Giant phonon-induced effective magnetic fields in 4f paramagnets

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(Dated: July 22, 2020)

We present a mechanism by which circularly driven phonon modes in the rare-earth trihalides generate giant effective magnetic fields acting on the paramagnetic 4f spins. With cerium trichloride (CeCl₃) as our model system, we calculate the coherent phonon dynamics in response to the excitation by an ultrashort terahertz pulse using a combination of first-principles calculations and phenomenological modeling. We find that effective magnetic fields of over 100 T can possibly be generated that polarize the spins for experimentally accessible pulse energies. This mechanism potentially creates a way to control the magnetic and electrical order of ferromagnets and ferroelectrics through interfacial coupling with the phonon-induced magnetization in heterostructures.

Ultrashort laser pulses are able to change the magnetic order of materials within picosecond timescales, orders of magnitude faster than conventional spin-based devices operating on nanosecond timescales [1, 2]. Usually, the electromagnetic field components of a laser pulse couple to electronic degrees of freedom of the magnetic ions, leading to the notion of ultrafast opto-magnetism [3–7]. Recent studies have demonstrated that light can also couple to the spins indirectly by exciting coherent vibrational modes of the crystal lattice (phonons) that transfer angular momentum to the magnetic ions [8–13] or modulate the crystal structure into a transient state of modified magnetic order [14–22]. These phono-magnetic methods promise higher selectivity and lower dissipation than techniques based on opto-magnetic effects due to the lower energy of the excitation. A central challenge is to produce effective magnetic fields that are strong enough to induce qualitative changes in the magnetic order, and common fields for optical and phononic driving have so far ranged in the order of milli to a few tesla [8, 9, 13, 23].

Here, we propose that circularly driven phonons in the rare-earth trihalides produce effective magnetic fields that exceed those previously seen by several orders of magnitude. We predict at the example of CeCl₃ that effective magnetic fields of over 100 tesla, which polarize the paramagnetically disordered spins, should be achievable for laser energies well within the damage threshold of the crystal. The mechanism allows for bidirectional control of the induced magnetization and possibly creates a way to control the magnetic and electrical order of ferroic materials through interfacial coupling with the phonon-induced magnetization in heterostructures.

I. PROPERTIES OF CERIUM TRICHLORIDE

Rare-earth trihalides are a class of 4f paramagnets with formula unit RH₃. CeCl₃ (R = Ce, H = Cl) is a representative of this class of materials that crystallizes in the hexagonal P6₃/m structure with an electronic band gap of 4.2 eV [24]. We chose CeCl₃ as our model system, because the primitive unit cell consists of only 8 atoms (Fig. 1(a)), resulting in a small number of 21 optical phonon modes characterized by the irreducible representations 2A_u + 1A_g + 2B_u + 2B_g + 1E_g + 3E_u + 2E_u + 1E_g in its 6/m point group. Early Raman studies have shown that the polarization of the 4f electrons in an external magnetic field leads to a splitting of the doubly degenerate E₁g and E₂g phonon modes into left- and right-handed circularly polarized [25, 26], see Fig. 1(b).

The splitting is given by ∆Ω(B) = ∆Ω sat sinh(sµB/(2k_BT)), where ∆Ω sat is the saturation splitting, s is the spectroscopic splitting factor of the ground state, µ_B is the Bohr magneton, B is the external magnetic field, k_B is the Boltzmann constant, and T is the temperature [27, 28]. It has been suggested that also the infrared-active E₁u phonon modes split in the same way [29], yet no experimental infrared spectroscopy measurements had been performed at that time. The infrared-active E₁u modes map into the same E’ representation at the local 6 symmetry of the Ce ions as the Raman-active E₂g modes, for which phonon splittings have been measured, and should therefore have the same effect on the paramagnetic spins.

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FIG. 1. Structure and properties of CeCl₃. (a) Hexagonal P6₃/m structure of paramagnetic CeCl₃. (b) Schematic splitting of a doubly degenerate phonon mode with frequency Ω₀ into right and left-handed circularly polarized components in an external magnetic field at liquid helium temperatures, saturating at frequencies Ω₊ and Ω₋. At higher temperatures, the phonon splitting saturates at higher magnetic fields.
Infrared-active phonon modes possess an electric dipole moment and can therefore be resonantly excited by the electric field component of a laser pulse to yield large vibrational amplitudes. We will explore in this work how circularly driven \( E_{1u} \) phonons act on the spins through the inverse of the spin-phonon coupling.

