Ultrafast spectroscopy and control of correlated quantum materials

By

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Abstract

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Acknowledgements

Preface

The physics of solids is, to me, one of the most important and fundamental fields of modern science. This might seem, to some, a bit of a hot take. After all, by studying condensed matter physics, one learns next to nothing about, say, the formation of the stars and planets, or the origin of the universe. Nor does one learn about life, death, consciousness, disease, ethics, God, or any other question that perhaps puzzled humanity prior to about five hundred years ago. Certainly no one would argue that condensed matter physics is quite *useless*, given that nearly every device we interact with in modern life required some condensed matter physicist somewhere along the way to make one brilliant discovery or another—yet when the human mind starts to wander, and our thoughts turn to the metaphysical, we tend to look up, not down.

In my work I have taken a quite different view. Condensed matter physics, to me, is ultimately the study of how *truly boring* objects, when brought together in large quantites, *become* interesting, seemingly in spite of themselves. When electrons are put together in a lattice and allowed to interact slightly with the massive nuclei, at low enough temperatures they pair, the low-energy excitations become gapped, and current can flow for infinite times and with absolutely zero energy loss. Those same electrons, with some other set of interactions, may instead ionize (the opposite of pairing!) to create an electrically insulating state, whose low-energy excitation spectrum is nevertheless gapless and consisting of charge-neutral spin-$^1/_2$ particles. In all such cases, these systems exist in otherwise ordinary-looking rocks, fit in the palm of a hand[1], and are more or less indistinguishable from something you might find sticking into the bottom of your shoe.

While such systems may not tell us a lot[2] about the early universe, considering these and related problems lets us ask deep, fundamental questions about the world we live in—like, why is this thing a metal, but this thing is an insulator? What do those terms even mean?—that I don't think we would try to ask otherwise. To me, focusing our attention on

[1]Hopefully, gloved.

[2]This discussion is obviously intentionally reductive. In truth there is still quite a bit one can learn about, e.g. the early universe by studying condensed matter physics, see the Kibble and Pickett [18].

these problems, despite their obviously terrestrial nature, is not a waste of time; rather, I think they remind us that even the most mundane aspects of the human experience involve a level of complexity far beyond what we are capable of understanding absent the pursuit of science.

Throughout the seven years of my Ph.D., I hope to have made a few contributions to this pursuit. As the title of this work implies, I have mainly focused on the application of ultrafast techniques to the study of correlated quantum materials, which I loosely define as those materials in which the interaction between particles is large enough so as to compete with the kinetic energy of those particles. It is in these materials that I think lies the true frontier of condensed matter physics; here, much of our basic intuition about non- or weakly-interacting theory fails, and more complicated notions of phase competition, phase separation, disorder, pairing, coherence, etc. are needed to property describe the relevant physics.

In my own view, and in the view of many scientists in this field[2], the main question for strongly correlated physics amounts to: "Given a correlated system with some defined combination of different interaction strengths, is there a general theory which allows us to predict the phase diagram of this system *a priori*?" Related of course are questions about the origins of high-T_c superconductivity, strange metallicity, quantum spin liquids, and other exotic phases that we find emerging from strongly interacting systems. Since such a theory does not currently exist, at least with the level of predictive power that I think most would find satisfactory, new advances in this field typically come directly from experiment. Ultrafast optics plays a special role in this regard, for reasons that I will explain in **??**.

Progress thus happens in this field somewhat unsystematically, with small pieces of the puzzle added at random, but not infrequenct, intervals. Usually it is either new techniques or new materials that are the driving force here. To this end, I have tried to pursue both directions in my Ph.D. Appearing also in **??** is thus a description of the materials I studied the most during my thesis, two of them, $CuBr_2$ and $CaMn_2Bi_2$ I consider criminally understudied. On the technique side, almost all of the work presented in this thesis was done using time-resolved second harmonic generation (tr-SHG), a relatively new, nonlinear optical technique which, at the most basic level, probes the point group assumed by the charge distribution function $\rho(\boldsymbol{x})$ at any given point in time. Second harmonic generation (SHG) and tr-SHG are tricky techniques, with many pitfalls both practically and theoretically; **????** are thus devoted to what I hope is a useful, if not fully compehensive, description of the technique. **??** is devoted to work that we did developing a new way to control the polarization of the light in a tr-SHG experiment using stepper motors. My hope is that these chapters are useful not only for the new student trying to build their own setup or analyze their own SHG data, but also for people for whom SHG is not a focus but nevertheless want to learn about it in slightly more detail than one would get from a typical paper or review article.

What follows, then, is a description of the three main research works I contributed during my Ph.D.. The first, which I describe in **??**, involves work that I did during my second and third years on $1T$-TaS_2, a very interesting charge density wave (CDW) material that, among other things, undergoes a mirror symmetry breaking CDW transition at $350\ \mathrm{K}$ that shows up in the SHG as a sudden distortion of the flower pattern at that temperature. Since this transition breaks mirror symmetry, two energetically degenerate domains should be present, corresponding to two opposite planar chiralities; in this work, we showed that SHG could differentiate between these two domains (i.e. the flower pattern in either domain looks different).

The second and third works, which I describe in **??** and chapter 1, in contrast to the $1T$-TaS_2 work, both involve taking the system out of equilibrium to study the dynamics. In $CaMn_2Bi_2$ (**??**), we discovered that photoexcitation causes the antiferromagnetic (AFM) order in that compound to reorient (relative to equilibrium) to a metastable state which is impossible to reach from the equilibrium state thermodynamically. Light is thus used to *control* the magnetic order in this material.

In $CuBr_2$ (chapter 1), light is not used to control the order parameter like in $CaMn_2Bi_2$, but it does excite coherent oscillations of the collective modes of the multiferroic order (electromagnons), whose frequency, amplitude, damping, etc. may be probed in tr-SHG as a function of temperature—a methodology referred to as ultrafast *spectroscopy*. In doing so, we found that one of these collective modes is actually quite special, as it is in fact the analogue of the Higgs mode of particle physics in the context of a multiferroic material.

