Total angular momentum conservation in laser-induced femtosecond magnetism

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Total angular momentum conservation is a fundamental law in classical and quantum mechanics, but since spin momentum is not a classical quantity, it is far from obvious how the law affects spin momentum change in laser-induced femtosecond magnetization. Here it is shown that if a system has full rotational symmetry, the law requires that the spin and orbital momenta are coupled, but there is no genuine magnetization change for linearly polarized light. To induce such a change, this very symmetry has to be broken. In solids, the rotational symmetry is lifted by the translational symmetry, and the spin and orbital momenta components of different total angular momenta mix to some extent. This mixing is the origin of the time-dependent total angular momentum as observed by Bartelt *et al.* [Appl. Phys. Lett. **90**, 162503 (2007)]. The remaining unmixed portion accounts for an extra spin change in three independent circularly polarized laser experiments.

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It is a fundamental law in classical and quantum mechanics that the total angular momentum of a system must be conserved in absence of external torques. In the laser-induced femtosecond magnetization process,[1] the system involves a lattice, electron spins, electron orbitals, and photons. The total angular momentum change $\Delta\vec{J}$ is[2] $\Delta\vec{J}=\Delta\vec{L}_{\text{photon}}+\Delta\vec{L}_{\text{lattice}}+\Delta\vec{L}_e+\Delta\vec{S}_e=0$, where $\vec{L}_{\text{photon}}$, $\vec{L}_{\text{lattice}}$, $\vec{L}_e$, and $\vec{S}_e$ are the photon, lattice, electron orbital, and spin momenta, respectively. The spin change can be computed as $\Delta\vec{S}_e=-\Delta\vec{L}_{\text{photon}}-\Delta\vec{L}_{\text{lattice}}-\Delta\vec{L}_e$. On the ultrafast time scale when the lattice is silent or $\Delta\vec{L}_{\text{lattice}}=0$, $\Delta\vec{S}_e=-\Delta\vec{L}_{\text{photon}}-\Delta\vec{L}_e$. In solids, due to translational symmetry, $\vec{L}_e$ is partially quenched, but the amount of quenching is unknown classically. On the other hand, if the orbital angular momentum were completely quenched, $\Delta\vec{S}_z=-\Delta\vec{L}_{\text{photon}}$ would provide a simple explanation of how spin is changed in the laser-induced femtosecond magnetism, which has attracted enormous interest.[1,3–7] However doing so would imply not enough photons available to demagnetize a sample.[4] This equation also suggests two incorrect results: without spin-orbit coupling (SOC), (i) the magnetization can be changed, and (ii) light is coupled to spin, both of which contradict the quantum mechanical results.[8]

To circumvent the above problem, recently Stamm *et al.*[9] suggested an efficient exchange channel between the spin and lattice systems on a time scale of 150 fs, while still maintaining the total momentum conservation. However this leads to another complication: one must assume an unprecedented large spin-lattice interaction.[7,10] To this end, there has been no consensus on the role of the angular momentum conservation law in femtosecond magnetism.[4,11] All the existing theoretical investigations,[8,12,13] including our previous investigations,[14] have not addressed the law explicitly. This greatly hampers current efforts in femtosecond magnetism.

In this paper, we aim to clarify the role of total angular momentum through an analytical investigation. This excludes any potential ambiguity that may occur in a numerical calculation. We first show that if the total angular momentum is a good quantum number, there is no genuine demagnetization for linearly polarized light. It is necessary to break the rotational symmetry to induce genuine demagnetization, where the spin and orbital momenta of different total angular momenta are mixed to some extent. In solids it is this mixing that accounts for the total angular momentum change in a recent experiment.[15] The remaining unmixed portion, though small, upholds the coupling between spin and orbital momenta. This allows the circularly polarized light to introduce an extra change in spin momentum, though the average of signals induced by left and right circularly polarized lights is exactly the same as that of linearly polarized light. Both findings are consistent with three independent experiments.[2,16,17]

To begin with, we should first point out that the spin degree of freedom is a pure quantum mechanical quantity. While it is possible to represent spin by a classical vector in a thermal or magnetic field driven process, in an optically excited process one must adopt a quantum-mechanical picture of spin. The fundamental reason for this difference is because the selection rule plays a dominant role in optical excitation, not in a thermal or magnetic excitation.

If a system has spherical symmetry, the total angular momentum is conserved. Without SOC, the dipole selection rule is $\Delta l=\pm 1, \Delta m_l=0,\pm 1$ and $\Delta s=0$, where l, m_l, and s are orbital, magnetic orbital, and spin quantum numbers, respectively. The light only couples to the orbital part of the angular momentum, leaving the spin momentum unchanged, which is also true in solids.

With SOC, states have mixed spin-up and spin-down states, and the selection rule becomes $\Delta j=0,\pm 1$ and $\Delta m_j=0,\pm 1$, excluding the transition of $j=0\leftrightarrow j=0$. Two eigenfunctions of $\vec{J}$ are

TABLE I. Spin momentum change $\Delta\langle s_z\rangle$ (in units of $\hbar/2$) for three possible transitions. The contribution from the radial wave function is not included.

