Ab Initio Investigation of the Elliott-Yafet Electron-Phonon Mechanism in Laser-Induced Ultrafast Demagnetization

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The spin-flip (SF) Eliashberg function is calculated from first principles for ferromagnetic Ni to accurately establish the contribution of Elliott-Yafet electron-phonon SF scattering to Ni's femtosecond laser-driven demagnetization. This is used to compute the SF probability and demagnetization rate for laser-created thermalized as well as nonequilibrium electron distributions. Increased SF probabilities are found for thermalized electrons, but the induced demagnetization rate is extremely small. A larger demagnetization rate is obtained for nonequilibrium electron distributions, but its contribution is too small to account for femtosecond demagnetization.

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Ultrafast demagnetization of ferromagnetic metals through excitation by a femtosecond laser pulse was discovered 15 years ago by Beaurepaire *et al.* [1]. In spite of intensive investigations, the microscopic origin of the ultrafast demagnetization could not be disclosed and continues to be controversially debated (see [2] for a recent review). Several mechanisms have been proposed to explain the observed ultrafast phenomenon [3–11]. Most of these theories assume the existence of an ultrafast spin-flip (SF) channel, which would cause dissipation of spin angular momentum within a few hundred femtoseconds.

Elliott-Yafet electron-phonon SF scattering has been proposed as a mechanism for ultrafast spin dissipation [4]. Strong support in favor of electron-phonon-mediated spin flips as the actual mediator of the femtosecond demagnetization was supplied in a very recent work, in which *ab initio* calculated SF probabilities for thermalized electrons compared favorably to SF probabilities derived from pump-probe demagnetization measurements [8]. While these results definitely favor the Elliott-Yafet SF scattering mechanism, the calculation of the electron-phonon scattering involved several serious approximations. Applying the so-called Elliott approximation [12], only spin mixing due to spin-orbit coupling in the *ab initio* wave functions was included, but no electron-phonon matrix elements and no real phonon dispersion spectrum were considered. The thus-obtained SF probability is, however, not a direct measure of demagnetization. Recent model simulations for thermalized hot electrons [9] using the Landau-Lifshitz-Bloch equation [13] and assuming a fitted SF parameter did reproduce the experimental magnetization response but could not assign the SF origin. Hence, it remains a crucial, open question whether laser-induced demagnetization can indeed be attributed to electron-phonon-mediated SF scattering.

Here, we report an *ab initio* investigation to accurately establish the extent to which the Elliott-Yafet electron-phonon SF scattering contributes to fs demagnetization. To this end, we perform *ab initio* calculations for ferromagnetic Ni, in which ultrafast magnetization decay is well-documented [1,8,14]. We include the full electron-phonon matrix elements and phonon dispersions in our calculations. Introducing an energy-dependent SF Eliashberg function, we compute SF probabilities and demagnetization rates for laser-heated thermalized electrons as well as laser-induced nonequilibrium electron distributions, from which we draw qualified conclusions on the phonon-mediated SF scattering, which we find to be too small to be accountable for ultrafast demagnetization.

To treat phonon-mediated SF scattering at variable electron energies, we define a *generalized* energy- and spin-dependent Eliashberg function,

$$\alpha^2_{\sigma\sigma'}F(E,\Omega)=\frac{1}{2M\Omega}\sum_{\nu,n,n'}\iint d\mathbf{k}\,d\mathbf{k}'\,g^{\nu\sigma\sigma'}_{\mathbf{k}n,\mathbf{k}'n'}(\mathbf{q})\times\delta(\omega_{\mathbf{q}\nu}-|\Omega|)\delta(E^{\sigma}_{\mathbf{k}n}-E)\delta(E^{\sigma'}_{\mathbf{k}'n'}-E), \quad (1)$$

