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Resolving photon-shortage mystery in femtosecond magnetism

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Abstract

For nearly a decade, it has been a mystery why the small average number of photons absorbed per atom from an ultrashort laser pulse is able to induce a strong magnetization within a few hundred femtoseconds. Here we resolve this mystery by directly computing the number of photons per atom layer by layer as the light wave propagates inside the sample. We find that for all the 24 experiments considered here, each atom has more than one photon. The so-called photon shortage does not exist. By plotting the relative demagnetization change versus the number of photons absorbed per atom, we show that, depending on the experimental condition, 0.1 photon can induce about 4%–72% spin moment change. Our perturbation theory reveals that the demagnetization depends linearly on the amplitude of the laser field. In addition, we find that the transition frequency of a sample may also play a role in magnetization processes. As long as the intensity is not zero, the intensity of the laser field only affects the matching range of the transition frequencies, but not whether the demagnetization can happen or not.

1. Introduction

The pioneering discovery by Beaurepaire *et al* [1], that a femtosecond laser pulse can demagnetize Ni on a subpicosecond timescale or femtomagnetism, has inspired enormous scientific activities both experimentally and theoretically [2–7]. The significance of this discovery is that it demonstrates a possibility for nonthermal writing in a ferromagnetic medium. One prominent example is the inverse Faraday effect, where the laser can nonthermally switch spins [8]. This process works even better when the temperature is lowered [9]. Its potential applications require a good understanding of the underlying excitation mechanism. In spite of extensive investigations in this field [10, 11], how the light transfers the photon energy to the system and subsequently demagnetizes the sample is still puzzling though new theoretical and experimental investigations have emerged [7]. At the center of the debate is whether there are enough photons absorbed per atom (estimated at 0.01 [12]) for magnetic moment change [8, 12–21]. On the one hand, Koopmans *et al* argue that the excitation density is too low to induce any substantial change in magnetization [13–15]. If the number of photons is not enough in the first place, the excitation density must be very low. On the other hand, nearly all the

experiments report the magnetization change either directly or indirectly. A strong demagnetization is incompatible with a shortage of photons. This puzzle affects our confidence in femtomagnetism [22, 23]. Therefore, resolving this apparent contradiction is of paramount importance to femtomagnetism and its future applications.

To this end, there are very few detailed investigations. Koopmans *et al* [15, 24] stated that an effective photon number for demagnetization can only quench the magnetic moment of $10^{-4} \mu_B/\text{atom}$, which is much lower than the observed demagnetization of $0.003 \mu_B/\text{atom}$. In 2007, Dalla *et al* [16] investigated the influence of photon angular momentum on the ultrafast demagnetization in Ni. Their results excluded direct transfer of angular momentum to be relevant for the demagnetization process and showed that the photon contribution to demagnetization is less than 0.01%. This motivated them and others to search for alternative mechanisms for strong demagnetization besides the spin-orbit coupling-based mechanism [25]. Such an argument is reiterated by Stanciu *et al* in [8, 12]. Very recently, Hertel [22], in a viewpoint on [23], claimed that the apparently simple assumption of a direct transfer of the photon spin to the magnetic system is not the solution. The majority of the research merely avoids this contradiction by referring to

the theoretical argument made in the early work [15] and the circularly polarized experiment in nickel [16]. As we pointed out in a previous paper [17], the insensitivity of the magnetization change to light polarization is not a sufficient condition to rule out the direct involvement of photons in the first place. A laser affects the spin moment change in two ways. One is that the light changes the magnetic angular momentum m_s . This is the case for the circularly polarized light. The other way is that the light changes the angular momentum l . This is the case for both circularly and linearly polarized light. If the magnetization change is not sensitive to circularly polarized light, it only means that the m_s channel is not effective, but it cannot exclude the l channel. As a result, one cannot exclude the photon mechanism. A very recent theoretical investigation by Woodford [18] reinforces this concept. Nevertheless it is important to note that, irrespective of the underlying mechanisms, such a low photon number is unlikely to induce a substantial magnetization in a sample.

In this paper, we develop a generic scheme to compute the average photons absorbed per atom and show that, for all 24 sets of experimental data considered here, each atom has more than 1 photon. For a weak laser field, we examine the relation between the relative demagnetization change and the mean photons absorbed per atom. Our results show that the small number of photons absorbed per atom can induce a strong magnetization. Moreover, the linear dependence of demagnetization change on the amplitude of the laser field is consistent with our perturbation result. For the strong laser intensity, we resort to the two-level model system. We find that an effective demagnetization change occurs even with a weak laser field as far as the system is at resonance.

