Phenomenological theory for coherent magnon generation through impulsive stimulated Raman scattering

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We present a phenomenological theory for impulsive generation of coherent magnons in magnetic dielectrics through impulsive stimulated Raman scattering. Consideration is given to a lattice which has an arbitrary number of localized spins in each magnetic unit cell. A general expression for the modulation of optical dielectric permittivity by light-induced spin waves is derived and its symmetry properties with respect to time reversal are analyzed. A simple cubic ferromagnet and a rutile-type structure antiferromagnet are treated as examples of application of the theory. It is shown that the ellipticity of the spin precession, inherent to spin waves in antiferromagnets, leads to an enhancement of the amplitude of light-induced spin waves.

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I. INTRODUCTION

In recent years, ultrafast pump-probe magneto-optical spectroscopy has been used to study the coherent spin dynamics triggered by femtosecond optical pulses in magnetically ordered media.[1–12] A common feature of the observed phenomena is a periodic modulation of the optical dielectric permittivity of a medium at frequencies of one- or two-magnon excitations. This fact unambiguously shows that in these experiments, coherent collective spin excitations were induced by light pulses. Understanding of microscopic processes governing the ultrafast generation of spin coherence in magnetically ordered media would be helpful for fundamental science and technical applications.

The most known microscopic mechanism of generation of coherent collective excitations by light is the stimulated Raman scattering (RS).[13] The possibility of exciting coherent one-magnon excitations (spin waves) by cw light in magnetically ordered media was discussed for the first time in Ref. 14. However, only developments in time-resolved femtosecond technique made it possible to monitor the coherent spin precession triggered by a short light pulse.

Applications of ultrafast time-resolved spectroscopy to the generation and detection of coherent collective excitations started with investigations of the phonon generation and raised a number of questions about microscopic mechanisms of the photoexcitation. In transparent media, the process of the phonon generation by a short light pulse is known to be impulsive stimulated Raman scattering (ISRS).[15] In absorbing media, in addition to ISRS, the generation of coherent excitations can occur also due to non-Raman mechanisms.[16–20] Obviously, the same is true for the magnon generation. However, an extension of the theory of the phonon generation to the magnon generation is not straightforward and requires some modification. The main difference between both cases comes from the fact that electron spins participating in the spin precession are involved in optical transitions, while phonons are not, because of relatively large nuclear masses.

Note that spin system in magnetic solids possesses many different types of collective excitations. Apart from one-magnon excitations, two-magnon excitations are well known.[21] Unlike spin waves, coherent two-magnon excitations are characterized by the oscillation of the magnetization fluctuations but not by the spin precession. The impulsive generation of coherent two-magnon excitations was studied in Ref. 22. In this paper, we will consider only one-magnon coherent excitations.

In most experimental works published so far, the generation of coherent magnons was interpreted as a result of a modification of the magnetic anisotropy by a light pulse, i.e., the non-Raman mechanism of the photoexcitation was proposed. This result is quite natural because not only magnetic metals but also most part of magnetic dielectrics possess significant optical absorption. A possible contribution of ISRS to the magnon generation was noted in Refs. 6 and 7 but no evidence for this was given. Only in the recently published paper,[12] it was convincingly demonstrated that in the transparent antiferromagnet $FeBO_3$, the generation of the spin waves occurs through ISRS. It was shown that the amplitude of the light-induced spin wave in the antiferromagnet depends on the exchange and anisotropy constants through specific combinations characterizing the ellipticity of the spin precession. It might be helpful for further experimental investigations in this field to elaborate a theory of the impulsive generation of spin waves by light in transparent magnetic crystals to understand the role of the equilibrium magnetic structure of a crystal in the process of the spin-wave generation.

In this paper, we develop a phenomenological theory for the impulsive generation of spin waves through ISRS. The light-induced spin-wave normal coordinates are calculated from the Hamilton equations in Sec. II. We also derive a general expression for the modulation of the dielectric permittivity by the induced spin waves and analyze its symmetry with respect to the time-reversal operation. In Sec. III, we apply the theory to a simple cubic ferromagnet and a rutile-type structure antiferromagnet.

II. THEORY

In this section, we present a formal mathematical theory of the light-induced spin-wave dynamics in a transparent magnetic dielectric, having $N \geq 1$ magnetic ions per unit cell.

Assuming a small wave vector $\mathbf{k}\approx 0$ of the pump light, we will consider only $\mathbf{k}=0$ spin-wave modes; i.e., we assume the localized magnetic moment $\mathbf{M}_r(t)$ at site r $(r=1,\ldots,N)$ to be independent of unit cell.