which comprises initial and final electron states with quantum numbers $\mathbf{k}n$ and $\mathbf{k}'n'$ that interact through a phonon with frequency $\Omega=\omega_{\mathbf{q}\nu}$; ν and $\mathbf{q}$ denote its mode and wave vector. M is the ionic mass, and $\sigma=\uparrow,\downarrow$ denote the spin majority and minority components. For $E=E_F$ (the Fermi energy), the SF part $\alpha^2_{\uparrow\downarrow}F(E_F,\Omega)$ gives the SF Eliashberg function [15] and the sum over all $\sigma\sigma'$ corresponds to the standard Eliashberg function, $\alpha^2F(E_F,\Omega)$ [16]. The (squared) electron-phonon matrix elements are

$$g^{\nu\sigma\sigma'}_{\mathbf{k}n,\mathbf{k}'n'}(\mathbf{q})=|\mathbf{u}_{\mathbf{q}\nu}\cdot\langle\Psi^{\sigma}_{\mathbf{k}n}|\nabla_{\mathbf{R}}V|\Psi^{\sigma'}_{\mathbf{k}'n'}\rangle|^2, \quad (2)$$

where V is the potential, $\mathbf{u}_{\mathbf{q}\nu}$ is the phonon polarization vector, and $|\Psi^{\sigma}_{\mathbf{k}n}\rangle$ are the eigenstates in the ferromagnet. Momentum conservation requires $\mathbf{q}=\mathbf{k}'-\mathbf{k}$. SF scattering becomes possible through the relativistic spin-orbit coupling. The majority and minority Bloch states $|\Psi^{\uparrow}_{\mathbf{k}n}\rangle$ and $|\Psi^{\downarrow}_{\mathbf{k}n}\rangle$ can be decomposed in pure spinor components

$$|\Psi^{\uparrow}_{\mathbf{k}n}\rangle = a^{\uparrow}_{\mathbf{k}n}\begin{pmatrix}1\\0\end{pmatrix} + b^{\uparrow}_{\mathbf{k}n}\begin{pmatrix}0\\1\end{pmatrix},$$
$$|\Psi^{\downarrow}_{\mathbf{k}n}\rangle = a^{\downarrow}_{\mathbf{k}n}\begin{pmatrix}0\\1\end{pmatrix} + b^{\downarrow}_{\mathbf{k}n}\begin{pmatrix}1\\0\end{pmatrix}, \quad (3)$$

where the components $b^{\sigma}_{\mathbf{k}n}$ are nonzero only if spin-orbit coupling is present and represent the degree of spin mixing, which is a precondition for nonzero $g^{\nu\uparrow\downarrow}_{\mathbf{k}n,\mathbf{k}'n'}$.

To study demagnetization, we consider two quantities, SF probabilities and spin-resolved transition rates. The latter are defined as [17]

$$S^{\sigma\sigma'} = \iint \alpha^2_{\sigma\sigma'}F(E,\Omega)f_{\sigma}(E)[1 - f_{\sigma'}(E+\hbar\Omega)] \times [\Theta(\Omega) + N(\Omega)]d\Omega dE. \quad (4)$$

Here, $N(\Omega)$ is the phononic Bose-Einstein distribution, f_{σ} the Fermi distribution, and $\Theta(\Omega)$ the Heaviside function. Important for the effective demagnetization is the spin-decreasing rate S^-, which corresponds to $S^{\uparrow\downarrow}$, while the increasing one S^+ corresponds to $S^{\downarrow\uparrow}$.

An approximation of Eq. (4) is helpful to achieve a faster evaluation and provide more insight in the process. Energy conservation during electron-phonon scattering requires $E_{\mathbf{k}'n'} - E_{\mathbf{k}n} = \hbar\Omega$, but the phonon energy $\hbar\Omega$ is usually very small (< 0.04 eV) compared to electron-related properties. Already in the standard Eliashberg formulation, Eq. (1), an energy difference between initial and final states is neglected while the δ functions $\delta(E^{\sigma}_{\mathbf{k}n} - E)$ are broadened with a parameter (0.03 eV, here). Similarly, one can neglect the energy variation due to $\hbar\Omega$ in the Fermi function $f_{\sigma}(E+\hbar\Omega)$, as long as the temperature is high enough. We can then rewrite spin-resolved transition rates in the form

$$S^{\sigma\sigma'} = \int w_{\sigma\sigma'}(E)f_{\sigma}(E)[1 - f_{\sigma'}(E)]dE, \quad (5)$$

where we introduced the energy- and spin-dependent specific scattering rate for electrons $w_{\sigma\sigma'}$ given by

$$w_{\sigma\sigma'}(E) = \int_0^{\infty} d\Omega\, \alpha^2_{\sigma\sigma'}F(E,\Omega)[1 + 2N(\Omega)]. \quad (6)$$

