PHYSICAL REVIEW B **79**, 140401(R) (2009)

Elliott-Yafet mechanism and the discussion of femtosecond magnetization dynamics

D. Steiauf and M. Fähnle*
Max-Planck-Institut für Metallforschung, Heisenbergstraße 3, 70569 Stuttgart, Germany
(Received 10 February 2009; published 3 April 2009)

In the past it has been questioned whether the Elliott-Yafet mechanism of spin-flip scattering of electrons at phonons may explain the ultrafast demagnetization of a ferromagnet (mostly Ni) after femtosecond laser excitation. The reason has been that the value of the Elliott-Yafet spin-mixing parameter b^2 of Ni has not been known and has been taken as the one of Cu (which is the nearest neighbor in the periodic table) which is very small. We present calculations of b^2 by the *ab initio* density-functional electron theory for Co, Fe, Ni, Cu, Au, and K. For Co, Fe, and Ni b^2 is about a factor of 25 larger than the value commonly assumed for Cu. This result should support the Elliott-Yafet mechanism in the discussion of fs magnetization dynamics.

DOI: 10.1103/PhysRevB.79.140401 PACS number(s): 75.40.Gb, 76.20.+q, 78.47.J−

It has been shown experimentally (see Refs. 1–3 and references therein) that the spin angular momentum of a metallic ferromagnetic film can be quenched in a few hundred femtoseconds (fs) by exciting the ferromagnet with a strong optical fs laser pulse. A simple estimate[4] for the case of Ni (for which many of the fs demagnetization experiments have been performed) revealed that the direct exchange of angular momentum between the film and the photon field is very small. Therefore there must be an extremely fast transfer of angular momentum from the electronic spin degrees of freedom either to the electronic orbital degrees of freedom—this could be excluded by fs time-resolved x-ray absorption spectroscopy[1]—or to the atomic orbital moments of the lattice. One possible mechanism for this transfer is discussed in the Elliott-Yafet scattering theory.[5,6] In this theory it is shown that a scattering event of an excited electron at a phonon changes the probability to find that electron in one of the spin states $|\uparrow\rangle$ or $|\downarrow\rangle$, thus delivering angular momentum from the electronic system to the lattice. Albeit there is no doubt that such a mechanism exists, and although it is quite popular (see, e.g., Refs. 7–10) to consider the Elliott-Yafet mechanism in the context of femtosecond magnetization dynamics, there have been always strong discussions on whether this mechanism can indeed explain these experiments quantitatively. In the present Rapid Communication it will be shown by means of the *ab initio* density-functional electron theory that the Elliott-Yafet mechanism in fact is a possible candidate for the quantitative explanation of the fs demagnetization experiments.

Because of the spin-orbit coupling, an electronic state in a solid is always a mixture of the two spin states, e.g., a dominant spin-up contribution $a_{\mathbf{k}}(\mathbf{r})\exp(i\mathbf{k}\cdot\mathbf{r})|\uparrow\rangle$ and a small spin-down contribution $b_{\mathbf{k}}(\mathbf{r})\exp(i\mathbf{k}\cdot\mathbf{r})|\downarrow\rangle$, defining the dominant "spin-up" crystal wave function,

$$\psi_{\mathbf{k},\uparrow} = [a_{\mathbf{k}}(\mathbf{r})|\uparrow\rangle + b_{\mathbf{k}}(\mathbf{r})|\downarrow\rangle]e^{i\mathbf{k}\cdot\mathbf{r}}. \tag{1}$$

