Unexpectedly marginal effect of electronic correlations on ultrafast demagnetization after femtosecond laser-pulse excitation

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The treatment of ultrafast demagnetization after femtosecond laser-pulse excitation of a ferromagnetic film is usually done by a theory based on Fermi's golden rule which neglects the effects of electronic correlations. In the present paper the contribution of spin-flip electron-phonon scatterings to the ultrafast demagnetization of Ni is calculated by this theory and by the density-matrix theory in which the correlations are taken into account. The unexpected result is that the correlations which are essential for the ultrafast dynamics of nonmagnetic phenomena have only a marginal effect for the considered magnetic problem. From this point of view the use of Fermi's golden rule in all former papers on ultrafast demagnetization is justified.

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I. INTRODUCTION

When a ferromagnetic film is excited by an optical femtosecond (fs) laser pulse, an ultrafast demagnetization on a time scale of about 100 fs can be observed, followed by a remagnetization back to the initial equilibrium state on a longer time scale [1–3]. In spite of many experimental and theoretical investigations the detailed mechanisms for this effect are not yet totally clear. Spin-flip processes during scattering of electrons at other electrons [4,5] or at quasiparticles (phonons [6–8], magnons [9]) are possible mechanisms.

So far these scatterings have been described in all former papers worldwide by a combination of Fermi's golden rule with Boltzmann's rate equation (FB theory). There are three preconditions for the application of Fermi's golden rule. First, it is a first-order perturbation theory which neglects the memory of the system at time t to states at former times, i.e., it makes a Markov approximation. It has been shown in the literature on ultrafast dynamics of nonmagnetic phenomena that non-Markovian effects may be relevant [10,11]. Second, it is valid for a time scale considerably larger than the oscillation time of the time-periodic perturbation because only then the Sinc function occurring in the perturbation theory can be replaced by Dirac's delta functional. However, the time scale of about 100 fs is shorter than typical oscillation times of the quasiparticles. Third, it is a theory for the occupation numbers (leading to rate equations) and does not take into account the effect of electronic correlations which we will introduce below and which have been shown in the literature to be essential for the ultrafast dynamics of nonmagnetic situations [12,13]. In the present paper we calculate the demagnetization and remagnetization due to spin-flip electron-phonon scatterings, once using the FB theory and once using a density-matrix theory (DM) [14]. In the density-matrix theory the effect of electronic correlations is taken into account. From this point of view it improves the FB theory. In our density-matrix theory we also use a Markov approximation and a long-time limit which replaces the Sinc function by a δ functional. This means that we do not study the influence of these two latter approximations. Instead, we will investigate the influence of the electronic correlations. We will show that this influence is unexpectedly marginal for the magnetic problem of ultrafast demagnetization. This is the first study of electronic correlations in the sense of a DM theory for a magnetic system. In the literature it has been shown [6–8] that spin-flip electron-phonon scatterings yield a non-negligible contribution to the ultrafast demagnetization after laser pulses although they cannot explain the whole effect quantitatively. We confine ourselves to study the effect of electronic correlations just for these electron-phonon scatterings, for which a rather simple form of the scattering matrix elements can be derived (see below). The scope of the paper is not to give a complete theory of ultrafast magnetization dynamics, but to investigate the effect of electronic correlations for an important and mathematically tractable contribution to it. There are papers on magnetic systems in which the effect of electronic correlations in the sense of a band theory are studied, by using a Hubbard Hamiltonian which introduces larger electronic correlations in the sense of a band theory than those included in a density functional theory. The electronic correlations of the density-matrix theory are different quantities from the electronic correlations in the sense of a band theory (see Sec. III). Therefore we investigate in our paper f the effect of electronic correlations in the sense of the density-matrix theory on dynamic magnetic processes.