**II. SPIN-PHONON COUPLING AND PHONON DYNAMICS**

Microscopically, the motions of the ions along the eigenvectors of the doubly degenerate phonon modes (illustrated in Figs. 2(a) and (b)) modify the crystal electric field around the Ce ions and induce virtual transitions between the \( J = 5/2 \) ground state doublet and higher-lying states \( [27] \) (Fig. 2(c)) that act as an effective magnetic field on the Ce spins. In addition, the circular motions act as atomistic current loops that produce local orbital magnetic moments with opposite directions for Ce and Cl, respectively (Fig. 2(d)). The orbital magnetic moments produced by the motions of the lighter Cl ions overcompensate those of the Ce ions, leading to a net magnetic field produced by the doubly degenerate phonon modes \([9, 12, 30]\). Our calculations reveal that the contribution from the phonon orbital magnetic moments is negligible compared to that from the modulation of the crystal electric field.

The phenomenological interaction Hamiltonian, \( H^{s-ph} \), can be written as

\[
H^{s-ph} = KM \cdot (Q \times \dot{Q}).
\]  

Here \( Q = (Q_a, Q_b, 0) \) contains the normal mode coordinates (amplitudes) of the two components of a doubly degenerate phonon mode, \( Q_a \) and \( Q_b \), in the \( ab \) plane of the crystal, see Fig. 1(a). \( Q \times \dot{Q} \) is the phonon angular momentum, \( M \) is the magnetization, and \( K \) is the spin-phonon coupling \([31–33]\), containing both contributions from the crystal electric field and phonon orbital magnetic moments, \( K = K_{CEF} + K_{POM} \). As light couples to phonon modes close to the center of the Brillouin zone, we may neglect any wavevector dependence. The phonon mode acts as an effective magnetic field, \( B \), on the spins of the material and can be written as

\[
B = V_c^{-1} \partial H^{s-ph} / (\partial M) = KV_c^{-1}Q \times \dot{Q},
\]  

where \( V_c \) is the volume of the unit cell. Phenomenologically, this interaction is a phonon analogue of the inverse Faraday effect in optics \([13]\), which is known to induce magnetizations in paramagnets \([34–37]\). Here, we assume that the thermodynamic relation between the effective magnetic field and paramagnetic magnetization is valid. For the opto-magnetic inverse Faraday effect, the relation holds for nanosecond timescales \([34, 35]\), however additional diamagnetic effects from the unquenching of electronic orbital moments come into play on femtosecond timescales that cannot be described by an effective magnetic field picture \([36–38]\). The phonon analogue to the inverse Faraday effect lies between these limits and we therefore estimate that the effective magnetic field picture is a reasonable approximation for the induced magnetization.

We obtain the amplitudes of the phonon mode components by solving the equation of motion

\[
\ddot{Q} + 2\kappa \dot{Q} + \Omega^2 Q = ZE(t).
\]  

Here \( \kappa \) is the linewidth of the phonon mode, \( \Omega \) its eigenfrequency, and \( Z = \sum_n Z_n^a q_n / \sqrt{M_n} \) its mode effective charge, where \( Z_n \) is the Born effective charge tensor, \( q_n \) the eigenvector, and \( M_n \) the atomic mass of ion \( n \), and the sum runs over all ions in the unit cell. We model the circularly polarized terahertz pulse as \( E(t) = (E(t), E(t - 2\pi / (4\Omega)), 0) / \sqrt{2} \), where \( E(t) = E_0 \exp(-t^2 / (2(\tau / \sqrt{8\ln 2})^2)) \cos(\omega_0 t) \). \( E_0 \) is the peak electric field, \( \omega_0 \) the center frequency, and \( \tau \) is the full width at half maximum duration of the pulse. Here, the two perpendicular components of the doubly degenerate phonon mode are excited with a quarter-period difference, resulting in circular polarization. The effective peak electric field in each component of the circular polarization is \( E_0 / \sqrt{2} \).

**COMPUTATIONAL DETAILS**

We calculate the phonon eigenfrequencies and eigenvectors, and Born effective charges from first principles, using the density functional perturbation theory formalism \([39, 40]\) as implemented in the Vienna ab-initio simulation package (VASP) \([41, 42]\). We use the VASP projector augmented wave (PAW) pseudopotentials with
valence electron configurations Ce (6s²5s²5p⁶5d⁴4f¹) and Cl (3p³3s²) and converge the Hellmann-Feynman forces to 25 µeV/Å. For the 8-atom unit cell, we use a plane-wave energy cut-off of 600 eV, and a 4×4×7 gamma-centered k-point mesh to sample the Brillouin zone. For the exchange-correlation functional, we choose the Perdew-Burke-Ernzerhof revised for solids (PBEsol) form of the generalized gradient approximation (GGA) [43]. We perform nonmagnetic calculations to obtain the structural and dynamical properties of CeCl₃, which is an approximation to its paramagnetic state. The lattice constants of our fully relaxed hexagonal structure (space group P6₃/m, point group 6/m) of a = 4.21 Å and c = 7.38 Å with a unit-cell volume of V = 199 Å³ match reasonably well with experimental values [44]. The phonon eigenfrequencies that we obtain using a nonmagnetic unit cell match the experimental values within approximately 10% [26, 29].