I conclude with various remarks in chapter 2, as well as an appendix, in which I enumerate briefly all of the null-result experiments I performed during my Ph.D., in the hopes that future scientists don't have to waste time on what we already know are fruitless pursuits. If you have any questions about this or any other section of this thesis, please do not hesitate to reach out via email.

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Chapter One

Amplitude-mode electromagnon in the spin-spiral multiferroic $CuBr_2$

1.1 Preface

This chapter is based on a manuscript intended for standalone publication and modified to fit the format of this thesis. It was coauthored by myself and Baiqing Lv (as co-first authors), along with Karna Morey, Zongqi Shen, Changmin Lee, Elizabeth Donoway, Alex Liebman-Peláez, Anshul Kogar, Takashi Kurumaji, Martin Rodriguez-Vega, Rodrigo Humberto Aguilera del Toro, Mikel Arruabarrena, Batyr Ilyas, Tianchuang Luo, Peter Müller, Aritz Leonardo, Andres Ayuela, Gregory A. Fiete, Joseph G. Checkelsky, Joseph Orenstein, and Nuh Gedik. It was coauthored by myself, Ajesh Kumar, and Baiqing Lv (as co-first authors), as well as Zongqi Shen, Karna Morey, Qian Song, Riccardo Comin, Todadri Senthil, and Nuh Gedik. Myself, Baiqing Lv, Zonqi Shen, and Karna Morey took the tr-SHG measurements, under the supervision of Nuh Gedik. Myself and Ajesh Kumar did the theory and analyzed the data, under the supervision of Nuh Gedik and Todadri Senthil. Qian Song grew the samples, under the supervision of Riccardo Comin. Myself and Ajesh Kumar wrote the paper, and Nuh Gedik supervised the project.

1.2 Abstract

Below a spontaneous symmetry breaking phase transition, the relevant collective excitations may be described as fluctuations in the amplitude and phase of the order parameter, referred to as amplitude and Goldstone modes, respectively. In solids, these modes may take on a different character than the equivalent excitations in particle physics due to the diverse vacuum states accessible in condensed matter. However, the amplitude mode in particular is quite difficult to observe experimentally as it decays quickly into the lower-energy Goldstone bosons and thus has a negligible lifetime in most systems. In this work, we report evidence for a novel amplitude mode in the multiferroic material $CuBr_2$, which shows up as a coherent oscillation in the time-resolved SHG signal upon excitation with a femtosecond light pulse. Since the spiral spin order in $CuBr_2$ induces a nonzero electric dipole moment in equilibrium, the amplitude mode—which is due to fluctuations in the amplitude of the on-site spin expectation value—is an electromagnon, and thus acquires an inversion quantum number of -1. This is in stark contrast to the amplitude boson of particle physics, which has even parity. Moreover, the excitation described here represents an entirely new type of electromagnon, distinct from the traditional electromagnon in linear spin wave theory which is due to the Goldstone mode. We argue that the amplitude mode in $CuBr_2$ acquires a nontrivial lifetime due to the combination of two features: (i) the quasi-1D nature of the material, and (ii) proximity at zero temperature to a quantum critical point separating the multiferroic ground state from a topological Haldane dimer phase.

1.3 Introduction

When the ground state of a given theory fails to respect one of its symmetries, that symmetry is said to have been be broken spontaneously[8, 24]. The low-energy excitations of this ground state may then be described as excitations of the order parameter either within the subspace of degenerate ground states, or perpendicular to it; these excitations are referred to as Goldstone and amplitude modes, respectively[25] (see Fig. 1.1(a)). This paradigm describes many fundamental phenomena in both particle physics and condensed matter, and the study of these modes has thus

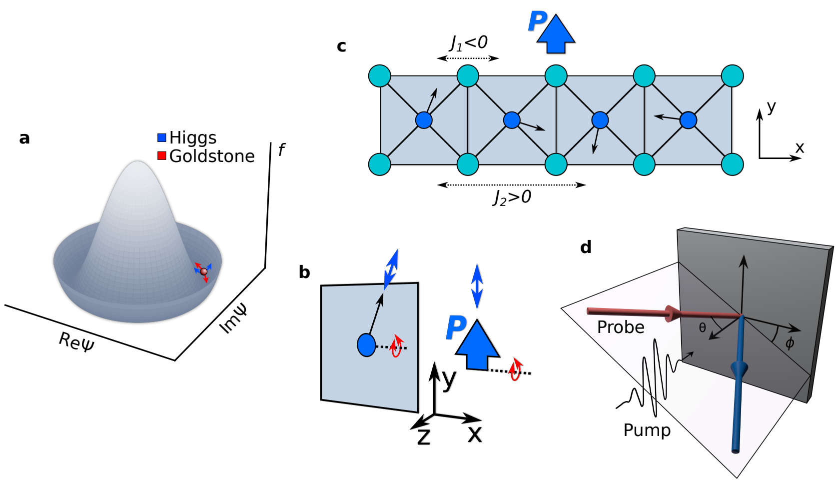

Figure 1.1: (a) Mexican hat potential with amplitude and Goldstone modes indicated. (b) $q = 0$ electromagnons in the quasi-1D spin-spiral in the spin (left) and charge (right) sectors. The amplitude and Goldstone modes are shown in blue and red, respectively. A second Goldstone mode, corresponding to uniform rotations of the spins about the z axis (which does not affect the polarization $\vec{P}$), is not shown. (c) Magnetic ground state of $CuBr_2$. The macroscopic polarization due to the spin order is depicted with a blue arrow. The axis labelled x is parallel to the nominal b axis of the crystal structure. (d) Schematic of the tr-SHG experimental geometry.

emerged as an essential pursuit in both contexts.

