PHYSICAL REVIEW B **92**, 195109 (2015)

Transition matrix elements for electron-phonon scattering: Phenomenological theory and *ab initio* electron theory

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(Received 9 February 2015; revised manuscript received 8 May 2015; published 5 November 2015)

Transition matrix elements for electron-phonon scattering are discussed. The approximate expression for the matrix element derived by the combination of a phenomenological theory of electronic and ionic screening of the electron-electron interaction with a microscopic perturbation theory for the matrix element is compared with unscreened matrix elements from the *ab initio* electron theory. The matrix elements calculated in these two ways differ very strongly. A situation is investigated (ultrafast demagnetization of a ferromagnetic film after excitation with a femtosecond laser pulse) in which nevertheless both types of matrix elements yield similar results for the demagnetization rate.

DOI: 10.1103/PhysRevB.92.195109 PACS number(s): 63.20.kd, 75.78.Jp

I. INTRODUCTION

Many phenomena in solid state physics are determined by the scattering of electrons at phonons. Examples are the electrical and thermal conductivity, the lifetime of excited electron states [1], and a possible contribution to the ultrafast demagnetization after irradiation of a ferromagnetic film by a femtosecond laser pulse (see below). For a monatomic crystal the transition rate $W^{\lambda}_{j\mathbf{k},j'\mathbf{k}'}$ from an electronic state $|j\mathbf{k}\rangle$ with energy $\epsilon_{j\mathbf{k}}$ to a state $|j'\mathbf{k}'\rangle$ with energy $\epsilon_{j'\mathbf{k}'}$ (j, j' denote the band indices, $\mathbf{k}$, $\mathbf{k}'$ the wave vectors, and λ is the index of the three polarization vectors $\mathbf{p}_{\mathbf{q}\lambda}$ of a phonon) is given by Fermi's golden rule,

$$W^{\lambda}_{j\mathbf{k},j'\mathbf{k}'} = \frac{2\pi}{\hbar}\left|M^{\lambda}_{j\mathbf{k},j'\mathbf{k}'}\right|^2\{n_{\mathbf{q}\lambda}\delta[\epsilon_{j'\mathbf{k}'} - (\epsilon_{j\mathbf{k}} + \hbar\omega_{\mathbf{q}\lambda})] + (n_{-\mathbf{q}\lambda}+1)\delta[\epsilon_{j'\mathbf{k}'} - (\epsilon_{j\mathbf{k}} - \hbar\omega_{-\mathbf{q}\lambda})]\}. \quad (1)$$

The phonon with wave vector $\mathbf{q}$ has the frequency $\omega_{\mathbf{q}\lambda}$, and $n_{\mathbf{q}\lambda} = [\exp(\hbar\omega_{\mathbf{q}\lambda}/k_B T_p) - 1]^{-1}$ is the Bose distribution function with the phonon temperature T_p. Absorption of phonons and both induced and spontaneous emission of phonons are included in Eq. (1). $M^{\lambda}_{j\mathbf{k},j'\mathbf{k}'}$ is the transition matrix element

$$M^{\lambda}_{j\mathbf{k},j'\mathbf{k}'} = \langle\Psi_{j'\mathbf{k}'}|V_{\text{ph}}|\Psi_{j\mathbf{k}}\rangle, \quad (2)$$

where V_{ph} is the electron-phonon scattering operator and $\mathbf{q} = \pm(\mathbf{k}' - \mathbf{k} + \mathbf{G})$, where the $+(-)$ sign holds for phonon absorption (emission) and where $\mathbf{G}$ is a reciprocal lattice vector which brings $\mathbf{k}'$ back to the first Brillouin zone if $\mathbf{k}' = \mathbf{k} + \mathbf{q}$ is outside. Equation (2) includes spin-flip transitions although the spin does not explicitly occur in the wave functions $\Psi_{j\mathbf{k}}$. The reason is that the dominant spin character of the state is determined by j and $\mathbf{k}$, and it may well be different for $\Psi_{j\mathbf{k}}$ and $\Psi_{j'\mathbf{k}'}$. The operator V_{ph} has two contributions [2], one resulting from the phonon-induced distortion of the lattice potential (which is often called Elliott part, standard part, or Fröhlich part) and one resulting from the phonon-induced distortion of the spin-orbit coupling (Yafet part). In the Elliott part only the phonon-induced modifications of the spin-diagonal part of the effective potential which the electrons feel enters, which is called deformation potential. Elliott has shown [3] that this spin-diagonal part nevertheless leads to spin-flip scatterings in systems with spin-orbit coupling. Yafet has shown [2] that one must take into account both contributions to get the correct $q = |\mathbf{q}| \to 0$ limit for the behavior of the matrix element, and this has been confirmed by Grimaldi and Fulde [4].

It is well known that the modification of the lattice potential and of the spin-orbit coupling due to the phonon is changed by the reaction of the electrons (mainly the valence electrons) and the ions on the displacement of the ions by the phonon, i.e., there is electronic and ionic screening of the electron-phonon transition matrix element, especially for small $|\mathbf{q}|$. The screening of the Elliott part has been discussed by Kittel [5] within a complicated perturbation theory and in the book of Giuliani and Vignale [6] with Green function based methods. A simplified version of a phenomenological screening theory is given in the textbooks of Ashcroft and Mermin [7] and of Czycholl [8]. In this simplified version it is assumed (see Sec. II) that the matrix element (which is in general $M^{\lambda}_{j\mathbf{k},j'\mathbf{k}'}$, see above) depends only on $\mathbf{q}$ and on the phonon frequency $\omega_{\mathbf{q}}$, yielding the approximation

$$|M(\mathbf{q})|^2 = \frac{4\pi e^2}{V\left(q^2 + k^2_{\text{TF}}\right)}\frac{1}{2}\hbar\omega_{\mathbf{q}}. \quad (3)$$

Here V is the volume of the system, and k_{TF} is the Thomas-Fermi wave vector of the system.