The deviation of $\mathbf{M}_r(t)$ from equilibrium can be characterized by the components M_{r,x_r} and M_{r,y_r} defined in a local coordinate system (x_r,y_r,z_r), in which the z_r axis is directed along the equilibrium magnetic moment $\mathbf{M}_r^{(0)}$. For the following consideration, it is convenient to introduce the canonical variables $\{b_r\}$ through the linearized Holstein–Primakoff transformation,

$$b_r=(2\gamma_r M_r)^{-1/2}(M_{r,x_r}+iM_{r,y_r}), \tag{1}$$

where $M_r=|\mathbf{M}_r|$ is the magnitude of the sublattice magnetization and γ_r is the gyromagnetic ratio for the ion at site r. In the harmonic approximation, the Hamiltonian H_0 of the spin system is a quadratic function of the variables $\{b_r,b_r^*\}$.

The electric field of a pump pulse exciting the spin system can be represented in the form $\mathbf{E}(t)=\mathrm{Re}\,\boldsymbol{\mathcal{E}}(t)e^{i\omega t}$, where ω is the central frequency of the pulse and $\boldsymbol{\mathcal{E}}(t)$ is the time-dependent amplitude.

Phenomenologically, the interaction between magnons (or other low-energy collective excitations) and the pump pulse in a transparent medium is given by[24]

$$V=-\frac{\delta\epsilon_{ik}}{16\pi}\mathcal{E}_i^*(t)\mathcal{E}_k(t), \tag{2}$$

where $\delta\epsilon_{ik}$ is the modulation of the optical dielectric permittivity by magnons. V can be expressed as a power series in b_r and b_r^*. When considering spin-wave excitations, one should take into account only linear in b_r and b_r^* terms in $\delta\epsilon_{ik}$. For a transparent medium, the tensor $\delta\epsilon_{ik}$ is Hermitian: $\delta\epsilon_{ik}=\delta\epsilon_{ki}^*$.

In impulsive limit, when the optical pulse width $\tau\ll\Omega^{-1}$, where Ω is the frequency of an induced spin wave, one may use the formal representation,

$$\mathcal{E}_i^*(t)\mathcal{E}_k(t)=\frac{4\pi I_0}{nc}e_i^*e_k\delta(t), \tag{3}$$

where n is the refractive index, c is the speed of light, I_0 is the integrated pulse intensity, and e_i is a component of the unit vector defining the polarization of light. The delta function $\delta(t)$ accounts for the impulsive character $(\tau\ll\Omega^{-1})$ of pump light. It is convenient to decompose the product $e_i^*e_k$ into the symmetric and antisymmetric parts,

$$e_i^*e_k=\Sigma_{ik}+\frac{i}{2}\mathfrak{e}_{ikl}h_l, \tag{4}$$

where $\Sigma_{ik}=1/2(e_i^*e_k+e_k^*e_i)$, $\mathfrak{e}_{ikl}$ is the unit antisymmetric tensor, and h_l is a component of the vector $\mathbf{h}=i(\mathbf{e}\times\mathbf{e}^*)$. $|\mathbf{h}|$ is a measure of light helicity and varies in the range from -1 to 1.

Under the action of the light pulse, b_r varies with the time according to the Hamilton equation,[25]

$$\frac{db_r}{dt}=-i\frac{\partial}{\partial b_r^*}(H_0+V). \tag{5}$$

The advantage of the classical approach which we use here is that there is no need to specify exactly the optical transitions caused by the photoexcitation. Optical properties of a medium relevant to the generation of coherent excitations are contained in the modulation of the optical dielectric permittivity. This is a consequence of the fact that all optical transitions are virtual for a transparent medium. When optical absorption is significant, such approach does not work and a microscopic consideration is necessary.[19]

Equations (2) and (5) are rather general and can be applied to pump pulses of arbitrary duration including cw light as a limiting case, provided that the electric field entering the interaction potential [Eq. (2)] is the total electric field, i.e., is the sum of the incident $\boldsymbol{\mathcal{E}}^I$ and scattered $\boldsymbol{\mathcal{E}}^S$ electric fields. Despite the smallness of the scattering field in comparison with the incident one, it is not always allowed to neglect $\boldsymbol{\mathcal{E}}^S$ in Eq. (2). When considering the stimulated RS of cw light, it is the term containing the product $(\mathcal{E}_i^I)^*\mathcal{E}_k^S$ is responsible for the scattering (and for the magnon generation) because $\boldsymbol{\mathcal{E}}^I$ and $\boldsymbol{\mathcal{E}}^S$ oscillate at different frequencies. Quite different situation takes place in the case of impulsive stimulated RS.[15] In this case, the spectral width of excitation pulses exceeds the spin-wave frequency. For this reason, in the impulsive stimulated scattering process, higher-frequency photons from a pump pulse are coherently scattered into lower-frequency photons within the pump pulse bandwidth and propagated in the same direction but with slightly smaller wave vector magnitude. Thus, ISRS is a forward-scattering process which is stimulated because the Stokes frequency is contained within the bandwidth of the incoming pulse. The pump pulse is asymmetrically redshifted through ISRS when it leaves the sample. Since the intensity of the scattered light is small as compared to the incident one, the scattered electric field can be neglected in the interaction potential. For this reason, we will neglect the scattered electric field in the following consideration.

