

Nonlinear optical quantum theory of demagnetization in $L1_0$ FePt and FePd

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(Dated: Thursday 30th October, 2025)

Abstract

It is now well established that a laser pulse can demagnetize a ferromagnet. However, for a long time, it has not had an analytic theory because it falls into neither nonlinear optics (NLO) nor magnetism. Here we attempt to fill this gap by developing a nonlinear optical theory centered on the spin moment, instead of the more popular susceptibility. We first employ group theory to pin down the lowest order of the nonzero spin moment in a centrosymmetric system to be the second order, where the second-order density matrix contains four terms of sum frequency generation (SFG) and four terms of difference frequency generation (DFG). By tracing over the product of the density matrix and the spin matrix, we are now able to compute the light-induced spin moment. We apply our theory to FePt and FePd, two most popular magnetic recording materials with identical crystal and electronic structures. We find that the theory can clearly distinguish the difference between those two similar systems. Specifically, we show that FePt has a stronger light-induced spin moment than FePd, in agreement with our real-time ultrafast demagnetization simulation and the experimental results. Among all the possible NLO processes, DFGs produce the largest spin moment change, a manifestation of optical rectification. Our research lays a solid theoretical foundation for femtomagnetism, so the light-induced spin moment reduction can now be computed and compared among different systems, without time-consuming real-time calculations, representing a significant step forward.

I. INTRODUCTION

Using the light to change magnetic properties in semiconductors, antiferromagnets and ferrites¹ can be traced back to several decades ago². Using an ultrafast laser pulse to demagnetize magnetic materials³ launched a new frontier, femtosecond magnetism, or femtomagnetism. The pioneering work by Beaurepaire and coworkers inspired several decades of intense investigations (see reviews^{4,5}). The laser pulses can also switch spins from one direction to another permanently^{6,7}. These discoveries are potentially applicable to spintronics, with improved speed and efficiency (see reviews^{5,7,8}).

However, in contrast to femtochemistry in molecules^{9,10} and femtobiology in photoisomerization in rhodopsins and yellow proteins^{11–15}, femtomagnetism lacks a firm grounding at a quantitative level in neither nonlinear optics (NLO) nor magnetism. The traditional spin wave theory¹⁶ is based on the Heisenberg spin exchange model or Stoner’s spin wave model without a light field^{16–20}, where spin-wave excitation (magnon) is at the center of demagnetization^{17,21}. On the other hand, NLO centers on the light generation, not the spin moment change, and is formulated around different orders of susceptibilities^{22–25}. For instance, the traditional magneto-optics^{26–29} investigates how a magnetic field affects the generated light signal. The inverse Faraday effect (IFE)²³ is probably the only exception, but there is no guarantee for demagnetization instead for magnetization. A prior study³⁰ focused on the helicity-dependent Faraday constants in simple $3d$ ferromagnets, without real-time-dependent simulation, so this does not explain the above ultrafast demagnetization experiments, where the spin moment is always decreased. To the best of our knowledge, a nonlinear optical theory that can yield a negative spin moment change does not exist.

In this paper, we aim to develop a nonlinear optical quantum theory for demagnetization perturbatively, by focusing on the spin moment change in ferromagnets. We employ two centrosymmetric magnetic materials, FePt and FePd, with similar crystal and electronic structures. We start with the symmetry analysis and find that the first-order light-induced spin moment for a centrosymmetric system is zero, and the lowest nonzero spin moment is the second order. This sets the framework for our NLO theory. There are in total eight NLO processes: four are sum frequency generation (SFG) and the other four are difference frequency generation (DFG). SFGs contribute a tiny spin change, but the contribution from DFGs is very large. Physically, DFGs correspond to the optical rectification. It is the com-

petition between two DFGs that leads to the negative spin moment. Under the same fluence, FePt has a stronger spin moment reduction than FePd, since the former has a stronger spin-orbit coupling. To verify this result, we carry out the time-dependent simulation of spin moment change and find that dynamically, FePt demagnetizes more, regardless of the laser photon energy, pulse duration, and fluence, fully consistent with the experimental results by Iihama *et al.*³¹. Our study represents a serious attempt to understand ultrafast demagnetization, without actual time-dependent simulation. Our theory will have a significant impact on future research, by providing a means to compute and contrast different materials at the quantitative first-principles level, greatly enhancing accessibility and reproducibility to the broader research community of ultrafast spintronics and all-optical spin switching.

The rest of the paper is arranged as follows. In Sec. II, we present our theoretical formalism and the ground-state properties of FePt and FePd. Section III is devoted to the nonlinear optical quantum theory for spin response, where we start from the symmetry analysis, and then move on to an analytic theory for the spin change. In Sec. IV, we carry out the real-time simulation of ultrafast demagnetization under laser excitation to realistically test the results of nonlinear optical quantum theory. We conclude this paper in Sec. V. We provide a detailed derivation of our main formulas in Appendix A. Since our theory uses a good number of symbols which the reader may find difficult to follow, we list them in our Table I.

II. THEORETICAL FORMALISM

There is no better example than FePt and FePd. They are among the most studied materials for magnetic recording^{32–34}, with a large magnetic anisotropy³⁵. The dependence of ultrafast demagnetization on the Mn doping³⁶, temperature and fluence³⁷ have been thoroughly examined. Yamamoto *et al.*³⁸ investigated the ultrafast demagnetization at the Pt-edge. Liu *et al.*³⁹ employed a single pump pulse to excite FePt, and found that the pump reduces their Kerr hysteresis loop but not the coercivity. Shi *et al.*⁴⁰ utilized two pump pulses of the same fluence, and showed that the coercivity is reduced once the laser fluence is above 4 mJ/cm². This important result reveals a possible onset for magnetic domain changes that are crucial for all-optical spin switching. All-optical spin switching was reported in FePt nanoparticles⁴¹ and Cr- and Mn-doped FePt films⁴².

Theoretically, we employ the state-of-the-art density functional theory as implemented in the Wien2k code⁴³. We numerically solve the Kohn-Sham equation

$$\left[-\frac{\hbar^2 \nabla^2}{2m_e} + V_{Ne} + V_H + V_{xc} \right] \psi_{n\mathbf{k}}(\mathbf{r}) = E_{n\mathbf{k}} \psi_{n\mathbf{k}}(\mathbf{r}), \quad (1)$$

where the terms on the left are the kinetic energy operator, the attraction between the nuclei and electrons, the Hartree term, and the exchange-correlation at the PBE level, respectively. The spin-orbit coupling is included using a second-variational method in the same self-consistent iteration. FePt and FePd share similar structures and crystallize in a face-centered tetragonal L1₀ structure⁴⁴, with space group No. 123, P4/mmm. Figure 1(a) illustrates that Fe occupies two inequivalent sites 1a(0, 0, 0) and 1c($\frac{1}{2}, \frac{1}{2}, 0$), and Pt/Pd takes two equivalent sites (2e)(0, $\frac{1}{2}, \frac{1}{2}$), ($\frac{1}{2}, 0, \frac{1}{2}$), so there are four atoms in the conventional tetragonal face-centered unit cell. They also have similar lattice constants $a = b = 3.859$ Å, $c = 3.7088$ Å for FePt^{45,46} and $a = b = 3.8564$ Å, $c = 3.7400$ Å for FePd. As shown by Laughlin *et al.*⁴⁴, this conventional unit cell can be further reduced, so the primitive cell is a base-centered tetragonal cell⁴⁴, where the in-plane lattice constant is $a_t = a/\sqrt{2}$, Fe takes (0,0,0) and Pt/Pd takes ($\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$). All our calculations use this primitive cell. We should point out an error in a prior publication by Ke⁴⁷, where the primitive cell is mischaracterized as a body-centered tetragonal lattice.

We use a dense $\mathbf{k}$ mesh of $25 \times 25 \times 18$, and set the quantization axis along the z axis. The spin moment for FePt/FePd is 3.26780/3.31417 μ_B , which agree with those prior studies^{48–51} under the same condition. We carry out two separate calculations with and without spin-orbit coupling (SOC) for the same $\mathbf{k}$ mesh and same functional. Then we compute the total energy difference between these two cases, $\Delta E(\text{FePt}) = E_{\text{soc}} - E_{\text{nosoc}} = -1.23$ eV, while $\Delta E(\text{FePd}) = E_{\text{soc}} - E_{\text{nosoc}} = -0.136$ eV. This shows that the 5d-Pt has a much stronger SOC than the 4d-Pd. This has an important consequence as seen below. Figure 2(a) is our band structure of FePt along seven high symmetry lines. Consistent with the prior study⁴⁷, FePt features multiple bands crossing the Fermi level (at 0 eV), which opens many channels for laser excitation. Figure 2(b) is the density of states (DOS) $\sigma^{(0)}$ in the ground state^{48,52}, where occupied states are between the Fermi energy (set at 0 eV) and -5 eV, and are integrated to 18 electrons, since Fe has $3d^6 4s^2$ and Pt has $5d^9 6s^1$ valence electrons. Note that the DOS for the spin minority states is plotted on the negative axis. The band

structure of FePd (Fig. 2(c)) is remarkably similar to that of FePt between -10 and 5 eV (see Fig. 2(a)). The major difference is that FePd starts at -8 eV, while FePt at -10 eV. As a result, their DOS is also similar (compare Figs. 2(b) and (d)). These similarities represent a stringent test for our theory.