Note that $w_{\uparrow\downarrow}(E) = w_{\downarrow\uparrow}(E)$. All calculations were checked against a more accurate numeric implementation not involving this approximation. The SF probability for an electron with energy E is defined as the ratio of the SF part to the corresponding total counterpart, $p_S(E) = 2w_{\uparrow\downarrow}(E)/\sum_{\sigma\sigma'}w_{\sigma\sigma'}(E)$. Analogously, the total SF probability during a scattering event can be defined as

$$P_S = (S^- + S^+)/\sum_{\sigma\sigma'}S^{\sigma\sigma'}. \quad (7)$$

Although the SF probability has been used in recent discussions of laser-induced demagnetization [8,18], it is actually not the crucial quantity (as a high but equal SF probability for both spin channels would not cause a demagnetization). We define therefore the normalized demagnetization ratio, $D_S = (S^- - S^+)/\sum_{\sigma\sigma'}S^{\sigma\sigma'}$, which tracks the difference of magnetic moment increasing and decreasing SF contributions.

To investigate phonon-induced demagnetization in laser-excited Ni, we proceed now in three steps. First, we compute the *ab initio* SF probability P_S for equilibrium Ni, i.e., for $E = E_F$. Second, we compute SF probabilities P_S for laser-heated Ni, by treating a range of electron energies that correspond to those in a hot, thermalized electron gas after laser excitation. Thermalization to electron temperatures T_e of a few thousand K occurs quickly within about 200 fs after the laser pulse, but the hot electrons are not in equilibrium with the lattice, and the lattice temperature is not altered significantly. In the third step, we consider the SF probability for nonequlibrium (NEQ) electron distributions [19] that are expected to be present within ~100 fs after laser stimulation. Demagnetization ratios D_S are subsequently evaluated for these three situations. The results obtained in these steps are furthermore compared to values which we compute with the so-called Elliott relation (see below).

An *ab initio* evaluation of the SF probability of equilibrium Ni requires calculated phonon dispersions and a relativistic electronic structure. Such calculation has previously been done for paramagnetic Al [15] but has not yet been accomplished for ferromagnets. An approximation was introduced years ago by Elliott [12], who pointed out a possible source of SF scattering arising from the spin mixing of eigenstates. Employing several assumptions, *viz.* a paramagnetic metal, nearly constant electron-phonon matrix elements, $b_{\mathbf{k}n}$ constant in the Brillouin zone, and $b^{\sigma}_{\mathbf{k}n} \ll a^{\sigma}_{\mathbf{k}n}$, Elliot derived a relation between the spin lifetime τ_S for a general kind of scattering event with lifetime τ. This so-called Elliott relation uses the Fermi-surface-averaged spin mixing of eigenstates $\langle b^2\rangle = \sum_{\sigma,n}\int d\mathbf{k}|b^{\sigma}_{\mathbf{k}n}|^2\delta(E^{\sigma}_{\mathbf{k}n} - E_F)$ and predicts the SF probability $P^{b^2}_S = (\tau_S/\tau)^{-1} = 4\langle b^2\rangle$.