Equation (5) can be applied to any instant of time including time within the pump pulse, $t<\tau$. However, experimentally, light-induced spin polarization can be reliably measured only at times after the pump pulse, $t>\tau$. For this reason, it takes sense to calculate the induced magnetization at these times. In this case, it is allowed to take the limit of zero pump pulse duration, $\tau\to 0$.

Integrating Eq. (5) over the pulse width and accounting for Eq. (3), we obtain b_r just after the photoexcitation $(t=0^+)$,

$$b_r(0^+)=i\frac{I_0}{4nc}\sum_{ik}\frac{\partial\delta\epsilon_{ik}}{\partial b_r^*}e_i^*e_k. \tag{6}$$

Thus, in general, immediately after the photoexcitation, the spins are rotated away from the equilibrium directions. This is simply a consequence of the fact that the electron spins can straightforwardly participate in the optical transitions induced by the light pulse.

Equations (5) and (6) are applicable to all coherent collective excitations, which are characterized by nonzero values of canonical variables. (This is not the case for coherent two-magnon excitations.[22]) Since the impulsive generation of coherent phonons was studied most fully in comparison with other collective excitations, it is pertinent to establish a relation between the approach based on the Hamiltonian equations [Eq. (5)] and the traditional one based on the harmonic-oscillator equations for ion displacements.[15–20] First, we note that the nonzero $b_r(0^+)$ predicted by Eq. (6) is not in contradiction with the physically evident fact that the displacements of the ions are zero at $t=0^+$. Equation (5) can be used to describe the phonon generation, if the canonical variables are defined as $b_{r,j}=(q_{r,j}+ip_{r,j})/\sqrt{2}$, where $q_{r,j}$ and $p_{r,j}$ are the Cartesian components $(j=x,y,z)$ of the displacement and momentum and r enumerates ions (magnetic and nonmagnetic) in the unit cell. The phonon-induced $\delta\epsilon_{ik}$ depends only on the displacements $\{q_{r,j}\}$ and for a transparent medium is real and symmetric. Then, replacing b_r in Eq. (6) by $b_{r,j}$, we obtain that $b_{r,j}(0^+)$ is purely imaginary. This means that the ion displacements are zero at $t=0^+$ but the momenta are finite in accordance with the intuitive physical picture and rigorous considerations.[18,19] For magnons, $\delta\epsilon_{ik}$ depends, in general, on both $\mathrm{Re}\, b_r \sim M_{r,x_r}$ and $\mathrm{Im}\, b_r \sim M_{r,y_r}$. Consequently, both real and imaginary parts of $b_r(0^+)$ are finite at $t=0^+$, i.e., each spin acquires a finite rotation during a pump pulse.

In order to study the temporal evolution of the spin system, it is convenient to transform $\{b_r\}$ to the normal coordinates $\{Q_\alpha\}$,

$$Q_\alpha = \sum_r (u_{r\alpha} b_r + v_{r\alpha} b_r^*), \tag{7}$$

where $\alpha=1,\ldots,N$ is the branch index and $u_{r\alpha}$ and $v_{r\alpha}$ are matrices, satisfying the following relations:

$$\sum_r (u_{r\alpha} u^*_{r\alpha'} - v_{r\alpha} v^*_{r\alpha'}) = \delta_{\alpha\alpha'}, \tag{8}$$

$$\sum_r (u_{r\alpha} v_{r\alpha'} - v_{r\alpha} u_{r\alpha'}) = 0. \tag{9}$$

These relations guarantee that transformation (7) is canonical. Two additional relations follow from the condition that Q_α is the normal coordinate,

$$\sum_\alpha (u_{r'\alpha} u^*_{r\alpha} - v_{r\alpha} v^*_{r'\alpha}) = \delta_{rr'}, \tag{10}$$

$$\sum_\alpha (u^*_{r\alpha} v_{r'\alpha} - v_{r\alpha} u^*_{r'\alpha}) = 0. \tag{11}$$

By using Eqs. (10) and (11) one can inverse Eq. (7),

$$b_r = \sum_\alpha (u^*_{r\alpha} Q_\alpha - v_{r\alpha} Q^*_\alpha). \tag{12}$$

After the transformation to the normal coordinates, the Hamiltonian of the spin system has the simple form,

$$H_0 = \sum_\alpha \Omega_\alpha Q_\alpha Q^*_\alpha, \tag{13}$$

where Ω_α is the frequency of the spin-wave mode α. Being the canonical variables, the normal coordinates also obey the Hamilton equations,

$$\frac{dQ_\alpha}{dt} + i\Omega_\alpha Q_\alpha = -i\frac{\partial V}{\partial Q^*_\alpha}. \tag{14}$$