A rich interplay exists between these two fields due to the fact that in particle physics we are limited to a single theory (the standard model), but in condensed matter, the theory is determined by the particular system of interest and may differ dramatically from one material to another. Thus, various exotic species of amplitude modes may be studied simply by exploring different material systems with spontaneous symmetry breaking. An important example is in multiferroics, where it has been predicted[21, 22] that the amplitude mode of the magnetic order (corresponding to modulations in the amplitude of the on-site spin excitation value, see Fig. 1.1(b)) should couple to the macroscopic polarization as an electromagnon, and thus acquire a negative parity eigenvalue. This is not the case for the Higgs boson of the standard model, which is of even parity[3]. In addition to its connection to particle physics, the excitation described here is is also fundamentally different from the traditional electromagnon in multiferroics (which is due to the (pseudo-)Goldstone mode[17]), and is thus of great interest for magnetoelectric device applications. Unfortunately, like in particle physics, the amplitude mode in condensed matter is difficult to observe since it may quickly decay into Goldstone bosons upon excitation[15], and the existence of this mode in real multiferroic systems has thus remained an important open question.

In this work, we report evidence for this mode in $CuBr_2$ (a quasi-1D, spin-spiral multiferroic, see Fig. 1.1(c)), observed by launching a coherent oscillation of this mode with a near-infrared light pulse and measuring the induced modulations in the electric polarization using a delayed SHG probe pulse (Fig. 1.1(d)). We find, as expected, that the mode modulates the macroscopic polarization only along the static ordering direction, and that the frequency of the mode decreases on approaching the critical temperature of the multiferroic order. These results provide conclusive evidence for the existence of this electromagnon in $CuBr_2$, solving a decade-old puzzle and paving the way for future study of the amplitude mode in novel condensed-matter contexts.

1.4 Results

1.4.1 Equilibrium

The low-energy spin Hamiltonian of $CuBr_2$ is well approximated by the so-called frustrated 1D XXZ spin chain, where localized spin-$1/2$ electrons interact ferromagnetically ($J_1 < 0$) with nearest neighbors but antiferromagnetically ($J_2 > 0$) with next-nearest neighbors (see Fig. 1.1(c)). When these interaction strengths are of comparable magnitude, the ground state is an incommensurate magnetic spiral, where the ordering wavevector is directed along the chain direction and has the appropriate magnitude so as to balance the two competing interaction terms. According to theory developed by Katsura, Nagaosa, and Balatsky[16], when spin-orbit coupling is strong this ground state induces an electric polarization at each site n given by

$$\vec{P}^n \propto \hat{x} \times (\vec{S}^n \times \vec{S}^{n+1}), \tag{1.1}$$

where we have set the chain direction to lie along $\hat{x}$. If the spins lie in the xy plane, then Eq. 1.1 induces a macroscopic electric polarization which is equal for each bond and directed purely along the $\hat{y}$ direction (Fig. 1.1(c)).

According to powder neutron diffraction, this spiral magnetic phase is realized in $CuBr_2$ below $T_c = 75\,\mathrm{K}$ [33**?** –35], with the propogation vector (in reciprocal lattice units) given by $\vec{k} = (0, k_y, 0.5)$, with $k_y \sim 0.235$[20, 35]. A pyroelectric current turns on at this temperature as well, indicating a macroscopic electric polarization density $|\vec{P}_0|$ of about $8\,\mu\mathrm{C/m}^2$ at $10\,\mathrm{K}$[35].

In a generalized Ginzburg-Landau theory, the SHG susceptibility tensor χ_{ijk} is linearly proportional to this polarization:

$$\chi_{ijk}(T < T_c) = \chi_{ijkl}(T > T_c)P_{0l} = \chi_{ijky}(T > T_c)P_0, \tag{1.2}$$

where χ_{ijkl} is some unknown tensor with the symmetry of the high temperature phase[27], and we have used that $\vec{P}_0||\hat{y}$. Fig. 1.2 shows the temperature dependence of the SHG intensity in $CuBr_2$, indicating a pronounced, order parameter-like enhancement of the SHG intensity at T_c due to Eq. 1.2. Note that other contributions to the SHG intensity due to e.g. magnetic dipole, surface electric dipole, and electric quadrupole terms are allowed above and below T_c and thus cannot explain the intensity increase below T_c. In addition, the c-type electric dipole term purely due to the magnetic order[4] is also not allowed by the magnetic point group of the incommensurate spin spiral (see Supplementary material, section 1.8.6.3).

1.4.2 Nonequilibrium

Having thus demonstrated that the SHG intensity is a direct probe of the electric polarization in $CuBr_2$, we proceed to investigate the low-energy collective excitations in this phase. To do so, we excite the sample with a $150\,\mathrm{fs}$ near-infrared pump pulse, and then probe the SHG intensity with a second pulse delayed in time by an amount Δt. We carry out this procedure in each of four independent polarization channels ($P_{\mathrm{in}}P_{\mathrm{out}}$, $P_{\mathrm{in}}S_{\mathrm{out}}$, $S_{\mathrm{in}}P_{\mathrm{out}}$, and $S_{\mathrm{in}}S_{\mathrm{out}}$, see section 1.7), where each channel probes a different linear combination of the tensor elements χ_{ijk}. The results are shown in Fig. 1.3. Two oscillations, with different dependencies on the