II. MODELS FOR ELECTRON AND PHONON SPECTRA

For fcc Ni we model the electronic bands by a Stoner-split modified tight-binding model which contains only two bands, one for spin-up majority electrons ($j = 1$) and one for spin-down minority electrons ($j = 2$) which are constantly shifted against each other by the exchange splitting. For the energies $\epsilon_{\mathbf{k},j}$ of the single-electron states $|\mathbf{k}, j\rangle$ with the wave vector $\mathbf{k}$ we use

$$\epsilon_{\mathbf{k},j} = \xi_k\left(-t\sum_{\gamma} e^{-i\mathbf{k}\cdot\mathbf{R}_\gamma}\right) + U\frac{n_j}{N}. \tag{1}$$

Here the $\mathbf{R}_\gamma$ are the position vectors of the nearest-neighbor atoms of an arbitrarily picked out central atom, U is the

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Hubbard parameter describing the on-site Coulomb interaction between the electrons, n_j is the number of electrons per atom in band j, and $N = n_1 + n_2$. In reality the exchange splitting is different for different $\mathbf{k}$. However, it would be far too time-consuming to take this into account in the quantum kinetic calculations by an *ab initio* density functional calculation. In this sense we compare the results of the density-matrix theory with those of the FB theory for this electronic model. For the parameter t we use the value $t = 2.7$ eV [15]. The quantity ξ_k is a correction factor which enlarges the number of available states close to the Fermi level ϵ_F [16]. The correction factor is required to fulfill for our model the Stoner criterion for a stable ferromagnetic state. We assume that in Ni we have ten valence states per atom which are hybridizations between two atomic s states and eight atomic d states. For our model with two bands, therefore, each band can be filled with a maximum of ten electrons. During demagnetization the magnetization decreases, i.e., the exchange splitting decreases. This is taken into account in the calculation, i.e., the band structure is dynamically adapted (n_j changes in time). We fit the parameter U to the value 0.41 eV in order to get the correct Curie temperature $T_C = 627.24$ K for fcc Ni. The magnetic moment per atom at 293 K then is $0.55\mu_B$ which is only a bit different from the experimental result of $0.616\,\mu_B$ (which includes also a small contribution of the electron orbital moment, neglected in our model).

For the phonons we use a model [16] where all branches of the vibrational spectrum are replaced by three branches (one longitudinal and two transversal) with the same linear dispersion relation $\omega_{\mathbf{q},\lambda} = c|\mathbf{q}|$ where λ is the polarization index, $\lambda = 1,2,3$. We insert for the velocity the value found in the literature for the transversal phonons in magnetic Ni.

III. CALCULATIONS BY DENSITY-MATRIX THEORY

The time dependence of the demagnetization is obtained by calculating the occupation numbers $f(\epsilon_{\mathbf{k},j},t) = \langle \hat{C}^\dagger_{\mathbf{k},j}\hat{C}_{\mathbf{k},j}\rangle(t)$. Neglecting the contribution of the electronic orbital moments, the z component of the magnetic moment per atom is given by

$$\begin{aligned} m_z(t) &= [n_1(t) - n_2(t)]\mu_B \\ &= \sum_{\mathbf{k}} (\langle \hat{C}^\dagger_{\mathbf{k},1}\hat{C}_{\mathbf{k},1}\rangle(t) - \langle \hat{C}^\dagger_{\mathbf{k},2}\hat{C}_{\mathbf{k},2}\rangle(t))\mu_{\mathrm{B}}. \end{aligned} \tag{2}$$

The $\hat{C}^\dagger_{\mathbf{k},j}$ and $\hat{C}_{\mathbf{k},j}$ are operators which create and annihilate an electron in state $|\mathbf{k},j\rangle$. We now discuss the equation of motion for $\langle \hat{C}^\dagger_{\mathbf{k},j}\hat{C}_{\mathbf{k},j}\rangle$ using Heisenberg's picture of quantum mechanics where the operators are time dependent, yielding

$$\frac{d}{dt}\langle \hat{C}^\dagger_{\mathbf{k},j}\hat{C}_{\mathbf{k},j}\rangle = \left\langle \frac{d\hat{C}^\dagger_{\mathbf{k},j}}{dt}\hat{C}_{\mathbf{k},j}\right\rangle + \left\langle \hat{C}^\dagger_{\mathbf{k},j}\frac{d\hat{C}_{\mathbf{k},j}}{dt}\right\rangle, \tag{3}$$

with

$$\frac{d\hat{C}^\dagger_{\mathbf{k},j}}{dt} = \frac{i}{\hbar}[\hat{H},\hat{C}^\dagger_{\mathbf{k},j}], \quad \frac{d\hat{C}_{\mathbf{k},j}}{dt} = \frac{i}{\hbar}[\hat{H},\hat{C}_{\mathbf{k},j}]. \tag{4}$$