The modulation of the dielectric permittivity by the spin waves is given by

$$\delta\epsilon_{ik} = \sum_\alpha (P^\alpha_{ik} Q_\alpha + R^\alpha_{ik} Q^*_\alpha), \tag{15}$$

where

$$P^\alpha_{ik} = \partial\delta\epsilon_{ik}/\partial Q_\alpha, \quad R^\alpha_{ik} = \partial\delta\epsilon_{ik}/\partial Q^*_\alpha \tag{16}$$

are scattering matrices for the mode α. Note that only relative phases of P_{ik} and R_{ik} are physically significant. The Hermiticity of $\delta\epsilon_{ik}$ gives

$$P^\alpha_{ik} = R^{\alpha *}_{ki}. \tag{17}$$

Solving Eq. (14) for Q_α with account of Eqs. (2), (3), and (15), we obtain

$$Q_\alpha = i\frac{I_0}{4nc}\sum_{ik} R^\alpha_{ik} e_i e^*_k e^{-i\Omega_\alpha t}. \tag{18}$$

The effect of the spin waves on the probe beam relies on the modulation of the dielectric permittivity by the pump-induced spin precession. Using Eqs. (15) and (18), we obtain

$$\delta\epsilon^{ind}_{ik} = i\frac{I_0}{4nc}\sum_{nm\alpha} (P^\alpha_{ik} R^\alpha_{nm} e^{-i\Omega_\alpha t} - P^\alpha_{nm} R^\alpha_{ik} e^{i\Omega_\alpha t}) e_n e^*_m. \tag{19}$$

Up to this point, we did not take into consideration that the time-reversal symmetry in magnetically ordered media is broken. To reveal features of $\delta\epsilon^{ind}_{ik}$, specific for the magnetic medium, we use the symmetry properties of the scattering matrices P^α_{ik} and R^α_{ik} with respect to time reversal. For the transparent magnetic crystal, Onsager's principle gives[26]

$$P_{ik}(\{\mathbf{M}^{(0)}_r\}) = R_{ki}(\{-\mathbf{M}^{(0)}_r\}). \tag{20}$$

From here on, we omit the branch index α for brevity.

By using relations (17) and (20), the scattering matrices can be represented as

$$P_{ik} = iA_{ik} + iS_{ik} + a_{ik} + s_{ik}, \tag{21}$$

$$R_{ik} = iA_{ik} - iS_{ik} - a_{ik} + s_{ik}, \tag{22}$$

where S_{ik} and s_{ik} (A_{ik} and a_{ik}) are symmetric (antisymmetric) in the indices i and k tensors. The tensors A_{ik} and S_{ik} (a_{ik} and s_{ik}) are time odd (time even). Thus, the distinctive feature of magnetic media is that the scattering matrices contain time-odd parts.

Substituting Eqs. (21) and (22) into Eq. (19), we obtain

$$\delta\epsilon^{ind}_{ik} = \frac{I_0}{2nc}[(d^a_{ik} + D^a_{ik} + D^s_{ik} + d^s_{ik})\sin\Omega t + (C^a_{ik} + c^a_{ik} + c^s_{ik} + C^s_{ik})\cos\Omega t], \tag{23}$$

where the superscript s (a) denotes symmetric (antisymmetric) under the permutations of the indices i and k parts of the tensors. The time-odd tensors are given by

$$D^s_{ik} = (S_{ik}\mathbf{a} - s_{ik}\mathbf{A}) \cdot \boldsymbol{h}, \tag{24}$$

$$D^a_{ik} = i\sum_{nm} (A_{ik}s_{nm} - a_{ik}S_{nm})e_n e^*_m, \quad (25)$$

$$C^s_{ik} = \sum_{nm} (s_{ik}S_{nm} - S_{ik}s_{nm})e_n e^*_m, \quad (26)$$

$$C^a_{ik} = i(A_{ik}\mathbf{a} - a_{ik}\mathbf{A}) \cdot \boldsymbol{h}, \quad (27)$$

where the vectors **a** and **A** are defined through the relations $a_{ik}=e_{ikl}a_l$ and $A_{ik}=e_{ikl}A_l$. Similar expressions hold for the time-even tensors,

$$d^s_{ik} = \sum_{nm} (S_{ik}S_{nm} + s_{ik}s_{nm})e_n e^*_m, \quad (28)$$

$$d^a_{ik} = - i(A_{ik}\mathbf{A} + a_{ik}\mathbf{a}) \cdot \boldsymbol{h}, \quad (29)$$

$$c^s_{ik} = (S_{ik}\mathbf{A} + s_{ik}\mathbf{a}) \cdot \boldsymbol{h}, \quad (30)$$

$$c^a_{ik} = i\sum_{nm} (A_{ik}S_{nm} + a_{ik}s_{nm})e_n e^*_m. \quad (31)$$

It follows from Eq. (23) that optical effects originated in $\delta\epsilon^{ind}_{ik}$ and detected by a probe pulse can be classified with respect to the time-reversal operation as time odd and time even. The former change sign under the transformation $\{\mathbf{M}^{(0)}_r\} \rightarrow \{-\mathbf{M}^{(0)}_r\}$, while the latter remain unchanged. The usefulness of representations (21) and (22) for the scattering matrices is that they allow one to determine time parity of different contributions to $\delta\epsilon^{ind}_{ik}$ by using Eqs. (24)–(31).