The spin-phonon coupling can be extracted from the experimentally found splittings of the doubly degenerate phonon modes as \( K_{CEF} M_s = \Delta \Omega_s / (4 \pi) \), where \( M_s \) is the saturation magnetization [33]. Experimental saturation splittings in the rare-earth trihalides for the Raman-active modes range between 10 cm⁻¹ and 25 cm⁻¹ [25, 26], equivalent to 0.3 THz and 0.75 THz. Because the infrared-active modes change the local symmetry of the magnetic Ce ion in the same way, we expect a similar strength of the spin-phonon coupling as for the Raman-active modes and use an average of the experimentally found values of \( \Delta \Omega_s / (2 \pi) = 0.52 \) THz. With a saturation magnetization of \( M_s = 2 \mu_B / V_c \) for CeCl₃, the spin-phonon coupling is \( K_{CEF} = 0.13 \) THz/\( \mu_B \). In comparison, we find the spin-phonon coupling arising from the phonon orbital magnetic moments, following the formalism of Refs. [9, 12], to be \( K_{POM} < 10^{-6} \) THz/\( \mu_B \) for CeCl₃ and we may therefore neglect this effect here. For the phonon linewidth, \( \kappa \), we assume a phenomenological value of 5% of the phonon frequency that matches those typically found in rare-earth trihalides [25, 26].

III. PHONON-INDUCED EFFECTIVE MAGNETIC FIELDS

In the following, we evaluate the effective magnetic fields produced by the two doubly degenerate infrared-active \( E_{1u} \) modes in CeCl₃ with eigenfrequencies of 5.9 and 4.8 THz. We find the mode effective charges of these modes to be 0.24\( e \) and 0.66\( e \), respectively, where \( e \) is the elementary charge. Fig. 3 shows the coherent phonon dynamics following the excitation by a circularly polarized terahertz pulse with a duration of \( \tau = 350 \) fs and an effective peak electric field of \( E_0 / \sqrt{2} = 5.5 \) MV/cm (corresponding to a fluence of 30 mJ/cm²). The center frequency \( \omega_0 \) is chosen to be resonant with the eigenfrequencies of the respective phonon modes. In Fig. 3(a), we show the evolutions of the phonon amplitudes \( Q_a \) according to Eq. (3). The evolutions of the \( Q_a \) components are shifted by a quarter period, respectively. The maximum amplitude of the \( E_{1u}(5.9) \) mode of \( Q_a = 0.6 \) Å/\( \sqrt{\text{amu}} \), where amu denotes the atomic mass unit, is roughly three times smaller than that of the \( E_{1u}(4.8) \) mode of \( Q_a = 2 \) Å/\( \sqrt{\text{amu}} \) due to the smaller mode effective charge and higher phonon frequency. In Fig. 3(b), we show the evolutions of the effective magnetic fields produced by the two phonon modes according to Eq. (2). We obtain a maximum effective magnetic field of \( B = 2.9 \) T for the \( E_{1u}(5.9) \) mode and 27 T for the \( E_{1u}(4.8) \) mode. This order-of-magnitude difference comes from the quadratic scaling of the effective magnetic field with the phonon amplitudes. The direction of the effective magnetic field is determined by the handedness of the phonon circular polarization, which can straightforwardly be controlled by inverting the circular polarization of the pulse.

We now investigate the magnitude of the effect on the strength of the excitation. We show the maximum amplitudes of the effective magnetic fields for a range of experimentally accessible fluences of the terahertz pulse in Fig. 3(c), where we fix the pulse duration at \( \tau = 350 \) fs [45]. The fluence \( F \) is connected...
to the peak electric field and the duration of the pulse through \( F = \tau/\sqrt{8\ln(2) c_0 c_0 \sqrt{\pi/2} E_0^2} \), where \( c_0 \) and \( c_0 \) are the speed of light and permittivity of vacuum. The effective magnetic fields depend linearly on the fluence and reach 12 T for the \( E_{1u}(5.9) \) mode and 108 T for the \( E_{1u}(4.8) \) mode at a fluence of 120 mJ/cm\(^2\) (corresponding to \( E_0/\sqrt{2} = 11 \text{ MV/cm} \)). In order to ensure experimental feasibility, we evaluate the atomic displacements along the eigenvectors of the phonon modes. The Lindemann stability criterion predicts melting of the crystal lattice when the root mean square displacements reach only 2.3% of the interatomic distance of 2\(^9\) \AA, for the \( E_{1u}(5.9) \) mode and 6.7% for the \( E_{1u}(4.8) \) mode, well below the vibrational damage threshold. Note that other effects may occur, e.g., Zener tunneling, that are not accounted for here. At these high fields, nonlinear couplings between coherently excited infrared-active modes and other vibrational degrees of freedoms come into play [47, 48]. These modes do not contribute directly to the spin-phonon coupling however, and we therefore neglect the effect of nonlinear phonics in this context. Furthermore, the centrosymmetry of CeCl\(_3\) prevents nonlinear optical effects, such as second-harmonic generation, to occur at high fluences.