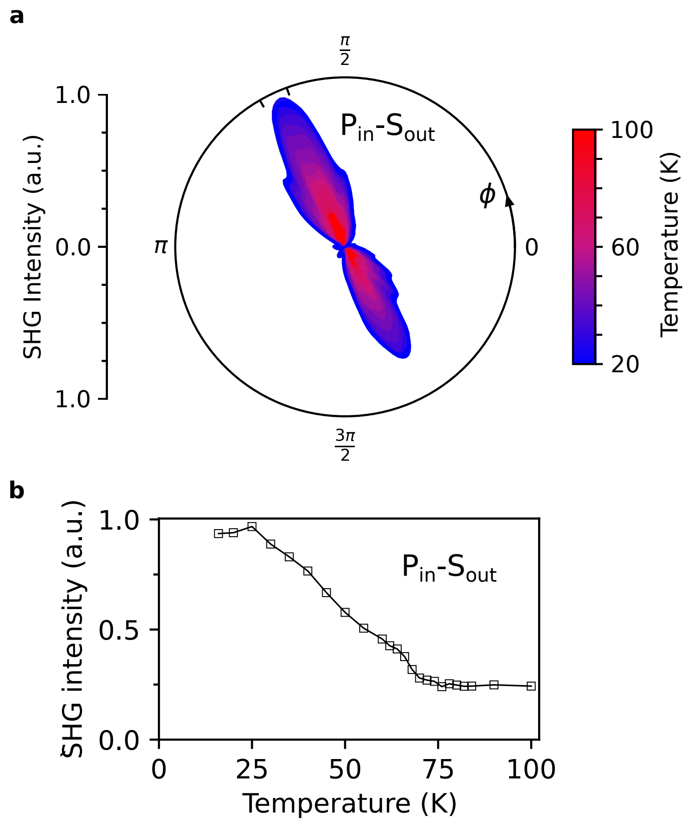

Figure 1.2: (a) SHG intensity as a function of temperature in the $P_{\text{in}}S_{\text{out}}$ polarization channel. (b) Integrated SHG intensity in the region near $\pi/2$ of **??** marked by the tick marks.

SHG polarization channel, may be observed: one high-frequency mode ($\nu \sim 0.23\,\text{THz}$, $\hbar\omega \sim 1.0\,\text{meV}$), which is only observed in the crossed polarization channels $P_{\text{in}}S_{\text{out}}$ and $S_{\text{in}}P_{\text{out}}$, and one low-frequency mode ($\nu \sim 0.05\,\text{THz}$, $\hbar\omega \sim 0.20\,\text{meV}$), which occurs in all polarization channels equally. Both of these are too low to be observed with typical THz or neutron spectroscopies, yet they are readily apparent in the tr-SHG data due to the pump-probe nature of the experiment.

To show that these two modes are directly related to the multiferroic transition at T_c, we measure the pump-induced change in the SHG intensity as a function of Δt for a series of temperatures approaching T_c

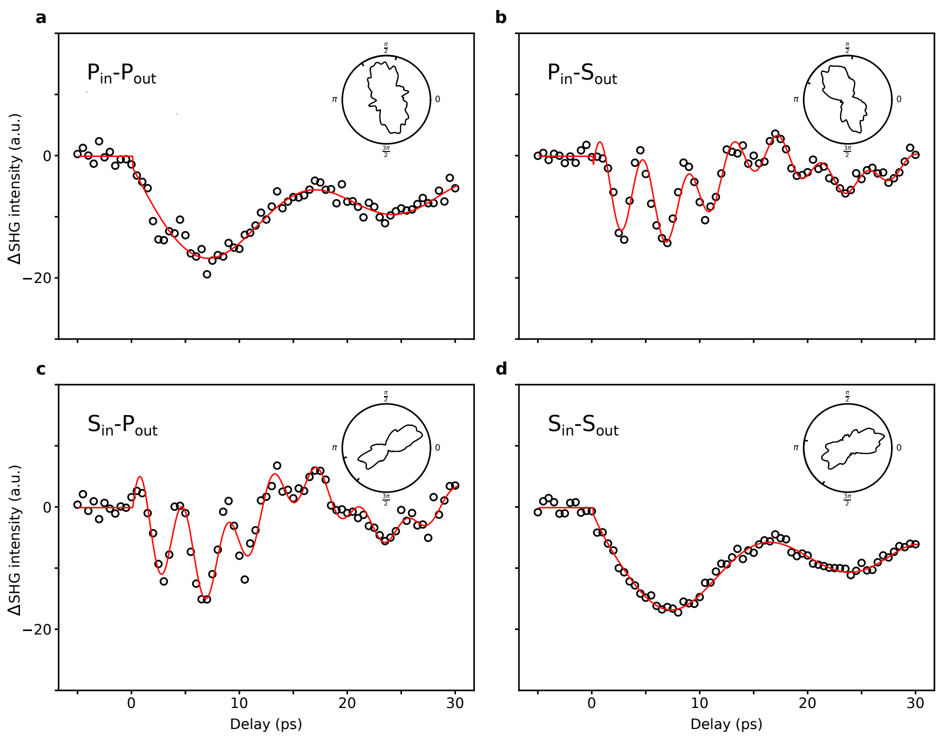

Figure 1.3: Pump-induced change in the SHG intensity at $15\,\mathrm{K}$ in the four polarization channels (a) $P_{\mathrm{in}}P_{\mathrm{out}}$ (b) $P_{\mathrm{in}}S_{\mathrm{out}}$ (c) $S_{\mathrm{in}}P_{\mathrm{out}}$, and (d) $S_{\mathrm{in}}S_{\mathrm{out}}$. Insets depict the static SHG intensity in each polarization channel. The time-domain signals are computed by performing an azimuthal integration at each delay of the full SHG pattern over the angles specified by the additional tick marks in each inset.

Fig. 1.4. By fitting the respective time-domain traces to damped harmonic oscillators (see Supplementary material, section 1.8.3), we can extract the natural frequency of each collective mode as a function of temperature (Fig. 1.4). Both modes exhibit a pronounced softening on approaching T_c, confirming their direct involvement in the multiferroic transition. We also note that both modes disappear above T_c, which is sensible given that the macroscopic polarization $\vec{P}_0$ also disappears above this temperature.

