger ions induce a stronger anisotropy, cf. Figs. 2(c) and 2(d). This can also be seen from Eq. (23) by approximating \( (j_2(qr)) \approx q^2(r^2)/15 \), which leads to \( n_{\text{ani}} \sim (r^2) \). The sign of \( Q_2 \) can enlarge or decrease the total conduction electron spin polarization, as shown in Figs. 2(c) and 2(e).

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\[
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\]

while \( \rho_i(r) \rightarrow \chi_i(r) \). In this limit all crystal field effects vanish.
describes spin flip scattering on the time scale $\tau_s$ and spin precession in the exchange torque exerted by the local moment. $\delta^i_\nu(r)$ is the spin current tensor, where the indexes $\sigma$ and $\nu$ refer to the spin polarization and current directions, respectively [27]. We obtain explicit expressions for the spin current divergence by substituting $s_i$ from Eq. (25), in the clean limit of the metal and slow magnetization dynamics ($\tau_s \to \infty$ and $\mathbf{S} \to 0$):

$$\nabla \cdot \mathbf{j}(r) = \frac{J^2}{\hbar^2} \rho_i(r) \mathbf{n}_{3d}(r) \mathbf{S} \times \mathbf{S} - \frac{J}{\hbar^2} \rho_i(r) \mathbf{S}.$$  

(30)

By writing the spin current in terms of a vector spin potential $\Phi(r)$ as $\mathbf{j}_\nu(r) = -\partial_t \Phi(r)$, Eq. (30) is reduced to a Poisson equation. The spin current direction is governed by the gradient of the spin potential, while its polarization is proportional to its direction. The solution of our Poisson equation is

$$\Phi(r,t) = \Phi_i(r) \mathbf{s}(t) \times \mathbf{S}(t) + \Phi_0(r) \mathbf{S}(t).$$  

(31)

where we defined dissipative ($\Phi_i$) and reactive ($\Phi_0$) scalar potentials. To leading order in the quadrupole moment

$$\Phi_0(r) = \frac{G_i^{\text{iso}}}{4\pi r^2} \left(1 + \frac{3 \cos^2 \theta - 1}{4r^2} Q_2 \right).$$  

(32)

where

$$G_i^{\text{iso}} = \frac{J^2}{\hbar^2} \int \frac{d\mathbf{q}}{(2\pi)^3} g_{\sigma\nu}(q) n_{\sigma\nu}(q)^2,$$

(33)

$$= G_i \left[ f_0 + \left( \frac{Q_2}{r^2} \right)^2 F_2 \right].$$  

(34)

with

$$G_i = \frac{\pi J^2 g^2}{8\hbar^2}.$$  

(35)

The dimensionless parameters can be obtained analytically as:

$$F_0 = \frac{11D(1208 - 5D(27D(3D - 16) + 682)) - 1627}{31185D(D + 1)^{11}} + \frac{1627}{31185D};$$  

(36)

$$F_2 = \frac{-44D(27D(12D - 43) + 985) + 197) - 788}{31185D(D + 1)^{11}} + \frac{788}{31185D},$$  

(37)

where $D = k_F^2(r^2)/4$, are given in Fig. 3 for various transition metal atoms. With increasing ionic radius, $F_0$ decreases, but $F_2$ increases up to half of $F_0$ for lighter ions, because the ratio of the anisotropic contribution $(j_{2z}(qr))/j_{0}(qr))$ is suppressed for small $r^2$ [see Eq. (A11)].

When $k_F^2(r^2) \to \infty$, both $F_0$ and $F_2$ converge to zero as $\sim(k_F^2(r^2)^{-1})$. While for small $k_F^2(r^2) \ll 1$, $F_0 \approx 1$, $F_2 \approx 4(k_F^2(r^2)^{-1})/135$ and $G_i^{\text{iso}}$ reduces to $G_i$. $\Phi_i(r)$ decays monotonically with $r$, but with an anisotropic component. The “reactive” spin potential in the “far field” $r^2 \gg r^2$ reads to leading order in $r^{-1}$

$$\Phi_i(r) = \left(1 + 2 \cos^2 \theta - \frac{3}{2} \right) G_i \cos 2k_F r$$  

(38)