Next, we look at the magnetization that can be induced through these effective magnetic fields in CeCl\(_3\), which can be detected in standard Faraday rotation experiments. The magnetization is given by \( M(B) = M_L g \mu_B B / (2k_B T) \), where \( M(x) = \coth(x) - x^{-1} \) is the Langevin function [49]. For \( B \), we input the phonon-induced effective magnetic fields of the \( E_{1u}(4.8) \) mode. We show the dependence of the induced magnetization on the fluence of the terahertz pulse and on the temperature in Fig. 4. In Fig. 4(a), we show the fluence dependence for four technologically important temperatures, the boiling temperatures of helium (4.2 K), hydrogen (20.3 K), and nitrogen (77 K), as well as room temperature (295 K). In Fig. 4(b), we show the temperature dependence for four different fluences between 30 and 120 mJ/cm\(^2\). The higher the temperature, the higher the fluences that are required to polarize the spins. At low temperatures < 10 K, even small fluences of 30 mJ/cm\(^2\) are sufficient to induce magnetizations close to the saturation value, \( M_s \). At room temperature, only a fraction of the saturation magnetization can be reached. We obtain a value of 0.17\(\mu_B\) for a fluence of 30 mJ/cm\(^2\) and 0.6\(\mu_B\) for the highest fluence of 120 mJ/cm\(^2\) investigated here.

**IV. DISCUSSION**

Our predictions can be experimentally realized in state-of-the-art tabletop setups that provide terahertz pulses in the required frequency range [45], where the phonon-induced magnetization of the material can be probed by Faraday rotation measurements. Tuning the frequency of the terahertz pulse in and out of resonance with the phonon modes can distinguish a possible contribution of the optical inverse Faraday effect to the magnetization (that should have negligible frequency dependence in this spectral range) from the phonon-induced mechanism. While we have chosen CeCl\(_3\) as our model system, the mechanism described here should be general to the entire class of rare-earth trihalides, in which strong spin-phonon couplings have been detected [25, 26], and possibly to 4f magnets in general. As an example, a similar magnitude of the spin-phonon coupling has been found in the 4f ferromagnet LiTbF\(_4\) [28] and in the 4f paramagnet Tbf\(_8\)Ga\(_5\)O\(_{12}\) [32, 33]. A future question to answer will be whether the spin-phonon couplings in 3d ferro- and antiferromagnets can reach similar magnitudes as those in 4f paramagnets. The additional functionality of giant phonon-induced effective magnetic fields in the paramagnetic phases of 3d magnets would directly impact a large variety of materials that are already being used in magnetoelectronic technologies [50].

We finally sketch possible applications, in which the phonon-induced magnetization can be exploited. The paramagnet can be incorporated into a heterostructure with materials that possess different ferroic orders, for example ferro- and antiferromagnets, or ferro- and antiferroelectrics, as is commonly done in multiferroic materials [51]. Circular driving of phonon modes then polarizes the paramagnet and the interfacial spins subsequently couple to the order of the adjacent material through interfacial exchange interaction, or interfacial magnetoelectric or Zeeman-type coupling with the induced magnetic fringe field. Through this mechanism, entirely new ways of ferroic and antiferroic switching can potentially be created.
ACKNOWLEDGMENTS

We are grateful to T. Neuman and C. Tzschaschel (Harvard University), J. Lehmann, S. Pal and N. Spaldin (ETH Zurich), and M. Fechner, A. Disa, A. von Hogen and A. Cavalleri (MPSD Hamburg) for useful discussions. This project was supported by the Swiss National Science Foundation (SNSF) under Project ID 184259 and the DARPA DRINQS Program under Award No. D18AC00014. P.N. is a Moore Inventor Fellow and gratefully acknowledges support from the Gordon and Betty Moore Foundation through Grant No. GBMF8048. Calculations were performed at the National Energy Research Scientific Computing Center (NERSC), supported by the Office of Science of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

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