The screening of the Yafet part has been discussed for paramagnetic systems by Yafet [2] using an *a priori* screened lattice potential, and by Grimaldi and Fulde [4] within a self-consistent Hartree approximation. For magnetic systems Rajagopal and Mochena [9] have discussed screening effects beyond the theory of Grimaldi and Fulde for paramagnetic systems. However, no explicit results for the screened electron-phonon matrix element have been given which could be used in electron-theoretical treatments of electron-phonon scattering. To get a simple phenomenological approximation for the screened matrix elements $M^{\lambda}_{j\mathbf{k},j'\mathbf{k}'}$ which can be used

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in such treatments, one must try to find explicit results of the theory of Ref. [9] and to approximate them as strongly as possible. In the literature on electron-theoretical treatments of electron-phonon scattering, the matrix elements $M^{\lambda}_{j\mathbf{k},j'\mathbf{k}}$ have been calculated on the one hand by the *ab initio* density functional electron theory and on the other hand by using phenomenological approximations based on Eq. (3).

II. *AB INITIO* TREATMENT AND PHENOMENOLOGICAL APPROACH

A. *Ab initio* treatment

These treatments included the Elliott and the Yafet part (see, e.g., Refs. [10–12]). Part of these treatments, e.g., Ref. [12], used the rigid-ion approximation which neglects the screening and which is a good approximation for transition metals [11,13]. Thereby the atoms which build the effective potential (which an electron at position $\mathbf{r}$ and with spin orientation $\alpha = \uparrow,\downarrow$ feels)

$$V(\mathbf{r},\{\mathbf{R}_n\}) = \sum_{n=1}^{N} \begin{pmatrix} v^{\uparrow}_{0,n}(\mathbf{r}-\mathbf{R}_n) & 0 \\ 0 & v^{\downarrow}_{0,n}(\mathbf{r}-\mathbf{R}_n) \end{pmatrix} \tag{4}$$

are rigidly displaced upon a deformation, without a reaction of electrons and ions on a displacement. Here N is the number of atoms in the system and $\mathbf{R}_n = \mathbf{R}_{0,n} + \delta\mathbf{R}_n$ is the position of the nth lattice atom, where $\mathbf{R}_{0,n}$ is the equilibrium position and $\delta\mathbf{R}_n$ is the displacement (in the present paper caused by the phonon). In this approximation one gets

$$V_{\mathrm{ph}} = -\sum_{n=1}^{N} \sqrt{\frac{\hbar}{2NM\omega_{\mathbf{q},\lambda}}}\exp(i\mathbf{q}\cdot\mathbf{R}_{0,n})(\mathbf{p}_{\mathbf{q}\lambda}\cdot\nabla_{\mathbf{r}}) \times \begin{pmatrix} v^{\uparrow}_{0,n}(\mathbf{r}-\mathbf{R}_{0,n}) & 0 \\ 0 & v^{\downarrow}_{0,n}(\mathbf{r}-\mathbf{R}_{0,n}) \end{pmatrix}, \tag{5}$$

where M is the atomic mass. In contrast to this the papers of Carva *et al.* [11,14] determine the effective potential of the system with phonons not just in rigid-ion approximation but also by a supercell calculation for phonons with wavelengths compatible with the supercell geometry, i.e., they take into account the screening in a self-consistent manner. The spin-flip Eliashberg functions calculated by the rigid-ion approximation thereby agreed well with those obtained by the supercell calculation, demonstrating that the rigid-ion approximation works rather well.

In Sec. III we compare the *ab initio* calculated matrix elements for the Elliott part with those from the phenomenological approximations using Eq. (3). The *ab initio* matrix elements are determined in rigid-ion approximation, Eq. (5), i.e., they neglect screening effects. Because these effects are important especially for small $|\mathbf{q}|$, we test whether there is an agreement of the two approaches for not too small $|\mathbf{q}|$. As mentioned above, there are so far no explicit simple phenomenological approximations for the screened Yafet part, therefore we cannot compare phenomenology and *ab initio* approach for the Yafet part.

Grimaldi and Fulde [4] have shown that there are long-range and short-range contributions to the matrix element. We will see in Sec. III that the electronic screening is described in the phenomenological approach by Thomas-Fermi screening which is valid only for small $|\mathbf{q}|$. In the *ab initio* matrix elements based on Eq. (5) there are long-range and short-range contributions. The short-range contributions arise from the fact that $(\mathbf{p}_{\mathbf{q}\lambda}\cdot\nabla_{\mathbf{r}})v^{\alpha}_{0,n}$ is not constant within an atomic shell around atom n, so that a Fourier transform of this would require large wave vectors, even for small phonon wave vectors $|\mathbf{q}|$. Unfortunately, it is not possible to disentangle these contributions in the *ab initio* matrix elements (2), because the calculations are done in a real-space representation of $\Psi_{j\mathbf{k}}$ and V_{ph}. The neglect of short-range contributions in the phenomenological matrix elements based on Eq. (3) will be one reason for the deviations of the phenomenologically calculated matrix elements from the *ab initio* calculated ones.

In the present paper we calculate the *ab initio* matrix elements by a relativistic version [15] of the linear-muffin-tin-orbital theory [16] (LMTO) in atomic-sphere approximation (ASA) and the spin-density functional electron theory [17,18]. We are convinced that the test of the phenomenological approximation (3) for the Elliott matrix element is of great interest for the community of people who deal with problems related to electron-phonon scattering but who do not want to perform *ab initio* calculations.