FIG. 2. Conduction electron spin densities induced by a time-dependent anisotropic magnetic moment, $\rho_i(r)$ (left panels) and $\rho_0(r)$ (right panels) as defined in Eq. (25) are plotted for several parameter values in the $y = 0$ plane. $\rho_i(r)$ and $\rho_0(r)$ are normalized to $2k_F^2 g_i$, and $\chi_i(r = 0)$, respectively. The values of $(k_F^2, Q_2, r^2)$ are (a) $(1/4, 1/7)$, (b) $(1, 1/7)$, (c) $(4, 1/7)$, (d) $(4, 2/1)$, (e) $(4, -1/7)$.

where the source term

$$\left( \frac{d\mathbf{s}_i(r,t)}{dt} \right)_{source} = -\frac{s_i(r,t)}{\tau_s} + \frac{J}{\hbar^2} \mathbf{s}_s(r,t) \times \mathbf{s}_s(r,t),$$  

(29)

with

$$\mathbf{s}_s(r,t) = \frac{1}{k_F^2} \mathbf{S}_i(r,t) \mathbf{S}_i(r,t).$$  

(30)

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(38)

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(35)
spin pumping currents are not manifest in the magnetization in the absence of spin pumping, respectively. The anisotropic spin pumping currents are not manifest in the magnetization dynamics because their torques vanish when integrated over the local moment. \( G_r^{iso} \) and \( G_r^0 \) play roles equivalent to the real and imaginary part of the spin mixing conductance at interfaces [1]. \( G_r^{iso} \) parameterizes the dissipative angular momentum and energy loss implied by spin pumping, just as the real part of the spin mixing conductance at interfaces.

The imaginary part \( G_i \) is sometimes referred to as an “effective magnetic field.” It apparently accelerates or decelerates the precessional motion but conserves energy. The present results offer a simple picture of the physics of spin pumping: The coherent motion of the proximity RKKY spin density is locked to the precessing screening spins that precesses causally. The magnetic screening cloud therefore that can escape attention because it is hidden in the scattering theory formulation of spin pumping. The coherent motion of the moment. The Zeeman energy of the uncoupled system acts only on the local magnetic moments

\[
H_Z^{0} = \gamma_0 \mathbf{B} \cdot \int \mathbf{S}(\mathbf{r},t) \, d\mathbf{r} = \gamma_0 \mathbf{B} \cdot \mathbf{S}(t).
\]

On the other hand, it is the entire magnetic moment including the screening spins that precesses

\[
\mathbf{M} = -\gamma_0 \int [\mathbf{s}(\mathbf{r},t) + \mathbf{s}(\mathbf{r},t)] \, d\mathbf{r},
\]

where, in the adiabatic limit,

\[
\int s_z(\mathbf{r},t) \, d\mathbf{r} = S(t) \frac{J}{\hbar^2} \int \rho_z(\mathbf{r}) \, d\mathbf{r} = G_i S(t).
\]

so \( \mathbf{M}(t) = -\gamma_0 (1 + G_i) \mathbf{S}(t) \). The Zeeman energy of the coupled systems therefore reads

\[
H_Z = \frac{1}{1 + G_i} \mathbf{B} \cdot \mathbf{M}(t).
\]

The renormalization field \( \mathbf{B} \rightarrow \mathbf{B}(1 + G_i)^{-1} \) is therefore caused by the magnetic screening cloud therefore that can equivalently be written in terms of a new gyromagnetic ratio \( \gamma_0 \rightarrow \gamma_{eff} \equiv \gamma_0 (1 + G_i)^{-1} \).