The operator $\hat{H}$ is the Hamiltonian,

$$\hat{H} = \hat{H}_{\mathrm{e}} + \hat{H}_{\mathrm{ph}} + \hat{H}_{\mathrm{e\text{-}ph}}, \tag{5}$$

with the electronic part

$$\hat{H}_e = \sum_{\mathbf{k},j} \epsilon_{\mathbf{k},j}\hat{C}^\dagger_{\mathbf{k},j}\hat{C}_{\mathbf{k},j}, \tag{6}$$

the phononic part

$$\hat{H}_{\mathrm{ph}} = \sum_{\mathbf{q},\lambda} \hbar\omega_{\mathbf{q},\lambda}\hat{b}^\dagger_{\mathbf{q},\lambda}\hat{b}_{\mathbf{q},\lambda}, \tag{7}$$

and with the Hamiltonian for the scattering of electrons at phonons (which will be introduced below). The operators $\hat{b}^\dagger_{\mathbf{q},\lambda}$ and $\hat{b}_{\mathbf{q},\lambda}$ create and annihilate phonons with wave vectors $\mathbf{q}$ and polarization vectors $\mathbf{e}_{\mathbf{q},\lambda}$, respectively.

The equations for $\langle \hat{C}^\dagger_{\mathbf{k},j}\hat{C}_{\mathbf{k},j}\rangle$ and for $\langle \hat{b}^\dagger_{\mathbf{q},\lambda}\hat{b}_{\mathbf{q},\lambda}\rangle$ contain expectation values of products of three operators. For them also equations of motion can be derived which contain expectation values of products of four operators, etc. This hierarchy of infinitely many coupled equations is terminated by a decoupling procedure [10,17]. We apply the decoupling by approximating the expectation values of products of four operators by products of all nonvanishing expectation values of lower-order products of these operators.

The electronic part $\hat{H}_e$ of $\hat{H}$ is specified by Eq. (1). The phononic part $\hat{H}_{\mathrm{ph}}$ is specified by the above discussed vibrational model with three branches and $\omega_{\mathbf{q},\lambda} = c|\mathbf{q}|$. For the electron-phonon operator we use

$$\begin{aligned} \hat{H}_{\mathrm{e\text{-}ph}} = &\sum_{\mathbf{k},j,j'}\sum_{\mathbf{q},\lambda} g_\lambda(\mathbf{q})[(1-asf)\delta_{jj'} + asf]\hat{C}'^\dagger_{\mathbf{k}+\mathbf{q},j}\hat{b}_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k},j} \\ &+ \sum_{\mathbf{k},j,j'}\sum_{\mathbf{q},\lambda} g^*_\lambda(\mathbf{q})[(1-asf)\delta_{jj'} + asf]\hat{C}^\dagger_{\mathbf{k},j'}\hat{b}^\dagger_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k}+\mathbf{q},j}, \end{aligned} \tag{8}$$

with $j,j' = 1,2$, which describes scattering processes from $|\mathbf{k},j\rangle$ to $|\mathbf{k}+\mathbf{q},\mathbf{j}'\rangle$ by absorbing a phonon and from $|\mathbf{k}+\mathbf{q},\mathbf{j}\rangle$ to $|\mathbf{k},j'\rangle$ by emitting a phonon. In Eq. (8) the quantity $g_\lambda(\mathbf{q})$ is the scattering matrix element calculated in Ref. [18] for scatterings without spin flips. This electron-phonon scattering operator is generalized in Eq. (8) because $\hat{H}$ includes the dependence of the matrix element on the spin states of the initial and final electronic states. If the scattering process changes the electron state from the spin-up band ($j = 1$) to the spin-down band ($j = 2$) or the other way around, then $g_\lambda(\mathbf{q})$ is multiplied by $asf = \sqrt{a_{sf}}$ where a_{sf} is in the probability for a spin-flip scattering. If the electron remains in the same band ($j = j'$), then $g_\lambda(\mathbf{q})$ is multiplied by 1. We use $\sqrt{a_{sf}}$ because the scattering rate is determined by the absolute square of the matrix element. The value of a_{sf} is determined [2] from a fit of the microscopic three-temperature model of ultrafast demagnetization to experimental data, yielding [2] $a_{sf} = 0.19$ for fcc Ni.