B. Phenomenological approximations

Examples for the use of phenomenological approximations for the Elliott part of the screened matrix elements are (among others), e.g., Refs. [19,20]. There are various steps in the derivation of Eq. (3). In Refs. [7] and [8] in part different arguments have been used, but the final result is the same. We just will summarize the various steps in the derivation of Eq. (3). The phenomenological theory starts with the matrix element $V_{\mathbf{k},\mathbf{k}'}$ for the Coulomb interaction between two free electrons which are approximated for valence electrons in a crystal with wave vectors $\mathbf{k}$ and $\mathbf{k}'$. Considering this Coulomb potential as being an "external potential," there is a screening (described by the use of dielectric constants) of $V_{\mathbf{k},\mathbf{k}'}$ due to the reaction of the other electrons (which is approximated by the Thomas-Fermi screening for a homogeneous and isotropic system of free electrons) and by the ions leading to an effective screened matrix element $V^{\mathrm{eff}}_{\mathbf{k},\mathbf{k}'} = V^{\mathrm{el}}_{\mathbf{k},\mathbf{k}'}(\mathbf{q},k_{\mathrm{TF}}) + V^{\mathrm{ion}}_{\mathbf{k},\mathbf{k}'}(\mathbf{q},k_{\mathrm{TF}},\omega_{\mathbf{q}},\omega)$ with $\mathbf{q} = |\mathbf{k}-\mathbf{k}'|$, the phonon frequency $\omega_{\mathbf{q}}$, and $\hbar\omega = \epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}$ with the energies $\epsilon_{\mathbf{k}}$, $\epsilon_{\mathbf{k}'}$ of the free electrons. In a second step, the used approximation for $V^{\mathrm{ion}}_{\mathbf{k},\mathbf{k}'}$ is represented as a matrix element for the transition between an initial state $|n\rangle$ to a final state $|m\rangle$ of a system of free electrons and ions. Thereby, the electrons interact mutually via Coulomb interaction, and they interact with the ions. Thereby, the transition is done by exciting two electrons with wave vector $\mathbf{k}$ and $\mathbf{k}'$ and energies below the Fermi energy ϵ_{F} to electrons with $\mathbf{k}+\mathbf{q}$ and $\mathbf{k}'-\mathbf{q}$ and energies above ϵ_{F}, and the ions remain in the same state. The idea is that $V^{\mathrm{ion}}_{\mathbf{k},\mathbf{k}'}$ is for a real system better approximated by a matrix element including the single-electron crystal states $|\mathbf{k}\rangle$, $|\mathbf{k}'\rangle$, $|\mathbf{k}+\mathbf{q}\rangle$, $|\mathbf{k}-\mathbf{q}\rangle$ instead of free electron states. The eigenstates $|n\rangle$ and $m\rangle$ are calculated in second-order perturbation theory where the perturbation is given by the electron-phonon interactions and by the Coulomb interaction between the electrons. Thereby a simple electron-phonon scattering operator is used which describes only intraband transitions and which contains transition matrix elements $M(\mathbf{q})$ which do not depend on $\mathbf{k}$ and on the phonon polarization. Equating the so obtained $V^{\mathrm{ion,perturb}}_{\mathbf{k},\mathbf{k}'}$ to the phenomenological expression $V^{\mathrm{ion}}_{\mathbf{k},\mathbf{k}'}$ from above yields Eq. (3).

III. COMPARISON OF THE APPROXIMATED MATRIX ELEMENTS WITH *AB INITIO* MATRIX ELEMENTS

We now compare the approximated electron-phonon transition matrix elements of Eq. (3) with electron-phonon matrix elements calculated from Eqs. (2) and (5) with crystal wave functions $\Psi_{j\mathbf{k}},\Psi_{j'\mathbf{k}'}$ determined by the *ab initio* density functional electron theory in local-spin-density approximation and the LMTO-ASA method. In Eq. (3) the volume of the crystal may be written as $V = N\Omega$, where N is the number of elementary cells in a crystal and Ω is the volume of the elementary cell. The *ab initio* matrix elements are already normalized to N, i.e., they contain $\frac{1}{N}$ as prefactor [see Eq. (20) of Ref. [12]], therefore they have to be divided by Ω for a comparison with the approximated matrix element, Eq. (3). Equations (2) and (5) show that the *ab initio* matrix elements do not depend just on the phonon wave vector $\mathbf{q}$ but both on $\mathbf{k}$ and $\mathbf{k}'$ from which $\mathbf{q} = \pm(\mathbf{k}' - \mathbf{k} + \mathbf{G})$ can be calculated. Furthermore, they depend on band indices j and j' whereas for the derivation of Eq. (3) it has been assumed (see Sec. II B) that there are only intraband transitions ($j = j'$). The matrix elements also depend on the index λ which denotes the three polarization vectors $\mathbf{p}_{\mathbf{q}\lambda}$ of the phonon, whereas this dependence has been neglected in Eq. (3). This is of course a very strong approximation, because according to Eq. (5) V_{ph} contains the scalar product between $\mathbf{p}_{\mathbf{q}\lambda}$ and the gradient of the potential $v^{\alpha}_{0,n}$, and this leads for the case of free electrons to a scalar product $\mathbf{q}\cdot\mathbf{p}_{\mathbf{q}\lambda}$ [see Eq. (6.14) of Ref. [8]]. For high-symmetry wave vectors $\mathbf{q}$ the polarization vectors are longitudinal ($\mathbf{p}_{\mathbf{q}\lambda} \parallel \mathbf{q}$) and transversal ($\mathbf{p}_{\mathbf{q}\lambda} \perp \mathbf{q}$)—for arbitrary $\mathbf{q}$ the polarization vectors are pseudolongitudinal or pseudotransversal. This clearly demonstrates that the transition matrix element depends strongly on $\mathbf{q}$. Finally, in systems with spin-orbit coupling the electronic states $\Psi_{j\mathbf{k}}$ are no pure spin states but spin-mixed states according to [12]

$$\Psi_{j\mathbf{k}} = [a_{j\mathbf{k}}(\mathbf{r})|\uparrow\rangle + b_{j\mathbf{k}}(\mathbf{r})|\downarrow\rangle]\exp(i\mathbf{k}\mathbf{r}), \tag{6}$$