V. MAGNETIC INSULATOR/NORMAL METAL INTERFACE

The present results are relevant for an understanding of the anisotropy at interfaces between normal metals and ferromagnetic/ferrimagnetic insulators [29], such as garnets and ferrites. The magnetism is then carried by local atomic moments that are ordered by superexchange interactions, usually via oxygen anions. Since localized on an atomic scale, only moments directly at the interface have a significant exchange interaction with the conduction electrons in the metal. Depending on the crystal direction and the interface cut, the number of...
contributing magnetic moments varies, as does the spin mixing conductance [4]. Here we focus on the effects of the crystal field on the spin pumping and the interface spin mixing conductance. The interface can be modelled in terms of independent local moments [4] whose motion is locked by the exchange coupling. The results for the single moments discussed above can then be applied. Cubic sites, such as symmetric octahedrals, do not deform the 3d electron density and suppress all anisotropies in cubic ferromagnets. However, at the interface the bulk point symmetry is broken and deformations normal to the interfaces may be expected, although we could not find estimates for the magnitude of such interface crystal fields.

Following Ref. [2], we model the metallic contact as a sheet of magnetic ions in a free electron gas, see Fig. 4(b). Spins can be pumped only in one direction, so we are only interested in the results for $z > 0$, i.e., the metallic side. We introduce the angle $\beta$ between the interface normal and the local symmetry axes, see Fig. 4(b). For example, when the crystal surface is in (001) and (111) directions, the local symmetry axis is tilted by angles $\beta = 0$ and $\beta \approx 55^\circ$, respectively. The equilibrium magnetization is assumed to lie in the interface plane by the thin-film easy-plane form anisotropy that is taken to dominate any perpendicular crystalline magnetic anisotropy. The coordinate along the interface normal $z$ is, in general, not parallel to the coordinate $Z$ that points along the local crystal symmetry axis, see Fig. 4. We adapt Eq. (10) to model the exchange interaction at an interface

$$H_{\text{ex}} = -\frac{J}{\hbar^2} \sum_n \int \mathbf{s}_n(r,t) \cdot \mathbf{S}_d(r - r_n,t) dr,$$

where the moments are at $r_n = (x_n,y_n,0)$ in the interface plane and $\mathbf{S}_d(r,t)$ has been defined in Eq. (1). Under FMR conditions all moments precess in phase. We expand the magnetic moment density at the interface into plane waves with reciprocal lattice vectors $\mathbf{G} = G_x \mathbf{x} + G_y \mathbf{y}$

$$\sum_n n_d(r - r_n) = \frac{N_d}{A} \sum \mathcal{G}^{G_x G_y} G \frac{d \mathbf{q}}{2\pi} n_d(q, \mathbf{\hat{z}} + \mathbf{G}) e^{i q z},$$

where $N_d$ is the number of magnetic ions. The proximity conduction electron spin density $\mathbf{s}_e(r,t)$ in linear response is $\mathbf{s}_e(r) = J\hbar^{-2} [\rho_e(r)\mathbf{S}(t) - \rho_e(r)\mathbf{S}(t)]$, where the densities $\rho_e,i$ are also periodic in the plane large,

$$\rho_{e,i}(r) = \frac{N_d}{A} \sum \mathcal{G}^{G_x G_y} G \frac{d \mathbf{q}}{2\pi} \chi_{e,i}(\sqrt{q^2 + G^2}) n_d(q, \mathbf{\hat{z}} + \mathbf{G}).$$

The spin conservation equation in the metal reads

$$\nabla \cdot \mathbf{J} = \frac{J^2}{\hbar^2} \mathbf{S}(t) \times \dot{\mathbf{S}}(t) \sum_n \rho_e(r) n_d(r - r_n) - \frac{J}{\hbar^2} \mathbf{S}(t) \rho_e(r).$$

In Appendix B we show that the in-plane components of the spin current are exponentially suppressed with distance from the interface with typical decay length of the order of the inverse of the (primitive) reciprocal vector, $G^{-1}$, which can be estimated as $1 \text{ nm} / (2\pi) = 1.6 \text{ Å}$, for a lattice constant parameter of 1 nm. The net spin current flow that leaves the magnet is therefore normal to the interface direction as illustrated in Fig. 4(b).