III. NONLINEAR OPTICAL QUANTUM THEORY FOR SPIN RESPONSE

Charge dynamics in a material is different from spin dynamics⁵³ because their respective operators are different. Charge dynamics is characterized by the electric polarization $\mathbf{P}$, a polar vector, defined as $\mathbf{P} = \frac{1}{V}\text{Tr}[\rho\mathbf{D}]$, where Tr is a trace, $\mathbf{D}$ is the dipole moment, V is the volume of the sample, and ρ is the density matrix. Spin dynamics is characterized by the magnetization $\mathbf{M}$, an axial vector^{54,55} defined as $\mathbf{M} = \frac{1}{V}\text{Tr}[\rho\mathbf{S}]$, where $\mathbf{S}$ is the spin. FePt and FePd are centrosymmetric, and have inversion symmetry $\mathcal{I}$. Under the inversion symmetry, $\mathbf{D} \rightarrow -\mathbf{D}$ but $\mathbf{S} \rightarrow \mathbf{S}$. Figure 1(b) shows this difference. ρ does not have a simple expression under $\mathcal{I}$, but its n th order¹³ $\rho^{(n)} \propto \mathbf{D}^n$, so $\mathbf{P} \propto \mathbf{D}^{n+1}$. It is helpful to examine a few lower orders. If $n = 1$, under $\mathcal{I}$, $\mathcal{I}\mathbf{P}^{(1)} = \mathbf{P}^{(1)}$, so $\mathbf{P}^{(1)} \neq 0$, but $\mathcal{I}\mathbf{M}^{(1)} = -\mathbf{M}^{(1)}$, so $\mathbf{M}^{(1)} = 0$. If $n = 2$, $\mathcal{I}\mathbf{P}^{(2)} = -\mathbf{P}^{(2)}$, so $\mathbf{P}^{(2)} = 0$, but $\mathcal{I}\mathbf{M}^{(2)} = \mathbf{M}^{(2)}$, so $\mathbf{M}^{(2)} \neq 0$. Therefore, $\mathbf{M}^{(2)}$ is the lowest possible magnetization for a centrosymmetric system, consistent with our prior numerical results⁵⁴ but proved here in a much simpler way than done before⁵⁶. This underlines a fundamental difference between the traditional nonlinear optical and magnetic responses, and will guide us through all the following presentations.

A. Second-order density matrix

We employ the standard perturbation theory and expand the density matrix $\rho = \rho^{(0)} + \rho^{(1)} + \rho^{(2)} + \dots$, and then get a hierarchy of equations for each order n . The n th order $\rho^{(n)}$ depends on the $(n-1)$ th order density matrix. We start with the first-order time-dependent Liouville equation⁵⁷

$$i\hbar\dot{\rho}^{(1)} = [H_0, \rho^{(1)}] + [H_I^{(a)}, \rho^{(0)}], \quad (2)$$

where $\rho^{(0)}$ and $\rho^{(1)}$ are the zeroth- and first-order density operators, respectively, and the dot over $\rho^{(1)}$ is the time derivative. Here H_0 is the unperturbed Hamiltonian and includes the effect of the exchange functionals. The interaction Hamiltonian is $H_I^{(a)} = \frac{e\mathbf{p}\cdot\mathbf{A}^{(a)}(t)}{2m_e}$, where

$\mathbf{p}$ is the electron momentum and m_e is the electron's mass. We choose a cw field with the vector potential being $\mathbf{A}^{(a)}(t) = \mathbf{A}_0^{(a)}(e^{i\omega_a t} + e^{-i\omega_a t})/2$, where ω_a is the carrier frequency of the laser field a and $\mathbf{A}_0^{(a)}$ is the field amplitude. The light fluence is $F^{(a)} = \frac{1}{2}n\epsilon_0 c(A_0^{(a)}\omega_a)^2 T = \pi n c \epsilon_0 (A_0^{(a)})^2 \omega_a$, where T is the laser period, n is the index of refraction, and c is the speed of light. Since our materials are crystallines, all the physical observables are labeled by crystal momentum $\mathbf{k}$, but for brevity, we hide it from all the quantities below.

Introducing two band states $|n\rangle$ and $|m\rangle$ of H_0 allows us to cast the density operator in Eq. 2 into a matrix form. Then we integrate over time to find $\rho^{(1)}$ as

$$\rho^{(1)}(n, m) = \frac{e\mathbf{A}_0^{(a)} \cdot \mathbf{p}(n, m)}{2\hbar m_e} (\rho^{(0)}(n) - \rho^{(0)}(m)) \left(\frac{e^{-i\omega_a t}}{\omega_{nm} - \omega_a - i\Gamma_{nm}} + \frac{e^{i\omega_a t}}{\omega_{nm} + \omega_a - i\Gamma_{nm}} \right), \quad (3)$$

where $\rho^{(0)}(n)$ is a shorthand notation of $\rho^{(0)}(n, n)$, $\omega_{nm} = (E_n - E_m)/\hbar$, E_n is the band energy, Γ_{nm} is a lifetime broadening, and $\mathbf{p}(n, m)$ is the momentum matrix element between bands n and m . Different from the traditional nonlinear optics treatment²⁵, Eq. 3 includes both the resonant and off-resonant terms (the first and second terms in Eq. 3), so $\rho^{(1)}(n, m)$ is Hermitian, i.e., $\rho^{(1)}(n, m) = \rho^{(1)*}(m, n)$, and can be used to compute the spin moment change $m^{(1)} = \text{Tr}(\rho^{(1)} S_z) = \sum_{n,m} (\rho^{(1)}(n, m) S_z(m, n))$, where $S_z(n, m)$ is the spin matrix. Equation 3 further reveals that if $n = m$, $\rho^{(1)}(n, n) = 0$, then only the off-diagonal first-order density matrix elements are nonzero. To have a nonzero $m^{(1)}$, $S_z(m, n)$ must have off-diagonal elements, i.e., the spin symmetry broken. We note in passing that since our calculation always contains the spin-orbit coupling, density matrix elements are represented in the spin-mixed states, where the spin index cannot be used any longer.

With the first-order density matrix in hand, we are ready to work out the second-order density matrix. We introduce a second light field with the vector potential $\mathbf{A}^{(b)}(t) = \mathbf{A}^{(b)}(e^{i\omega_b t} + e^{-i\omega_b t})/2$, where ω_b is its carrier frequency. The second-order $\rho^{(2)}$ obeys the second-order time-dependent Liouville equation

$$i\hbar\dot{\rho}^{(2)} = [H_0, \rho^{(2)}] + [H_I^{(b)}, \rho^{(1)}]. \quad (4)$$

We multiply both sides by band $\langle n|$ from the left and band $|m\rangle$ from the right, and use the completeness relation⁵⁸ to find

$$\begin{aligned}
i\hbar\dot{\rho}^{(2)}(n, m) &= \langle n|[H_0, \rho^{(2)}]|m\rangle + \langle n|[H_I^{(b)}, \rho^{(1)}]|m\rangle \\
&= (E_n - E_m)\rho^{(2)}(n, m) + \sum_l \langle n|H_I^{(b)}|l\rangle \langle l|\rho^{(1)}|m\rangle - \langle n|\rho^{(1)}|l\rangle \langle l|H_I^{(b)}|m\rangle \\
&= (E_n - E_m)\rho^{(2)}(n, m) + \sum_l H_I^{(b)}(n, l)\rho^{(1)}(l, m) - \rho^{(1)}(n, l)H_I^{(b)}(l, m). \quad (5)
\end{aligned}$$

A lengthy but straightforward calculation (see Appendix A) yields the second-order density matrix $\rho_{ab}^{(2)}(n, m)$ between two band states $|n\rangle$ and $|m\rangle$,

$$\begin{aligned}
\rho_{ab}^{(2)}(n, m) &= \frac{e^2}{4m_e^2\hbar^2} \times \\
&\sum_l \left[\overbrace{\mathcal{Q}_l(n, m, \omega_a, \omega_b) + \mathcal{Q}_l^*(m, n, \omega_a, \omega_b)}^{\text{SFG}_1} + \overbrace{\mathcal{Q}_l(n, m, -\omega_a, -\omega_b) + \mathcal{Q}_l^*(m, n, -\omega_a, -\omega_b)}^{\text{SFG}_2} \right. \\
&\quad \left. + \underbrace{\mathcal{Q}_l(n, m, \omega_a, -\omega_b) + \mathcal{Q}_l^*(m, n, \omega_a, -\omega_b)}_{\text{DFG}_1} + \underbrace{\mathcal{Q}_l(n, m, -\omega_a, \omega_b) + \mathcal{Q}_l^*(m, n, -\omega_a, \omega_b)}_{\text{DFG}_2} \right] \quad (6)
\end{aligned}$$

where the first four terms are from the sum frequency generation (SFG), and the last four terms are from the difference frequency generation (DFG), e is the elementary charge, $\hbar$ is the reduced Planck constant, and the summation over the crystal momentum $\mathbf{k}$ is implied. These frequency generation terms are identified through the frequency variables, i.e., ω_a and ω_b in $\mathcal{Q}_l$. If ω_a and ω_b have the same sign, we have a sum frequency generation; otherwise a difference frequency generation. There are four possible combinations of $\pm\omega_a \pm \omega_b$. $\mathcal{Q}_l$ itself is given by,