The equation of motion for $\langle \hat{C}^\dagger_{\mathbf{k},1}\hat{C}_{\mathbf{k},1}\rangle$ obtained after decoupling contains for our two-band model on the right-hand side the expectation values $\langle \hat{C}^\dagger_{\mathbf{k}+\mathbf{q},1}\hat{b}_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k},1}\rangle$, $\langle \hat{C}^\dagger_{\mathbf{k}+\mathbf{q},2}\hat{b}_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k},1}\rangle$, $\langle \hat{C}^\dagger_{\mathbf{k}-\mathbf{q},1}\hat{b}^\dagger_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k},1}\rangle$, and $\langle \hat{C}^\dagger_{\mathbf{k}-\mathbf{q},2}\hat{b}^\dagger_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k},1}\rangle$ and the Hermitian conjugates. The equation of motion for $\langle \hat{C}^\dagger_{\mathbf{k},2}\hat{C}_{\mathbf{k},2}\rangle$ has the same form, but the index "1" is replaced everywhere by "2" and "2" is replaced everywhere by "1". We denote these expectation values by the symbol $y(t)$. The equations of motion

for $y(t)$ have the general form of a first-order inhomogeneous differential equation with, respectively, an inhomogeneity $F(\mathbf{q},\lambda,\mathbf{k},\mathbf{j},\mathbf{j}',\mathbf{t})$,

$$\frac{dy(t)}{dt}t - i\Omega y(t) = F(\mathbf{q},\lambda,\mathbf{k},\mathbf{j},\mathbf{j}',\mathbf{t}), \tag{9}$$

where the inhomogeneity F and the frequency Ω is different for the different expectation values introduced above, with, e.g., $\Omega = (1/\hbar)(\epsilon_{\mathbf{k}+\mathbf{q},1} - \epsilon_{\mathbf{k},1} - \hbar\omega_{\mathbf{q},\lambda})$ for $\langle \hat{C}^{\dagger}_{\mathbf{k}+\mathbf{q},1}\hat{b}_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k},1}\rangle$.

In the following we do not try to get the exact solution of Eq. (9), but we solve this equation by the use of a Markov approximation. Thereby we write the expectation values occurring in $\frac{dy(t)}{dt}t$ (we show representative examples below) as products of a factor which varies slowly in time with an exponential:

$$\langle \hat{C}^{\dagger}_{\mathbf{k},i}\hat{C}_{\mathbf{k},j}\rangle(t) = \tilde{f}^{ij}_{\mathbf{k}}(t) = \tilde{f}^{ij}_{\mathbf{k}}(t)e^{(i/\hbar)(\epsilon_{\mathbf{k},i}-\epsilon_{\mathbf{k},j})(t-t_0)}, \tag{10}$$

$$\begin{aligned}\langle \hat{C}^{\dagger}_{\mathbf{k}',i}\hat{b}_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k},j}\rangle(t) &= s^{ij\lambda}_{\mathbf{k}',\mathbf{q},\mathbf{k}}(t) \\ &= \tilde{s}^{ij\lambda}_{\mathbf{k}',\mathbf{q},\mathbf{q},\mathbf{k}}(t)e^{(i/\hbar)(\epsilon_{\mathbf{k}',i}-\epsilon_{\mathbf{k},j}-\hbar\omega_{\mathbf{q},\lambda})(t-t_0)}.\end{aligned} \tag{11}$$

