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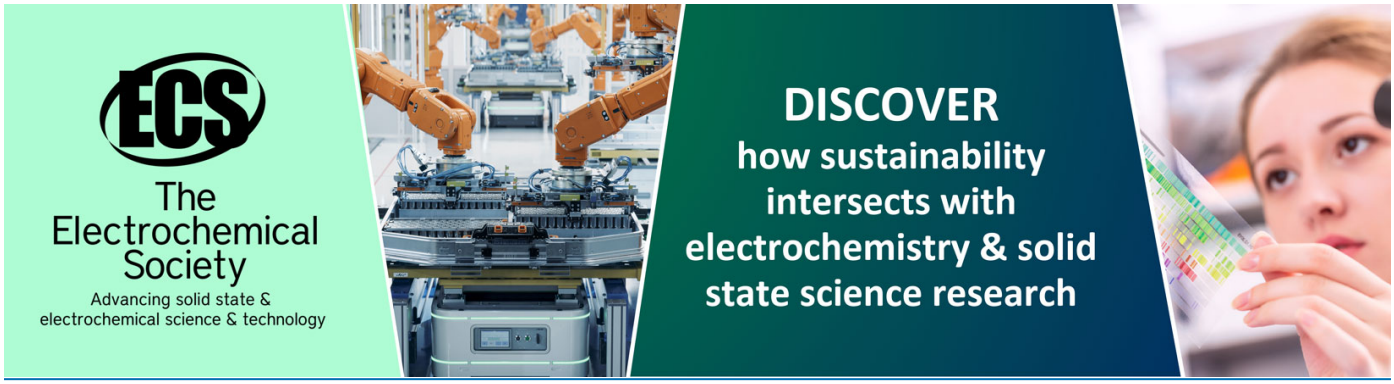

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Demagnetization on the fs time-scale by the Elliott-Yafet mechanism

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Abstract. An ultrashort laser pulse can change the magnetization of ferromagnetic metals such as Ni in less than a picosecond. Thereby, angular momentum is transferred from the spin system to the lattice. One possible candidate for this transfer is the Elliott-Yafet mechanism of spin-orbit mediated spin-flip scattering of electrons at phonons. Former ab-initio calculations have shown that for Ni and Co the Elliott-Yafet spin-mixing parameter which describes the degree of mixing of the two spin states for the electronic eigenstates – averaged over all states involved in the demagnetization process – is large enough to explain the experimentally observed demagnetization rates. In the present paper we calculate in addition the spin-mixing for the individual electronic states as function of their wavevector. Furthermore, the theory is extended to the case of Gd for which experiments have revealed a slower demagnetization dynamics than for Ni.

Experiments have shown (see Ref. [1, 2, 3] and references therein) that upon exposure of a ferromagnetic metal film to a strong optical femtosecond (fs) laser pulse the spin magnetization can be quenched within a few hundred fs. Simple estimates [4] can be given that the angular momentum transferred directly from the photons to the sample is too small to cause this finding. Thus the angular momentum is transferred extremely fast from the spin system to either electronic degrees of freedom (orbital moments) or to lattice degrees of freedom. Experiments using the fs time-resolved x-ray absorption spectroscopy have excluded the first case [1]. The Elliott-Yafet mechanism [5, 6] that is to be discussed here is one possible candidate for the latter case.

In this mechanism the elementary process is a scattering of an electron at a phonon in a system with spin-orbit coupling. This scattering event changes the probability to find the electron in one of the spin states $|\uparrow\rangle$ or $|\downarrow\rangle$, thus delivering angular momentum from the electronic system to the lattice. In the discussion of femtosecond magnetization dynamics the Elliott-Yafet mechanism is used often (e. g., Ref. [7, 8]), however, it was unclear whether it can describe the experimental findings also quantitatively. In the present paper we present numerical results from the ab-initio density functional electron theory that will support the use of the Elliott-Yafet mechanism as one possibility to understand the experiments quantitatively.