The second term (with $\cos \Omega t$ dependence) in Eq. (23) is a distinctive feature of coherent collective excitations associated with electron degrees of freedom because these degrees of freedom (unlike phonons) straightforwardly participate in optical transitions. Such dependence is consistent with Eq. (6), which shows that immediately after the photoexcitation, the spin rotations are finite.

For a medium, possessing the time-reversal symmetry, only the tensors s_{ik} and a_{ik} are nonzero in Eqs. (21) and (22). For phonons, in addition, $a_{ik}=0$ and Eq. (23) reduces to

$$\delta\epsilon^{ind}_{ik} = \frac{I_0}{2nc} s_{ik} \sum_{nm} s_{nm} e_n e^*_m \sin \Omega t. \quad (32)$$

Note that in real experiments, even in transparent crystals, in addition to the precessional spin motion induced through ISRS, there are another contributions to the nonequilibrium magnetization induced through the inverse Faraday effect (IFE) and inverse Cotton–Mouton effect (ICME).[23] These effects are nonlinear optical effects creating nonequilibrium magnetization (but not magnons) in nonabsorbing media without scattering of light (unlike ISRS). This means that after the interaction with spins, the spectral distribution of light does not change. Only average number of photons decreases thus reflecting the energy transfer between light and spin system. Microscopic mechanisms of these effects essentially differ from that of ISRS. IFE and ICME originate in optical (dipole) coherence.[13] The magnetization induced through these effects arises due to a light-induced change in the ground state spin population rather than in the ground state spin coherence (Raman coherence). As a consequence, these mechanisms do not lead to the excitation of the spin precession and spin waves. IFE and ICME can play appreciable role only when the optical dephasing time $T_2 \gg \tau$. This is not the case for magnetic solids. For this reason, we expect that contributions of these effects to the photoinduced magnetization are negligibly small for femtosecond light pulses.

The scattering matrices P_{ik} and R_{ik} contain all information for the dependence of the spin-wave generation on the light polarization, crystal orientation, and magnetic structure. In order to calculate these matrices, one need to know the modulation of the dielectric permittivity $\delta\epsilon_{ik}=\epsilon_{ik}-\epsilon^{(0)}_{ik}$ induced by the deviation of the spin system from equilibrium. $\delta\epsilon_{ik}$ can be expanded in powers of the sublattice magnetizations.[21] To reveal distinctive features of the magnon generation in more detail, we will perform the explicit calculations of the light-induced spin precession for a simple cubic ferromagnet and a rutile-type structure antiferromagnet.

III. EXAMPLES

A. Cubic ferromagnet

For the simple cubic ferromagnet $(N=1)$, the magnetization-dependent part of ϵ_{ik} is given by[21]

$$\epsilon_{ik} = iK\sum_{l} e_{ikl}M_l + \sum_{ln} G_{ikln}M_l M_n, \quad (33)$$

where the first and second terms in the right-hand side describe the Faraday and the Cotton–Mouton effects, respectively.

For simplicity, we consider the special case when an external magnetic field **H** and the equilibrium magnetization $\mathbf{M}_0$ are directed along the [001] z axis. Then, the change in the permittivity as a function of the spin-wave components $M_x(t)$ and $M_y(t)$ reads

$$\delta\epsilon_{ik} = \sum_{l=x,y} (iKe_{ikl}M_l + 2G_{iklz}M_0 M_l). \quad (34)$$

The ferromagnet possesses only one spin-wave eigenmode with the frequency $\Omega=\gamma H$ and the normal coordinate,

$$Q = (2\gamma M_0)^{-1/2}(M_x + iM_y). \quad (35)$$

Using Eqs. (34) and (35), we obtain the scattering matrices,

$$P_{ik} = \sqrt{\frac{\gamma M_0}{2}}[K(ie_{ikx} + e_{iky}) + 2M_0(G_{ikxz} - iG_{ikyz})], \quad (36)$$

$$R_{ik} = \sqrt{\frac{\gamma M_0}{2}}[K(ie_{ikx} - e_{iky}) + 2M_0(G_{ikxz} + iG_{ikyz})]. \quad (37)$$