In a similar way as introduced above, the influence of spin mixing on the SF probability in laser-heated Ni can be evaluated. We define a SF density of states (DOS) as

$$n_{\uparrow\downarrow}(E) = \sum_{n,\sigma}\int d\mathbf{k}|b^{\sigma}_{\mathbf{k}n}|^2\delta(E^{\sigma}_{\mathbf{k}n} - E). \quad (8)$$

A generalized Elliott SF probability for an electron with energy E is then given as $P^{b^2}_S(E) = 4n_{\uparrow\downarrow}(E)/n(E)$ [with $n(E)$ the total DOS] which yields the standard Elliott expression $\langle b^2\rangle$ in the limits $b^{\sigma}_{\mathbf{k}n} \ll a^{\sigma}_{\mathbf{k}n}$ and $E = E_F$. The total SF probability $P^{b^2}_S$ of a laser-heated system with electron distribution $f_{\sigma}(E)$ is obtained from Eqs. (7) and (5), where $w_{\uparrow\downarrow}(E)$ is replaced by $n_{\uparrow\downarrow}(E)$ and $w(E)$

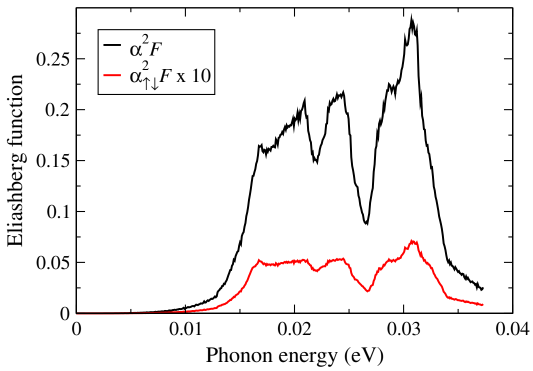

FIG. 1 (color online). *Ab initio* calculated Eliashberg function $\alpha^2F(E_F, \Omega)$ and SF Eliashberg function $\alpha^2_{\uparrow\downarrow}F(E_F, \Omega)$ of Ni in equilibrium.

by $n(E)$. Note that, although the treatment is intended for phonon scattering, the Elliott relation in fact does not take the character of scattering involved into account. Also, the assumption of a paramagnetic material is essential in Elliott's derivation, as this permits SF scattering *in each* **k** *point* in the spin-degenerate majority and minority bands at E_F. Experimentally, the Elliott relation was found to be valid up to a multiplication by a material-specific constant with variation smaller than 1 order of magnitude for various paramagnetic metals [20]. Recently, it has also been applied to ferromagnetic metals [8,18], even though for exchange-split ferromagnetic bands there exist far less **k** points at which spin-degenerate band crossings occur.

We have tested the implementation by computing first Al and Ni in equilibrium at low temperature (< 300 K). Our calculations are based on the density functional theory within the local spin-density approximation; see [21] for details. For Al, our calculated $\alpha^2_{\uparrow\downarrow}F$ is of the order of 10^5 smaller than α^2F and in good agreement with the previous result [15]. The *ab initio* calculated SF and non-SF Eliashberg functions of equilibrium Ni are shown in Fig. 1. For Ni, the computed SF $\alpha^2_{\uparrow\downarrow}F$ function is only about 50 times smaller than the ordinary α^2F function; this is due to the larger spin-orbit coupling. The resulting total SF probability, $P_S = 0.04$, is given in Table I. To estimate the accuracy of the Elliott approximation, we have calculated the Elliott SF probability and obtain $P_S^{b^2} = 0.07$. This value is in rough agreement with $P_S^{b^2} = 0.10$, computed in Ref. [18]. Thus, we find that the Elliott relation overestimates the SF probability in equilibrium Ni by about a factor of 2.

TABLE I. Calculated spin-flip probabilities P_S and demagnetization ratios D_S for laser-pumped Ni. Results are given for equilibrium (low T), for thermalized electrons at a high Fermi temperature T_e, and for the NEQ electron distribution created by fs laser excitation. Results obtained for the approximate Elliott SF probability $P_S^{b^2}$ (this work and [18]) are given for comparison.