$$\mathcal{Q}_l(n, m, \omega_a, \omega_b) = \frac{[\mathbf{A}^{(a)} \cdot \mathbf{p}(n, l)][\mathbf{A}^{(b)} \cdot \mathbf{p}(l, m)][\rho^{(0)}(n) - \rho^{(0)}(l)]e^{i(\omega_a + \omega_b)t}}{(\omega_{nm} + \omega_a + \omega_b - i\Gamma_{nm})(\omega_{nl} + \omega_a - i\Gamma_{nl})}, \quad (7)$$

where $\mathbf{p}(n, l) = \langle n|\hat{\mathbf{p}}|l\rangle$ is the momentum matrix element between bands $|n\rangle$ and $|l\rangle$, $\omega_{nm} = (E_n - E_m)/\hbar$, E_n is the band energy, $\rho^{(0)}(l)$ is the ground-state occupation of band l , and t is the time. All Γ 's are broadening, and they represent the disorder in the sample, electron-phonon scattering and other scattering processes that are not explicitly treated in our theory. If we compare Eq. 6 with Eq. 3.6.7 on page 135 of²⁵, we see that his equation only contains $\mathcal{Q}_l(n, m, -\omega_a, -\omega_b)$ and $\mathcal{Q}_l^*(m, n, \omega_a, \omega_b)$. One can easily verify that $\rho_{ab}^{(2)}(n, m)$ with only $\mathcal{Q}_l(n, m, -\omega_a, -\omega_b)$ and

$\mathcal{Q}_l^*(m, n, \omega_a, \omega_b)$ is not even Hermitian, so $\rho_{ab}^{(2)}(n, m)$ cannot be used to compute the spin moment. By contrast, our $\rho_{ab}^{(2)}(n, m)$ is Hermitian, which ensures the second-order spin moment $m_{ab}^{(2)} = \text{Tr}[\rho_{ab}^{(2)} S_z]$ to be real. So our theory represents a fundamental departure from the traditional NLO theory. Since the third order is zero, i. e., $m^{(3)} = 0$, the truncation at $m^{(2)}$ is a good start and is adequate when the vector potential is below 0.01 Vfs/Å as seen in our numerical calculation below (Fig. 5(f)). On the other hand, the fourth order $m^{(4)}$ is analytically difficult to compute since it contains 128 terms. Microscopically, Eq. 7 represents a process that the magnet is kept irradiated by a continuous light wave, so the magnetization of the magnet can be affected. This is the essence of our theory.

B. Second-order density of states

The second-order density matrix has been extensively studied in conjugated polymers¹³, but in solids, it remains largely unexplored, partly because the focus has often been on the second-order susceptibility. A quick inspection of the diagonal elements in Eq. 6 reveals that in contrast to its first-order counterpart ($\rho^{(1)}(n, n) = 0$ in Eq. 3), it is nonzero, i.e., $\rho^{(2)}(n, n) \neq 0$, and obeys the sum rule, $\sum_n \rho^{(2)}(n, n) = 0$. To see what it entails, we disperse it into the energy domain and introduce the second-order density of states as

$$\sigma_{ab}^{(2)}(E; \omega_p, \omega_q) = \sum_n \frac{\rho_{ab}^{(2)}(n, n)}{E_n - E + i\gamma}, \quad (8)$$

which is not a simple second-order derivative of the ground-state DOS. Rather, it represents how electrons are excited out of the Fermi sea under two light fields. For this reason, it depends on both the excited-state property of a material, and the laser parameters such as laser polarization, photon energy and vector potential amplitude. This is fundamentally different from the ground-state DOS.

We consider that two light fields a and b are both polarized along the x axis, i.e., collinear configuration, and have the same photon energy $h\nu$. We fix the incident fluence at $F = 10$ mJ/cm², typically used in experiments⁵⁹. Since the vector potential A_0 is inversely proportional to $\hbar\omega$, at $\hbar\omega = 1.6$ eV we have $A_0 = 0.222$ Vfs/Å. The thin solid line in Fig. 3(a) is our $\sigma_{xx}^{(2)}(E)$ in FePt. Different from the regular DOS (see Figs. 2(b) and (d)), for $E < E_F$, $\sigma_{xx}^{(2)}(E)$ is negative, but becomes positive if $E > E_F$. This stems from the population difference $\rho^{(0)}(n) - \rho^{(0)}(l)$ in Eq. 7, where the Pauli exclusion principle is realized through the permutation relation in Eq. 4. The

population difference differs from zero only when one band is occupied or partially occupied and the other band is unoccupied or partially unoccupied. This makes sense physically as during excitation the valence bands only lose electrons and the conduction bands only receive electrons. We find the trough at -0.64 eV and the peak at 0.72 eV, which are intrinsic to FePt, nearly independent of $h\nu$. This shows that those states close to the Fermi surface make significant contributions to laser excitation in terms of intraband transitions⁶⁰. Because of the sum rule, the positive and the negative $\sigma_{xx}^{(2)}$ cancel to zero. Increasing $h\nu$ to 2.0 eV reduces the amplitude of $\sigma_{xx}^{(2)}(E)$ because the vector potential is inversely proportional to $h\nu$ (see the thick dashed line in Fig. 3(a)). Figure 3(c) shows that in FePd the trough is moved to -1.00 eV and the peak is moved to 0.75 eV, also independent of $h\nu$. If we compare the amplitudes of $\sigma_{xx}^{(2)}(E)$ between FePt and FePd, we notice that FePt has a larger amplitude, because FePt has stronger momentum transition matrix elements.

Using the cross-polarized light fields, one along the x -axis and the other along the y -axis, alters the picture completely. The dashed lines in Figs. 3(a) and 3(c) are $\sigma_{xy}^{(2)}$ for FePt and FePd, respectively. They are significantly smaller than $\sigma_{xx}^{(2)}$. This feature is generic, as we find same results in other materials. This difference can be traced back to Eq. 7. Under cross-polarization excitation, microscopically it is the off-diagonal matrix elements $\mathbf{p}_x(n, l)\mathbf{p}_y(l, n)$ that contribute to $\sigma_{xy}^{(2)}$. In contrast to $\mathbf{p}_x(n, l)\mathbf{p}_x(l, n)$ which is always positive, $\mathbf{p}_x(n, l)\mathbf{p}_y(l, n)$ can change signs. This explains why $\sigma_{xy}^{(2)}$ can be positive or negative in different energy regimes. However, one thing remains: the peak and trough positions are also independent of $h\nu$.

C. Light-induced spin moment change

It is a known experimental fact that under a weak laser excitation, the spin moment reduction is linearly proportional to the incident laser fluence. We can first check whether our theory is able to reproduce this. As mentioned above, our light-induced second-order spin moment is defined as

$$m_{ab}^{(2)} = \text{Tr}[\rho_{ab}^{(2)} S_z], \quad (9)$$

where $\rho_{ab}^{(2)}$ is from Eq. 6, S_z is the spin matrix in the spin-mixed states, not diagonal in general, and a and b denote the light polarizations. We see immediately that since $\rho_{ab}^{(2)}$ in Eq. 7 is proportional to $\mathbf{A}^{(a)}\mathbf{A}^{(b)}$, the induced spin moment is proportional to the fluence. Now here comes to the most difficult question: How can one be sure that $\rho^{(2)}$ delivers a negative $m^{(2)}$ to have demagnetization?

Under the cw approximation, $\rho_{ab}^{(2)}$ in Eqs. 6 and 7 is time dependent, as is $m_{ab}^{(2)}$. To be definitive, we choose a time instant at $t = 0$. Figure 3(b) has six curves for FePt, but only three are visible with a good reason. The filled circles, partially hidden behind the empty circles, denote $m_{xx}^{(2)}$ which is negative and real across the entire region of $h\nu$, also true for FePd (see Fig. 3(d)). The reason for a negative $m^{(2)}$ lies in how $\rho^{(2)}$ changes across the Fermi energy. As shown in Figs. 3(a) and 3(b), during excitation, the spin majority states lose electrons and the spin minority states gain electrons, so one has a negative spin moment, a net angular momentum loss. This demonstrates for the first time that our nonlinear optical theory is capable of describing the demagnetization. Using a denser $\mathbf{k}$ mesh of $34 \times 34 \times 25$ yields nearly identical results (the empty circles), which overlap with the filled circles obtained at $25 \times 25 \times 18$. This shows that our results are well converged. We also employ the LDA functional instead of GGA used above. The thin star and empty box lines are $m_{xx}^{(2)}(\text{LDA})$ and $m_{xy}^{(2)}(\text{LDA})$. They again overlap with the above data strongly. This shows that different functionals do not produce a significant change. Across the same energy regime investigated, FePd has a weaker $m_{xx}^{(2)}$ of around $-0.02 \mu_B$ than FePt of around $-0.1 \mu_B$.