This paper is structured as follows. Section 2 presents a formal algorithm to compute the photon number in femtosecond magnetism. In section 3 we show the relation between the relative demagnetization change and the mean photons absorbed per atom. Then we present a theoretical investigation on the demagnetization change from the perturbation theory in section 4. For a strong field, results are presented in a two-level model in section 5. Finally, the main conclusions of our study are summarized in section 6.

2. Photon number in femtosecond magnetism

We start with a laser pulse propagating along the positive z axis with the field along the x axis:

$$\mathcal{E}_x(\omega, \tau, z; t) = A_0 \exp(-t^2/\tau^2) \exp\left[i\omega\left(\frac{z}{v} - t\right)\right], \quad (1)$$

where A_0 is the amplitude of the laser field, τ is the pulse duration, v is the phase velocity, ω is the laser frequency and t is the time. Since $v = c/(n + ik)$, we can rewrite the above equation as

$$\begin{aligned} \mathcal{E}_x(\omega, \tau, z; t) &= A_0 \exp(-t^2/\tau^2) \exp\left[i\omega\left(\frac{nz}{c} - t\right)\right] \\ &\times \exp\left(-\frac{\omega k}{c}z\right), \end{aligned} \quad (2)$$

where n and k are real and imaginary parts of the index of refraction, respectively, and both are wavelength-dependent. c is the speed of light.

The laser intensity I for the linearly polarized light is computed from [26]

$$\begin{aligned} I(\omega, \tau, z; t) &= 2n\epsilon_0 c |\mathcal{E}_x(\omega, \tau; t)|^2 \\ &= 2n\epsilon_0 c A_0^2 \exp(-2t^2/\tau^2) \exp\left(-\frac{2\omega k}{c}z\right) \\ &\equiv I_0(\omega, \tau; t) \exp\left(-\frac{z}{d}\right), \end{aligned} \quad (3)$$

where ϵ_0 is the permittivity of free space and $I_0(\omega, \tau; t)$ is the laser intensity before penetrating the sample. This is the well-known Beer–Lambert law. Here d is the penetration depth, defined as $d = c/2\omega k = \lambda/4\pi k$, which describes how the light intensity falls off starting from the surface of a sample. It is clear that d itself is intensity-independent. Since we are interested in the pulse energy fluence $F(\omega, \tau, z)$, we integrate the above equation over time and find

$$F(\omega, \tau, z) = 2nc\epsilon_0 \int_{-\infty}^{\infty} |\mathcal{E}_x(\omega, \tau, z; t)|^2 dt, \quad (4)$$

which can be simplified as

$$F(\omega, \tau, z) = 2nc\epsilon_0 A_0^2 \sqrt{\frac{\pi}{2}} \tau \exp\left(-\frac{z}{d}\right) \equiv F_{\max} \exp\left(-\frac{z}{d}\right), \quad (5)$$

where F_{\max} is the initial laser fluence given in experiments. This is the exact expression for the laser energy fluence at depth z , if the pulse is a Gaussian function.

If a laser pulse of fluence $F(\omega, \tau, z)$ shines on a spot with area A , the total number of photons within A at a depth z is

$$N_{\text{photon}}(\omega, z) = \frac{F_{\max} A}{\hbar\omega} \exp\left(-\frac{z}{d}\right), \quad (6)$$

where $\hbar\omega$ is the photon energy and \hbar is Planck's constant over 2π . This equation reveals some crucial information: (i) N_{photon} is a surface quantity and (ii) the number of atoms illuminated by these photons must be a surface quantity as well. In other words, we must compute the photon numbers provided per atom layer by layer, since the light wave propagates uniaxially. Consider an fcc structure with lattice constant a and surface area A : the number of atoms in each layer is

$$N_{\text{atom}} = \frac{2A}{a^2}, \quad (7)$$

where 2 comes from the fact that each unit cell has two atoms per layer. The mean number of photons available to each atom at different depths z is

$$\begin{aligned} \Xi(z) &= \frac{N_{\text{photon}}(\omega, z)}{N_{\text{atom}}} = \frac{F_{\max} a^2}{\hbar v} \frac{1}{2} \exp\left(-\frac{z}{d}\right) \\ &\equiv \Xi_{\max} \exp\left(-\frac{z}{d}\right), \end{aligned} \quad (8)$$

where Ξ_{\max} is the maximum mean number of photons provided to each atom in the top layer. This equation gives the true mean number of photons provided to each atom for demagnetization at depth z .