The reason for this is the fact that without $\hat{H}_{\text{e-ph}}$ in the Hamiltonian the time developments would be given by the fast oscillating exponentials in Eqs. (10) and (11), whereas when including the small part $\hat{H}_{\text{e-ph}}$ we get additional slow time dependencies expressed by $\tilde{f}^{ij}_{\mathbf{k}}(t)$ and $\tilde{s}^{ij\lambda}_{\mathbf{k}',\mathbf{q},\mathbf{q},\mathbf{k}}(t)$ in the sense of an interaction picture. The expectation values $\langle \hat{C}^{\dagger}_{\mathbf{k},i}\hat{C}_{\mathbf{k},j\neq i}\rangle$ are called correlations. In the band theory based, e.g., on the density functional theory the electronic correlation energy describes the difference between the true total electronic energy and the electronic energy in Hartree-Fock approximation. It results from the Coulomb interactions between the electrons. This interaction is taken into account by two contributions. The first contribution describes the mean effect of the Coulomb interactions of an electron with all the other electrons, and this contribution is represented by the Hartree energy in the Hartree-Fock approximation. The second contribution is the deviation of the real Coulomb interaction energy from the mean Coulomb interaction energy, and this contribution is represented by the electronic correlation energy. The electronic energy thereby is written in terms of the distributions $\langle \hat{C}^{\dagger}_{\mathbf{k},j}\hat{C}_{\mathbf{k},j}\rangle$. In contrast, electronic correlations in the density-matrix theory are defined as $\langle \hat{C}^{\dagger}_{\mathbf{k},i}\hat{C}_{\mathbf{k},j}\rangle$ for $i \neq j$. These expectation values do not appear in the energy expression of the band theory. Their values are also affected by the electronic correlations in the sense of a band theory, but they do not enter the energy expression of this theory. In our density-matrix theory the electronic correlations result from the spin-flip scatterings of electrons at phonons which yield a nonzero $\langle \hat{C}^{\dagger}_{\mathbf{k},i}\hat{C}_{\mathbf{k},j}\rangle$ for $i \neq j$. So, altogether, the electronic correlations in the sense of a band theory and those in the sense of our density-matrix theory have different physical origins, and they are described by different expectation values.

Using Eq. (9) the equation of motion for $\tilde{s}^{ij\lambda}_{\mathbf{k}+\mathbf{q},\mathbf{q},\mathbf{k}}(t)$ may be derived. It contains on the right-hand side an expression with the expectation values $f^{11}_{\mathbf{k}}$, $f^{11}_{\mathbf{k}+\mathbf{q}}$, $f^{21}_{\mathbf{k}}$, and $f^{12}_{\mathbf{k}+\mathbf{q}}$, and the whole expression is multiplied by $\exp[-(i/\hbar)(\epsilon_{\mathbf{k}+\mathbf{q},1} - \epsilon_{\mathbf{k},1} - \hbar\omega_{\mathbf{q},\lambda})(t-t_0)]$. Inserting Eq. (10) introduces further exponentials. Altogether this gives

$$\begin{aligned}\tilde{s}^{11\lambda}_{\mathbf{k}+\mathbf{q},\mathbf{q},\mathbf{k}}(t) = \tilde{s}^{11\lambda}_{\mathbf{k}+\mathbf{q},\mathbf{q},\mathbf{k}}(t_0) + \frac{i}{\hbar}\int_{t_0}^{t} dt'[&\tilde{A}(t')e^{-(i/\hbar)(\epsilon_{\mathbf{k}+\mathbf{q},1}-\epsilon_{\mathbf{k},1}-\hbar\omega_{\mathbf{q},\lambda})(t'-t_0)} + \tilde{B}(t')e^{-(i/\hbar)(\epsilon_{\mathbf{k}+\mathbf{q},1}-\epsilon_{\mathbf{k},2}-\hbar\omega_{\mathbf{q},\lambda})(t'-t_0)} \\ &+ \tilde{C}(t')e^{-(i/\hbar)(\epsilon_{\mathbf{k}+\mathbf{q},2}-\epsilon_{\mathbf{k},1}-\hbar\omega_{\mathbf{q},\lambda})(t'-t_0)} + \tilde{D}(t')e^{-(i/\hbar)(\epsilon_{\mathbf{k}+\mathbf{q},2}-\epsilon_{\mathbf{k},2}-\hbar\omega_{\mathbf{q},\lambda})(t'-t_0)}],\end{aligned} \tag{12}$$

with $\tilde{A}(t)$, $\tilde{B}(t)$, $\tilde{C}(t)$, and $\tilde{D}(t)$ which are slowly varying in time because they contain as time-dependent quantities only the various $\tilde{f}^{ij}_{\mathbf{k}}(t)$, and which can therefore be put in front of the integral (Markov approximation). We now make the same long-time approximation as in Fermi's golden rule, i.e., we represent the Sinc functions which result from the integrations over the exponentials in Eq. (12) after the tilded quantities have been put in front of the integral by δ functionals. The final result is