Then, using Eqs. (18) and (35), we calculate the light-induced spin-wave components,

$$M_x(t) = - c_1 \cos \Omega t + c_2 \sin \Omega t, \quad (38)$$

$$M_y(t) = c_2 \cos \Omega t + c_1 \sin \Omega t, \tag{39}$$

where

$$c_1 = \beta(Kh_y + GM_0\Sigma_{yz}), \tag{40}$$

$$c_2 = \beta(Kh_x + GM_0\Sigma_{xz}), \tag{41}$$

with $G=G_{xzxz}=G_{yzyz}$ and $\beta=\gamma M_0 I_0/(4nc)$. Note that for the cubic ferromagnet, the tensor G_{ikln} has three independent components, but only one of them determines symmetric one-magnon RS when $\mathbf{M}_0 \| [001]$ (see, e.g., Ref. 21). According to Eqs. (38)–(41), the "initial" magnetization rotational displacement $\Delta\mathbf{M}=\{M_x(0^+), M_y(0^+)\}=\{-c_1, c_2\}$ consists of two contributions: $\Delta\mathbf{M}=\Delta\mathbf{M}_K+\Delta\mathbf{M}_G$, which are proportional to the Faraday and the Cotton–Mouton magneto-optical constants, respectively. $\Delta\mathbf{M}_K$ vanishes for linearly polarized light ($\mathbf{h}=0$), while $\Delta\mathbf{M}_K$ can be nonzero for any light polarization. Typically, $K \gg G$, so that for circularly polarized light, $\Delta\mathbf{M}_K$ dominates. It can be detected by measuring the Faraday rotation of a linearly polarized probe pulse. However, the detection is possible only when the pump and probe beams propagate in different directions. As follows from Eqs. (38)–(41), $\Delta\mathbf{M}_K \cdot \mathbf{h}=0$, i.e., $\Delta\mathbf{M}_K$ is perpendicular to the direction of the pump pulse propagation. This can be interpreted as follows. In the cubic ferromagnet, a pulse of circularly polarized light acts on spins as an impulsive effective magnetic field directed along the pulse propagation, i.e., along $\mathbf{h}$. Such magnetic field induces the magnetization rotation $\Delta\mathbf{M}_K \sim \mathbf{M}_0 \times \mathbf{h}$. It follows from this relation that $\Delta\mathbf{M}_K$ is perpendicular to $\mathbf{h}$. For this reason, $\Delta\mathbf{M}_K$ does not cause the Faraday rotation of the probe pulse polarization when the probe propagates along the pump pulse. In contrast, $\Delta\mathbf{M}_G$ can be detected in the collinear geometry, however, the Faraday angle will be smaller ($\sim KG$) in this case.

Substituting Eqs. (38) and (39) into Eq. (34), we obtain the change in the permittivity, induced by the spin wave,

$$\delta\epsilon_{ik}^{ind} = \beta w_{ik}^{s} \sin \Omega t + \beta w_{ik}^{c} \cos \Omega t, \tag{42}$$

where

$$w_{ik}^{s} = iK^2(h_x e_{ikx} + h_y e_{iky}) + 2iKGM_0(e_{ikx}\Sigma_{xz} + e_{iky}\Sigma_{yz}) + 2KM_0(G_{ikxz}h_x + G_{ikyz}h_y) + 4GM_0^2(G_{ikxz}\Sigma_{xz} + G_{ikyz}\Sigma_{yz}), \tag{43}$$

$$w_{ik}^{c} = iK^2(h_y e_{ikx} - h_x e_{iky}) + 2iKGM_0(e_{ikx}\Sigma_{yz} - e_{iky}\Sigma_{xz}) + 2KM_0(G_{ikxz}h_y - G_{ikyz}h_x) + 4GM_0^2(G_{ikxz}\Sigma_{yz} - G_{ikyz}\Sigma_{xz}). \tag{44}$$

The order of terms in Eqs. (43) and (44) corresponds to that in Eq. (23). So, the first terms in Eqs. (43) and (44) (proportional to K^2) correspond to the antisymmetric tensors d_{ik}^a and C_{ik}^a, respectively, and have different time parity, i.e., behave differently under the transformation $\mathbf{M}_0 \to -\mathbf{M}_0$. As we have already noted, these terms, potentially, give main contribution to the rotation of the probe pulse polarization. The rotation angle ϕ scales as $ie_{ikl}\delta\epsilon_{ik}^a k_l$, where k_l is a component of the wave vector of the probe light. Then, the contribution of the K^2 terms to ϕ depends on time as

$$\phi \propto K^2[(\mathbf{k}\cdot\mathbf{h})\sin \Omega t + (\mathbf{k}\times\mathbf{h})_z \cos \Omega t]. \tag{45}$$

It follows from this equation that in the cubic ferromagnet, the spin rotation at $t=0^+$ ($\cos \Omega t$ contribution) can be detected only when the pump and probe beams are noncollinear. This restriction relaxes in crystals of lower symmetry. However, in general, light is elliptically polarized in such crystals ($h<1$) and ϕ becomes smaller. As we have already noted, the two terms in Eq. (45) behave differently under time reversal: the first term reverses sign, but the second does not. This follows from different time parity of the tensors d_{ik}^a and C_{ik}^a. However, it is difficult to establish these symmetry properties from explicit expressions (43)–(45).