	$P_S^{b^2}$	P_S	D_S
Ni (low T)	0.07 (0.10 [18])	0.04	0
Ni (T_e = 1500 K)	0.08	0.05	0.002
Ni (T_e = 3000 K)	0.11	0.07	0.003
Ni (T_e = 5000 K)	0.12	0.10	0.004
Ni (NEQ)	0.12	0.09	0.025

Next, we turn to the topic of current controversy, the actual amount of phonon-induced demagnetization in laser-excited Ni. In Fig. 2 (top), we show calculated energy-resolved SF and non-SF scattering rates [$w_{\uparrow\downarrow}(E)$ and $w(E)$]. Note the strong energy variations of $w(E)$. In Fig. 2 (bottom), we compare the computed electron-phonon SF probability $P_S(E)$ to that obtained from the Elliott relation. At some energies, e.g., 0.5–1 eV, these two quantities are nearly the same, but, at other energies, there is no direct relation other than that SF probability is large where band states are present. An interesting difference in the context of ultrafast demagnetization is the suppression of $P_S(E)$ around E_F, which is not captured by $P_S^{b^2}(E)$. The features of $P_S(E)$ that are not captured by $P_S^{b^2}(E)$ can be understood by comparing Eqs. (1) and (8). One of the differences is the presence or absence of summation over destination eigenstates $\mathbf{k}'n'$. The latter are restricted in Eq. (1) by the construction of $g^{\nu\uparrow\downarrow}_{\mathbf{k}n,\mathbf{k}'n'}$ to correspond to a *different* spin than the source state $\mathbf{k}n$. The number of available end states is, however, not taken into account in Elliott formula (which, derived for a paramagnetic metal, assumes that the same number of states is available for both spins and hence suppresses this distinction). The mentioned discrepancy between P_S and $P_S^{b^2}$ above E_F is thus easily explained by the lack of states with the same energy and opposite spin in the Ni DOS (see

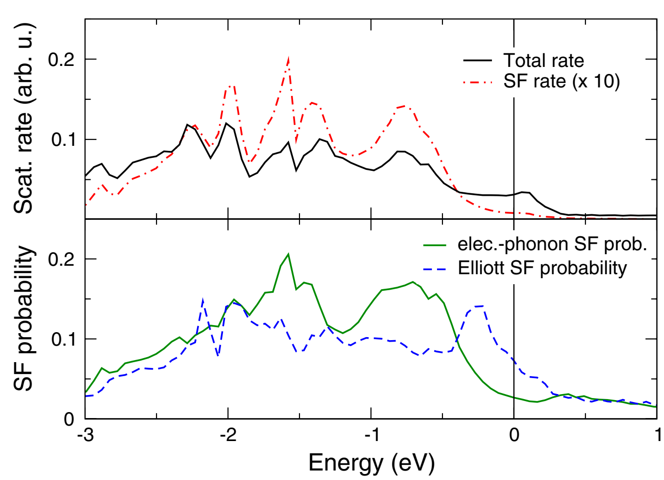

FIG. 2 (color online). Energy-resolved electron-phonon total and SF scattering rates $w(E)$ and $w_{\uparrow\downarrow}(E)$ of Ni (top), and normalized SF probability $P_S(E)$ and approximate SF probability $P_S^{b^2}(E)$ obtained from the Elliott relation (bottom).

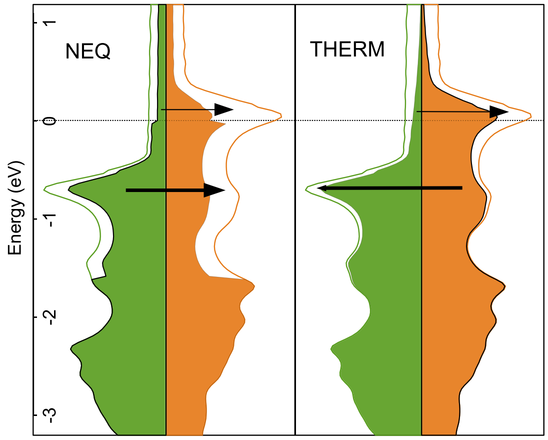

FIG. 3 (color online). Spin-resolved DOS (filled areas) and phonon-induced spin flips (arrows) of NEQ and electron thermalized Ni. The equilibrium DOS is shown by thin lines. SF transitions are significantly different at energies above and below $E_F (= 0\ \text{eV})$. The arrow's thickness corresponds to the transition rate; its direction and length give which direction is dominant and by how much. The amount of laser-redistributed electrons has been enlarged to improve visibility.