$$\begin{aligned}\tilde{s}^{11\lambda}_{\mathbf{k}+\mathbf{q},\mathbf{q},\mathbf{k}}(t) = \tilde{s}^{11\lambda}_{\mathbf{k}+\mathbf{q},\mathbf{q},\mathbf{k}}(t_0) &+ \pi\frac{i}{\hbar}\tilde{A}(t)\delta(\epsilon_{\mathbf{k}+\mathbf{q},1} - \epsilon_{\mathbf{k},1} - \hbar\omega_{\mathbf{q},\lambda}) + \pi\frac{i}{\hbar}\tilde{B}(t)\delta(\epsilon_{\mathbf{k}+\mathbf{q},1} - \epsilon_{\mathbf{k},2} - \hbar\omega_{\mathbf{q},\lambda}) \\ &+ \pi\frac{i}{\hbar}\tilde{C}(t)\delta(\epsilon_{\mathbf{k}+\mathbf{q},2} - \epsilon_{\mathbf{k},1} - \hbar\omega_{\mathbf{q},\lambda}) + \pi\frac{i}{\hbar}\tilde{D}(t)\delta(\epsilon_{\mathbf{k}+\mathbf{q},2} - \epsilon_{\mathbf{k},2} - \hbar\omega_{\mathbf{q},\lambda}).\end{aligned} \tag{13}$$

To calculate $m_z(t)$ according to Eq. (2) with the final results for $\langle \hat{C}^{\dagger}_{\mathbf{k},i}\hat{C}_{\mathbf{k},j}\rangle$ which contain the $\tilde{s}^{11\lambda}_{\mathbf{k}+\mathbf{q},\mathbf{q},\mathbf{k}}(t)$ with the δ functionals of Eq. (13) and the expectation values of products of the other three operators listed before Eq. (9) (which are calculated in the same way as $\langle \hat{C}^{\dagger}_{\mathbf{k}+\mathbf{q},1}\hat{b}_{\mathbf{q},\lambda}\hat{C}_{\mathbf{k},1}\rangle$), we use a discrete $\mathbf{k}$-point grid. As a result we cannot just replace the δ functionals by the corresponding Kronecker deltas, because then more or less all Kronecker deltas would yield zero. Therefore the deltas are replaced by Gaussian curves with a smoothing parameter σ [19]. In principle one then should test for convergence of the results for $m_z(t)$ with respect to the number of $\mathbf{k}$ points in the grid and σ, thereby keeping σN_1 constant, where N_1 is the number of $\mathbf{k}$ points in one direction of the Brillouin zone. However, this would be too time-consuming. We therefore have done the calculation only for $N_1 = 40$ and $\sigma = 4$ mRy, which are typical values used in the literature.

We now perform two types of calculations. In the first type we start with a Fermi-Dirac distribution at $T = 300$ K for $\langle \hat{C}^{\dagger}_{\mathbf{k},j}\hat{C}_{\mathbf{k},j}\rangle$, a Bose-Einstein distribution at $T = 300$ K for $\langle \hat{b}^{\dagger}_{\mathbf{q},\lambda}\hat{b}_{\mathbf{q},\lambda}\rangle$, and zero correlations $\langle \hat{C}^{\dagger}_{\mathbf{k},j}\hat{C}_{\mathbf{k},j'\neq j}\rangle$. Then the code runs until we reach the stationary thermodynamic equilibrium state for all expectation values. In this stationary state the