To clarify the microscopic origin of these polarization oscillations, we begin by performing density functional theory (DFT)+U and finite-displacement lattice dynamics calculations[] (see Supplementary material, section 1.8.6.2) to compare their energies with those of the zone-center phonon modes. The lowest zone-center optical phonon in this calculation appears at $7.4\,\mathrm{meV}$, in excellent agreement with Raman spectroscopy[32], and the calculated acoustic phonon branches (which agree with inelastic neutron scattering (INS)[32]) disperse too rapidly to form a zone-folded acoustic phonon mode at the Γ point with an energy low enough to match the frequencies observed in the tr-SHG experiment. Thus, the modes observed in Fig. 1.3 and **??** are not phonons. The only remaining possibility is that these modes are magnons of the incommensurate spin spiral, which imprint themselves on the polarization via Eq. 1.1; i.e., they are electromagnons.

In linear spin wave theory (LSWT), there is only one spin boson which couples to the polarization in the spiral magnetic phase of $CuBr_2$ (see Supplementary material, section 1.8.1); it is the so-called pseudo-goldstone mode of the magnetic order[17], which corresponds to a rotation of the spin plane about the chain direction (Fig. 1.1(b)). This mode has zero energy if the system is isotropic about the chain axis, but in the presence of an anisotropy term it acquires a finite energy. In $CuBr_2$, this energy is expected to lie around $1.25\,\mathrm{meV}$ (see Supplementary material, section 1.8.2), which is close to the value observed for our high-frequency oscillation ($1.0\,\mathrm{meV}$). Additionally, since this mode involves a rotation of the spin plane about the chain direction, the effect of this mode (from Eq. 1.1) on the polarization is to tilt the vector $\vec{P}_0$ into the $\hat{z}$ direction (Fig. 1.1(b)). Since the equilibrium polarization lies along $\hat{y}$, Eq. 1.2 implies that a canting of the polarization $\delta\vec{P}||\hat{z}$ involves new elements of the tensor $\chi_{ijkz}(T > T_c)$ which are not present in equilibrium. The result is that this mode may appear in different polarization channels with different magnitudes; we

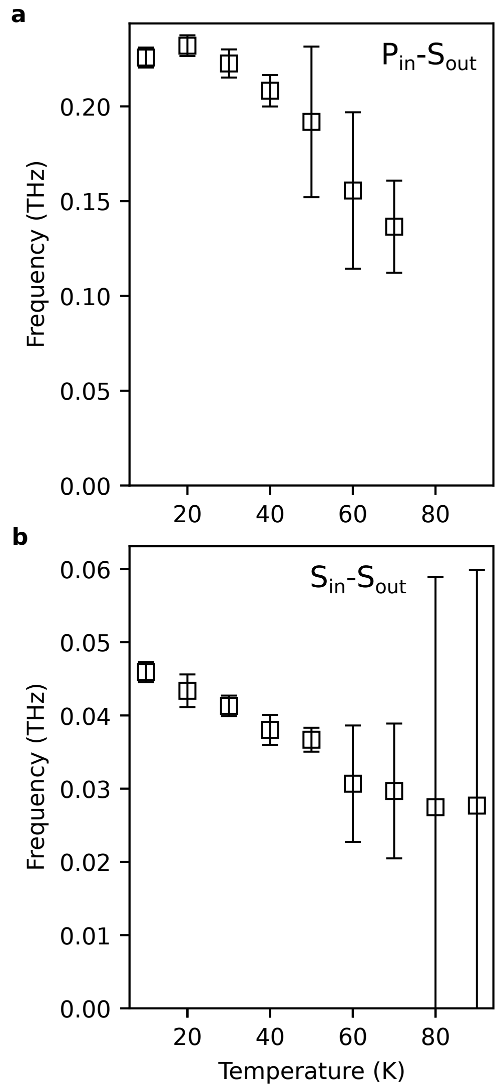

Figure 1.4: Temperature dependece of the frequencies extracted from the (a) $P_{\rm in}S_{\rm out}$ and (b) $S_{\rm in}S_{\rm out}$ time-domain signals (Supplementary material, section 1.8.3) in a damped harmonic oscillator model. Error bars denote $95\,\%$ confidence intervals estimated within a parametric bootstrap (see Supplementary material, section 1.8.4).

thus identify the fast, $0.23\,\mathrm{Thz}$ oscillation in Fig. 1.3 with this mode.

The observation of a *second* mode in the tr-SHG, however, is impossible to explain in LSWT, and represents the most striking aspect of this work. To understand the origin of this second mode, we note that there are only three normal modes of the polarization which occur at the Γ point in the Brillouin zone, corresponding to polarization oscillations $\delta\vec{P}$ along $\hat{x}$, $\hat{y}$, and $\hat{z}$. Since the $\delta\vec{P}||\hat{z}$ mode is already accounted for by the pseudo-goldstone mode, that leaves only $\delta\vec{P}||\hat{x}$ and $\delta\vec{P}||\hat{y}$ as possibilities. The $\delta\vec{P}||\hat{x}$ mode does not couple to the spin order in this compound[17], and in any case is not observable in the geometry of our experiment (see Supplementary material, section 1.8.6.1). Thus, the only polarization oscillation which is consistent with the observation of a second mode is an oscillation with $\delta\vec{P}||\hat{y}$. Since the equilibrium polarization is also directed along $\hat{y}$, this mode simply corresponds to an oscillation in the total amplitude of the polarization, and is thus expected to modulate the overall SHG intensity irrespective of the polarization channel – in excellent agreement with Fig. 1.3, which shows the low-frequency oscillation appearing in all four polarization channels with equal magnitude.

Naively, electromagnons with $\delta\vec{P}||\hat{y}$ do not exist in LSWT. The key insight, however, is that LSWT neglects dynamics associated with the magnitude of the onsite spin expectation value. By Eq. 1.1, such dynamics change the magnitude of the induced polarization only, not its direction; i.e., they induce oscillations $\delta\vec{P}||\hat{y}$. In fact, it is possible to show (see Supplementary material, section 1.8.1) that oscillations along $\hat{y}$ of the induced polarization *necessarily* involve modulations in the amplitude of the onsite spin excpectation value; that is, the only mode which couples to $\delta\vec{P}||\hat{y}$ is the amplitude mode of the magnetic spiral. Naturally, this mode should soften on approaching T_c, in agreement with Fig. 1.4. The $0.05\,\mathrm{THz}$ oscillation observed in our experiment (which is of similar energy to the amplitude mode in non-ferroelectric quantum magnetsHong et al. [14]) is therefore direct evidence for this mode in $CuBr_2$, with the additional information that it couples to the electric polarization (i.e., it is an electromagnon) via Eq. 1.1.