where $a_{j\mathbf{k}}(\mathbf{r})$ and $b_{j\mathbf{k}}(\mathbf{r})$ are lattice periodic functions and $|\uparrow\rangle$, $|\downarrow\rangle$ are the two spinor eigenfunctions of $\widehat{S}_z$. The wave function is denoted as "dominant spin up" or "dominant spin down" if $|a_{j\mathbf{k}}|^2 = \int |a_{j\mathbf{k}}(\mathbf{r})|^2 d^3r$ is larger or smaller than $|b_{j\mathbf{k}}|^2 = \int |b_{j\mathbf{k}}(\mathbf{r})|^2 d^3r$. Usually one denotes the dominant spin character by $\widetilde{m}_s$, and this index (which is determined by $j\mathbf{k}$) is added to the wave function $\Psi^{\widetilde{m}_s}_{j\mathbf{k}}$. In *ab initio* theories of electron-phonon scatterings, one distinguishes between spin flip transitions if in $\Psi^{\widetilde{m}_s}_{j\mathbf{k}}$ and $\Psi^{\widetilde{m}'_s}_{j'\mathbf{k}'}$ we have $\widetilde{m}_s \neq \widetilde{m}'_s$ and non-spin-flip transitions for $\widetilde{m}_s = \widetilde{m}'_s$. In Eq. (3) the dominant spin character is not considered, i.e., it gives the same result independent of the question of whether the considered electron-phonon interaction is a spin-flip transition or a non-spin-flip transition. In many materials, however, there is a big difference between these two types of transitions, because spin mixing is small for most $\Psi_{j\mathbf{k}}$. To estimate the difference between the two types of transitions we assume that we can write approximately

$$\begin{aligned}\Psi^{\widetilde{m}_s}_{j\mathbf{k}}(\mathbf{r}) &= \varphi_{j\mathbf{k}}(\mathbf{r})[a^{\widetilde{m}_s}|\uparrow\rangle + b^{\widetilde{m}_s}|\downarrow\rangle]\exp(i\mathbf{k}\mathbf{r})\\ &= \varphi_{j\mathbf{k}}(\mathbf{r})|\chi^{\widetilde{m}_s}\rangle\exp(i\mathbf{k}\cdot\mathbf{r}),\end{aligned} \tag{7}$$

with

$$\begin{aligned}|\chi^{\text{dominant up}}\rangle &= \begin{pmatrix}\sqrt{1-b^2}\\ b\end{pmatrix},\\ |\chi^{\text{dominant down}}\rangle &= \begin{pmatrix}b\\ \sqrt{1-b^2}\end{pmatrix}.\end{aligned} \tag{8}$$

Thereby b^2 is the averaged spin-mixing factor calculated by the average $\langle|b_{j\mathbf{k}}|^2\rangle$ over all states involved in the electron-phonon scattering processes [21]. Therefore the square of the transition matrix element is for $\widetilde{m}_s \neq \widetilde{m}'_s$

$$\begin{aligned}\left|\left\langle\Psi^{\widetilde{m}'_s}_{j'\mathbf{k}'}\middle|V_{\mathrm{ph}}\middle|\Psi^{\widetilde{m}_s}_{j\mathbf{k}}\right\rangle\right|^2 &= \left|2b\sqrt{1-b^2}\langle\varphi_{j'\mathbf{k}'}|V_{\mathrm{ph}}|\varphi_{j\mathbf{k}}\rangle\right|^2\\ &\approx 4b^2|\langle\varphi_{j'\mathbf{k}'}|V_{\mathrm{ph}}|\varphi_{j\mathbf{k}}\rangle|^2,\end{aligned} \tag{9}$$

whereas for $\widetilde{m}_s = \widetilde{m}'_s$ we have

$$\left|\left\langle\Psi^{\widetilde{m}'_s}_{j'\mathbf{k}'}\middle|V_{\mathrm{ph}}\middle|\Psi^{\widetilde{m}_s}_{j\mathbf{k}}\right\rangle\right|^2 = |\langle\varphi_{j'\mathbf{k}'}|V_{\mathrm{ph}}|\varphi_{j\mathbf{k}}\rangle|^2, \tag{10}$$

i.e., the squares of spin-flip transition matrix elements are typically a factor of $4b^2$ smaller than the squares of non-spin-flip transition matrix elements. For Ni, which we consider in the following, the *ab initio* calculated value is [21] $b^2 = 0.025$.

In the following we do not consider transition matrix elements from all conceivable $\Psi_{j\mathbf{k}}$ to all conceivable $\Psi_{j'\mathbf{k}'}$, but we make a preselection of $\Psi_{j\mathbf{k}}$ and $\Psi_{j'\mathbf{k}'}$ in such a way that only transitions are considered which contribute significantly [12] to the ultrafast demagnetization of a ferromagnetic film after excitation with a fs laser pulse (see Introduction). These are states $\Psi_{j\mathbf{k}}$ (or $\Psi_{j'\mathbf{k}'}$, respectively) with energies $\epsilon_{j\mathbf{k}}$ close to the Fermi energy ϵ_{F} and for which $|\epsilon_{j\mathbf{k}} - \epsilon_{j'\mathbf{k}'}|$ is not too large. We consider matrix elements with $|\epsilon_{j\mathbf{k}} - \epsilon_{\mathrm{F}}| < 10$ mRy and $|\epsilon_{j\mathbf{k}} - \epsilon_{j'\mathbf{k}'}| < 3.5$ mRy, whereby $\mathbf{k}$ and $\mathbf{k}'$ are points from a $20 \times 20 \times 20$ Monkhorst-Pack $\mathbf{k}$-point grid [22].