Transition	$\phi^a \to \phi^{a'}$		$\phi^b \to \phi^{b'}$		$\phi^a \to \phi^b$
$j'-j$	-1	1	-1	1	0
$m_{j'}-m_j$					
1	$+\frac{1}{j-1}+\frac{m_j}{j(j-1)}$	$+\frac{1}{j+1}-\frac{m_j}{j(j+1)}$	$-\frac{1}{j}-\frac{m_j}{j(j+1)}$	$-\frac{1}{j+2}+\frac{m_j}{(j+1)(j+2)}$	$-\frac{1}{j+1}-\frac{(2j+1)m_j}{j(j+1)}$
0	$\frac{m_j}{j(j-1)}$	$-\frac{m_j}{j(j+1)}$	$-\frac{m_j}{j(j+1)}$	$\frac{m_j}{(j+1)(j+2)}$	$-\frac{(2j+1)m_j}{j(j+1)}$
-1	$-\frac{1}{j-1}+\frac{m_j}{j(j-1)}$	$-\frac{1}{j+1}-\frac{m_j}{j(j+1)}$	$+\frac{1}{j}-\frac{m_j}{j(j+1)}$	$+\frac{1}{j+2}+\frac{m_j}{(j+1)(j+2)}$	$+\frac{1}{j+1}-\frac{(2j+1)m_j}{j(j+1)}$

$$\phi^a_{jm_j}=\frac{1}{\sqrt{2l+1}}\begin{bmatrix}\sqrt{l+m+1}\,Y_{lm}\\ \sqrt{l-m}\,Y_{lm+1}\end{bmatrix};\quad j=l+\frac{1}{2},\quad m_j=m+\frac{1}{2}, \tag{1}$$

$$\phi^b_{j'm_{j'}}=\frac{1}{\sqrt{2l'+1}}\begin{bmatrix}-\sqrt{l'-m'}\,Y_{l'm'}\\ \sqrt{l'+m'+1}\,Y_{l'm'+1}\end{bmatrix};$$
$$j'=l'-\frac{1}{2},\quad m_{j'}=m'+\frac{1}{2}, \tag{2}$$

where different quantum numbers l,j,m_j are used for ϕ^a and ϕ^b to highlight that those numbers may differ. Here Y_{lm} is the spherical harmonic function. The expectation values of s_z are $\langle\phi^a|s_z|\phi^a\rangle=[m_j/j]\hbar/2$ and $\langle\phi^b|s_z|\phi^b\rangle=[-m_{j'}/j'+1]\hbar/2$.[18,19] There is no need to compute $\langle l_z\rangle$ separately since $\langle l_z\rangle=m_j\hbar-\langle s_z\rangle$. How the spin momentum changes during the transitions between the eigenfunctions ϕ^a and ϕ^b holds the key to femtosecond magnetism.

First, the spin momentum change depends on the type of transitions. Table I shows all the possible changes in the spin momentum. To get a rough estimate for each transition, we consider transitions of $m_{j'}-m_j=\Delta m_j=0$. For the $\phi^a\to\phi^{a'}$ and $\phi^b\to\phi^{b'}$ transitions, the percentage change $\Delta\langle s_z\rangle/\langle s_z\rangle$ ranges from $-1/(j+1)$ to $1/(j-1)$ and from $-1/(j+1)$ to $1/(j+2)$, respectively, and for $\phi^a\to\phi^b$ it is just $-(2j+1)/(j+1)$. These results suggest that it is possible to induce a huge spin change in ferromagnets, and even the sign can be changed. Such a big change has been observed in $CoPt_3$,[20] where a nearly complete demagnetization was demonstrated.

Second, the light polarization is not a necessary condition for spin momentum change. Both linearly and circularly polarized light can change the spin momentum. The middle row of Table I ($m_{j'}-m_j=0$) shows that linearly polarized light can change the spin momentum since the spin and orbital momenta can exchange. For circularly polarized light with $\Delta m_j=\pm 1$, all of the spin momentum change contains two contributions (see Table I): the first term is from the light polarization, and the second is from the internal exchange between orbital and spin momenta. Compared to linearly polarized light, right/left circularly polarized light contributes an extra "$1/j$" term. This is a manifestation of the angular momentum conservation. If the orbital momentum is partially quenched and the total angular momentum is no longer a good quantum number, the circularly polarized light, via the remaining coupling between spin and orbital momenta, still induces an extra contribution to the spin momentum. This is the origin of the extra peak around the zero time delay as observed by three independent experimental groups,[2,16,17] and the polarization effect becomes even more pronounced in semiconductors.[21] The table also shows that the average of the spin momentum changes induced by right/left circularly polarized light is exactly the same as that by the linearly polarized light, which is verified experimentally.[16,17] This finding is very different from the simple classical consideration above.