1.5 Discussion

Let us make two important remarks about this mode in $CuBr_2$. First, we note that this mode is fundamentally distinct from the amplitude mode in non-multiferroic magnets – since the multiferroic order breaks inversion symmetry, the amplitude mode of this phase has a parity quantum number $l = -1$ rather than $+1$. The static polarization $\vec{P}_0$ which appears at T_c can thus be viewed as arising from the odd-parity nature of its amplitude mode. Second, we remark that the amplitude mode in $CuBr_2$ can in principle decay – as in non-multiferroic magnets – quite rapidly into the Goldstone modes of the magnetic order (which in the spin-spiral phase of Fig. 1.1(c) are gapless and correspond to uniform rotations of each spin about the $\hat{z}$ direction), and thus should not exist as a well-defined quasiparticle unless these decay channels are quenched by some mechanism.

In non-multiferroic magnets, two such mechanisms have been identified. First, the amplitude mode may be stabilized by bringing the system close to a quantum critical point (QCP)[14, 15, 26, 29], which suppresses the Goldstone bosons and thus stabilizes the amplitude mode. A second option is to lower the dimensionality[1, 5, 9, 28, 36]; in 1D, enhanced fluctuations weaken the long range magnetic order and also reduce the spectral weight of the Goldstone bosons[36]. In $CuBr_2$, not only is the system fundamentally one-dimensional, but is thought also to lie in close proximity to a zero-temperature QCP[11] separating the spiral phase considered here and a paraelectric Haldane dimer phase. Both of these mechanisms thus likely contribute to stabilizing the amplitude electromagnon in $CuBr_2$.

1.6 Conclusion

To summarize, we have presented evidence of a novel electromagnon arising from the amplitude mode of the spiral magnetic order in $CuBr_2$. This mode appears alongside the pseudo-Goldstone mode in the tr-SHG data as a low-frequency oscillation in the longitudinal component of the electric polarization, which softens on warming close to T_c. Looking forward, we note that the two mechanisms we identified for stabilizing this mode in $CuBr_2$ – low dimensionality and potential proximity to a QCP – are not necessarily unique to this material. Thus, the amplitude electromagnon presented here may in fact be a common feature of 1D multiferroics, and its observation could indicate a wealth of new opportunities to explore the amplitude mode of particle physics in novel condensed-matter contexts.

1.7 Methods

Tr-SHG measurements were carried out using a fast-rotating optical grating setup described previously[10, 13, 31]. $100\,\mathrm{fs}$ ultrashort pulses from a regeneratively amplified $5\,\mathrm{kHz}$ Ti:Sapphire laser were used to pump an optical parametric amplifier (OPA), producing $1300\,\mathrm{nm}$ (Fig. 1.3) or $1650\,\mathrm{nm}$ (Fig. 1.4) pump pulses which were delayed with an optical delay line and focused at normal incidence to a $300\,\mathrm{um}$-diameter spot on the sample. The pump fluence was $\sim 1\,\mathrm{mJ}\cdot\mathrm{cm}^{-2}$ for all measurements. A small portion of the Ti:Sapphire output was used for the SHG probe experiment, the output of which was spectrally filtered with a $400\,\mathrm{nm}$ bandpass filter, collected by a photomultiplier tube, filtered with a lock-in amplifier, and correlated with the plane of incidence angle using an optical rotary encoder. To measure the pump-induced change in the SHG signal, the pump pulses were chopped at a frequency of $2.5\,\mathrm{kHz}$, and the lock-in amplifier was set to that frequency so as to measure $I_{\mathrm{pump+probe}} - I_{\mathrm{probe}}$. For the pump-probe rotational anisotropy SHG (RA-SHG) measurements, the plane of incidence was rotated while the delay stage was moved and the polarizers were controlled automatically using homebuilt polarization rotators described in Morey et al. [23]. For the single-angle tr-SHG measurements, the plane of incidence was parked at the angle which maximized the static SHG intensity in the respective polarization channel.

1.8 Supplementary material

1.8.1 Electromagnons in $CuBr_2$

The observed co-existence of spiral magnetic order and ferroelectricity is due to the spin-orbit coupling enabled interaction term between the spins

$\vec{S}$ and the electronic polarization $\vec{P}$ []:

$$H_{s-P} = \lambda \sum_i \vec{P}_i \cdot (\hat{x} \times \vec{S}_i \times \vec{S}_{i+1}) \tag{1.3}$$

The ordered state for $T < T_N$ is a multiferroic with spontaneous polarization

$$\left\langle \vec{P}_i \right\rangle = P_0 \hat{y} \tag{1.4}$$

and spiral spin ordering

$$\left\langle \vec{S}_i \right\rangle \equiv \vec{S}_{0,i} = S_0 \left(\cos(\vec{Q} \cdot \vec{R}_i)\hat{x} + \sin(\vec{Q} \cdot \vec{R}_i)\hat{y} \right), \tag{1.5}$$

where $\vec{Q}$ is the spin-ordering wavevector and $\vec{R}_i$ are the spatial coordinates of the Cu atoms. Let us consider fluctuations about this ordered state and ask which fluctuations are detectable via SHG. Representing fluctuations in the polarization by $\delta\vec{P}_i$ and spin by $\delta\vec{S}_i$, we get the following fluctuation Hamiltonian:

$$H^f_{s-P} = \lambda \sum_i \delta\vec{P}_i \cdot (\hat{x} \times \delta\vec{S}_i \times \vec{S}_{0,i+1} + \hat{x} \times \vec{S}_{0,i} \times \delta\vec{S}_{i+1}) + \mathcal{O}(\delta\vec{P}^2, \delta\vec{S}^2). \tag{1.6}$$