As discussed above it is difficult to compare the *ab initio* matrix elements with Eq. (3), because one must be careful not to compare the incomparable. First, we compare the *ab initio* results for a situation which resembles most closely the situation described by Eq. (3). Thereby we use for k_{TF} two different values for Ni. The Thomas-Fermi wave vector is generally defined by [7]

$$k^2_{\mathrm{TF}} = 4\pi e^2\frac{\partial n_e}{\partial\mu}, \tag{11a}$$

where $n_e(\mu)$ is the density of valence electrons

$$n_e(\mu) = \sum_j\int\frac{d^3k}{8\pi^3}\frac{1}{\exp[(\epsilon_{j\mathbf{k}} - \mu)/k_BT_{\mathrm{el}}] + 1}, \tag{11b}$$

with the chemical potential μ which depends on the temperature T_{el} of the electronic system. For $T_{\mathrm{el}} = 0$, Eq. (11a) yields $k^2_{\mathrm{TF}} = 4\pi e^2Z(E_{\mathrm{F}})$, where $Z(E_{\mathrm{F}})$ is the density of states at the Fermi energy. For a free electron gas this gives for $T_{\mathrm{el}} = 0$

$$k^2_{\mathrm{TF}} = \frac{6\pi e^2 n_{\mathrm{e}}}{\epsilon_{\mathrm{F}}}, \tag{11c}$$

where ϵ_F is the Fermi energy of free electrons with density n_e. This gives for Ni a value of $k_{TF} = 1.15\frac{2\pi}{a}$, where $a = 3.521$ Å denotes the lattice constant of the fcc lattice in Ni. For real Ni with an electronic density of states $Z(\epsilon)$, Eq. (11a) gives at arbitrary temperature T_{el}

$$k_{TF}^2 = 4\pi e^2 \int d\epsilon\, Z(\epsilon) \frac{\partial f(\epsilon, T_{el}, \mu)}{\partial \mu}\bigg|_{\mu=\epsilon_F(T_{el})} \tag{11d}$$

with the chemical potential $\epsilon_F(T_{el})$ which has to be calculated from Eq. (11b). This expression is of course more accurate than the free electron expression (11c), because it takes into account band-structure effects of the real material. For an electron temperature of $T_{el} = 300$ K this yields $k_{TF} = 6.67\frac{2\pi}{a}$ (see Table I of Ref. [23], where, however, k_{TF} is given in other units). The two values differ a lot, and we will see that only for the second value we get some agreement of the phenomenological theory with the *ab initio* theory.

We then consider only non-spin-flip intraband transitions (in the *ab initio* calculations from dominant up to dominant up), which are caused by nearly longitudinal phonons (with angles between $\mathbf{p}_{\mathbf{q}\lambda}$ and $\mathbf{q}$ smaller than $\frac{\pi}{4}$). Furthermore we consider only values of $|\mathbf{q}|$ which are not too small ($|\mathbf{q}| > 0.8 \cdot \frac{2\pi}{a}$), so that the screening is not very strong, because the *ab initio* matrix elements are unscreened matrix elements whereas Eq. (3) describes screened matrix elements. Figure 1 is a histogram showing the absolute occurrence of squared matrix elements with magnitude $|M|^2$ for the above discussed k-point grid. Thereby we consider the above preselected wave vectors $\mathbf{k}$ and $\mathbf{k}'$ which give different values of $\mathbf{q}$ and therefore also different values of the approximated $|M(\mathbf{q})|^2$.

Figure 2 shows the histogram for a situation in which in the *ab initio* calculations' non-spin-flip matrix elements from dominant down to dominant down are considered, which are more frequent than dominant up to dominant up transitions in Ni. It becomes obvious that the values for the *ab initio* matrix elements are considerably smaller than the results according to Eq. (3).

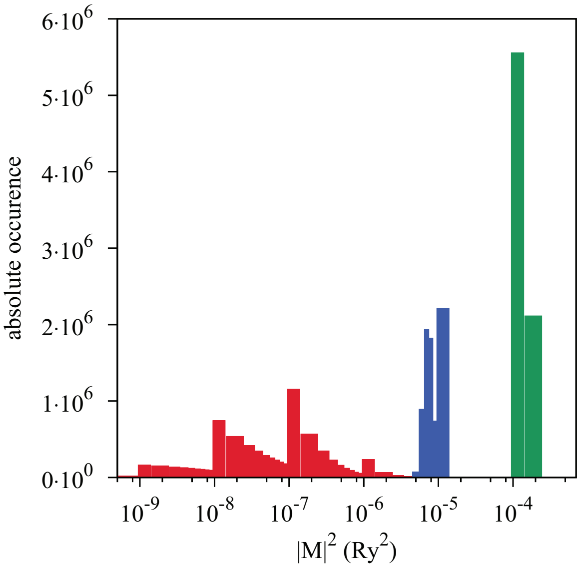

FIG. 2. (Color) Same as Fig. 1, but now with non-spin-flip transitions from dominant down to dominant down in the *ab initio* calculations.

Second, we give a histogram (Fig. 3) for a situation which differs from the first situation just because now only spin-flip transitions are considered. For each $\Psi_{j\mathbf{k}}$, $\Psi_{j'\mathbf{k}'}$ which gives a preselected *ab initio* matrix element corresponding to a spin-flip transition, we calculate $|M(\mathbf{q})|^2$ according to Eq. (3) and

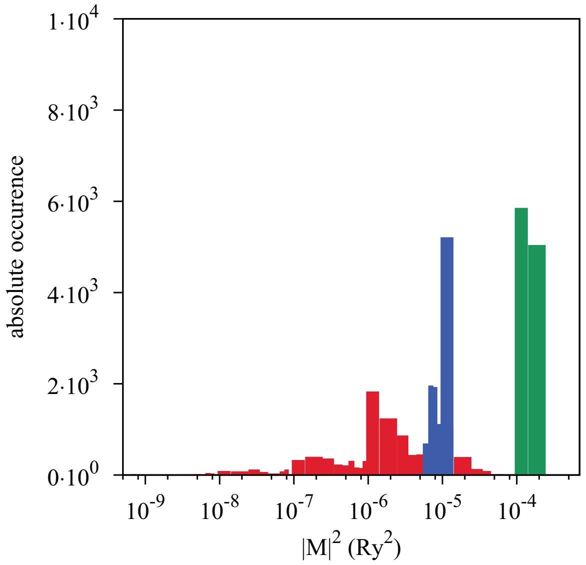

FIG. 1. (Color) Comparison of the magnitudes of the squares of the matrix elements between the *ab initio* calculated results (red) and results according to Eq. (3) with $k_{TF} = 1.15 \cdot \frac{2\pi}{a}$ (green) and with $k_{TF} = 6.67 \cdot \frac{2\pi}{a}$ (blue). Thereby the *ab initio* calculations are for intraband, quasilongitudinal, large q, non-spin-flip transitions (dominant up to dominant up).