Under the low pulse energy fluence, ignoring small reflected photons, which is justified in highly absorbed metals, we estimate the photons absorbed per atom at the j th layer as

$$\begin{aligned} \eta_j &= \Xi_{\max} \left[\exp\left(-\frac{(j-1)a}{2d}\right) - \exp\left(-\frac{ja}{2d}\right) \right], \\ j &= 1, 2, 3, \dots \end{aligned} \quad (9)$$

Table 1. The calculated number of photons per atom in Ni. Ξ_{\max} is the maximum photon number per atom. d is the optical penetration depth. $\Xi(d)$ is the photon number at d . η_1 is the photons absorbed per atom in the first top layer. The other parameters are taken from the literature. n and k are real and imaginary parts of the index of refraction [27], respectively.

No.	F (mJ cm ⁻²)	λ (nm)	$n + ik$	d (nm)	Ξ_{\max}	$\Xi(d)$	η_1	References
1	0.6	729	2.28 + 4.18i	13.93	1.368	0.503	0.017	[14]
2	0.76	729	2.28 + 4.18i	13.93	1.741	0.641	0.022	[13]
3	1.4	790	2.46 + 4.35i	14.45	3.457	1.272	0.042	[19]
4	1.45	798	2.476 + 4.375i	14.52	3.617	1.331	0.044	[7]
5	1.8	800	2.48 + 4.38i	14.45	4.501	1.656	0.055	[28]
6	2.04	729	2.28 + 4.18i	13.93	4.644	1.708	0.058	[13]
7	2.0	785	2.45 + 4.34i	14.50	4.908	1.805	0.059	[16]
8	2.0	800	2.48 + 4.38i	14.45	5.001	1.840	0.061	[29]
9	2.5	800	2.48 + 4.38i	14.45	6.252	2.300	0.076	[11]
10	2.83	800	2.48 + 4.38i	14.45	7.075	2.603	0.086	[30]
11	3.5	800	2.48 + 4.38i	14.45	8.752	3.220	0.106	[11]
12	4.4	800	2.48 + 4.38i	14.45	11.003	4.048	0.133	[11]
13	5.3	800	2.48 + 4.38i	14.45	13.254	4.876	0.161	[11]
14	7.0	620	1.93 + 3.65i	13.53	13.566	4.991	0.176	[1, 31]
15	6.0	800	2.48 + 4.38i	14.45	15.004	5.520	0.182	[32]
16	8.0	620	1.93 + 3.65i	13.52	15.504	5.704	0.201	[31]
17	7.1	800	2.48 + 4.38i	14.45	17.755	6.532	0.215	[28]
18	12.0	800	2.48 + 4.38i	14.45	30.008	11.039	0.364	[33]
19	12.73	827	2.53 + 4.47i	14.73	32.897	12.102	0.391	[10]
20	13	827	2.53 + 4.47i	14.73	33.588	12.356	0.399	[34]
21	13.3	827	2.48 + 4.38i	14.73	34.363	12.642	0.409	[35]
22	15.72	800	2.48 + 4.38i	14.45	39.308	14.461	0.476	[36]
23	20	800	2.48 + 4.38i	14.45	50.014	18.400	0.606	[37]
24	35	827	2.13 + 4.17i	14.73	90.430	33.267	1.075	[35]

To appreciate how large Ξ and η_j are, in table 1 we list the results from 24 different sets of experimental data increasing from top to bottom. This table is very telling. The maximum photon number Ξ_{\max} ranges from 1.368 to 90.430. The photon number at the penetration depth $\Xi(d)$ varies from 0.503 to 33.267. Note that, since the majority of samples are thinner than the penetration depth, the number of photons available to each atom exceeds 1. Here we emphasize that there are enough photons for each atom. The absorbed photons in the first layer η_1 range from 0.017 to 1.075. The mean photons absorbed per atom within the sample is less than η_1 . Koopmans' group [14] has the smallest value of 0.017. Beaurepaire *et al*'s [1] η_1 value is 0.176, ten times higher than that of Koopmans' η_1 . Cheskis *et al* [35] have the largest η_1 value of 1.075 or roughly one photon per atom, and importantly they already found that the demagnetization is saturated. This provides a first indication that one does not need a large amount of photons to demagnetize the sample. The small number of photons absorbed does not necessarily mean that they cannot induce a strong magnetization change.