Expanding the spin fluctuations along all directions, we find that they couple only to polarization fluctuations along $\hat{y}$ and $\hat{z}$. Focusing on zero-momentum polarization fluctuations (since they are sensitive to SHG),

$$\begin{aligned} H^f_{s-P} =& i \sin(\vec{Q} \cdot \vec{a}) \delta P_z(\vec{q} = 0) \left(\delta S_z(-\vec{Q}) - \delta S_z(\vec{Q}) \right) \\ &+ \sin(\vec{Q} \cdot \vec{a}) \delta P_y(\vec{q} = 0) \left(-\delta S_x(-\vec{Q}) + i\delta S_y(-\vec{Q}) - \delta S_x(\vec{Q}) - i\delta S_y(\vec{Q}) \right) \\ &+ \mathcal{O}(\delta\vec{P}^2, \delta\vec{S}^2) \end{aligned} \tag{1.7}$$

where $\vec{a}$ is the lattice vector along the chain. Transverse polarization fluctuations $\delta P \sim \hat{z}$ couple to a uniform rotation of the spin-plane about the x axis. These are the electromagnons discussed in Katsura et al. [17]. The longitudinal fluctuations, on the other hand, couple to longitudinal fluctuations of the magnetization on each site.

1.8.2 Energy of the pseudo-Goldstone mode

An expression for the frequency of the pseudo-Goldstone mode in the presence of an easy-plane anisotropy is given by Katsura et al. [17] as

$$\omega_- = \sqrt{A(Q)B(Q)}, \tag{1.8}$$

where

$$A(q) = 2S\left[\frac{2J(Q) - J(Q+q) - J(Q-q)}{2}\right], \tag{1.9}$$

$$B(q) = 2S\left[J(Q) - J(q) + D\right], \tag{1.10}$$

$$J(q) = 2\left[J_1\cos(qa) + J_2\cos(2qa)\right], \tag{1.11}$$

D is the anisotropy energy, and $2S$ is the amplitude of the spin in units of μ_B.

Using $Q = 0.235\,\mathrm{rlu}$[35], $J_1 = 8.8\,\mathrm{meV}$[19], $J_2 = -22.2\,\mathrm{meV}$[19], $D = 0.15\,\mathrm{meV/Cu}$[20], and $2S = 0.38$[20]), we have

$$\omega_- = 1.3\,\mathrm{meV} \tag{1.12}$$

in good agreement with the experiment.

1.8.3 Fits of time domain signals

Time-domain plots corresponding to the frequencies in Fig. 1.4 are illustrated in Fig. 1.5. Each plot is a least-squares fit of the data to a damped harmonic oscillator model

$$I_p^{\mathrm{SHG}}(t,\theta) = P_p^0\,\delta P_p(t,\theta) + [\delta P_p(t)]^2, \tag{1.13}$$

where

$$\delta P_p = A_p e^{-\gamma_p t}\cos\left(\sqrt{(2\pi\nu_p)^2 - \gamma_p^2}\,t + \psi_p\right), \tag{1.14}$$

$p \in \{P_{\mathrm{in}}S_{\mathrm{out}}, S_{\mathrm{in}}S_{\mathrm{out}}\}$, and θ denotes the set of free parameters to be estimated.

The main conclusion of these fits is that the frequency of the two modes (most notably, the low-frequency $S_{\mathrm{in}}S_{\mathrm{out}}$ mode) soften on approaching T_c. This may also be seen heuristically from the time-domain signals without doing any fits. Fig. 1.6 shows an enlarged (i.e., scaled to account for the

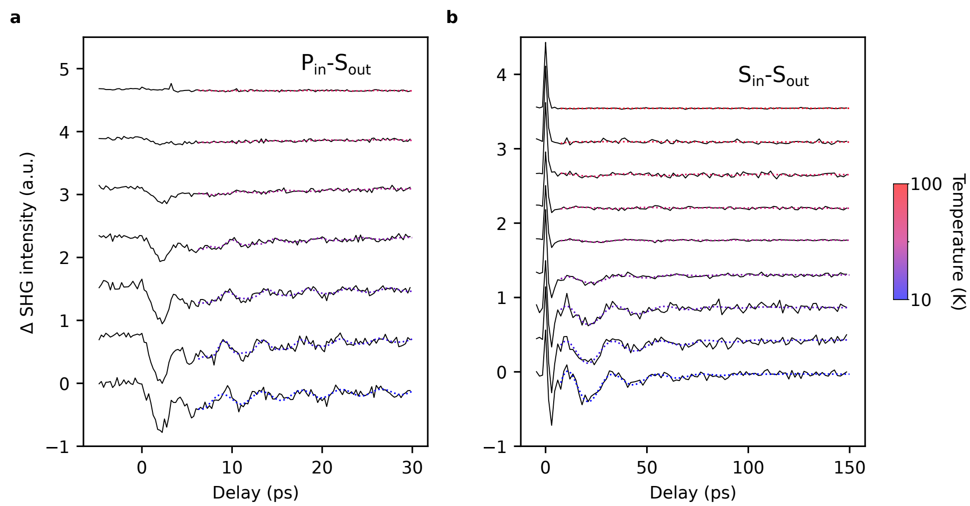

Figure 1.5: Time-domain signals corresponding to (a) Fig. 1.4(a) and (b) Fig. 1.4(b). Dashed lines depict least-squares fits to the data in a damped harmonic oscillator model, see Supplementary material, section 1.8.3.

decrease in signal amplitude) view of the $S_{\mathrm{in}}S_{\mathrm{out}}$ signal for three temperatures below T_c, showing a clear decrease in the oscillation frequency at high temperature. Fig. 1.7 shows an alternative fit where the frequency parameter ν_{SS} is constrained to be constant as a function of temperature, showing that our data is not consistent with a hypothetical model where the frequency shift with temperature in Eq. 1.14 is only attributed to the damping term γ_{SS}.