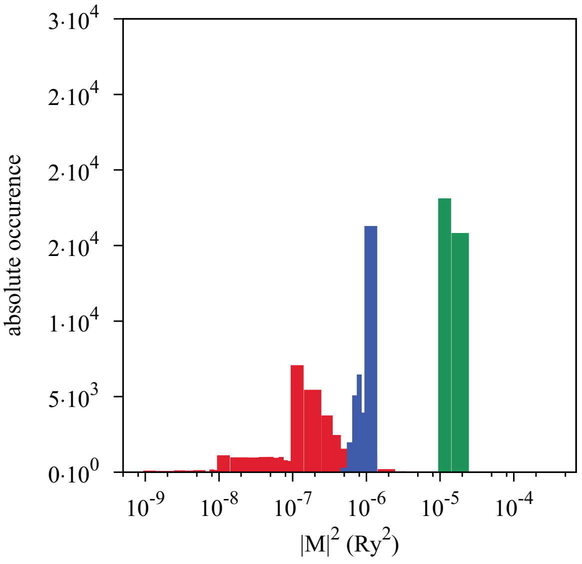

FIG. 3. (Color) Comparison of the magnitudes of the squares of the matrix elements between the *ab initio* calculated results (red) and the results according to Eq. (3) with $k_{TF} = 1.15\frac{2\pi}{a}$ (green) and $k_{TF} = 6.67\frac{2\pi}{a}$ (blue). The *ab initio* calculations are for intraband, quasilongitudinal, large q, spin-flip transitions, and $|M(\mathbf{q})|^2$ of Eq. (3) is multiplied by $4b^2 \approx 0.1$ (see text).

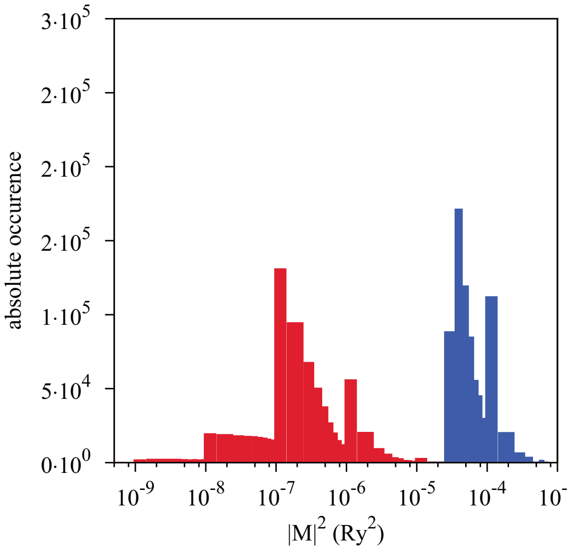

FIG. 4. (Color) Comparison of the magnitudes of the squares of the spin-flip matrix elements between *ab initio* calculated results (red) and results (blue) according to $|M(\mathbf{q})|^2$ from Eq. (3) multiplied by $4b^2$, whereby in Eq. (3) $k_{\rm TF}$ is set to zero.

multiply it with $4b^2 \approx 0.1$, see Eq. (9). Again the values for the *ab initio* matrix elements are considerably smaller than those according to the unscreened case of Eq. (3).

Third, we consider all spin-flip matrix elements (Fig. 4), whereby we switch off the electronic screening in Eq. (3) by setting $k_{\rm TF} = 0$. Again the results of the two calculations differ strongly; the values for the *ab initio* matrix elements are considerably smaller than those according to the modified Eq. (3).

Fourth, we try to take into account approximate electronic screening also for the *ab initio* matrix elements. To do this, we multiply in Eq. (5) $v^\alpha_{0,n}(\mathbf{r}-\mathbf{R}_{0,n})$ by $\exp(-k_{\rm TF}\cdot r)$. For the Coulomb potential $V \propto \frac{1}{r}$, this would account for the electronic screening for free electrons in real space. For the more complicated position dependence of $v^\alpha_{0,n}(\mathbf{r}-\mathbf{R}_{0,n})$, this is certainly an approximation. The corresponding histogram is shown in Fig. 5. Again, the results of the two calculations differ strongly. Now the *ab initio* matrix elements are larger than those calculated from the modified Eq. (3), which seems to be surprising. The reason is that in Eq. (5) we now have

$$\nabla_{\mathbf{r}}\big(\exp(-k_{\rm TF}\cdot r)v^\alpha_{0,n}(\mathbf{r}-\mathbf{R}_{0,n})\big) = \exp(-k_{\rm TF}\cdot r)\left[\nabla_{\mathbf{r}}v^\alpha_{0,n}(\mathbf{r}-\mathbf{R}_{0,n}) - \frac{\mathbf{r}}{|\mathbf{r}|}k_{\rm TF}\,v^\alpha_{0,n}(\mathbf{r}-\mathbf{R}_{0,n})\right]. \tag{12}$$

A nonzero $k_{\rm TF}$ has two effects. First, it reduces the matrix element via the exponential in Eq. (12); second, it enhances the matrix element via the second term in the square bracket which for Ni has the same sign as the first term. Taking into account that in the matrix element there are also position-dependent wave functions $\Psi_{j'\mathbf{k}'}(\mathbf{r})$ and $\Psi_{j\mathbf{k}}(\mathbf{r})$, it is conceivable that, depending on $k_{\rm TF}$, either the exponential prefactor or the second term in the square bracket dominates.