Changing the light polarization affects $m^{(2)}$ significantly. In FePt, when $h\nu$ is above 0.7 eV, $m_{xx}^{(2)}$ is larger than $m_{xy}^{(2)}$ (empty boxes). Just as in the inverse Faraday effect, different from $m_{xx}^{(2)}$, $m_{xy}^{(2)}$ is helicity-dependent, $m_{xy}^{(2)} = -m_{yx}^{(2)}$. In the figure, we choose a negative $m_{xy}^{(2)}$, so we can compare it with $m_{xx}^{(2)}$ easily. Quantitatively, at $h\nu = 1.6$ eV, $m_{xx}^{(2)}$ reaches $-0.09344 \mu_B$, 13.5 times larger than $m_{xy}^{(2)}$ of $-0.0069 \mu_B$. But below 0.7 eV, $m_{xy}^{(2)}$ is stronger. This shows that in the THz regime, the cross-polarization is equally effective to the spin change. In FePd, the crossing point where $m_{xy}^{(2)}$ is larger than $m_{xx}^{(2)}$ is at 1.3 eV. This is consistent with the differences seen in $\sigma^{(2)}$ in Figs. 3(a) and 3(c). To this end, we use the broadening $\Gamma = 0.05$ Ry. When we reduce it to 0.03 Ry (empty diamonds in Fig. 3(c)), $m_{xx}^{(2)}$ negatively increases as expected. Our theory can be applied to other magnets as well. Figures 3(e) and (f) show $m_{xx}^{(2)}$ and $m_{xy}^{(2)}$ for bcc Fe and fcc Ni, respectively. The trend is very interesting. Fe has a stronger response than Ni, in both $m_{xx}^{(2)}$ and $m_{xy}^{(2)}$, consistent with their native spin moments. What is different from FePt and FePd is that the cross-polarized spin moment $m_{xy}^{(2)}$ is in general stronger than collinearly-polarized spin moment $m_{xx}^{(2)}$. This reflects that $m^{(2)}$ is very sensitive to the intrinsic material properties.

D. Frequency-generation-resolved partial spin moment

To understand what and how optical processes underline the spin change, we compute the partial second-order spin moment,

$$m_{\text{DFG/SFG}}^{(2)} = \text{Tr}(\rho_{\text{DFG/SFG}}^{(2)} S_z), \quad (10)$$

where $\rho^{(2)}$ is from Eq. 6 and is now split into eight time-dependent terms. They form four groups: two subgroups of sum frequency generation (SFG₁ and SFG₂) and another two subgroups of difference frequency generation (DFG₁ and DFG₂). Each subgroup in Eq. 6 contains two terms. Using one term leads to a complex $m_{\text{DFG/SFG}}^{(2)}$, and the sum of both terms results in a real $m_{\text{DFG/SFG}}^{(2)}$. This highlights the importance of Hermitian in the density matrix. We take $\hbar\omega_a = \hbar\omega_b = 1.6$ eV as an example. Figure 4(a) shows all SFG terms have very small $m_{xx}^{(2)}$ and beat with time t at the frequency of 2ω because of their large denominator and the phase factor. By contrast, DFG₁ and DFG₂ are much larger and do not oscillate with time because the phase factor is zero. Their partial $m^{(2)}$ are not the same. When we sum them up, we obtain the total $m^{(2)}$ as shown in Fig. 4(b). The solid line is the result of FePt, where the total $m^{(2)}$ oscillates around a negative value. FePd has a smaller negative $m^{(2)}$ (dotted line). There is no other major change. Therefore, even at a particular time instant, our theoretical result can be compared with the experiments with a finite pulse duration. To be sure that our theory does not only apply to FePt and FePd, we also compute $m^{(2)}$ for bcc Fe (dashed line) and fcc Ni (long-dashed line). A trend is found. Whenever its ground-state spin moment is smaller, $m^{(2)}$ in general is smaller. Table II lists $m_{\text{DFG}_1}^{(2)}$ and $m_{\text{DFG}_2}^{(2)}$ separately. One sees that $m_{\text{DFG}_1}^{(2)}$ and $m_{\text{DFG}_2}^{(2)}$ each are very large, but they differ by a sign. For FePd, $m_{\text{DFG}_1}^{(2)} = -0.30346\mu_B$, and $m_{\text{DFG}_2}^{(2)} = 0.28868\mu_B$. The net second-order spin moment is the competition between these two large numbers. This is true for each material that we investigated. In the table, we also include fcc Ni and bcc Fe. Physically, the DFG groups, with zero frequency $\omega_p - \omega_q = 0$, correspond to the optical rectification/shift current in nonlinear optics^{23,25}. The light-induced rectification manifests itself in the second-order spin moment.

IV. LASER-INDUCED ULTRAFAST DEMAGNETIZATION

Can all the predictions above be realized in real-time dynamic simulations? There have been several prior studies on FePt using time-dependent density functional theory (TDDFT)^{61,62}, but their demagnetization has been plagued by spurious rapid oscillations^{56,63}, which are absent from the experimental results³¹. We employ the time-dependent Liouville equation⁶⁰ which does not have this problem. We choose a laser pulse of 60-fs and 1.6-eV. The vector potential amplitude is $A_0 = 0.015$ Vfs/Å, corresponding to the fluence of 1.34 mJ/cm². In order to appropriately describe the collective excitation of conduction electrons in metals, we include the intraband transitions as described in our prior publication⁶⁰, where a bracket energy δ is used. Figure 5(a) shows that upon laser excitation, both FePt and FePd demagnetize quickly, where ΔM is the spin moment change $\Delta M = M(t) - M_0$, and M_0 is the initial spin moment. Quantitatively, Fig. 5(a) shows $\frac{\Delta M}{M_0}(\text{FePt}) = -26.8\%$ for FePt and $\frac{\Delta M}{M_0}(\text{FePd}) = -19.4\%$ for FePd. So for the same set of laser parameters, FePt demagnetizes more than FePd, by 1.38 times ($26.8/19.4$). A similar ratio was obtained experimentally. Iihama *et al.*³¹ used two comparable fluences 1.6 mJ/cm² for FePd and 1.4 mJ/cm² for FePt, and they found that FePt demagnetizes smoothly by 7.5% and FePd by 5% , or 1.5 times larger in FePt. This agreement is encouraging, given that there are many differences between the experiment and theory. To show that the demagnetization difference between FePt and FePd is intrinsic to the materials themselves, not related to a particular set of laser parameters. When we increase the photon energy to 2.0 eV, we find that FePt still demagnetizes more than FePd (see Fig. 5(b)). This remains true regardless of whether we increase the laser pulse duration to 120 fs (Fig. 5(c)) or increase the vector potential amplitude to 0.03 Vfs/Å (Fig. 5(d)). Quantitatively, our percentage differs from the experimental one since in our current study, we do not purposely tune our bracket energy δ ⁶⁰ to match the experimental one, as our goal here is to verify our above analytical theory. Figure 5(e) shows that the amount of demagnetization increases quickly with δ . To match the experimental one, we only use $\delta = 0.6$ eV. All the above results are obtained with $\delta = 1$ eV. Second, experimentally it is difficult to have the same efficiency as the theory, because many factors such as the sample surface reflection and surface roughness may lead to a smaller demagnetization. Figure 5(f) is the dependence of the demagnetization on the laser vector potential.

Finally, we wish to investigate the photon-energy dependence of ultrafast demagnetization, though we have provided the dependence for the above analytical results in Fig. 3(a). Figure 6(a) shows that for the fixed fluence and duration, the amount of demagnetization weakly depends on the photon energy. The reason why we have to fix the fluence and laser duration is because the vector potential A_0 depends on both the laser fluence and duration. Another interesting topic is the effects of the exchange-correlation functional on demagnetization. Figure 6(b) shows that the LDA functional leads to a stronger demagnetization, partly because the electronic states under the LDA functional have stronger transition matrix elements, as it builds upon the free electron gas model. We also investigate whether the sample orientation matters to demagnetization. In this case, we choose two directions, one along the [111] direction and the other along the [101] direction. We apply linearly polarized laser pulses along those two directions. Figure 6(c) shows their orientation dependence is rather weak, because FePt, although having a $L1_0$ structure, is still quite symmetric spatially. Our numerical studies prove that our nonlinear optical quantum theory for spin change agrees with our real-time demagnetization simulation.

V. CONCLUSION

We have developed the first nonlinear optical quantum theory of demagnetization. We start from the symmetry analysis and find that for centrosymmetric systems, the second-order spin moment is the lowest order. Our theory has two features. First, different from nonlinear optical theory, all the terms, regardless of whether they are resonant or off-resonant, must be included to ensure the Hermitian of the density matrix. Specifically, we show that the difference frequency generations (DFG) dominate over the sum frequency generations (SFG). This is the manifestation of optical rectification in spin moment change. The competition between two DFG terms determines the net spin moment change. Second, it allows one to compute and compare light-induced spin moment changes among different magnetic materials at the first-principles level. We find that FePt demagnetizes more than FePd, even though their crystal and electronic structures are very similar. This is confirmed in our real-time simulation and the experiment³¹. We expect that our finding will motivate further experimental and theoretical studies in femtomagnetism.

ACKNOWLEDGMENTS

GPZ was partly supported by the U.S. Department of Energy under Contract No. DE-FG02-06ER46304. The work was carried out on Indiana State University's Quantum Cluster and High Performance computers.