In [5, 6] the so-called Elliott-Yafet relation for the longitudinal relaxation time T_1 of the magnetic moment,

$$\frac{1}{T_1} = p\, b^2 \frac{1}{\tau} \,, \tag{1}$$

	Z	b^2_{thermal}	b^2_{optical}
Ni	28	0.025	0.045
Cu	29	0.001 4	0.013
Gd	64	0.059	0.062

Table 1. Spin-mixing parameter b^2 for various metals for states in the range of thermal ($\sigma = 25\,\text{meV}$) and optical ($\sigma = 1.4\,\text{eV}$) energies around ε_F. For details of the calculation and for results on other metals see [11]

is suggested where τ is the relaxation time from the electrical resistivity, b^2 is the spin-mixing parameter and p is a material-specific parameter which has been estimated to lie between 1 and 10 [9, 10]. The spin-mixing parameter b^2 ranges between 0 for pure spin states and 0.5 for totally mixed states (where the latter case is not included in the Elliott-Yafet theory) and is defined as the average of the spin-mixing $b^2_\mathbf{k}$ of the individual electronic states with wavevector $\mathbf{k}$ at or near the Fermi energy where the system is disturbed by the laser excitation. These can be defined as $b^2_\mathbf{k} = \min(p_{\mathbf{k}\uparrow}, p_{\mathbf{k}\downarrow})$ where the $p_{\mathbf{k}s}$ is the probability to find an electron in the spin state s,

$$\langle\psi_\mathbf{k}|\psi_\mathbf{k}\rangle = \langle\psi_\mathbf{k}|\uparrow\rangle\langle\uparrow|\psi_\mathbf{k}\rangle + \langle\psi_\mathbf{k}|\downarrow\rangle\langle\downarrow|\psi_\mathbf{k}\rangle = p_{\mathbf{k}\uparrow} + p_{\mathbf{k}\downarrow} = 1 \tag{2}$$

with $0 < p_{\mathbf{k}s} < 1$. An additional band index is omitted because in the theory of Yafet [6] only scattering events within the same band are considered.

Using a different and more complicated set of rate equations than in [6] and taking from the Elliott-Yafet-theory a spin-flip probability in an electron-phonon scattering event of $\alpha_\text{EY} = p\,b^2$ for each electron-phonon scattering event, it has been concluded [7] that a value of α_EY well below 1 may suffice to reproduce the experimentally observed ultrafast demagnetization. Moreover, as b^2 for Ni was not known, the value of Cu has been inserted which is its nearest neighbour in the periodic table. However, this value is too low to explain the experiments quantitatively [7].

In Ref. [11] we have calculated the values of b^2 for several metals by the ab-initio density functional electron theory in the local-spin-density approximation, for details see [11]. For the ferromagnetic transition metals (and Cu) the states that are excited by the laser and for which the spin-mixing is averaged over are the 3d4s4p valence electrons which also carry the itinerant magnetic moment. At present it is unclear whether the hot electrons are directly responsible for the demagnetization or whether there is a first redistribution of energy within the electronic system followed by a subsequent step of scattering events leading to demagnetization. Therefore, we calculated b^2 by averaging over all electronic states around ε_F with a Gaussian weight of width σ. For the smearing width we insert on the one hand $\sigma = 0.025\,\text{eV}$ (which corresponds to the situation that only excitation energies close to the thermal energies at room temperature would be relevant) and on the other hand $\sigma = 1.4\,\text{eV}$ (which corresponds to the full laser energy). For $\sigma = 25\,\text{meV}$, the value of b^2 (Table 1) for Ni is a factor of about 18 larger than the one for Cu, and about a factor of 100 larger than the commonly assumed value of $b^2 = \alpha_\text{EY}/4$ [7]. Similar values are obtained for Fe and Co [11]. Our value for Ni is large enough to explain the experiments [11].

In the present paper we extend the theory in two respects. First, we calculate for Ni in addition to the averaged quantity b^2 the spin-mixing $b^2_\mathbf{k}$ of the individual electronic states with wavevector $\mathbf{k}$. The objective of this calculation is to investigate whether there are regimes in the Brillouin zone for which the spin-mixing is much stronger than for states from other regions of the Brillouin zone. Second, we calculate b^2 for the case of Gd. Experiments have shown [12] that for this rare-earth metal the demagnetization dynamics is much slower than for Ni or Co. The question is whether this is related to a much smaller spin-mixing parameter b^2.

Table 2. Histogram of the spin-mixing of the states in an energy range of 4 mRy around the Fermi energy for Ni. The last line gives the contribution to the average spin-mixing by these states.