Altogether, we see that the single matrix element calculated either by *ab initio* or by the use of Eq. (3) differs very much.

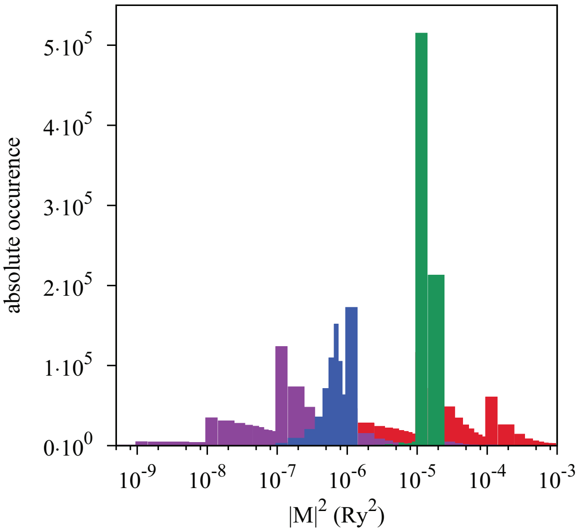

FIG. 5. (Color) Comparison of the magnitudes of the squares of the spin-flip matrix elements between *ab initio* calculated results and results according to $|M(\mathbf{q})|^2$ from Eq. (3). In the *ab initio* calculations electronically screened atomic potentials are used (see text) with $k_{\rm TF} = 1.15\frac{2\pi}{a}$ (red) and $k_{\rm TF} = 6.67\frac{2\pi}{a}$ (violet). The results according to $|M(\mathbf{q})|^2$ from Eq. (3), multiplied by $4b^2$, are also for $k_{\rm TF} = 1.15\frac{2\pi}{a}$ (green) and $k_{\rm TF} = 6.67\frac{2\pi}{a}$ (blue).

In the expression for macroscopic observables the matrix elements enter in a weighted way. For instance, the contribution of electron-phonon scatterings (which transfer angular momentum from the electrons to the lattice) to the demagnetization rate $\frac{dM}{dt}$ (which is the rate of the magnetic moment change per atom) of a ferromagnetic film after irradiation with a fs laser pulse is given within Elliott-Yafet theory [2] by [12]

$$\begin{aligned}\frac{dM}{dt} =& \frac{1}{\Omega^2_{\rm BZ}}\sum_{j,j',\lambda}\int_{\rm BZ} d^3k \int_{\rm BZ} d^3k'\, m_{j\mathbf{k}\uparrow,j'\mathbf{k}'\downarrow}\\ &\times \big\{ f_t(\epsilon^{\uparrow}_{j\mathbf{k}})[1-f_t(\epsilon^{\downarrow}_{j'\mathbf{k}'})]W^{\lambda}_{j\mathbf{k}\uparrow,j'\mathbf{k}'\downarrow}\\ &- f_t(\epsilon^{\downarrow}_{j'\mathbf{k}'})[1-f_t(\epsilon^{\uparrow}_{j\mathbf{k}})]W^{\lambda}_{j'\mathbf{k}'\downarrow,j\mathbf{k}\uparrow}\big\}.\end{aligned} \tag{13}$$

$m_{j\mathbf{k}\uparrow,j'\mathbf{k}'\downarrow}$ keeps track of the spin magnetic moment change for every spin-flip transition. For pure spin states, $m_{j\mathbf{k}\uparrow,j'\mathbf{k}'\downarrow}$ equals $2\,\mu_B$, but for spin-mixed states it is a bit less than $2\,\mu_B$. $\Omega_{\rm BZ}$ is the volume of the Brillouin zone, and $f_t(\epsilon^{\widetilde{m}_s}_{j\mathbf{k}})$ is the time-dependent Fermi distribution function

$$f_t(\epsilon^{\widetilde{m}_s}_{j\mathbf{k}}) = \left[\exp\left(\frac{\epsilon^{\widetilde{m}_s}_{j\mathbf{k}} - \epsilon^{\widetilde{m}_s}_{\rm F}(t)}{k_B T_{\rm el}(t)}\right) + 1\right]^{-1} \tag{14}$$

with the time-dependent chemical potential $\epsilon^{\widetilde{m}_s}_{\rm F}(t)$ and the time-dependent electron temperature $T_{\rm el}(t)$. Thereby it is assumed that the electrons which are excited by the laser beam thermalize extremely quickly via electron-electron scatterings so that their occupation numbers $f_t(\epsilon^{\widetilde{m}_s}_{j\mathbf{k}})$ at time t can be described by the time-dependent Fermi distribution function $f_t(\epsilon^{\widetilde{m}_s}_{j\mathbf{k}})$ of Eq. (14). The electron temperature $T_{\rm el}(t)$ depends on time because during demagnetization energy is transferred from the electronic system to the lattice by electron-phonon scatterings. Because the expression (13) for dM/dt contains via Eq. (14) the time-dependent electron temperature, the demagnetization rate also depends on t. In principle it is desirable to characterize the ultrafast demagnetization by one quantity which does not depend on time. Commonly, the demagnetization time T_1 is used for that purpose. However, it can be calculated only if the full time dependence of M is known, i.e., its calculation requires much more effort. T_1 cannot be uniquely determined by dM/dt directly after the laser pulse only. $T_{\mathrm{el}}(t)$ is not calculated *ab initio*, but it is taken from experimental fits to the three-temperature-model (see Ref. [24] and references therein). The electron temperature directly after the laser pulse and the thermalization depends on the laser fluence; it is typically between 350 and 1000 K. For the calculation of the transition rates $W^{\lambda}_{j\mathbf{k},j'\mathbf{k}'}$ according to Eq. (1), it is assumed that the phonon temperature T_{p} directly after the laser pulse is equal to the temperature at which the experiment is performed (we use room temperature).