correlations are nonzero; they have been built by the spin-flip electron-phonon scatterings. In the second type we start with the state directly after the action of the laser pulse. This state is described by the Elliott-Yafet scenario [2,3] in which it is assumed that the laser beam produces a nonequilibrium situation in which directly after the pulse the magnetic moment m_z is still the magnetic moment $m_z(t_0)$ we had before the pulse, and the $\langle \hat{C}^\dagger_{\mathbf{k},j} \hat{C}_{\mathbf{k},j} \rangle$ are given by nonequilibrium Fermi-Dirac distributions $f_{\mathrm{FD}}(\epsilon_{\mathbf{k},j}, \mu_j, T_e)$ where the chemical potentials are different for spin-up and spin-down electrons and where the electron temperature T_e is increased. In our simulation we use a typical value [2] of $T_e = 600$ K. The μ_j are determined from two equations. First we demand that $m_z(t_0) = \mu_B[n_1(t_0) - n_2(t_0)]$ where $n_{1,2}(t_0)$ can be calculated by using f_{FD} and the electron density of the states for our tight-binding model. Second, we demand $N = n_1(t_0) + n_2(t_0) = 10$ in the case of Ni. Optical photons have typical energies of about 2 eV, and therefore they can excite electrons with that excitation energy. The excited electron system then thermalizes very quickly by electron-electron scatterings, leading to the increased electron temperature T_e, for which we insert 600 K which corresponds to a thermal energy of about 0.05 eV. These thermalized excited electrons show spin-flip scatterings either at other electrons [4,5] or at phonons [6–8] or at magnons [9], and this leads to modifications of the lengths of the atomic magnetic moments. This is the accepted picture of the Elliott-Yafet scenario which has been used in most of the theoretical studies of ultrafast demagnetization, also in Ni. There may also be other contributions to the ultrafast demagnetization, for instance, electronic spin currents for the case of superdiffusion [20] of excited mainly spin-up electrons from the sample to a conducting substrate which we neglect in our calculations, i.e., we implicitly assume an insulating substrate. As soon as electrons are excited by transitions to other electronic bands, the electronic correlations in the sense of a DM theory come into play. To investigate their effect, it is therefore good to use the Elliott-Yafet scenario. One future project of research may be to question the validity of the so far widely accepted Elliott-Yafet scenario. However, this is not within the scope of the present paper. The scope is to study the effect of electronic correlations for cases of the magnetization dynamics in which electrons are excited by transitions to other electronic bands. For Ni the electronic correlations in the sense of a band theory are small because the Hubbard parameter is small. But this does not mean that the electronic correlations in the sense of the DM theory are automatically also small.

For the phononic correlations $\langle \hat{b}^\dagger_{\mathbf{q},\lambda} \hat{b}_{\mathbf{q},\lambda' \neq \lambda} \rangle$ we start with zero. We fix the phononic occupation numbers $\langle \hat{b}^\dagger_{\mathbf{q},\lambda} \hat{b}_{\mathbf{q},\lambda} \rangle$ to the Bose-Einstein distribution with $T_{\mathrm{ph}} = 300$ K. Thereby we assume that the laser photons do not directly excite phonons. Furthermore, the fixing of the phononic distributions mimics the fact that in reality the heat delivered by the laser pulse is transferred from the sample to the surroundings, so that for very long times the phonon and the electron temperatures are equal and given by the temperature $T = 300$ K at which the experiment is performed. Therefore at the end we have $m_z(t)/m_z(t_0) = 1$. For the initial values of the electronic correlations we use the values obtained at the end of the first type of calculation, i.e., we assume that the laser beam does not influence the correlations strongly. In reality there is some influence of the laser photons on the correlations. In principle this could be taken into account by explicitly including the electron- photon interaction in the Hamiltonian, e.g., in dipole approximation. However, the main purpose of our theory is to compare the results of the density-matrix calculations with those of the FB theory. As long as the density-matrix calculations give results which agree qualitatively with the experimental results, we can compare the results of the two theories for the model situation that the laser photons do not directly influence the correlations. We thus start with a situation directly after the action of the laser pulse with initial values of the distributions according to the Elliott-Yafet scenario and with initial values for the correlations resulting from our type 1 calculation after having reached the stationary situation. If for this model situation the density-matrix theory and the FB theory give very similar results, this would certainly also be the case for a situation in which we take into account the direct influence of the photons on the correlations, i.e., for which we start with slightly modified initial values for the correlations.