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Appendix A: Derivation of Eq. 6

Here we provide additional details of our derivation of Eq. 6. To simplify our expression, we introduce $\rho^{(2)}(n, m) = e^{-i\omega_{nm}t}Q(n, m)$, where $\omega_{nm} = (E_n - E_m)/\hbar$, and substitute it into Eq. 5 to find

$$\begin{aligned} & i\hbar(-i\omega_{nm}e^{-i\omega_{nm}t}Q(n, m) + e^{-i\omega_{nm}t}\dot{Q}(n, m)) \\ &= (E_n - E_m)e^{-i\omega_{nm}t}Q(n, m) + \sum_l [H_I^{(b)}(n, l)\rho^{(1)}(l, m) - \rho^{(1)}(n, l)H_I^{(b)}(l, m)] \end{aligned} \quad (\text{A1})$$

$$i\hbar e^{-i\omega_{nm}t}\dot{Q}(n, m) = \sum_l [H_I^{(b)}(n, l)\rho^{(1)}(l, m) - \rho^{(1)}(n, l)H_I^{(b)}(l, m)] \quad (\text{A2})$$

$$\dot{Q}(n, m) = \frac{1}{i\hbar}e^{i\omega_{nm}t} \sum_l [H_I^{(b)}(n, l)\rho^{(1)}(l, m) - \rho^{(1)}(n, l)H_I^{(b)}(l, m)], \quad (\text{A3})$$

where $H_I^{(b)}(n, l) = \frac{e}{2m_e}\mathbf{p}(n, l) \cdot \mathbf{A}_0^{(b)}(e^{i\omega_b t} + e^{-i\omega_b t})$ and $H_I^{(b)}(l, m) = \frac{e}{2m_e}\mathbf{p}(l, m) \cdot \mathbf{A}_0^{(b)}(e^{i\omega_b t} + e^{-i\omega_b t})$.

Now we substitute Eq. 3 into the first term of Eq. A3 to obtain

$$\begin{aligned} & \frac{e^{i\omega_{nm}t}}{i\hbar}H_I^{(b)}(n, l)\rho^{(1)}(l, m) = \frac{e^{i\omega_{nm}t}}{i\hbar} \frac{e}{2m_e}\mathbf{p}(n, l) \cdot \mathbf{A}_0^{(b)}(e^{i\omega_b t} + e^{-i\omega_b t}) \\ & \times \frac{e\mathbf{A}_0^{(a)} \cdot \mathbf{p}(l, m)}{2\hbar m_e}(\rho^{(0)}(l) - \rho^{(0)}(m)) \left(\frac{e^{i\omega_a t}}{\omega_{lm} + \omega_a - i\Gamma_{lm}} + \frac{e^{-i\omega_a t}}{\omega_{lm} - \omega_a - i\Gamma_{lm}} \right) \end{aligned} \quad (\text{A4})$$

$$\begin{aligned} &= \frac{e^2[\mathbf{p}(n, l) \cdot \mathbf{A}_0^{(b)}][\mathbf{A}_0^{(a)} \cdot \mathbf{p}(l, m)][\rho^{(0)}(l) - \rho^{(0)}(m)]}{4i\hbar^2 m_e^2} \\ & \times e^{i\omega_{nm}t}(e^{i\omega_b t} + e^{-i\omega_b t}) \left(\frac{e^{i\omega_a t}}{\omega_{lm} + \omega_a - i\Gamma_{lm}} + \frac{e^{-i\omega_a t}}{\omega_{lm} - \omega_a - i\Gamma_{lm}} \right). \end{aligned} \quad (\text{A5})$$

We focus on the second line of Eq. A5 and we multiply them out to have

$$\begin{aligned} & (e^{i(\omega_b + \omega_{nm})t} + e^{-i(\omega_b - \omega_{nm})t}) \left(\frac{e^{i\omega_a t}}{\omega_{lm} + \omega_a - i\Gamma_{lm}} + \frac{e^{-i\omega_a t}}{\omega_{lm} - \omega_a - i\Gamma_{lm}} \right) \\ &= \frac{e^{i(\omega_a + \omega_b + \omega_{nm})t}}{\omega_{lm} + \omega_a - i\Gamma_{lm}} + \frac{e^{-i(\omega_a + \omega_b - \omega_{nm})t}}{\omega_{lm} - \omega_a - i\Gamma_{lm}} + \frac{e^{i(-\omega_a + \omega_b + \omega_{nm})t}}{\omega_{lm} - \omega_a - i\Gamma_{lm}} + \frac{e^{i(\omega_a - \omega_b + \omega_{nm})t}}{\omega_{lm} + \omega_a - i\Gamma_{lm}}. \end{aligned} \quad (\text{A6})$$

Next, we integrate each term from $-\infty$ to t . We take the first term in Eq. A6 as an example

$$\int_{t'=-\infty}^{t'=t} \frac{e^{i(\omega_a+\omega_b+\omega_{nm})t'}}{\omega_{lm} + \omega_a - i\Gamma_{lm}} = \frac{e^{i(\omega_a+\omega_b+\omega_{nm})t}}{i(\omega_{nm} + \omega_a + \omega_b - i\Gamma_{nm})(\omega_{lm} + \omega_a - i\Gamma_{lm})}, \quad (\text{A7})$$

where we have introduced the decaying factor $e^{\Gamma_{nm}t}$ ($\Gamma_{nm} > 0$) so the integral at $t' = -\infty$ is zero. The remaining terms are obtained by changing (ω_a, ω_b) to $(-\omega_a, -\omega_b)$, $(-\omega_a, \omega_b)$, and $(\omega_a, -\omega_b)$, respectively, so the first term in $Q(n, m)$ in Eq. A3 is

$$\begin{aligned} & \sum_l \frac{e^2 [\mathbf{p}(n, l) \cdot \mathbf{A}_0^{(b)}][\mathbf{A}_0^{(a)} \cdot \mathbf{p}(l, m)][\rho^{(0)}(l) - \rho^{(0)}(m)]}{4i\hbar^2 m_e^2} \\ & \times \left(\frac{e^{i(\omega_a+\omega_b+\omega_{nm})t}}{i(\omega_{nm} + \omega_a + \omega_b - i\Gamma_{nm})(\omega_{lm} + \omega_a - i\Gamma_{lm})} + \frac{e^{-i(\omega_a+\omega_b-\omega_{nm})t}}{i(\omega_{nm} - \omega_a - \omega_b - i\Gamma_{nm})(\omega_{lm} - \omega_a - i\Gamma_{lm})} \right. \\ & \left. + \frac{e^{i(-\omega_a+\omega_b+\omega_{nm})t}}{i(\omega_{nm} - \omega_a + \omega_b - i\Gamma_{nm})(\omega_{lm} - \omega_a - i\Gamma_{lm})} + \frac{e^{i(\omega_a-\omega_b+\omega_{nm})t}}{i(\omega_{nm} + \omega_a - \omega_b - i\Gamma_{nm})(\omega_{lm} + \omega_a - i\Gamma_{lm})} \right) \\ & = -\frac{e^2}{4\hbar^2 m_e^2} \sum_l [\mathbf{p}(n, l) \cdot \mathbf{A}_0^{(b)}][\mathbf{A}_0^{(a)} \cdot \mathbf{p}(l, m)][\rho^{(0)}(l) - \rho^{(0)}(m)] \\ & \times \left(\frac{e^{i(\omega_a+\omega_b+\omega_{nm})t}}{(\omega_{nm} + \omega_a + \omega_b - i\Gamma_{nm})(\omega_{lm} + \omega_a - i\Gamma_{lm})} + \frac{e^{-i(\omega_a+\omega_b-\omega_{nm})t}}{(\omega_{nm} - \omega_a - \omega_b - i\Gamma_{nm})(\omega_{lm} - \omega_a - i\Gamma_{lm})} \right. \\ & \left. + \frac{e^{i(-\omega_a+\omega_b+\omega_{nm})t}}{(\omega_{nm} - \omega_a + \omega_b - i\Gamma_{nm})(\omega_{lm} - \omega_a - i\Gamma_{lm})} + \frac{e^{i(\omega_a-\omega_b+\omega_{nm})t}}{(\omega_{nm} + \omega_a - \omega_b - i\Gamma_{nm})(\omega_{lm} + \omega_a - i\Gamma_{lm})} \right) \end{aligned}$$

The second term in Q in Eq. A3 can be worked out similarly,

$$\begin{aligned} & -\frac{e^{i\omega_{nm}t}}{i\hbar} H_I(l, m) \rho^{(1)}(n, l) = -\frac{e^{i\omega_{nm}t}}{i\hbar} \frac{e}{2m_e} \mathbf{p}(l, m) \cdot \mathbf{A}_0^{(b)} (e^{i\omega_b t} + e^{-i\omega_b t}) \\ & \times \frac{e \mathbf{A}_0^{(a)} \cdot \mathbf{p}(n, l)}{2\hbar m_e} (\rho^{(0)}(n) - \rho^{(0)}(l)) \left(\frac{e^{i\omega_a t}}{\omega_{nl} + \omega_a - i\Gamma_{nl}} + \frac{e^{-i\omega_a t}}{\omega_{nl} - \omega_a - i\Gamma_{nl}} \right) \\ & = -\frac{e^2 [\mathbf{p}(l, m) \cdot \mathbf{A}_0^{(b)}][\mathbf{A}_0^{(a)} \cdot \mathbf{p}(n, l)][\rho^{(0)}(n) - \rho^{(0)}(l)]}{4i\hbar^2 m_e^2} \end{aligned} \quad (\text{A8})$$

$$\times e^{i\omega_{nm}t} (e^{i\omega_b t} + e^{-i\omega_b t}) \left(\frac{e^{i\omega_a t}}{\omega_{nl} + \omega_a - i\Gamma_{nl}} + \frac{e^{-i\omega_a t}}{\omega_{nl} - \omega_a - i\Gamma_{nl}} \right). \quad (\text{A9})$$