3. Demagnetization change versus photons absorbed

We need to map out the connection between the magnetization change and the number of the photons absorbed. We investigate the relative demagnetization changes $\Delta M/M$ which are extracted from their corresponding literatures versus the photons absorbed per atom for a weak laser intensity [1, 7, 13, 14, 19, 20, 35]. We purposely choose a weaker intensity in order to search for a one-to-one correspondence between the number of photons absorbed and

the amount of magnetization change. For a strong laser, due to the saturation, such a relation will become complicated. To find some valuable information on the demagnetization mechanism associated with the number of photons, it is necessary to estimate the mean photons absorbed per atom $\bar{\eta}$ more accurately. Based on equation (9), $\bar{\eta}$ is computed from

$$\bar{\eta} = \frac{1}{k} \sum_{j=1}^k \eta_j, \quad (10)$$

where k is the number of layers within the sample's thickness. If the sample's thickness is thinner than the penetration depth, we calculate $\bar{\eta}$ averaged within the sample's thickness, or else within the penetration depth. The $\bar{\eta}$ itself includes the reflected and absorbed photons.

Figure 1 shows $\Delta M/M$ as a function of $\bar{\eta}$. Four solid lines (as guides for the eyes) from top to bottom represent the results by Bigot *et al* [7], Beaurepaire *et al* [1], Cheskis *et al* [35] and Atxitia *et al* [20], respectively. The corresponding results from Koopmans *et al* [13, 14] and Wilks *et al* [19] are also shown in the bottom left corner of the figure. The vertical dotted line shows that 0.1 photon can induce about 4%–72% spin momentum change for different laser durations. This figure is very insightful. (i) The slope of these lines approximately characterizes the demagnetization ability of a laser pulse. Each solid line denotes the results obtained by an identical laser pulse which has the same pulse duration. (ii) The amplitude of laser A_0 plays a key role in the demagnetization change. For the same fluence, as predicted by equation (5), the smaller the pulse duration is, the bigger the laser amplitude becomes. Within the dipole approximation, the interaction between the laser and the system is dominated by the amplitude of the

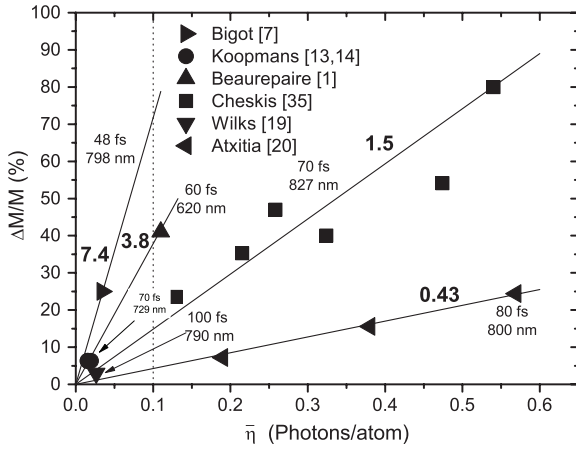


Figure 1. Dependence of the relative demagnetization changes on the average photons absorbed per atom for six different sets of experimental data. The solid lines are guides for the eye, whose slopes are shown on the lines. The laser pulse durations and the photon wavelengths are also given near their data.

laser. Therefore, for the same \bar{n} , the amplitudes increase from bottom to top. (iii) With the same pulse duration parameter, the demagnetization change increases along with the photons absorbed per atom or the laser fluence. The solid lines with slopes of 1.5, 3.8 and 7.4 indicate that one does not need a large amount of photons to induce a substantial moment change. For Koopmans *et al* [13, 14] and Wilks *et al* [19] data, because their laser energy fluences are very small and the pulse durations are long, the demagnetization changes are much smaller, but are still consistent with the above picture.