Note that the magneto-optical constants K and G define the Faraday rotation and magnetic linear birefringence, respectively, and can be determined by measurements of these effects. In magnetic dielectrics, $KM_0 \sim 0.001$ and GM_0/K usually lies in the range of 0.1–1. Thus, the magnetic linear birefringence is relatively strong in magnetically ordered media.[27] It follows from this fact that linearly and circularly polarized light can induce spin waves of comparable amplitude.

B. Two-sublattice antiferromagnet

As an example, we consider an antiferromagnet of rutile-type structure (of D_{4h} point group symmetry), which has two spins per unit cell ($N=2$) oriented along + and $-z$ directions of the fourfold [001] z axis. The magnon spectrum of the antiferromagnet consists of two branches, which are degenerate in zero magnetic field and have the frequency

$$\Omega = \gamma[H_A(H_A + 2H_E)]^{1/2}, \tag{46}$$

where H_E is the exchange field and H_A is the z-directed anisotropy field. Coefficients $u_{r\alpha}$ and $v_{r\alpha}$ defining the normal coordinates of the two spin-wave modes [Eq. (7)] are given by[28,29]

$$u_{11} = u, \quad v_{21} = v, \quad u_{21} = v_{11} = 0 \quad (\alpha = 1), \tag{47a}$$

$$u_{22} = u, \quad v_{12} = v, \quad u_{12} = v_{22} = 0 \quad (\alpha = 2), \tag{47b}$$

where

$$u = \{[\gamma(H_E + H_A) + \Omega]/2\Omega\}^{1/2}, \tag{48}$$

$$v = \{[\gamma(H_E + H_A) - \Omega]/2\Omega\}^{1/2}. \tag{49}$$

In accordance with Eqs. (8) and (10), u and v obey the relation $u^2-v^2=1$. From Eqs. (7), (47), and (12), we obtain the normal coordinates Q_1 and Q_2 for the two modes,

$$Q_1 = ub_1 + vb_2^*, \tag{50}$$

$$Q_2 = ub_2 + vb_1^*, \tag{51}$$

where b_1 and b_2 are defined by Eq. (1). The inverse relations have the form

$$b_1 = uQ_1 - vQ_2^*, \tag{52a}$$

$$b_2 = uQ_2 - vQ_1^*. \tag{52b}$$

Noting that $|b_r| \propto |\mathbf{M}_{r,\perp}|$, where $\mathbf{M}_{r,\perp} = \{M_{r,x}, M_{r,y}\}$, one can see that the spin precession is circular when only one of the two modes is excited, but it is elliptical when a coherent superposition of the modes is induced, as in the process of ISRS. Indeed, it follows from Eqs. (52) that

$$|b_r|^2 = C_r - 2uv \,\mathrm{Re}\, Q_1 Q_2, \tag{53}$$

where C_r is time independent, while the second term oscillates in time at the frequency 2Ω. Thus, $|b_1|$ and $|b_2|$ vary with the time when a coherent superposition of the two modes is excited and becomes time independent only when Q_1 or Q_2 equals zero. This means that an elliptical spin precession is induced in the process of ISRS. The product $uv = \gamma H_E/2\Omega \gg 1$ increases with decreasing of the anisotropy thus leading to an increase of the ellipticity. Note that in the case of spontaneous RS, contributions of both magnon modes to the intensity of scattered light are summed incoherently.

To describe the light-induced spin dynamics of the antiferromagnet in more detail, it is convenient to introduce the ferromagnetic and antiferromagnetic vectors: $\mathbf{M} = (\mathbf{M}_1 + \mathbf{M}_2)$ and $\mathbf{L} = (\mathbf{M}_1 - \mathbf{M}_2)$. The components M_x and M_y characterize mainly the relative orientation of the sublattice magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$, while L_x and L_y describe their deviations away from the z axis. To calculate these components, we will start with the calculation of the normal coordinate $Q_1(t)$ and $Q_2(t)$ from Eq. (18) and then, using Eqs. (1) and (52), determine the components of $\mathbf{M}$ and $\mathbf{L}$.