$b^2_{\mathbf{k}}$	$> 20\%$	$\ldots 10\%$	$\ldots 5\%$	$\ldots 2\%$	$\ldots 1\%$	$\ldots 0.5\%$	$\ldots 0.2\%$	$\ldots 0.1\%$	$< 0.1\%$
number	4%	3%	3%	5%	11%	12%	18%	29%	14 %
contrib.	54%	16%	9%	7%	6%	4%	2%	2%	$< 1\%$

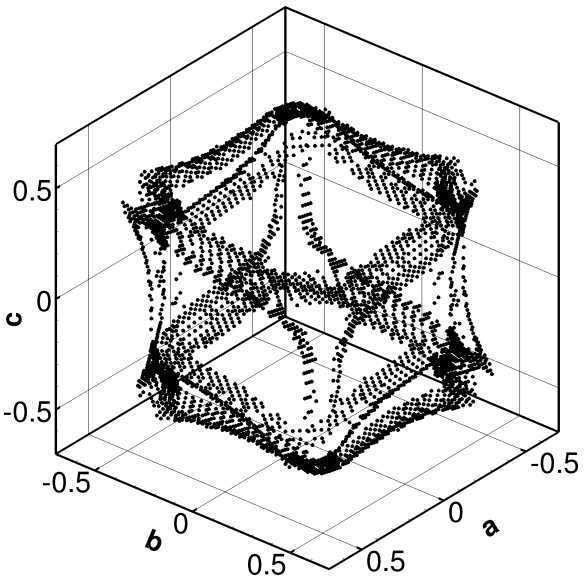

Figure 1. Positions of states with more than 20% spin-mixing in an energy range of 4 mRy around the Fermi energy in the Brillouin zone of fcc Ni. Note that the calculations have been performed on a discrete mesh of k-points in the Brillouin zone [11].
The directions of the axes are $\mathsf{a} = \frac{1}{2\sqrt{3}}(1 + \sqrt{3}, 1 - \sqrt{3}, -2)$, $\mathsf{b} = \frac{1}{2\sqrt{3}}(1 - \sqrt{3}, 1 + \sqrt{3}, -2)$ and $\mathsf{c} = \frac{1}{2\sqrt{3}}(2, 2, 2)$. The units are given in 2π/lattice constant.

Our results for the spin-mixing of the individual electronic states are presented in Table 2 and in Figure 1. Table 2 shows the distribution of the spin-mixing of the states in an energy range of 4 mRy around ε_{F} in Ni. Only 4% of the states near ε_{F} have a strong spin-mixing of more than 20%, however, they contribute more than 50% to the mean spin-mixing value. Therefore, these strongly mixed states are of great importance, although the connection between the rate equation and the relaxation of the magnetic moment becomes more difficult when the states have a broad spectrum of different mixing values. Figure 1 shows the distribution of these states in the Brillouin zone which is of interest for experimental investigations of the band-structure effects on the spin-dependent scattering, e. g., by angle-resolved photoemission spectroscopy. It is found here that these states are along the crossing lines of the (dominant) spin-up and spin-down Fermi surface sheets. Note that earlier investigations on Al found a more spot-like distribution of the states with high spin-mixing in the Brillouin zone [10].

We now extend the theory to the case of Gd where both the localized 4f states and the 5d6s6p valence states contribute to the magnetic behaviour. In this material the laser excites the valence states whereas the energetically lower-lying 4f states are primarily unaffected. Evidence has been given [13] that, nevertheless, there is a simultaneous demagnetization of the itinerant 5d6s6p moments and the 4f moments. We therefore assume that the Elliott-Yafet scattering processes appear entirely within the system of valence electrons, providing the driving mechanism for the demagnetization, whereas the 4f moments are rigidly coupled to the valence moments by the 4f-5d6s6p exchange coupling. It therefore suffices to calculate b^2 for the valence states. In line with this physical model, we calculate the occupied and unoccupied valence states of Gd for the

effective potential of the ferromagnetic ground state, thereby treating the 4f states as core states in the line described in Ref. [14].

As shown in Table 1, we find for the rare-earth metal Gd a spin-mixing parameter of similar magnitude as for the transition metal Ni. Obviously the different demagnetization time scales for Gd and Ni do not originate from drastically different primary spin flip rates of the conduction electrons in these two materials. Further investigations are needed, e. g., on the question whether 4f states and valence states are indeed rigidly coupled during the magnetization process, as assumed in our model calculation.

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