The origin of increase of damping in transition metals with rare earth impurities

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Abstract

The damping due to rare earth impurities in transition metals is discussed in the low concentration limit. It is shown that the increase in damping is mainly due to the coupling of the orbital moments of the rare earth impurities and the conduction p-electrons. It is shown that an itinerant picture for the host transition ions is needed to reproduce the observed dependence of the damping on the total angular moment of the rare earths.

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Magnetization dynamics has become one of the most important issues of modern magnetism. This development is driven by the technological demand to tailor magnetic responses on ever smaller length and shorter time scales. The importance of this issue manifests itself in a completely new area of research, spintronics, and a huge literature that cannot be cited here. Selected highlights include precessional switching by tailored field pulses [1, 2], spin-torque [3, 4], and laser-induced magnetization dynamics [5, 6].

In general, magnetization dynamics is described via the Landau-Lifshitz-Gilbert equation (LLG) [7] including additional terms to incorporate spin-torque effects [8] or those due to pulsed optical excitations [9]. All these descriptions account for energy dissipation via a phenomenological damping parameter which governs the time needed for a non-equilibrium magnetic state to return to equilibrium. Recently it has even been suggested that α determines the magnetic response to ultrafast thermal agitations [10].

Technological applications call for the ability to tailor α [11]. The most systematic experimental investigation on this topic was published by Bailey et al. [12] who studied the effect of rare-earth doping on the damping in permalloy. Most rare earth ions induced a large increase of α, but neither Eu nor Gd altered the damping of permalloy (cf. Fig. 2). Since Gd+++ and Eu+++ have no orbital momentum, this points immediately to the importance of the angular momentum in the damping process. Bailey et al. determined damping by reproducing their data via the LLG equation using α as a fit parameter. This widely used procedure points to a fundamental problem of this phenomenological approach. Though the LLG equation describes data well, a more microscopic approach is needed to understand the origin of damping.

It was Elliott [13] who first studied damping in semiconductors due to spin-orbit coupling. Later Kambersky [14] argued that the Elliot-Yafet mechanism should be also operable in magnetic conductors. Korenman and Prange [15] developed a more microscopic treatment and found that spin-orbit coupling should be important at low temperature in transition metals. Recent measurements of damping in magnetic multilayers at room temperature [16] suggest that the s-d interaction might also be at the origin of damping [17, 18]. However, all of the present models fail to reproduce the data of Ref. [12].

In this Letter, we explain the increase of damping in rare earth doped transition metals via a novel orbit-orbit coupling between the conduction electrons and the impurities. The well known s-f interaction [19] gives rise to a $(g_f - 1)^2$ dependence of the damping that is in contradiction to experimental observations [12]. In contrast, the orbit-orbit coupling considered here reproduces the measured $(g_f - 2)^4$ dependence of the damping. Both dependencies on the Lande g-factor $g_f$ follow directly from the fact that the rare earth ions are in their ground state. Hence, their angular momentum $L_f$, spin $S_f$, and total angular momentum $J_f$ are related by the Wigner Eckardt theorem: $L_f = (2 - g_f) J_f$ and $S_f = (g_f - 1) J_f$. Deriving the magnetic moments of the transition ions from the electronic degrees of freedom is essential to capture the correct behavior of damping as a function of $J_f$. For the uniform mode, the damping due to orbit-orbit coupling is of Gilbert form in the low frequency limit.

Taking the wave functions of the $d_-, f-_e$, and conduction electrons orthogonal, the Hamiltonian for the rare-earth doped transition metal in an external field $H$ is

$$H = H_e + H_f + H_d.$$  (1)

This approximation should be valid for the heavy rare earths but probably fails for elements like Cerium where valence fluctuations are important. The conduction electron Hamiltonian $H_e$ is the usual one, $H_e = \sum_{k,\sigma} \epsilon_k \hat{a}_{k,\sigma}^\dagger \hat{a}_{k,\sigma}$, where $\hat{a}_{k,\sigma}^\dagger$ and $\hat{a}_{k,\sigma}$ are the creation and annihilation operators of a conduction electron with momentum $k$ and spin $\sigma$. $\epsilon_k$ is the energy of the conduction electrons including a Zeeman term.