The demagnetization rate $\frac{dM}{dt}$ directly after the laser pulse is calculated for Ni for $T_{\mathrm{el}} = 1000$ K and $T_{\mathrm{p}} = 300$ K on the one hand with *ab initio* matrix elements $|M^{\lambda}_{j\mathbf{k},j'\mathbf{k}'}|^2$ in Eq. (1) and on the other hand with the values of $|M(\mathbf{q})|^2$ of Eq. (3) multiplied by $4b^2 \approx 0.1$. For the quantities $\epsilon_{j\mathbf{k}}$ and $m_{j\mathbf{k}\uparrow,j'\mathbf{k}'\downarrow}$ occurring in Eq. (13), we use in both cases the *ab initio* calculated values. Furthermore, for the calculation of dM/dt we consider states $\Psi_{j\mathbf{k}}$ and $\Psi_{j'\mathbf{k}'}$ for which $|\epsilon_{j\mathbf{k}} - \epsilon_{\mathrm{F}}| < 500$ mRy and $|\epsilon_{j\mathbf{k}} - \epsilon_{j'\mathbf{k}'}| < 100$ mRy, as in Ref. [12]. Thereby we calculate the *ab initio* matrix elements according to comparison 2 (Fig. 3), comparison 3 (Fig. 4), and comparison 4 (Fig. 5). The results are shown in Table I.

Note that we compare dM/dt for two types of calculations which both consider the contribution of electron-phonon scatterings to the ultrafast demagnetization after irradiation of a ferromagnetic film by a femtosecond laser pulse, whereby we insert in Eq. (13) the $W^{\lambda}_{j'\mathbf{k}'\downarrow,j\mathbf{k}\uparrow}$ as determined from Eq. (1), with the phenomenological approximation for the transition matrix elements for electron-phonon scattering on the one hand and with the *ab initio* calculated matrix elements on the other hand. In the experiment there may also be other contributions (e.g., electron-electron electron-magnon scattering), therefore it does not make sense to compare in detail with experimental data.

TABLE I. Demagnetization rates in units of $\mu_{\mathrm{B}}/(100$ fs atom) for the situation described in the figure captions.

Situation according to			
Fig. 3 red	Fig. 3 green	Fig. 3 blue	
0.009	0.2	0.009	
Situation according to			
Fig. 4 red	Fig. 4 blue		
0.009	1.1		
Situation according to			
Fig. 5 red	Fig. 5 violet	Fig. 5 green	Fig. 5 blue
1.4	0.013	0.2	0.009

In Table I we see that the demagnetization rate obtained by *ab initio* calculations with unscreened matrix elements is 0.009 $\mu_B/100$ fs, according to the situation of Fig. 3, red and of Fig. 4, red. We now want to figure out under what circumstances the phenomenological matrix elements yield similar results.

Astonishingly enough, the results for $\frac{dM}{dt}$ calculated with the *ab initio* matrix elements or with the approximated matrix element $4b^2|M(\mathbf{q})|^2$ are very similar (although the respective matrix elements differ strongly) if for k_{TF} the value of $6.67\frac{2\pi}{a}$ is inserted which is obtained by using the *ab initio* calculated density of states for Ni. The reason is that in Eq. (13) integrals over $\mathbf{k}$ and $\mathbf{k}'$ are performed and that the matrix elements appear in a weighted way. This justifies in retrospect the use of the matrix elements according to Eq. (3) as done in Ref. [19], in which $k_{\mathrm{TF}} = 6.67\frac{2\pi}{a}$ has been used as well, as $|M(\mathbf{q})|^2$ according to Eq. (3) has been used and multiplied by $4b^2$ (see above) with a value of $b^2 = 0.047$ (fitted to the experiments, the *ab initio* calculated value is 0.025–0.045, depending on the electron states used for averaging). Of course, this finding does not mean that the matrix elements (3) can also be safely used for the calculation of other macroscopic observables for which the matrix elements enter in another weighted way.

IV. CONCLUSIONS

Transition matrix elements for electron-phonon scatterings are important for many processes in solid state physics. The electron-phonon interaction has two contributions, one arising from the phonon-induced distortion of the lattice potential (Elliott part) and one resulting from the phonon-induced distortion of the spin-orbit coupling (Yafet part). In the present paper the Elliott part is considered.

In the literature the transition-matrix elements are calculated in two ways. First, there is an approximate expression for the matrix elements derived (for example in the textbooks of Ashcroft and Mermin [7] or of Czycholl [8]) by the combination of a phenomenological theory of electronic and ionic screening of the electron-electron interaction with a microscopic perturbation theory for the matrix elements. Second, the matrix elements are calculated in other papers by the *ab initio* electron theory. A comparison shows that the matrix elements calculated in these two ways differ very strongly, which in principle does not justify the use of the approximate expression. However, it is shown that possibly it can be used for the calculation of macroscopic observables which involve a weighted summation of matrix elements. This is shown for the example of the demagnetization rate of a ferromagnetic film after excitation with a fs laser pulse. However, this does not mean that the approximated matrix element can also be used for a calculation of other macroscopic observables for which they enter in another weighted way.

As a future project we will consider the transition matrix elements for the Yafet part of the electron-phonon interaction. In the literature there are already microscopic treatments of the electronic screening of the Yafet electron-phonon interaction by the spin-other-orbit interaction [4] or by the spin-same-orbit interaction [9]. In Ref. [9], a variety of contributions to the electronic screening of the Yafet part are found, however, no explicit results are given. We will try to figure out how large the various contributions are. In a second step we will try to figure out whether the unscreened *ab initio* matrix elements for the Yafet part can be modified in a way that these electronic screening effects are approximately taken into account, in the same sense as we have accounted for the electronic screening of the *ab initio* Elliott matrix elements in Sec. III. Finally, we will investigate whether there are also ionic screening effects for the Yafet part of the electron-phonon interaction.

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