Here $\mathbf{k}$ is the wave vector. We omit the band index because in the general theory of Yafet[6] only scattering events between two Bloch states belonging to the same band are considered. In the following we write the potential describing the interaction of an electron with a phonon of wave vector $\mathbf{q}$ as

$$W_{\mathbf{q}} = W_{\mathbf{q}}^{\text{ordinary}} + W_{\mathbf{q}}^{\text{spin-orbit}}, \tag{2}$$

where $W_{\mathbf{q}}^{\text{ordinary}}$ describes the spin-independent part and $W_{\mathbf{q}}^{\text{spin-orbit}}$ refers to the spin-dependent part of the scattering potential. The matrix element which is of interest for the depolarization is the spin-flip matrix element between a dominant spin-up state $\psi_{\mathbf{k},\uparrow}$ and a dominant spin-down state $\psi_{\mathbf{k+q},\downarrow}$,

$$M_{\mathbf{k+q},\downarrow;\mathbf{k},\uparrow} = \langle\psi_{\mathbf{k+q},\downarrow}|W_{\mathbf{q}}^{\text{ordinary}} + W_{\mathbf{q}}^{\text{spin-orbit}}|\psi_{\mathbf{k},\uparrow}\rangle. \tag{3}$$

It was a key observation of Elliott[5] that the first part of the matrix element $M_{\mathbf{k+q},\downarrow;\mathbf{k},\uparrow}$ of Eq. (3) originating from $W_{\mathbf{q}}^{\text{ordinary}}$ is already nonzero because it connects the large and small components of the two involved spinors, and he neglected the second part completely. Later on Yafet[6] noted that it is essential to take into account both parts of the matrix elements.

We denote the numbers of electrons in dominant spin-up states and in dominant spin-down states as $N_\uparrow$ and $N_\downarrow$. Assuming that all the electronic states $\psi_{\mathbf{k},\uparrow}$ or $\psi_{\mathbf{k},\downarrow}$ carry a magnetic moment of the same size, the total magnetic moment of the system is proportional to the population difference $D = N_\uparrow - N_\downarrow$. If $W_{\uparrow,\downarrow}$ is the number of transitions per unit time from spin-up to spin-down and if $W_{\downarrow,\uparrow}$ describes the analogous transition rate, then the relation

$$\frac{dD}{dt} = 2(W_{\uparrow,\downarrow} - W_{\downarrow,\uparrow}) \tag{4}$$

holds, where the factor 2 appears because a spin flip changes the population difference by 2.

In Ref. 6 a situation has been considered for which the system is close to equilibrium and where the deviations from the equilibrium can be accounted for by introducing two different Fermi energies, $\varepsilon_{F,\uparrow}(t) \neq \varepsilon_F^0$ and $\varepsilon_{F,\downarrow}(t) \neq \varepsilon_F^0$, where ε_F^0 is the common Fermi energy of the two spin channels for the case of equilibrium. Furthermore, it has been assumed that the system is invariant with respect to the inversion of both space and time, implying

$$\varepsilon_{\mathbf{k},\downarrow} = \varepsilon_{\mathbf{k},\uparrow}, \quad M_{\mathbf{k+q},\downarrow;\mathbf{k},\uparrow} = -(M_{\mathbf{k+q},\uparrow;\mathbf{k},\downarrow})^*. \tag{5}$$

Equation (4) then can be written as

$$\frac{dD}{dt} = -\frac{D - D_0}{T_1}, \tag{6}$$

with the spin-relaxation time T_1. An analytical expression for T_1 [Eq. (18.6) of Ref. 6] can be given which involves an average over all phonon wave vectors $\mathbf{q}$ of a function which in turn contains $|M_{\mathbf{k+q},\downarrow;\mathbf{k},\uparrow}|^2$. We have redone the calculation of Ref. 6 for a ferromagnet for which $\varepsilon_{\mathbf{k},\downarrow} \neq \varepsilon_{\mathbf{k},\uparrow}$ and where $M_{\mathbf{k+q},\downarrow;\mathbf{k},\uparrow}$ and $M_{\mathbf{k+q},\uparrow;\mathbf{k},\downarrow}$ are not interrelated via Eq. (5). This calculation yields again Eq. (6), and T_1 can be expressed once again as an average over all $\mathbf{q}$ of a slightly different function which contains $|M_{\mathbf{k+q},\downarrow;\mathbf{k},\uparrow}|^2$ and $|M_{\mathbf{k+q},\uparrow;\mathbf{k},\downarrow}|^2$. Thus, in both cases T_1 may be calculated if these matrix elements are known.