$H_f$ is the Kondo Hamiltonian [20] of the localized rare earth moment

$$H_f = \Gamma S_e \cdot S_f + \lambda L_e \cdot L_f - \mu_f \cdot H.$$  (2)

$S_{e/f}$ and $L_{e/f}$ are the spin and angular momentum of conduction and f electrons, respectively. $L_{e/f}$ are taken with respect to the position of the impurity. The spin-spin term is the well known s-f coupling used by...
\[ H_{LL} = i(2 - gJ) \sum_{k,k'} \lambda(k,k') \hat{\mathbf{k}} \times \hat{\mathbf{k}}' \cdot \mathbf{J} f_{k} a_{k'}^\dagger a_{k'} \]  

where the orbit-orbit coupling \( \lambda \) will be assumed to be a function of the relative angles of the \( \mathbf{k} \) vectors and is almost everywhere zero except for \( k \) close to the Fermi level \( k_F \). The magnitude of \( \lambda \) is not known but is expected to be of the same order as the spin-spin coupling constant \( \Gamma \). The crystalline electric field effect in transition metals is less than 0.1 meV which is small and hence the spin-orbit term \( \mathbf{S}_F \cdot \mathbf{L}_F \) is neglected. At room temperature all the rare earth ions studied in Ref. [12] are in their ground state making the term \( \mathbf{S}_F \cdot \mathbf{L}_F \) ineffective as damping mechanism. This follows immediately from the Wigner-Eckart theorem.

The Hamiltonian for the host transition ions is based on the Anderson Hamiltonian with explicit spin rotational invariance in the absence of a Zeeman term. It is

\[ H_d = \epsilon_d d^\dagger \sigma d + \sum_k V_{kd} (a_{k,\sigma}^\dagger d^\dagger \sigma a_{k,\sigma} + d^\dagger \sigma a_{k,\sigma}) + \frac{U}{2} S_d \cdot S_d - \mu_d \cdot \mathbf{H}, \]

where \( S_d \) is the spin operator of the local \( d \) electrons while their orbital angular momentum is assumed quenched. \( \rho \) is the charge density operator of the \( d \) electrons. In transition ions such as \( \text{Ni}, \ V_{kd} \approx 1.0 - 10.0 \text{ eV} \) is comparable to the Coulomb potential \( U \). The hybridization term between the conduction- \( d \)-electrons is essential to establish a spin-independent orbit-orbit coupling between the \( d \) and the \( f \)-ions. The degree of localization of the magnetic moments increases with decreasing \( V_{kd} \) and controls the extent to which rare earth impurities enhance damping.

The orbit-orbit coupling (cf. Eq.3) gives no contribution for \( \text{Gd}^{+++} \) (4f7) as observed in the experiment [12]. As for the element \( \text{Eu} \), it is believed from measurements of the paramagnetic susceptibilities that the ionic state is \( \text{Eu}^{++} \) (4f7) and not \( \text{Eu}^{++} \) (4f6) [19, 20]. If this is the case then clearly this is a state with \( f \) levels of the order 0.1 meV which is small and hence the damping. We finally compare the functional form of this result to that of LLG and discuss why the electronic (lattice) picture of the host transition ions is essential.

The fundamental quantity in our calculation is the generating functional

\[ \mathcal{Z} [\eta, \eta^*] = \text{Tr} e^{-\int_0^\beta d\tau (H - \eta^* \eta^0(\tau) \mathbf{m}^-(\tau) - \eta \mathbf{m}^+(\tau))}, \]

where \( \eta \) and \( \eta^* \) are external sources and \( \beta \) is inverse temperature. The propagator, i.e. the connected two-point Green’s function, of the volume mode of the transition metal ions is found by functional differentiations with respect to the external sources \( \eta^* \) and \( \eta \).

The resulting effective Lagrangian is now written in terms of \( m \) only

\[ \mathcal{L} = -\frac{1}{2} m_{ij} \mathcal{K}_{ijkl} m_{kl} - m \ln \left[ G_{d}^{-1} + \mathcal{K} m \right]. \]

where \( G_{d}^{-1}(\sigma_1, \sigma_2) = \partial_{\tau} - \bar{\epsilon}_d + V^2 G_c + \alpha_{\mathbf{r}} \{ G_{j} G_{c} B G_{c} A \} \) is the propagator of the \( d \)-electrons in the presence of...
the conduction electrons and the rare earth impurity ($\sigma_i = 1, 2$ for spin up and spin down respectively). The quadratic term in $m$ represents effective anisotropy and spin-charge interactions and is given by