The pumped spin current can be calculated by applying the Gauss theorem to a flat “pill box” with volume $V = A z$ as shown in Fig. 4(b). The spin current in the insulator vanishes, so

$$\int_J \nabla \cdot \mathbf{J} d\mathbf{r} = 0 \int_J dx dy,$$

where $J$ is a surface on the metal side at a distance $z$ parallel to the interface and $J/A$ is the current polarization. Then,

$$\int_J dx dy = \frac{J^2}{\hbar^2} \mathbf{S}(t) \times \dot{\mathbf{S}}(t) \int_J d\mathbf{r} \sum_n \rho_e(r) n_d(r - r_n) - \frac{J}{\hbar^2} \mathbf{S}(t) \int_J d\mathbf{r} \rho_e(r),$$

which has the solution

$$\mathbf{J} = g_j^\perp \mathbf{m} \times \mathbf{m} - g_i^\perp f(z) \mathbf{m},$$

where

$$g_j^\perp = \frac{N_d J^2 S^2}{A h^4} \sum \mathcal{G}^{G_x G_y} G \frac{d \mathbf{q}}{(2\pi)^2} \chi_{e,i}(\sqrt{q^2 + G^2}) n_d(q, \mathbf{\hat{z}} + \mathbf{G})^2,$$

and the function $f(z)$ represents the integrated RKKY density in Fig. 6 that can be expressed analytically for short and long distances $z$ from the interface:

$$f(z > k^\perp_F) \approx \frac{\cos 2k_F z}{(2k_F z)^2} \left(1 - 2Qz k^\perp_F \frac{3 \cos^2 \beta - 1}{3}\right),$$

$$f(z < k^\perp_F) \approx 1 + 2Qz k^\perp_F \frac{3 \cos^2 \beta - 1}{9}.$$
CRYSTAL FIELD EFFECTS ON SPIN PUMPING

FIG. 5. The dissipative spin current injected by a ferromagnetic insulator into a normal metal $g_{i}^{11}$ as a function of angle $\beta$ between crystal field direction and interface normal.

parts of the spin-mixing conductance, respectively. The sum over $G$ in Eq. (54) reflects interference effects that can be simplified in the limit of large density of magnetic moments, i.e., when $\alpha \ll \pi/k_{F}$. Since the susceptibility $\chi(q)$ is proportional to the step function, see Eq. (19), and modes with wave number $|G| \geq 2k_{F}$ do not contribute. In that limit

$$
g_{i}^{11} \approx \frac{N_{d}F^{2}S^{2}}{A h^{4}} \int \frac{d\mathbf{q}}{(2\pi)^{3}} \chi_{i}(q) n_{d}(q) n_{d}(-q)$$

$$
= \frac{N_{d}S^{2}G_{r}^{\text{iso}}}{A} \left( 1 - Q_{z}k_{F}^{2} \left( 3 \cos^{2} \beta - 1 \right) \right),
$$

where $G_{r}^{\text{iso}}$ is given by Eq. (34). The contribution by Bragg scattering with finite $G$ is relevant for materials with lower interface moment density. However, Eq. (54) as a function of the interface moment density can be calculated only numerically, but we estimate that correction terms suppress the anisotropy. In the dilute limit $N_{d} \rightarrow 0$ all interference and thereby anisotropies vanish, as discussed in Appendix C.

For a 3$d^{6}$ high-spin state of Fe$^{3+}$ or Co$^{3+}$: $S = 3h/2$ and $N_{d}/A = (0.5 \text{ nm})^{-2}$, the isotropic contribution $g_{i}^{11} = N_{d}S^{2}G_{r}^{\text{iso}}/A \sim 10^{18} \text{ cm}^{-2}$ is of the order of magnitude accepted for magnetic insulators, while $g_{i}^{11} = N_{d}S^{2}/A \sim 10^{18} \text{ cm}^{-2}$ appears to be rather large [6]. The anisotropy of the spin current pumped through transition metal ions in an elongated octahedral crystal field and high spin state with $Q_{2} = \pm 4/21$ is plotted in Fig. 5. The magnetic ions emit less spin current in the direction in which the 3$d$ subshell is elongated, because the spin current is generated by exchange in the overlap volume of the spin densities $s_{\downarrow}$ and $s_{\uparrow}$, see Eqs. (30) and (29), and $s_{\uparrow}$ is preferentially suppressed by proximity in that direction. Ions with positive $Q_{2}$ generate maximal (minimal) spin current for $\beta = \pi/2$ ($\beta = 0$), while this is opposite for negative $Q_{2}$. The anisotropic spin current depend on the relative angle between the 3$d$ subshell orientation and the interface orientation $\beta$, which should be observable at selected interfaces.