The interaction of the nonequilibrium spin system of the antiferromagnet with light is given by[21,30]

$$V = K_+(M_x h_x + M_y h_y) + K_-(L_x h_y + L_y h_x) + G_+(L_x \Sigma_{xz} + L_y \Sigma_{yz}) + G_-(M_x \Sigma_{xz} + M_y \Sigma_{yz}), \tag{54}$$

where $K_\pm$ and $G_\pm$ are the magneto-optical constants. Using Eqs. (1), (12), and (18), we obtain

$$M_x = -m_1 \cos \Omega t + l_2 (u-v)^2 \sin \Omega t, \tag{55}$$

$$M_y = m_2 \cos \Omega t + l_1 (u-v)^2 \sin \Omega t, \tag{56}$$

$$L_x = -l_1 \cos \Omega t + m_2 (u+v)^2 \sin \Omega t, \tag{57}$$

$$L_y = l_2 \cos \Omega t + m_1 (u+v)^2 \sin \Omega t, \tag{58}$$

where

$$m_1 = \beta(K_- h_x + G_+ \Sigma_{yz}), \tag{59}$$

$$m_2 = \beta(K_- h_y + G_+ \Sigma_{xz}), \tag{60}$$

$$l_1 = \beta(K_+ h_y + G_- \Sigma_{yz}), \tag{61}$$

$$l_2 = \beta(K_+ h_x + G_- \Sigma_{xz}). \tag{62}$$

Equations (55)–(58) show that, similar to the ferromagnet, the spins in the antiferromagnet are rotated away from the equilibrium directions just after the light pulse and this rotation is independent of H_E and H_A. As is seen from Eqs. (55) and (56), the parameters m_1 and m_2 determine the angle between the sublattice magnetizations just after the pump pulse, i.e., the degree of their noncollinearity. These parameters depend on the polarization of the pump pulse and the magneto-optical constants K_- and G_+ [Eqs. (59) and (60)]. However, unlike the ferromagnet, the induced spin precession depends on H_E and H_A through the parameters u and v. Equations (57) and (58) show that the initial (at $t=0^+$) noncollinearity leads to the enhancement of the spin rotation away from the z axis in the subsequent spin motion. When $H_A \ll H_E$,

$$(u-v)^2 \simeq (H_A/2H_E)^{1/2} \ll 1, \tag{63}$$

$$(u+v)^2 \simeq (2H_E/H_A)^{1/2} \gg 1, \tag{64}$$

and one can see again from Eqs. (57) and (58) that the ellipticity of the spin precession in the superposition of the modes of the antiferromagnet increases with the mode softening ($\Omega \to 0$). The ellipticity also depends on the light polarization through the parameters m_1 and m_2. Thus, large values of the magneto-optical constants K_- and G_+ in combination with the relatively small anisotropy field ($H_A \ll H_E$) enhance the deviations of the sublattice magnetizations from the equilibrium directions. Experimentally, this will manifest itself as an increase of $\sin \Omega t$ contribution to optical effects detected by probe light in comparison with $\cos \Omega t$ contribution. This effect can also be interpreted in terms of angular momentum transfer between the two magnetic sublattices, which is due to the intersublattice exchange interaction. Initially, due to the action of the light pulse, each spin sublattice acquires a small amount of angular momentum, which depends on the magneto-optical constants of the antiferromagnet and polarization of light, but is independent of the parameters of the spin precession itself, i.e., is independent of u and v. If the intersublattice exchange interaction was absent ($u=1, v=0$), but the intrasublattice one still preserved, the spin precession of each sublattice would be circular. However, due to the intersublattice exchange interaction, the angular momentum of each sublattice varies with the time, while the total angular momentum remains unchanged. This effect reveals itself in the magnification of $\sin \Omega t$ contributions in Eqs. (57) and (58) as compared to $\cos \Omega t$ ones.

In a nonzero external magnetic field, the degeneracy of the spin-wave modes is lifted and the spin precession in each mode becomes elliptical. The same is true about spin-wave modes of two-sublattice antiferromagnets of lower symmetry even in zero magnetic field. A simple extension of the above analysis to the nondegenerate case shows again that the ellipticity of the spin precession in a spin-wave mode leads to the increase of $\sin \Omega t$ contributions to the induced permittivity. This facilitates the detection of the mode by probe light.

The ellipticity increases when passing from high- to low-frequency modes of spin-wave spectrum of an antiferromagnet.[31] For this reason, low-frequency modes of antiferromagnets are best suited for studies of the spin-wave generation through ISRS. This conclusion is supported by recent experiments,[12] where the excitation of the low-frequency spin-wave mode in the easy-plane antiferromagnet

$FeBO_3$ was observed, but the high-frequency mode, which has 1 order of magnitude smaller ellipticity, was not detected. Note that the impact of the ellipticity on the intensity of spontaneous RS is known for a long time,[31] but to our knowledge has never been discussed in the context of the spin-wave generation. Though related on the microscopic level, ISRS and spontaneous RS are essentially different phenomena.

IV. CONCLUSION

In this paper, we have studied the ultrafast optical generation of spin waves in transparent magnetic dielectrics through ISRS. Our approach is based on the Hamilton equations for the normal coordinates of the spin system. We have calculated the modulation of the optical dielectric permittivity induced by the spin waves, which is straightforwardly related to optical effects detected by a probe pulse. The oscillating in time permittivity can be represented as a sum of cosine and sine contributions. The former depends on the magneto-optical constants and light polarization but is independent of the parameters of the spin precession, while the latter increases with the ellipticity of the spin precession.

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