The second line of Eq. A9 also contains four terms as

$$\begin{aligned} & (e^{i(\omega_b+\omega_{nm})t} + e^{-i(\omega_b-\omega_{nm})t}) \left(\frac{e^{i\omega_a t}}{\omega_{nl} + \omega_a - i\Gamma_{nl}} + \frac{e^{-i\omega_a t}}{\omega_{nl} - \omega_a - i\Gamma_{nl}} \right) \\ & = \frac{e^{i(\omega_a+\omega_b+\omega_{nm})t}}{\omega_{nl} + \omega_a - i\Gamma_{nl}} + \frac{e^{-i(\omega_a+\omega_b-\omega_{nm})t}}{\omega_{nl} - \omega_a - i\Gamma_{nl}} + \frac{e^{i(-\omega_a+\omega_b+\omega_{nm})t}}{\omega_{nl} - \omega_a - i\Gamma_{nl}} + \frac{e^{i(\omega_a-\omega_b+\omega_{nm})t}}{\omega_{nl} + \omega_a - i\Gamma_{nl}}, \end{aligned} \quad (\text{A10})$$

whose respective time-integrals are

$$\left(\frac{e^{i(\omega_a + \omega_b + \omega_{nm})t}}{i(\omega_{nm} + \omega_a + \omega_b - i\Gamma_{nm})(\omega_{nl} + \omega_a - i\Gamma_{nl})} + \frac{e^{-i(\omega_a + \omega_b - \omega_{nm})t}}{i(\omega_{nm} - \omega_a - \omega_b - i\Gamma_{nm})(\omega_{nl} - \omega_a - i\Gamma_{nl})} \right. \\ \left. + \frac{e^{i(-\omega_a + \omega_b + \omega_{nm})t}}{i(\omega_{nm} - \omega_a + \omega_b - i\Gamma_{nm})(\omega_{nl} - \omega_a - i\Gamma_{nl})} + \frac{e^{i(\omega_a - \omega_b + \omega_{nm})t}}{i(\omega_{nm} + \omega_a - \omega_b - i\Gamma_{nm})(\omega_{nl} + \omega_a - i\Gamma_{nl})} \right).$$

We then multiply it by the coefficient $-\frac{e^2[\mathbf{p}(l, m) \cdot \mathbf{A}_0^{(b)}][\mathbf{A}_0^{(a)} \cdot \mathbf{p}(n, l)][\rho^{(0)}(n) - \rho^{(0)}(l)]}{4i\hbar^2 m_e^2}$ to find

$$\frac{e^2[\mathbf{p}(l, m) \cdot \mathbf{A}_0^{(b)}][\mathbf{A}_0^{(a)} \cdot \mathbf{p}(n, l)][\rho^{(0)}(n) - \rho^{(0)}(l)]}{4\hbar^2 m_e^2} \\ \times \left(\frac{e^{i(\omega_a + \omega_b + \omega_{nm})t}}{(\omega_{nm} + \omega_a + \omega_b - i\Gamma_{nm})(\omega_{nl} + \omega_a - i\Gamma_{nl})} + \frac{e^{-i(\omega_a + \omega_b - \omega_{nm})t}}{(\omega_{nm} - \omega_a - \omega_b - i\Gamma_{nm})(\omega_{nl} - \omega_a - i\Gamma_{nl})} \right. \\ \left. + \frac{e^{i(-\omega_a + \omega_b + \omega_{nm})t}}{(\omega_{nm} - \omega_a + \omega_b - i\Gamma_{nm})(\omega_{nl} - \omega_a - i\Gamma_{nl})} + \frac{e^{i(\omega_a - \omega_b + \omega_{nm})t}}{(\omega_{nm} + \omega_a - \omega_b - i\Gamma_{nm})(\omega_{nl} + \omega_a - i\Gamma_{nl})} \right).$$

Since $\rho^{(2)}(n, m) = e^{-i\omega_{nm}t}Q(n, m)$, all we need to do is to remove $e^{i\omega_{nm}t}$ from the above expressions to get

$$\rho^{(2)}(n, m) = \frac{e^2}{4\hbar^2 m_e^2} \sum_l [\mathbf{p}(n, l) \cdot \mathbf{A}_0^{(a)}][\mathbf{A}_0^{(b)} \cdot \mathbf{p}(l, m)][\rho^{(0)}(n) - \rho^{(0)}(l)] \\ \times \left(\underbrace{\frac{e^{i(\omega_a + \omega_b)t}}{(\omega_{nm} + \omega_a + \omega_b - i\Gamma_{nm})(\omega_{nl} + \omega_a - i\Gamma_{nl})}}_{\mathcal{Q}_l(n, m, \omega_a, \omega_b)} + \underbrace{\frac{e^{-i(\omega_a + \omega_b)t}}{(\omega_{nm} - \omega_a - \omega_b - i\Gamma_{nm})(\omega_{nl} - \omega_a - i\Gamma_{nl})}}_{\mathcal{Q}_l(n, m, \omega_a, \omega_b)} \right. \\ \left. + \underbrace{\frac{e^{i(-\omega_a + \omega_b)t}}{(\omega_{nm} - \omega_a + \omega_b - i\Gamma_{nm})(\omega_{nl} - \omega_a - i\Gamma_{nl})}}_{\mathcal{Q}_l(n, m, -\omega_a, \omega_b)} + \underbrace{\frac{e^{i(\omega_a - \omega_b)t}}{(\omega_{nm} + \omega_a - \omega_b - i\Gamma_{nm})(\omega_{nl} + \omega_a - i\Gamma_{nl})}}_{\mathcal{Q}_l(n, m, \omega_a, -\omega_b)} \right) \\ - \frac{e^2}{4\hbar^2 m_e^2} \sum_l [\mathbf{p}(n, l) \cdot \mathbf{A}_0^{(b)}][\mathbf{A}_0^{(a)} \cdot \mathbf{p}(l, m)][\rho^{(0)}(l) - \rho^{(0)}(m)] \\ \times \left(\underbrace{\frac{e^{i(\omega_a + \omega_b)t}}{(\omega_{nm} + \omega_a + \omega_b - i\Gamma_{nm})(\omega_{lm} + \omega_a - i\Gamma_{lm})}}_{\mathcal{Q}_l^*(m, n, -\omega_a, -\omega_b)} + \underbrace{\frac{e^{-i(\omega_a + \omega_b)t}}{(\omega_{nm} - \omega_a - \omega_b - i\Gamma_{nm})(\omega_{lm} - \omega_a - i\Gamma_{lm})}}_{\mathcal{Q}_l^*(m, n, \omega_a, \omega_b)} \right. \\ \left. + \underbrace{\frac{e^{i(-\omega_a + \omega_b)t}}{(\omega_{nm} - \omega_a + \omega_b - i\Gamma_{nm})(\omega_{lm} - \omega_a - i\Gamma_{lm})}}_{\mathcal{Q}_l^*(m, n, \omega_a, -\omega_b)} + \underbrace{\frac{e^{i(\omega_a - \omega_b)t}}{(\omega_{nm} + \omega_a - \omega_b - i\Gamma_{nm})(\omega_{lm} + \omega_a - i\Gamma_{lm})}}_{\mathcal{Q}_l^*(m, n, -\omega_a, \omega_b)} \right),$$

where we identify each term with $\mathcal{Q}_l$. Caution must be taken that the actual $\mathcal{Q}_l$ includes the

coefficients (see Eq. 7).

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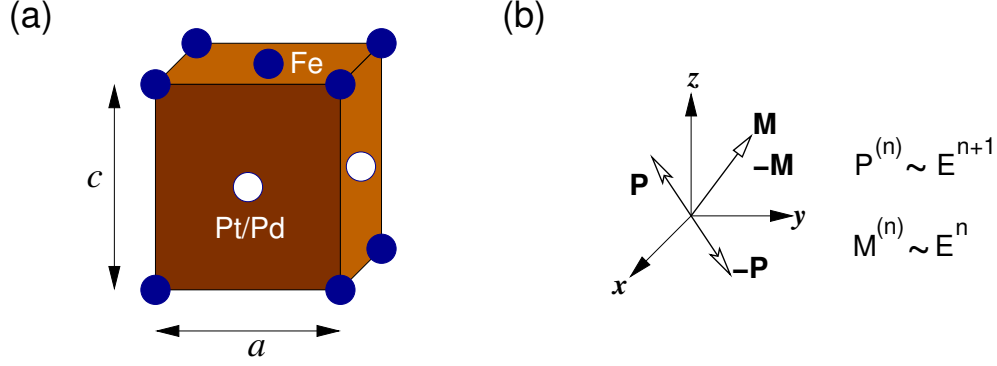


FIG. 1. (a) FePt/FePd crystal structures. The filled circles are Fe atoms, while the unfilled ones are Pt/Pd. (b) Symmetry difference between the polarization $\mathbf{P}$ and magnetization $\mathbf{M}$ determines how the n th-order polarization and magnetization depend on the external electric field $\mathbf{E}$ differently.

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TABLE I. Table of symbols used in this paper.