Instead of calculating the matrix elements explicitly for a given material, Yafet[6] gave a rough estimate for them which shows that they scale with the respective minor contribution to the wave function given by Eq. (1). Furthermore, he showed that his expression for T_1 for systems which obey Eq. (5) and the expression for the relaxation time τ for the electrical resistivity are closely related, suggesting the Elliott-Yafet relation[5,6,11–15]

$$\frac{1}{T_1} = pb^2\frac{1}{\tau}. \tag{7}$$

Here p is a material-specific parameter which has been estimated to be between 1 and 10. In Eq. (7) the spin-mixing parameter b^2 characterizes the degree of spin mixing for the involved states $\psi_{\mathbf{k}}$, where $\psi_{\mathbf{k}}$ stands symbolically for dominant spin-up states $\psi_{\mathbf{k},\uparrow}$ or dominant spin-down states $\psi_{\mathbf{k},\downarrow}$. It can be defined via the normalization

$$\langle\psi_{\mathbf{k}}|\psi_{\mathbf{k}}\rangle = \langle\psi_{\mathbf{k}}|\uparrow\rangle\langle\uparrow|\psi_{\mathbf{k}}\rangle + \langle\psi_{\mathbf{k}}|\downarrow\rangle\langle\downarrow|\psi_{\mathbf{k}}\rangle = p_{\mathbf{k}\uparrow} + p_{\mathbf{k}\downarrow} = 1, \tag{8}$$

as

$$b^2 = \overline{\min(p_{\mathbf{k}\uparrow}, p_{\mathbf{k}\downarrow})}, \tag{9}$$

where the bar denotes a suitably defined average over all involved states. Here $0 \leq p_{\mathbf{k}\uparrow,\downarrow} \leq 1$ and thus $0 \leq b^2 \leq 0.5$, where $b^2 = 0$ means that all relevant states are pure spin states and $b^2 = 0.5$ stands for totally mixed states. (The latter case is not included in the Elliott-Yafet reasoning.) Because the relation for T_1 which we found for ferromagnets is closely related to Yafet's expression [Eq. (18.6) of Ref. 6] for systems which obey Eq. (5), we adopt in the following the Elliott-Yafet relation also for the case of ferromagnets.

In principle, the Elliott-Yafet theory should not be applied to discuss ultrafast demagnetization experiments after a fs laser pulse because in these experiments the system is driven far out of the equilibrium, whereas the theory of Elliott and Yafet is valid for a situation close to equilibrium. Because of the lack of a better theory, a relation of the form of Eq. (7) or a theory in the spirit of the Elliott-Yafet theory[7–10] have often been used to estimate whether an Elliott-Yafet mechanism is able to explain the ultrafast demagnetization rates after a fs laser pulse has hit, e.g., a thin film of Ni. Assigning a spin-flip probability $\alpha_{\mathrm{EY}} = pb^2$ to each electron-phonon scattering event, it has been concluded[7,8] that a value of α_{EY} well below 1 may suffice to reproduce the experimentally observed ultrafast demagnetization. Because for Ni the value of b^2 was not known so far, it has been often assumed that it should be close to the one of Cu which is the nearest neighbor of Ni in the periodic table. The corresponding value of α_{EY}, however, is very low (about 10^{-3}, Ref. 7) so that it has been doubted that the Elliott-Yafet mechanism is able to explain the fs demagnetization experiments. However, it must be taken into account that the metals Ni and Cu are electronically very different. Whereas in Cu there are only very few holes in the d band (see, e.g., Ref. 16), Ni has a noncomplete d band, and this may have severe consequences for the value of b^2.