The reactive spin current depends on position by the function $f(z)$. It is not a transport spin current but is caused by the coherent precession of the proximity spin density. It can be obtained by applying Gauss’ theorem to an integral over the volume $V = A z$, as in Fig. 6. The magnitude of this current near the interface is estimated $g_{r}^{11} \sim 10^{18} \text{ cm}^{-2}$ and vanishes algebraically with distance from the interface with the RKKY oscillations. This spatial dependence $f(z)$ is lost in scattering theory in which only the transport of electrons between incoherent reservoirs are considered. The imaginary part of the spin mixing conductance $g_{i}^{11}$ has been found to be relatively small for most systems [30].

Cobalt ferrite (CoFe$_{2}$O$_{4}$ or CFO) is an iron-based spinel. Cobalt ferrites possess an inverse spinel structure, [Fe$^{3+}$]$_{T}$[Co$^{2+}$Fe$^{3+}$]$_{L}$O$_{4}$, where the subscripts $[T]$ and $[L]$ stand for the tetrahedral and octahedral sites, respectively. The ions have half filled subshell and an isotropic electronic cloud, regardless of the symmetries of their environment. However, the octahedrally coordinated Co$^{2+}$ ions occupy elongated octahedrals when grown on SrTiO$_{3}$ (STO) substrates. The unit cell lattice parameter of STO $a_{\text{STO}} = 3.906$ Å [20] is smaller than the corresponding lattice parameter of CFO $a_{\text{CFO}} = 4.195$ Å [20]. As a result of this lattice mismatch, CFO films are in-plane compressed and tetragonally distorted [7,21], depending on the growth direction of the sample. When CFO is grown in the (001) direction, the resultant crystal field is an elongated octahedral, while in the (111) growth direction the compression creates a slanted octahedral crystal field. The resultant crystal field can be described by that of an elongated octahedral with a small energy splitting (see Appendix D). Our model predicts that the exchange between the cobalt ions and the conduction electrons is stronger for a (001) CFO than for a (111) one. Indeed, replacing the cobalt quadrupole $Q_{2} = -4/21$ in Eq. (59) for the angles $\beta_{(001)} = 0$ and $\beta_{(111)} = \pi/4$, we find that $g_{r}^{11}(001)$ is 50% larger than $g_{r}^{11}(111)$, in agreement with the experiment of Ref. [7]. It should be mentioned that the magnetization and the surface Co$^{2+}$/Fe$^{3+}$ concentration ratio strongly depend on the preparation conditions [7], however.

Our model can be applied to other than ferromagnetic order of the local moments at the interface. The dissipative spin current emitted by each ion is proportional to $S \times S$ and thereby invariant to spin reversal $S \rightarrow -S$. The sum of all spin-current contributions, as well as the real part of the spin mixing conductance, does not depend on the (collinear) order of the sublattices (ferro, ferri, or antiferromagnetic) [4]. However, the imaginary part of the spin mixing conductance

FIG. 6. The RKKY-like spatial oscillation, represented by dimensionless $f(z)$ in the reactive spin current $g_{i}^{11}$, where $f = f_{\text{iso}} + f_{\text{aniso}}$ is decomposed into an isotropic ($f_{\text{iso}}$) and anisotropic ($f_{\text{aniso}}$) terms. $f_{\text{iso}}(z) = f_{\text{iso}}(z)Q_{2}^{1/2}k_{F}^{2}/(3\cos^{2}\beta - 1)$ is the normalized $f_{\text{iso}}$. For $z \gg k_{F}^{-1}$, $f_{\text{iso}}$ and $f_{\text{aniso}}$ approach $(2k_{F})^{-2}\cos 2k_{F}z$ and $-(2/3)(2k_{F})^{-2}\cos 2k_{F}z$, respectively. When $Q_{2} \neq 0$ the spin current depends on the angle $\beta$. 