Symbol	Meaning
$\mathbf{P}, \mathbf{P}^{(n)}$	electric polarization, n th order
$\mathbf{D}$	dipole moment
$\rho, \rho^{(n)}$	density matrix, n th order
$\mathbf{M}, \mathbf{M}^{(n)}$	magnetization, n th order
$M(t), \Delta M$	time-dependent spin moment, its change
$\mathcal{I}$	inversion symmetry operator
H_0, H_I	system Hamiltonian, interaction Hamiltonian
$\mathbf{p}(n, m)$	momentum matrix element between bands n and m
$F^{(a)}$	fluence of light field a
$\mathbf{A}^{(a)}, \omega_a, \nu_a$	vector potential, angular frequency and frequency of field a
ω_{nm}	angular frequency difference between bands n and m
Γ_{nm}	lifetime broadening difference for bands n and m
$\mathcal{Q}_l$	shorthand notation for sum and difference frequency generations in Eq. 7
$\sigma_{ab}^{(2)}$	second-order density of states for fields a and b (Eq. 8)
$m_{ab}^{(2)}$	light-induced second-order spin moment fields a and b

TABLE II. Partial second-order spin moment $m_{\text{DFG}}^{(2)}$ due to the difference frequency generation in FePd, FePt, bcc Fe and fcc Ni. They are computed from $m_{\text{DFG}_1}^{(2)} = \text{Tr}[\rho_{\text{DFG}_1}^{(2)} S_z]$ and $m_{\text{DFG}_2}^{(2)} = \text{Tr}[\rho_{\text{DFG}_2}^{(2)} S_z]$, where $\rho_{\text{DFG}_1}^{(2)}$ and $\rho_{\text{DFG}_2}^{(2)}$ are the third and fourth terms in Eq. 6. $m_{xx}^{(2)}$ is the net second-order spin moment, $m_{xx}^{(2)} = m_{\text{DFG}_1}^{(2)} + m_{\text{DFG}_2}^{(2)}$. $m^{(0)}$ is the ground-state spin moment. $m_{xx}^{(2)}/m^{(0)}$ is the percentage change. All SFG terms are small, so are not included. Here the photon energy is $h\nu = 1.6$ eV. All the results are calculated at $t = 0$.

Material	$m_{\text{DFG}_2}^{(2)}(\mu_B)$	$m_{\text{DFG}_1}^{(2)}(\mu_B)$	$m_{xx}^{(2)}(\mu_B)$	$m^{(0)}(\mu_B)$	$m_{xx}^{(2)}/m^{(0)}(\%)$
FePd	-0.30346	0.28868	-0.01678	3.3142	-0.445
FePt	-0.3420	0.24968	-0.09232	3.2678	-2.825
bcc Fe	-0.14066	0.13682	-0.00384	2.1770	-0.176
fcc Ni	-0.05052	0.04842	-0.00210	0.6389	-0.328

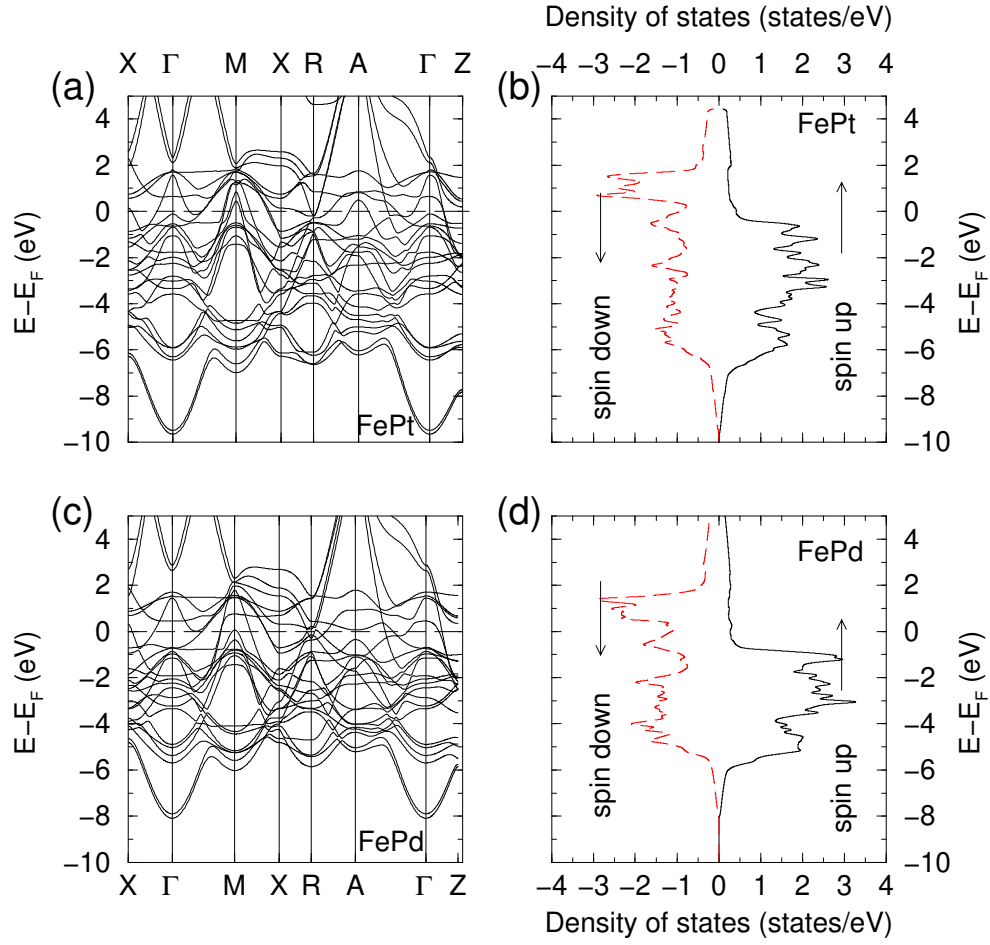


FIG. 2. (a) Band structure of FePt. (b) Density of states of the ground state in FePt, where the solid line denotes the spin majority states and the dashed line the spin minority states plotted on the negative axis. (c) Band structure of FePd. (d) Density of states of the ground state in FePd.

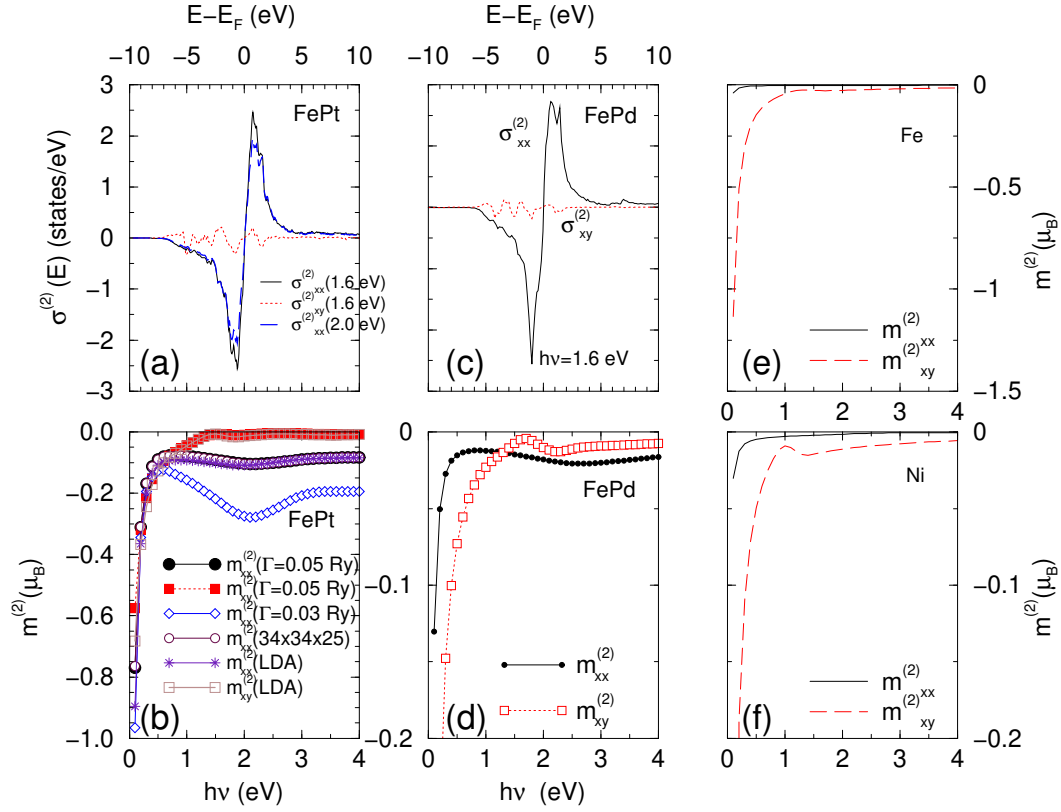


FIG. 3. (a) Second-order density of states in FePt. The solid line denotes $\sigma_{xx}^{(2)}$ with both laser polarizations along the x axis, while the dotted line denotes $\sigma_{xy}^{(2)}$ with cross-polarizations along the x and y axes. Here the photon energy is $h\nu_a = h\nu_b = 1.6$ eV. The long-dashed line is $\sigma_{xx}^{(2)}$ with $h\nu_a = h\nu_b = 2.0$ eV. (b) Second-order magnetic moment $m^{(2)}$ in FePt as a function of photon energy $h\nu$. The key feature is that except $m_{xx}^{(2)}$ with a different broadening of $\Gamma = 0.03$ Ry (empty diamonds), the convergences with the $\mathbf{k}$ points and functionals are reached within 1-3% as estimated from their overlaps. Here the filled circles denote $m_{xx}^{(2)}$ and the filled boxes denote $m_{xy}^{(2)}$ components, both with $\Gamma = 0.05$ Ry. The empty circles are $m_{xx}^{(2)}$ with a larger $\mathbf{k}$ mesh of $34 \times 34 \times 25$. The thin star line and empty boxes are $m_{xx}^{(2)}$ and $m_{xy}^{(2)}$ computed with LDA instead of GGA. All the calculations are done with one parameter changed, while the rest are fixed. (c) Same as (a) but for FePd. (d) Same as (c) but for FePd. (e) and (f) are $m_{xx}^{(2)}$ and $m_{xy}^{(2)}$ for bcc Fe and fcc Ni, respectively.