We therefore have calculated the values of b^2 for a variety of metals (Ni, Cu, Fe, Co, K, and Au) by the *ab initio* density-functional electron theory in the local-spin-density approximation[17] and by the tight-binding linear-muffin-tin-orbital method[18] in which the tools for the treatment of the spin-orbit coupling have been implemented.[19] Most of the laser-pulse experiments have been performed at room temperature, our calculations, however, are for zero temperature. We then can calculate the occupied and nonoccupied single-electron states for the effective potential of the ferromagnetic ground state. The laser pulse heats up the system more or less immediately, i.e., we can assume (sudden approximation) that after the pulse the excited electrons occupy electronic states of the effective ground-state potential which where unoccupied before the pulse. Thereby excited electrons in a range between the Fermi energy ε_F and $\varepsilon_F + \hbar\omega_{\mathrm{photon}}$ are produced, as well as holes in the range $\varepsilon_F - \hbar\omega_{\mathrm{photon}}$ to ε_F. Both excited electrons and holes may contribute to the demagnetization, and it is also unclear whether all of these quasiparticles contribute with equal efficiency or whether the excited electrons and holes close to ε_F are more important than the "hot" electrons or holes. To take all this into account, we calculated b^2 according to Eqs. (8) and (9) where the bar in Eq. (9) means that we average over all electronic states near ε_F which we can get by a Brillouin-zone sampling with a Gaussian smearing function for the occupation numbers which is located at ε_F and which has a smearing parameter σ. For the smearing parameter we insert on the one hand $\sigma = 0.025$ 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 $\sigma = 1.4$ eV (which would mean that all quasiparticles which can be produced by an optical laser pulse of 1.4 eV contribute). The effective potential is calculated for a sampling of the reciprocal space with 512.000 k points (163.840 in the case of Co) in the Brillouin zone.

At finite temperatures for which most of the experiments are performed there are already thermally activated spin waves which reduce the magnetic moment by $\approx 5\%$ in Ni. Due to these spin waves the atomic magnetic moments are no longer ferromagnetically aligned but they exhibit a noncollinearity. This noncollinearity produces a spin mixing of the same form as given by Eq. (1) even if the system had no spin-orbit coupling. However, the matrix elements for the spin-flip scattering evaluated with the corresponding amplitudes $a_{\mathbf{k}}(\mathbf{r})$ and $b_{\mathbf{k}}(\mathbf{r})$ then are necessarily zero as long as we neglect the spin-dependent part $W_{\mathbf{q}}^{\text{spin-orbit}}$ of the scattering potential because in a system without spin-orbit coupling the

TABLE I. Spin-mixing parameter b^2 for various metals for states in the range of thermal (σ=25 meV) and optical (σ=1.4 eV) energies around ε_F.

	Z	b^2_{thermal}	b^2_{optical}
K	19	0.000003	0.00025
Fe	26	0.024	0.037
Co	27	0.011	0.049
Ni	28	0.025	0.045
Cu	29	0.0014	0.013
Au	79	0.026	0.051

angular momentum of the spin system is conserved in a scattering process. When we switch on the spin-orbit coupling it may be that the spin-flip scattering probability is modified as compared to the spin-orbit-mediated spin-flip scattering in a ferromagnetically aligned system. Our guess is that it would even increase the probability for a spin-flip scattering as compared to the ferromagnetic situation. As a second effect, the reduced magnetic moment at room temperature will slightly change the effective potential which the electrons feel but we think that this is only a minor effect. Altogether, it would be desirable to perform experiments at various temperatures to figure out whether the thermal excitations have a sizeable effect on the spin-flip scattering.

Table I shows our results for b^2. For σ=25 meV, the value of b^2 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. For the three ferromagnetic metals the values of b^2 are similar to the one of Au which has—such as Cu—only very few holes in the d band[16] but which is a 5d metal for which the spin-orbit coupling is very large. In contrast, for K the b^2 value is extremely small. The spin mixing of states in the optical energy range is larger than in the thermal range for all metals considered here.