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or effective exchange field $g_{ij}^{\uparrow \downarrow}$ felt by the conduction electrons is governed by the sum of the local moments and vanishes for exactly compensated antiferromagnetic interface order. This is consistent with previous studies of the spin pumping by antiferromagnets [31,32].

VI. CONCLUDING REMARKS

The deformations of partially filled $3d$ shells of local moments in noncubic crystal fields are reflected by anisotropic RKKY spin-density oscillations and nonlocal exchange interactions in metallic hosts. We show that the spin current pumped by a magnetic moment with nonspherical spin density is anisotropic as well. The spin pumping leads to enhanced magnetization damping and a renormalized gyromagnetic ratio. The latter can be interpreted in terms of the coherent motion of the RKKY spin density oscillations.

The properties of interfaces between magnetic insulators and metals are governed by the local moments in the terminating monolayer. The spin mixing conductance and its asymmetry depends not only on the density of exposed moments but also on the local point symmetry. We applied the theory to analyze the spin pumping from a ferromagnetic insulator to an adjacent normal metal. Most anisotropies focus the spin currents into a direction normal to the interface, which is beneficial for spintronics. Spin pumping and spin transfer torque are each others Onsager reciprocals and governed by the same spin-mixing conductance. The crystal field effects in spin pumping addressed here therefore equally affect the spin-transfer torque efficiency.

The anisotropy of the pumped spin-current depends on the quadrupole moment $Q_2$, which in turns depends on the orbital occupation of interface magnetic atoms. While we focus here on CFO, the anisotropy should affect all transition metal based magnetic insulator with magnetic moments at the interface with nonspherical spin distribution. An interesting material to apply the present analysis could be La$_{1-x}$Sr$_x$MnO$_3$ (LSMO) in which the $3d$ shell of the Mn ions is not half filled. First principles band structure calculations can test our predictions and render them more quantitative.

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APPENDIX A: TESSERAL SPHERICAL HARMONICS

The $3d$ orbitals can be written as

$$\psi_j(r) = R_{3d}(r)Y_j(\Omega), \quad (A1)$$

where the radial function can be approximated by Slater-type wave functions [17,18],

$$R_{3d}(r) = \sqrt{\frac{1}{6!}} \left(\frac{2}{a}\right)^{7/2} e^{-r/a}, \quad (A2)$$

where the constant $a$ is related to the mean-square radius by $(r^2) = 14a^2 \sim 1 \text{Å}$. In crystals, the angular part of the $3d$ wave functions is described by the set of orbitals [9]

$$Y_{z^2} = \sqrt{\frac{5}{16\pi}(3\cos^2 \theta - 1)} = \sqrt{\frac{5}{16\pi}} \frac{2z^2 - x^2 - y^2}{r^2}, \quad (A3)$$

$$Y_{x^2-y^2} = \sqrt{\frac{15}{16\pi}} \sin^2 \theta \cos 2\phi = \sqrt{\frac{15}{16\pi}} \frac{x^2 - y^2}{r^2}, \quad (A4)$$

known as $e_g$ orbitals, and

$$Y_{xy} = \sqrt{\frac{15}{16\pi}} \sin^2 \theta \sin 2\phi = \sqrt{\frac{15}{4\pi}} \frac{xy}{r^2}, \quad (A5)$$

$$Y_{yz} = \sqrt{\frac{15}{16\pi}} \sin 2\theta \sin \phi = \sqrt{\frac{15}{4\pi}} \frac{yz}{r^2}, \quad (A6)$$

$$Y_{zx} = \sqrt{\frac{15}{16\pi}} \sin 2\theta \cos \phi = \sqrt{\frac{15}{4\pi}} \frac{zx}{r^2}, \quad (A7)$$

known as $t_{2g}$ orbitals. They are shown in Fig. 7.