From figure 1, it is obvious that the laser amplitude plays a crucial role in the demagnetization changes. However, is the laser amplitude the only deciding factor for demagnetization change? The answer is negative. In fact, using the concept of photons absorbed per atom to explain the strong demagnetization change obviously neglects two very important factors. First, it does not take into account the interaction between the light and the material. Light has energy (how strongly the field oscillates) and frequency (how fast the field oscillates with time). \bar{n} only takes into account the energy but not the frequency, nor the transition matrix elements between different states. Second, once the number of photons per atom becomes small, it is well known that the quasi-classical description of photons absorbed per atom for demagnetization becomes invalid. In particular, when the average number of photons provided to each atom is less than 40, the electric field behaves quantum mechanically [38], i.e. the field oscillates strongly around its average value. The fewer photons there are per atom, the stronger the oscillation is. Therefore, it is necessary to reconsider this hurdle from a different perspective, and we examine this issue from the perturbation theory and two-level model, respectively.

4. Perturbation theory and weak intensity

In section 3, it is found that the relative demagnetization change $\Delta M/M$ is proportional to the mean photons absorbed

per atom \bar{n} under a weak laser field. This allows us to treat the laser field perturbatively. The Hamiltonian of a system can be described by $H = H^0 + H^1$, where H^0 is the time-independent Hamiltonian of the unperturbed system and $H^1 = -e\vec{\mu} \cdot \vec{\mathcal{E}}(t)$ is the time-dependent perturbation. We start with the Liouville equation $i\hbar\dot{\rho} = [H, \rho]$ for the density matrix. We keep only the first-order term and have

$$i\hbar\dot{\rho}^{(1)} = [H^0, \rho^{(1)}] + [H^1, \rho^{(0)}], \quad (11)$$

where $\rho^{(0)}$ and $\rho^{(1)}$ are the zeroth and first order of the density matrices, respectively. If we make a unitary transformation as $\rho^{(1)} = e^{-iH^0 t/\hbar} Q e^{iH^0 t/\hbar}$, equation (11) can be written as

$$i\hbar\dot{Q} = e^{iH^0 t/\hbar} [H^1, \rho^{(0)}] e^{-iH^0 t/\hbar}. \quad (12)$$

We integrate this equation over time and take $\rho^{(1)}(-\infty) = 0$. Then we apply the eigenstate $|n\rangle$ on the left and $|m\rangle$ on the right of this equation, and we can obtain

$$\begin{aligned} \rho_{nm}^{(1)}(t) &= \frac{1}{i\hbar} (\rho_{nn}^{(0)} - \rho_{mm}^{(0)}) e\mu_{nm} e^{-i\omega_{nm}t - \Gamma t} \\ &\times \int_{-\infty}^t dt' \mathcal{E}(t') e^{i\omega_{nm}t' - \Gamma t'}, \end{aligned} \quad (13)$$

where $\omega_{nm} = (E_n - E_m)/\hbar$, μ_{nm} are the transition matrix elements and Γ is the damping factor. Theoretically, the demagnetization change is involved in the time-dependent density matrix $\rho_{nm}^{(1)}(t)$ through $M_z^{(1)}(t) = \text{Tr}[S_z \rho^{(1)}(t)]$, where S_z is the spin matrix. By investigating the density matrix, we can reveal some crucial details of the demagnetization. Next we discuss two typical cases.

Case 1. Continuum wave laser. Consider a periodic field perturbation $H^1 = -e\mu A_0 e^{-i\omega t}$. The density matrix becomes

$$\begin{aligned} \rho_{nm}^{(1)}(t) &= \frac{e\mu_{nm}}{i\hbar} (\rho_{nn}^{(0)} - \rho_{mm}^{(0)}) A_0 e^{-2\Gamma t} \\ &\times \frac{(\Delta\omega \sin \omega t - \Gamma \cos \omega t) + (\Delta\omega \cos \omega t + \Gamma \sin \omega t)i}{\Delta\omega^2 + \Gamma^2}, \end{aligned} \quad (14)$$

where $\Delta\omega = \omega - \omega_{nm}$. Note that its frequency dependence is exactly the same even if we treat photons quantum mechanically. It shows that the density matrix $\rho_{nm}^{(1)}(t)$ not only depends on the amplitude of the laser field A_0 , which is consistent with the conclusion obtained from figure 1, but also depends on the resonance term $(\frac{1}{\Delta\omega^2 + \Gamma^2})$. Since the spin moment change is directly proportional to $\rho_{nm}^{(1)}(t)$, this predicts that the magnetic moment change will depend linearly on the amplitude of the laser field in the weak field limit. On the other hand, the resonance term demonstrates that, even for exactly the same laser amplitude, the transition or the magnetic change can be very different for different frequencies. This has been largely ignored in the literature.