As outlined above, a value of α_{EY}=0.1 suffices to explain the experimentally observed demagnetization rate in Ni.[8] Our calculated value of b^2=0.025 for Ni leads exactly to this required value using p=4. This demonstrates that the Elliott-Yafet mechanism in principle is able to describe the fast transfer of angular momentum from the spin system to the lattice system in Ni.

To conclude, our calculations have shown that the spin-mixing parameter b^2 of Ni, Fe, and Co is much larger than the one of Cu which so far has been used for the discussion of fs demagnetization experiments on Ni. We therefore think that our calculations support the Elliott-Yafet mechanism for the discussion of the ultrafast demagnetization after fs laser pulses.

The authors are indebted to B. Koopmans, M. Aeschlimann, H. A. Dürr, and R. Singer for helpful discussions.

*faehnle@mf.mpg.de

[1] C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupulescu, E. F. Aziz, M. Wietstruk, H. A. Dürr, and W. Eberhardt, Nature Mater. **6**, 740 (2007).

[2] B. Koopmans, Nature Mater. **6**, 715 (2007).

[3] M. Cinchetti, M. Sánchez Albaneda, D. Hoffmann, T. Roth, J. P. Wüstenberg, M. Krauß, O. Andreyev, H. C. Schneider, M. Bauer, and M. Aeschlimann, Phys. Rev. Lett. **97**, 177201 (2006).

[4] B. Koopmans, M. van Kampen, and W. J. M. de Jonge, J. Phys.: Condens. Matter **15**, S723 (2003).

[5] R. J. Elliott, Phys. Rev. **96**, 266 (1954).

[6] Y. Yafet, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1963), Vol. 14.

[7] B. Koopmans, H. H. J. E. Kicken, M. van Kampen, and W. J. M. de Jonge, J. Magn. Magn. Mater. **286**, 271 (2005).

[8] B. Koopmans, J. J. M. Ruigrok, F. Dalla Longa, and W. J. M. de Jonge, Phys. Rev. Lett. **95**, 267207 (2005).

[9] G. M. Müller, J. Walowski, M. Djordjevic, G.-X. Miao, A. Gupta, A. V. Ramos, K. Gehrke, V. Moshnyaga, K. Samwer, J. Schmalhorst, A. Thomas, A. Hütten, G. Reiss, J. S. Moodera, and M. Münzenberg, Nature Mater. **8**, 56 (2009).

[10] J. Walowski, G. Müller, M. Djordjevic, M. Münzenberg, M. Kläui, C. A. F. Vaz, and J. A. C. Bland, Phys. Rev. Lett. **101**, 237401 (2008).

[11] J. Fabian and S. Das Sarma, Phys. Rev. Lett. **81**, 5624 (1998).

[12] J. Fabian and S. Das Sarma, Phys. Rev. Lett. **83**, 1211 (1999).

[13] F. Beuneu and P. Monod, Phys. Rev. B **18**, 2422 (1978).

[14] J. Fabian and S. Das Sarma, J. Vac. Sci. Technol. B **17**, 1708 (1999).

[15] I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).

[16] S. Schweizer, C. Elsässer, K. Hummler, and M. Fähnle, Phys. Rev. B **46**, 14270 (1992).

[17] Correlation part from J. P. Perdew and Y. Wang, Phys. Rev. B **45**, 13244 (1992); Exchange part from J. P. Perdew and Y. Wang, *ibid.* **33**, 8800 (1986).

[18] O. K. Andersen and O. Jepsen, Phys. Rev. Lett. **53**, 2571 (1984).

[19] C. Ederer, M. Komelj, M. Fähnle, and G. Schütz, Phys. Rev. B **66**, 094413 (2002).