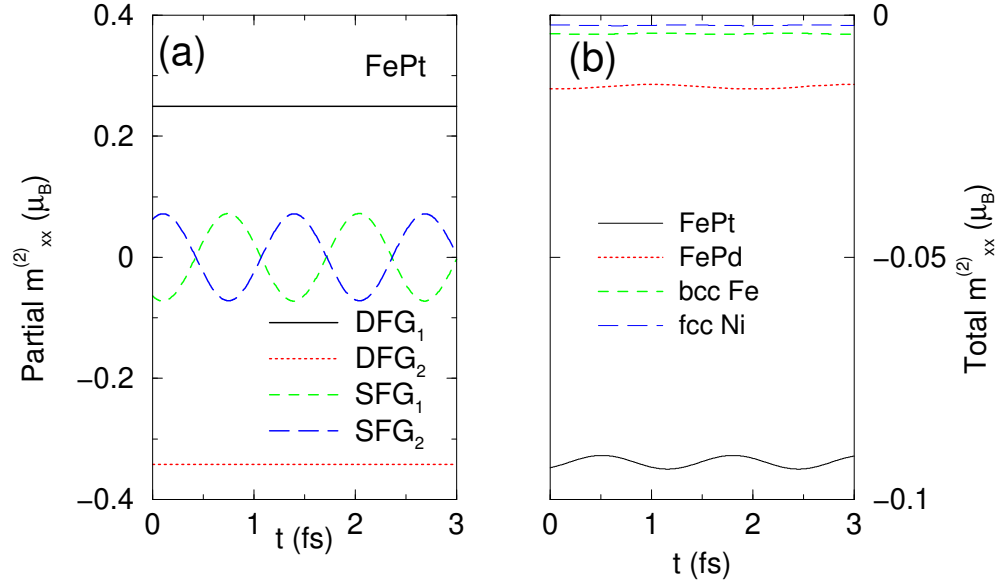


FIG. 4. (a) Contribution of the sum frequency [SFG₁ (dashed line), SFG₂ (long-dashed line)] and difference frequency generations [DFG₁ (solid line), DFG₂ (dotted line)] to $m^{(2)}$ as a function of time t . Here the photon energy is $h\nu_a = \hbar\nu_b = 1.6$ eV. (b) Total $m_{xx}^{(2)}$ as a function of time t for FePt (solid line), FePd (dotted line), bcc Fe (dashed line) and fcc Ni (long-dashed line).

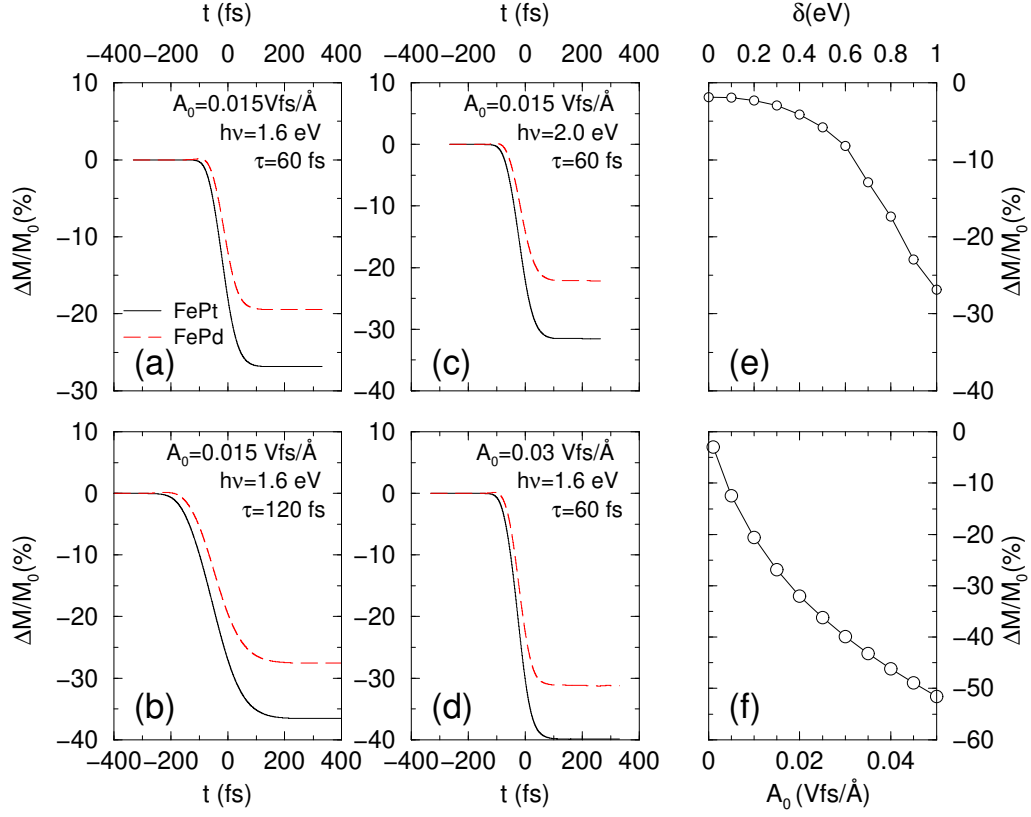


FIG. 5. Laser-parameter and intraband-bracket energy dependence of ultrafast demagnetization in FePt (black solid line) and FePd (red dashed line). (a) The laser photon energy is $h\nu = 1.6$ eV, vector field potential is $A_0 = 0.015$ Vfs/Å, and pulse duration $\tau = 60$ fs. (b) Same as (a) but with the pulse duration $\tau = 120$ fs. (c) Same as (a) but with $h\nu = 2.0$ eV. (d) A_0 is increased to 0.03 Vfs/Å. (e) The spin moment reduction in FePt as a function of the bracket energy δ which controls the contribution of the intraband transitions. (f) The spin moment reduction in FePt as a function of vector potential amplitude.

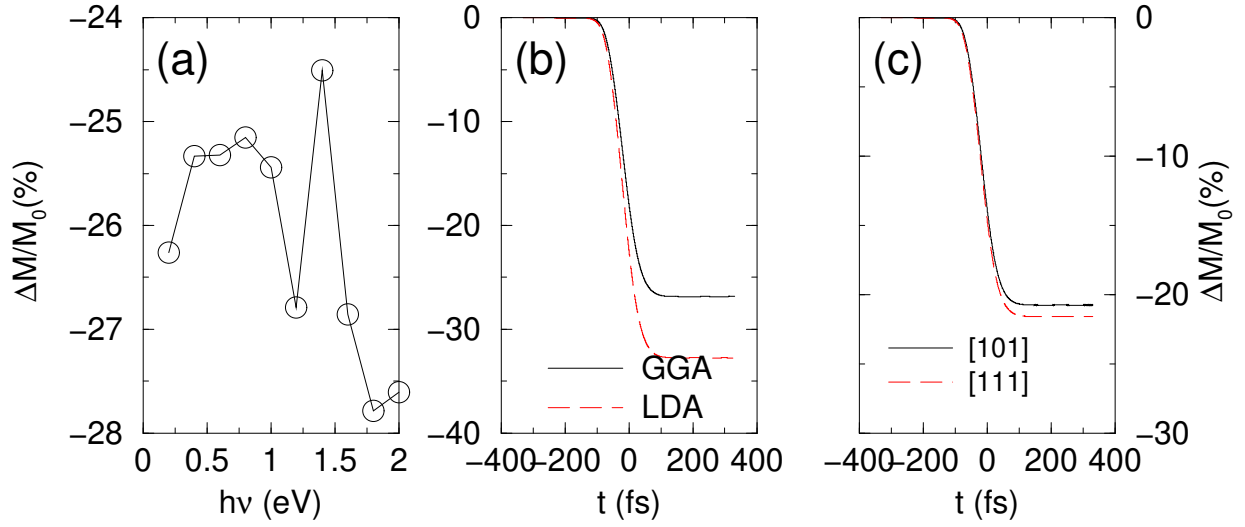


FIG. 6. (a) Photon energy dependence of demagnetization in FePt. Since our vector potential A_0 is photon-energy dependent, we fix the fluence at 1.34 mJ/cm^2 and pulse duration at $\tau = 60$ fs. (b) Comparison of the spin moment reduction between the GGA (solid line) and LDA results (long-dashed line), where $h\nu = 1.6 \text{ eV}$, $\tau = 60 \text{ fs}$ and $A_0 = 0.015 \text{ Vfs/\AA}$. Using the LDA functional produces a stronger demagnetization, but they agree within 10%. (c) Orientation dependence. The solid line denotes the result with the laser linear polarization along the [101] direction, and the long-dashed line is for the [111] direction. We find their difference is small.