$$K_{\sigma_1\sigma_2\sigma_3\sigma_4} = \frac{-U}{4} (\delta_{\sigma_1\sigma_2}\delta_{\sigma_3\sigma_4} - 2\delta_{\sigma_1\sigma_3}\delta_{\sigma_2\sigma_4})$$

$$-2V^4G_f(G_cBG_cAG_c)_{\sigma_1\sigma_2}\delta_{\sigma_3\sigma_4} \delta_{\sigma_3\sigma_4} \delta_{\sigma_2\sigma_2}$$

Integrations over momentum and spin are implied in all these expressions. The different terms that appear in $K$ are as follows: $G_c$ is the Green’s function of the conduction electrons in the mean field approximation

$$G_c^{-1}(k,\sigma_1, k',\sigma_2, \tau) = (\partial_{\tau} + \varepsilon_{k\sigma_1} - \mu_F)\delta_{kk'}\delta_{\sigma_1\sigma_2}$$

$$+i\lambda(k, k') (2 - g_f) \langle J_f^\sigma \rangle \left( k_x' k_y - k_y' k_x \right) \delta_{\sigma_1\sigma_2}, \quad (8)$$

which is off-diagonal in momentum due to the orbit-orbit coupling. $\varepsilon_{k\sigma}$ now includes Zeeman terms due to the external field and the $z$-component of the field due to impurity. The propagator $G_f$ is that of the $f$-ions in the presence of both the conduction electrons and the transition ions, $G_f^{-1}(\tau) = \partial_{\tau} + \mu_f H + Tr_{k\sigma} \{G_cAG_cB\}$. The $A$ and $B$ matrices are solely due to the presence of the impurity and represent the indirect coupling between the transition ions and the $f$-ions

$$A(k',\sigma_1; k,\sigma_2) = B(k,\sigma_1; k',\sigma_2)^* = \Gamma_0\delta_{\sigma_1\sigma_2} - i\lambda_0\Delta^+_{kk'}\quad (9)$$

where we have set $\Gamma_0 = \frac{\sqrt{2J_f}}{2} (g_f - 1)$, $\lambda_0 = \frac{\lambda\sqrt{2J_f}}{2} (2 - g_f)$, and $\Delta^+_{kk'} = \left(k' \times k\right)_{\pm}^\dagger$. In the trace log term of the effective Lagrangian, the first nontrivial contribution is of order $V^4$ and is given by Fig. 1. The diagram with a single insertion of an $f$-propagator does not contribute due to the antisymmetry of the orbit-orbit coupling in the momentum space. Varying the effective

$$C(\omega) = \sum_{n,m} m_{11}(\omega_n)m_{22}(\omega_n + \omega_l) \quad \text{for} \quad \omega_l = (2l + 1)\pi/\beta \text{ for integer } l.$$
virtual mixing parameter data is well reproduced by the orbit-orbit coupling and 2, without the orbit-orbit coupling.

The squares represent damping due to s-f coupling only, Eq. 13 (solid line) and Eq.19 (dashed line) to the data of Ref. 12. The experimental measurements (triangles) clearly show that at room temperature non-spin flip scattering is more important than spin-flip scattering which only becomes important close to the critical temperature. Again, the predicted increase of damping is proportional to 1-10 eV compared to 0.01 eV for spin-orbit coupling.

The orbit-orbit coupling between the conduction electrons and the impurity ions. For near equilibrium conditions and in the low frequency regime this leads to damping for the uniform mode that is of Gilbert form. The orbit-orbit mechanism introduced here is much stronger than the spin-orbit based Elliott-Yafet-Kambersky mechanism since the charge-spin coupling at the host ion is of the order of 1-10 eV compared to 0.01 eV for spin-orbit coupling. The predicted increase of damping is proportional to $V^4$ which in transition ions is of the same order as $U$ the Coulomb potential. A localized model for the d-moments based on the s-d exchange is unable to account for the increase in damping in these doped systems as a function of the orbital moment of the rare-earth impurities. An additional test of this damping theory would be to measure the effect of a single rare earth element on the damping in various transition metals. Such experiments will provide further insight into the dependence of damping on $V$ and will improve our understanding of the itinerant versus localized pictures of magnetism.

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