IV. RESULTS

Figure 1 shows the results for $m_z(t)/m_z(t_0)$. There is a fast demagnetization and a remagnetization on a larger time scale, as in the experiments. There are oscillations in time with a period of roughly 20 fs with amplitudes which increase in time so that they are more clearly visible in the remagnetization process. When the final stationary state is reached, then the amplitude does not increase any longer. Of course we cannot expect that the experimental results for Ni are perfectly reproduced by our theory, because we consider only the contribution of spin-flip electron-phonon scatterings, whereas in reality there may be other contributions (see the first paragraph), and because we use simple models for the electronic band structure and for the phonons. More important is the comparison between the results obtained with the density-matrix formalism and those we get by using the FB theory. The FB results are obtained by setting the various electronic correlations to zero at all times. When doing this, the density-matrix equations for $\langle \hat{C}^\dagger_{\mathbf{k},j} \hat{C}_{\mathbf{k},j} \rangle$ are then the same as those used in a FB theory [21]. As shown in Fig. 1, both types of calculations give very similar results. The main difference is the oscillations in $m_z(t)$ obtained by the density-matrix

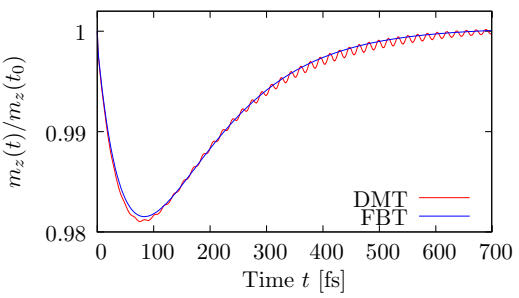

FIG. 1. $\frac{m_z(t)}{m_z(t_0)}$ of fcc Ni after excitation with a femtosecond optical laser pulse. Red: Results of the density-matrix calculation. Blue: Results of the Fermi-Boltzmann theory.

theory, which result from the correlations neglected in the FB theory. In the experiments no oscillations are seen. There may be three reasons for this. First, the time resolution of the measurement (e.g., by time-resolved magneto-optical Kerr effect measurements [1,2]) may not be good enough. Second, the very fast oscillations may be suppressed by damping of the magnetization dynamics which occur in reality but are not taken into account in our theory. Third, in reality the exchange splitting between the bands depends on $\mathbf{k}$, leading to different oscillation frequencies in the correlation terms, so that destructive interferences might reduce the oscillation amplitude of $m_z(t)$.

So far we have investigated the behavior of the total magnetic moment after femtosecond laser pulses. This quantity is related to the electronic spin moments averaged over all the electrons involved in the process. We have also investigated spectrally resolved quantities by looking at the $\langle \hat{C}^{\dagger}_{\mathbf{k},j} \hat{C}_{\mathbf{k},j} \rangle$ for values of $\mathbf{k}$ which correspond to energies $\epsilon_{\mathbf{k},j}$ in various ranges of about 0.01 Ry around the Fermi energy. For these quantities the results of the two calculations are also very similar, which shows that the correlations have only a very small influence on the occupations of the spin states.

V. CONCLUSIONS

To conclude, we have applied the density-matrix theory to a problem in magnetism by calculating the contribution of spin-flip electron-phonon scatterings to the demagnetization and the following remagnetization of the magnetic moment of fcc Ni after excitation with a femtosecond optical laser pulse. The results are very similar to those obtained by the Fermi-Boltzmann theory which neglects the quantum-kinetic electronic correlations. This shows that for the ultrafast demagnetization the electronic correlations are not very important, in contrast to other physical phenomena where the correlations are important (see above). From this point of view the use of the FB theory for a description of the ultrafast magnetization phenomena after laser-pulse excitation in all former papers in the literature is justified. This finding is a very important result from the viewpoint of fundamental physics which is of interest for all people working on ultrafast dynamic phenomena. In the Fermi-Boltzmann theory a Markov approximation is used. We used a Markov approximation also in our density-matrix theory. The hope is that the results of the two theories would be very similar also when avoiding the Markov approximation in the density-matrix theory. To test this is a future project of our research. We hope that our paper will initiate the following possible extensions of the theory: (a) The investigation whether the effect of electronic correlations is marginal also for the other contributions to ultrafast demagnetization (see the first paragraph). (b) The application of the theory to other systems, e.g., Fe [22], where the values of U are larger, i.e., the electronic correlations in the sense of a band theory are stronger. As mentioned before, this does not automatically mean that the electronic correlations in the sense of a DM theory are also stronger. These latter electronic correlations depend, among others, on the degree of excitations of the electrons to other bands which is of course different between Ni and Fe, for instance. (c) The application of the theory for other values of the duration and photon energy of the laser pulse.

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