Fig. 3). Hence, the Elliott relation fails for ferromagnets in strongly exchange-split energy regions.

After laser excitation, electrons equilibrate quickly due to electron-electron scattering at a high electron temperature T_e of the order of thousands of K. To describe this situation, we use appropriate $f_\sigma(E)$, but note that the chemical potential must also be adjusted. Spin conservation leads to differences between $f_\uparrow(E)$ and $f_\downarrow(E)$, namely, $f_\downarrow(E)$ has a lower chemical potential than $f_\uparrow(E)$ in Ni due to the shape of its DOS. SF probabilities P_S computed for several T_e are given in Table I. With increasing T_e, P_S increases, too. The Elliott SF probability $P_S^{b^2}$ also increases with T_e but it deviates still from P_S. A previous work [8] used a Gaussian smearing to simulate a thermalized system (without E_F adjustment) and obtained $P_S^{b^2} \approx 0.18$. Our values are smaller, but note that the way the thermalized distribution is described is different.

As mentioned before, a large SF probability does not necessarily imply a large demagnetization. Evaluating the demagnetization rate $dM/dt = 2\mu_B(S^- - S^+)$ for thermalized electron distributions, we obtain quite small values, of the order of $0.08\mu_B/\text{ps}$. The reason is that not just a large SF probability, but also an imbalance between $f_\uparrow(E)$ and $f_\downarrow(E)$ is essential for a magnetization change. The distributions of spin populations specific to Ni imply that, for thermalized electrons below E_F, most spin flips *increase* the spin moment; spin-reducing transitions occur only above E_F. In that region, the SF scattering rate is, however, very low (Fig. 2). The situation is illustrated in Fig. 3. As a consequence, the spin-decreasing rate $(S^- - S^+)$ is thus much lower than the SF rate $(S^- + S^+)$ and, in addition, it exhibits only a weak temperature dependence. Hence, we find that phonon-mediated SF scattering in thermalized Ni cannot be the mechanism of the observed ultrafast demagnetization.

One remaining possibility for a fast demagnetization is an enhanced SF rate in the NEQ distribution present immediately after the laser pulse. Previous *ab initio* calculations showed that minority-spin electrons are excited more than majority-spin ones; see [19]. Assuming a 1.5-eV pump laser and a simplified steplike electron distribution reduced by about 5% in the 1.5-eV energy window below E_F, the calculated demagnetization ratio D_S is higher than for thermalized distributions (Table I). A critical role is played here by holes deep below E_F with high SF probability as well as a significant difference between majority and minority occupations (see Fig. 3). An important yet unknown element in estimating the demagnetization is the laser fluence. Nonetheless, we find that phonon-mediated demagnetization in Ni is much more effective in the NEQ state than in the thermalized state, as was proposed recently for Gd [22]. An important aspect is the time scale on which the NEQ demagnetization is active. Electron thermalization proceeds fast in Ni and transforms the initial NEQ distribution into a thermalized one in ~200 fs. A rough estimate of the demagnetization in this time window is $0.1\mu_B$, i.e., smaller than the observed experimental demagnetization. The precise amount of the demagnetization depends, however, on the time evolution of the distributions, which requires further investigations.

Using relativistic *ab initio* calculations, we have evaluated the phonon-induced SF probability and demagnetization in laser-pumped Ni. A strong dependence of these quantities on the electron energy is observed, which is not tracked by the Elliott approximation. In the electron thermalized state, Elliott-Yafet phonon-mediated demagnetization is too small to explain the ultrafast demagnetization, despite reasonably large SF probabilities. We find that Elliott-Yafet SF scattering contributes more to the demagnetization for NEQ distributions immediately after the fs laser excitation. We note lastly that the existence of *other* fast SF channels [5–7,11] cannot be excluded.

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