Third, it is necessary to break the spherical symmetry for linearly polarized light to induce a true magnetization change. If j and m_j are both good quantum numbers and m_j takes the values $-j, -j+1,\ldots,$ and j, a sum of the spin momentum change over m_j is zero (see Table I, where all the terms for $\Delta m_j=0$ contain m_j). Compared to circularly polarized light, linearly polarized light is more sensitive to the magnetic ordering. This finding is not surprising. In all of magneto-optics, it is well known that the breakdown of spherical symmetry is a necessary condition. This result is valid for both ferromagnets and magnetic semiconductors. In solids, the breakdown of the symmetry is guaranteed thanks to the translational symmetry, where the spin and orbital momenta, belonging to different total angular momenta, mix to some extent, and the amount of mixing depends on the specific system. As will be seen below, if the spherical symmetry is broken, the coupling between spin and orbital momenta is only maintained at the basis-set level, not at the eigenstate level, which is a major cause of the current confusion in the literature. This argument becomes even clearer if we examine the impact of translational symmetry on the total angular momentum.

We start with a Bloch state in a regular crystal with translational symmetry and expand it in the basis set of eigenfunctions $\phi_{j,m_j;k}$ of J for each k point,

$$|\psi_{nk}\rangle=\frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{\Omega}}\sum_{j,m_j}c_{j,m_j;nk}\phi_{j,m_j;k}, \tag{3}$$

where Ω is the unit cell volume, and $\mathbf{k}$ and $\mathbf{r}$ are the electron wave vector and position, respectively. If a material has a

spherical symmetry, Eq. (3) has only one term in the summation. Because of translational symmetry, it contains many more terms. How spin and orbital momenta are mixed is determined by $c_{j,m_j;nk}$. Note that $\phi_{j,m_j;k}$ carries a k index since at each k point the eigenfunctions may differ.

Equation (3) is very insightful. (i) There is no limit on how large the orbital momentum change can be. First, the summation contains all the possible j and m_j, with its contribution weighted by the coefficients $c_{j,m_j;nk}$. Second, if we expand the plane wave factor as

$$e^{i\mathbf{k}\cdot\mathbf{r}} = 4\pi \sum_{l=0}^{\infty} \sum_{m=-l}^{+l} i^l j_l(kr) Y_{lm}(\hat{r}) Y_{lm}^*(\hat{k}), \tag{4}$$

where j_l is the spherical Bessel function, and $\hat{r}$ and $\hat{k}$ are the unit vectors of $\mathbf{r}$ and $\mathbf{k}$, we see that l and m can take any possible value as well. The availability of the orbital momentum is not a limiting factor for spin momentum change. If the laser intensity is strong, multiple excitations certainly increase/decrease the orbital momentum to a very high/low value.

(ii) The total angular momentum becomes time dependent. Since it does not permute with the crystal Hamiltonian, the expectation value of J is a superposition of J values among different states and becomes time dependent. To be more specific, we assume that the system is described by $|\Psi(t)\rangle = \Sigma_{nk} \xi_{nk}(t) e^{-iE_{nk}t/\hbar} |\psi_{nk}\rangle$, where $\xi_{nk}(t)$ is the population coefficient and E_{nk} is the band energy. The expectation value of J is

$$\langle \Psi(t)|J|\Psi(t)\rangle = \sum_{n_1,k_1;n_2,k_2} \xi_{n_1k_1}^*(t) \xi_{n_2k_2}(t) e^{i(E_{n_1k_1}-E_{n_2k_2})t/\hbar} \langle \psi_{n_1k_1}|J|\psi_{n_2k_2}\rangle, \tag{5}$$

with its time dependence from the state population change (first two terms behind the summation) and the energy phase factor (third term). $\langle \Psi(t)|J|\Psi(t)\rangle$ will be time dependent as long as $\langle \psi_{n_1k_1}|J|\psi_{n_2k_2}\rangle$ is not diagonal or J is not a good quantum number. This prediction is fully consistent with the experimental finding where the J_z value obtained by Bartelt *et al.*[15] indeed changes with time. A quantitative estimate on the time scale of angular momentum change from Eq. (5) is difficult since it requires a detailed band structure calculation, but determining factors can be easily seen. The exponent factor represents the state phase change. If both $\xi_{n_1k_1}^*(t)$ and $\xi_{n_2k_2}(t)$ were time independent, the dephasing among different states would determine the time scale of angular momentum change, where the shorter time scale dynamics would be determined by those pairs of states which have a larger energy difference, and the longer dynamics by states with a smaller energy difference. Since normally $\xi_{n_1k_1}^*(t)$ and $\xi_{n_2k_2}(t)$ are driven by the ultrafast laser pulse and change with time, the time scale will be jointly determined by the extrinsic laser pulse and intrinsic state dephasing.

In conclusion, we have investigated the influence of total angular momentum on the laser-induced femtosecond magnetization change. We show analytically if the total angular momentum is a good quantum number, the magnetization change is not possible for linearly polarized light. The lifting of the full rotational symmetry is a necessary condition for the magnetization change. As a result, the spin and orbital momenta of different total angular momenta mix and the total momentum changes with time. This is the origin of the experimental results by Bartelt *et al.*[15] The unmixed portion leads to an extra magnetization change by circularly polarized light. Our results show that an average of spin momentum change over left and right circularly polarized lights is exactly the same as the linearly polarized one. This explains the findings in three independent experiments by Longa *et al.*,[2] Comin *et al.*,[16] and Wilks *et al.*[17]

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