Case 2. Pulsed laser. We assume that the laser field has a form $H^1 = -e\mu A_0 e^{-t^2/\tau^2} e^{-i\omega t}$. Equation (13) becomes

$$\begin{aligned} \rho_{nm}^{(1)}(t) &= \frac{e\mu_{nm}}{i\hbar} (\rho_{nn}^{(0)} - \rho_{mm}^{(0)}) e^{-i\omega_{nm}t - \Gamma t} \\ &\times A_0 \int_{-\infty}^t dt' e^{-t'^2/\tau^2} e^{-i\Delta\omega t' - \Gamma t'} \\ &\equiv \frac{e\mu_{nm}}{i\hbar} (\rho_{nn}^{(0)} - \rho_{mm}^{(0)}) e^{-i\omega_{nm}t - \Gamma t} R(t), \end{aligned} \quad (15)$$

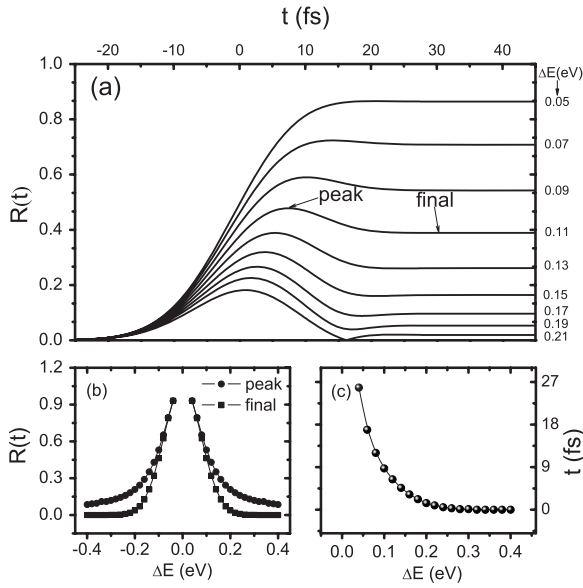


Figure 2. (a) The module of response function $R(t)$ as a function of time at different ΔE . (b) The dependence of the peak and final values on ΔE . (c) The time needed to reach the peak as a function of ΔE .

where $R(t)$ is defined as the module of the response function. To get some insightful information about the perturbation of the laser field, we choose $\tau = 12$ fs and $A_0 = 0.05$ V \AA^{-1} , and integrate the response function numerically.

Figure 2(a) shows the module of the response function $R(t)$ as a function of time for twelve different energy detunings ΔE ($\hbar\Delta\omega$) from 0.05 to 0.21 eV. We can see that, as time goes by, $R(t)$ first increases to a peak value and then settles down to its final value. Importantly the peak values of $R(t)$ vary a lot for different ΔE even with the same laser field amplitude. The biggest one is about 0.9 ($\Delta E = 0.05$ eV) and the smallest one is about 0.07 ($\Delta E = 0.15$ eV). Their difference is over an order of magnitude. It demonstrates that the transition probability or spin momentum change can be very large when ΔE becomes very small or the system is excited resonantly, even if the field intensity is weak.

Figure 2(b) compares the peak and final values of $R(t)$ as a function of ΔE . The final $R(t)$ values decrease more quickly with ΔE than that of peak ones. The maximum difference is 0.16 at $\Delta E = 0.22$ eV and the minimum difference is 0.09 at $\Delta E = 0.4$ eV. When ΔE reaches 0.07 eV, the peak and final values are almost the same. It implies that, regardless of the amplitude of the laser field, the transition from one state to another state is finished when ΔE goes to zero. Figure 2(c) shows the time needed reaching the peak value as a function of ΔE . This indicates that the peak time is reduced as ΔE becomes larger. All the above results are obtained within a weak field limit. Once the laser field becomes strong, the first-order perturbation becomes invalid. Under this situation, we resort to a two-level model system.