1. Mean value of spherical Bessel functions

The $n$th spherical Bessel function for $n = 0$ and $n = 2$ read

$$j_0(x) = \frac{\sin x}{x}, \quad (A8)$$

$$j_2(x) = \left(\frac{3}{x^2} - \frac{1}{x}\right) \frac{\sin x}{x} - \frac{3\cos x}{x^2}. \quad (A9)$$

Their mean values over Slater-type single-exponential orbitals are

$$\langle j_0(qr) \rangle = \int j_0(qr) n_d(r) dr \approx \frac{1 - \frac{5}{6} a^2 q^2 + \frac{1}{16} a^4 q^4}{(1 + \frac{1}{4} a^2 q^2)^2}$$

$$\approx \left(1 - \frac{q^2 (r^2)}{6}\right) \quad (A10)$$
\[ (j_z(r)) = \int j_z(r)n_d(r)dr = \frac{2a^2q^2(7 - 3a^2q^2/4)}{15(1 + a^2q^2/4)^2} \]
\approx \frac{q^2(r^2)}{15}. \quad (A11)

where the approximations are valid for \( q^2(r^2) \ll 1 \).

**APPENDIX B: SPIN CURRENT DIRECTION FOR INTERFACES**

A two-dimensional periodic lattice is specified by two independent primitive translation vectors, \( \mathbf{a}_1 \) and \( \mathbf{a}_2 \)

\[ \mathbf{r}_n = n_1\mathbf{a}_1 + n_2\mathbf{a}_2. \quad (B1) \]

Its reciprocal lattice is

\[ \mathbf{G} = n'_1\mathbf{b}_1 + n'_2\mathbf{b}_2, \quad (B2) \]

where \( \mathbf{b}_1 = 2\pi\mathbf{a}_2/|\mathbf{a}_1 \times \mathbf{a}_2| \) and \( \mathbf{b}_2 = 2\pi\mathbf{a}_1/|\mathbf{a}_1 \times \mathbf{a}_2| \). Equation (50) implies that translational symmetry of the density \( n_d \) and \( \rho_c \) carries over to the vector spin potential \( \Phi \) and spin current tensor \( \Phi_\mu(r) = -\partial_\mu \Phi(r) \). Therefore

\[ \Phi(r) = \sum_G \Phi_G(z)e^{i(G_z + G_y)q} \], \quad (B3)

where the Fourier coefficients \( \Phi_G \) depend on the distance \( z \) from the interface. Inserting this expansion into Eq. (50) leads to

\[ [G^2 - q_z^2]\Phi_G(z) = \frac{1}{A} \int dxdye^{-i(G_z + G_y)q}T(r), \quad (B4) \]

with

\[ T(r) = -J^1/r\rho_c(r)\hat{S} + J^2/r\rho_c(r)\sum_n n_d(r - r_n)\mathbf{S} \times \hat{S}. \quad (B5) \]

where \( A \) is the unit cell area. Fourier transforming with respect to \( z \) gives

\[ [G^2 + q_z^2]\Phi_G(q_z) = T_G(q_z). \quad (B6) \]

\[ T_G(q_z) = A^{-1} \int dxdye^{-iq_zz} \int dxdye^{-i(G_z + G_y)q}T(r). \]

Hence

\[ \Phi(r) = \sum_G e^{i(G_z + G_y)q} \int \frac{dq_z e^{iq_zz}}{2\pi(q_z^2 + G^2)} T_G(q_z) \quad (B8) \]

\[ = \sum_G e^{i(G_z + G_y)q} e^{-Gz} \frac{G}{2G} T_G(iG), \quad (B9) \]

using the residuals theorem for the pole \( q_z = iG \). \( J \) can be decomposed into the currents along \( z \) and \( \mathbf{G} \) as

\[ J^z = \frac{1}{2} \sum_G e^{-Gz}e^{i(G_z + G_y)q} \mathbf{S} \otimes T_G(iG) \quad (B10) \]

\[ J^G = \frac{-i}{2} \sum_G e^{-Gz}e^{i(G_z + G_y)q} \frac{G}{G} \mathbf{S} \otimes T_G(iG), \quad (B11) \]

where \( \otimes \) is the external product of the two subspaces (spin direction and current flow direction). Thus the spin current flowing in the in-plane directions \( (J^G) \) decays exponentially with distance \( z \) when \( G \neq 0 \). Only the contribution perpendicular to the interface \( (G = 0) \) propagates as

\[ J(z \gg r) = A^{-1} \int drT(r), \quad (B12) \]

which is the same result we obtain in the main text using the divergence theorem. On the other hand, \( J^G \) is not defined for \( G = 0 \).