5. Two-level model and strong intensity

The two-level model has been extensively used in atoms [39, 40], semiconductors [41–43] and ferromagnetic ma-

terials [44]. For a system excited by a laser, the two-level model can give us a quantitative understanding of the spin transition. Here we directly quote equation (7) of [44] in our previous work about the spin change ΔS_z for a transition from state $|a\rangle$ to state $|b\rangle$:

$$\Delta S_z = \aleph_{ab}(t; \omega)(\langle b|s_z|b\rangle - \langle a|s_z|a\rangle), \quad (16)$$

where t is time and ω is laser frequency. $\aleph_{ab}(t; \omega)$, the probability amplitude of finding the system at time t in state $|b\rangle$, is equal to

$$\aleph_{ab}(t; \omega) = \frac{|W_{ab}|^2}{|W_{ab}|^2 + \hbar^2(\omega - \omega_{ba})^2} \times \sin^2 \left[\sqrt{\frac{|W_{ab}|^2}{\hbar^2} + (\omega - \omega_{ba})^2} \frac{t}{2} \right], \quad (17)$$

where $W_{ab} = \mu_{ab}A_0$. The transition matrix elements μ_{ab} can be obtained by calculating the corresponding momentum operator \vec{p}_{ab} from the *ab initio* calculation [45–47].

The probability $\aleph_{ab}(t; \omega)$ is an oscillatory function of time; for certain values of t , $\aleph_{ab}(t; \omega) = 0$, meaning that the system returns to the initial state $|a\rangle$. At the same time it also reveals fast oscillation, expressed by the latter term of equation (17). If $|W_{ab}|$ is large enough, the value of $\aleph_{ab}(t; \omega)$ or ΔS_z can be very large. For the off-resonance excitation, the laser field amplitude A_0 controls the demagnetization change entirely. This happens in semiconductors. For the resonance excitation, or $\omega = \omega_{ba}$, regardless of how weak the perturbation is, the field can cause the system to transit from state $|a\rangle$ to state $|b\rangle$ [48]. Importantly, the field intensity only affects the time needed for the system to transit from $|a\rangle$ to $|b\rangle$, not whether a transition can occur or not. The smaller the field intensity is, the longer the time it takes.

For a ferromagnetic metal like nickel, the chance that the transition frequency ω_{ba} matches that of the laser field is very high [49]. This explains the observed demagnetization in those experiments, in spite of a relatively weak laser electric field. Figure 3 shows the detailed dependence of the spin change on the laser electric field. For a weak laser field intensity with 0.001 V \AA^{-1} , the spin change is possible as far as ω_{ba} matches ω . This demonstrates that the strong demagnetization change is achievable in experiments even if the laser field intensity is very weak. If the laser field becomes larger, the range of matching frequencies becomes broad (see figure 3). In semiconductors, Pavlov *et al* [50] showed that increasing temperature reduces the GaAs bandgap. This may be a test case for our theory.

6. Conclusion

We have clarified a long-standing conceptual puzzle of the photon shortage in femtosecond magnetism by comparing the relative demagnetization change versus the mean photons absorbed per atom. In the weak laser field, it increases along with the mean photons absorbed per atom \bar{n} , which is consistent with perturbation theory. Importantly, the results show that a few photons absorbed per atom can induce

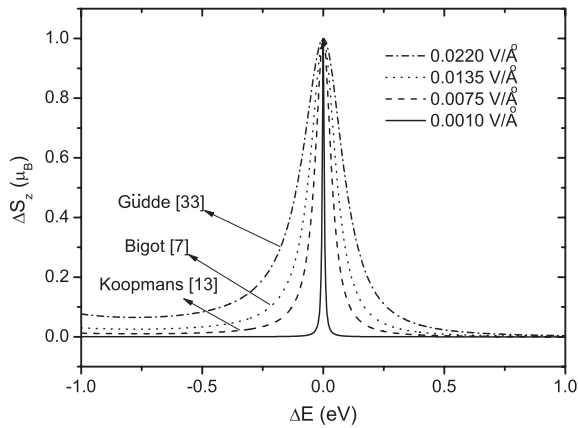


Figure 3. Amplitude of spin momentum change as a function of the detuning $\Delta E = \hbar(\omega - \omega_{ba})$ for three groups at different laser electric fields. The amplitude of the laser field is deduced from the experimental laser fluence based on equation (5). All the curves are shifted to zero at resonance.

considerable spin moment changes. The spin moment can be reduced at resonance even if the field is weak. Our findings overcome a big hurdle in ferromagnetism and should inspire new experimental and theoretical investigations into the role of photons and their interactions with electrons in the magnetization change.

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