**APPENDIX C: FINITE WAVELENGTH CONTRIBUTIONS TO THE UNIFORM SPIN CURRENT**

Corrections for finite \( G = |\mathbf{G}| < 2k_F \) can be calculated by using the susceptibility \( \chi_r(q) \) and spin density \( n_d(q) \) in Eqs. (19), (21), and (23) in the spin-mixing conductance formula (54). Equation (54) can be rewritten in terms of lattice vector \( \mathbf{a}_{n_1,n_2} = n_1a_x + n_2a_y \),

\[ g_{r}^{\perp} = \frac{N_sJ^2\pi^2}{4\pi^4} \int \frac{dq}{(2\pi)^2} \chi_r(q)[n_d(q)]^2 \sum_{n_1,n_2} e^{iqa_{n_1,n_2}.} \quad (C1) \]

The numerical integration of the above equation is tedious. However, two natural limit cases are analytically accessible, namely the dense \( (a_x,a_y \rightarrow 0) \) and dilute \( (a_x,a_y \rightarrow \infty) \) local moments approximations. While the former is used in the main text, we address here the second one by the following expansion,

\[ e^{iqa_{n_1,n_2}} = 4\pi \sum_i j_i(qa_{n_1,n_2}) Y_{lm}(\hat{q})Y_{l'm}(\hat{a}_{n_1,n_2}). \]

For garnets with large lattice constants (large \( a_0 \)), we can use the asymptotic properties of the spherical Bessel functions,

\[ \lim_{qa_{n_1,n_2} \gg 1} j_i(qa_{n_1,n_2}) = \frac{\sin(qa_{n_1,n_2} - \frac{i\pi}{2})}{qa_{n_1,n_2}}. \quad (C2) \]

Therefore the contribution of the \( n_1,n_2 \neq 0 \) terms decays as \( \propto (k_Fa_{n_1,n_2})^{-1} \) and for a moment-to-moment distance of a nm and an elemental metal typically smaller than 0.1. The sum of Eq. (C1) is then dominated by the isotropic \( n_1 = n_2 = 0 \) term and the interface spin-mixing conductance is just the sum of the (isotropic) single ion contributions, \( g_{r}^{\perp} = N G_{mm}^i \). The spin current generated by well-separated magnetic moments does cause interference effects on the perpendicular spin current and the anisotropies vanish.

**APPENDIX D: CRYSTAL FIELD FIELD OF DISTORTED OCTAHEDRAL SITES**

Growing CFO films on lattice mismatched substrates [7] causes magnetostriction [33] that leads to a distortion of the octahedral environment of the cobalt moments. An elongation or contraction in the crystal directions (001) and (111) shifts the oxygen ion positions along the direction \((\sin \beta, \cos \beta, \sin \beta)\), where \( \beta_{001} = 0 \) and \( \beta_{111} = 55^\circ \). Similar to Eq. (6), the point charge model leads to a crystal field splitting

\[ \Delta H_{cf}^{\beta}(r) = \Delta_0 \frac{2z^2 - x^2 - y^2}{(r^2)}, \quad (D1) \]
\[
\Delta_0 = \frac{3(r^2)\delta \cos(\beta + \beta_0)}{4\pi \epsilon_0 R_1^2 \cos \beta_0},
\]

(2D)

\(\delta\) is the strain, and \(\beta_0 \approx 35^\circ\). This distortion creates an effective quadrupolar crystal field. The lattice constants of CFO, SrTiO\(_3\) substrate, and Pt overlayer are \(a_{\text{STO}} = 3.906\) Å, \(a_{\text{CFO}} = 4.195\) Å, \(a_{\text{Pt}} = 3.912\) Å [34]. With strain \(\delta \approx 2\%\), we estimate \(\Delta_0 \approx 0.06\) eV.