1.8.4 Error bars in Fig. 1.4

In this section, we describe how the uncertainties in the least square estimates of the frequency parameter ν of Eq. 1.14, which are depicted as a function of temperature in Fig. 1.4, were calculated from the the time-domain signals in Fig. 1.5. For each temperature and polarization channel, a Levenberg-Marquardt (LM) algorithm was used to find the

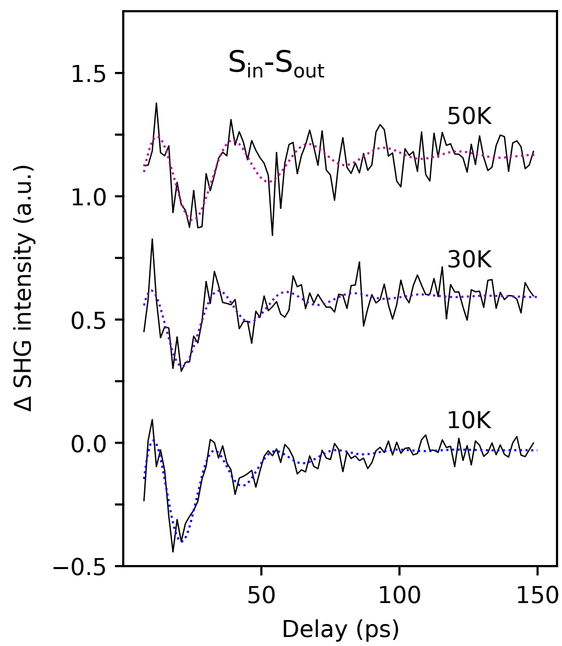

Figure 1.6: Rescaled $S_{\rm in}S_{\rm out}$ time-domain signals (see Fig. 1.5(b)) for select temperatures below T_c.

minimum θ_0 of the objective function

$$f_p(\theta) \propto \sum_{n=0}^{N-1} \left(I_p^{\rm SHG}(t_n, \theta) - I_{p,n}^{\rm SHG}\right)^2, \tag{1.15}$$

where $\{(t_n, I_{p,n}^{\rm SHG}), n \in (0, 1, \ldots, N-1)\}$ are the data points in Fig. 1.5, and we have assumed the noise level is independent of delay. The uncertainty in each parameter is estimated within a parametric bootstrap[6]: for each temperature, 1000 bootstrap samples are generated by adding noise (normally distributed, with variance given by the variance of data points at long times where the signal is constant) to the LM estimate $I_p^{\rm SHG}(t_n, \theta_0)$. For each bootstrap sample s, an estimate θ_s is computed by minimizing Eq. 1.15 as above, and the $95\,\%$ confidence interval reported in Fig. 1.4 is taken to be 1.96 times the standard deviation of the distribution $\{\theta_s - \theta_0\}$.

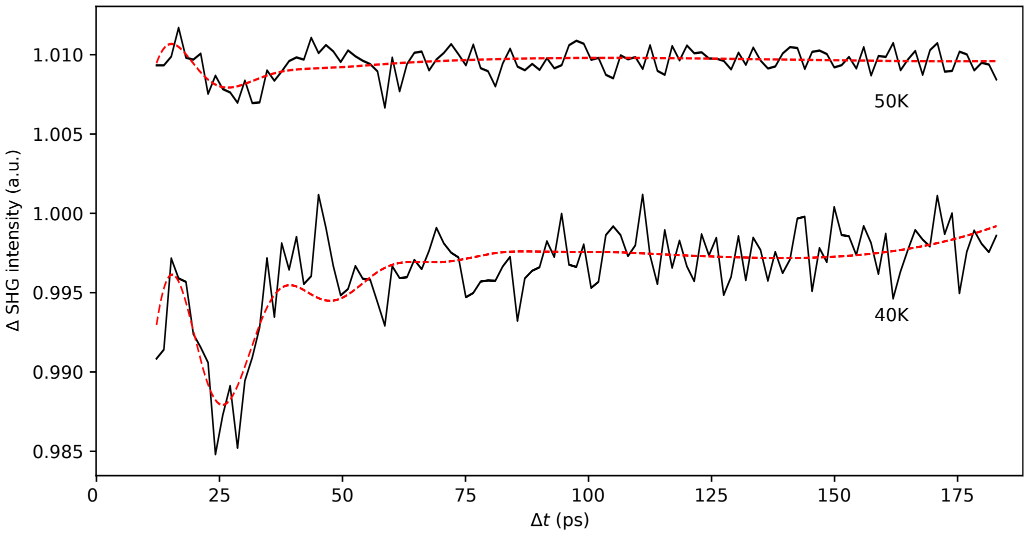

Figure 1.7: $S_{\text{in}}S_{\text{out}}$ time-domain signals (see Fig. 1.5(b)) for select temperatures approaching T_c. Dashed lines depict least-squares fits to the data in a variant of Eq. 1.14 where ν_{SS} is constrained not to vary with temperature.

1.8.5 Fits to static RASHG data

The static SHG intensity was fit by Eq. 1.18. The susceptibility tensor was taken to be the form Eq. 1.19, plus an additional C_1 component (likely due to surface adsorbates). The result is shown in Fig. 1.8.

1.8.6 Excluded possibilities for observed results

1.8.6.1 $\delta\vec{P}||\hat{x}$ oscillation

Without loss of generality, let the maximum of the SHG in $S_{\text{in}}P_{\text{out}}$ occur when the incoming electric field is along $\hat{x}$. Then, we have:

$$\Delta I^{\text{SHG}}_{\text{SP}} \propto |\hat{e}^{\text{out}}_i \chi_{ijkl} \hat{e}^{\text{in}}_j \hat{e}^{\text{in}}_k [P_{0l} + \delta P_l]|^2 - |\hat{e}^{\text{out}}_i \chi_{ijkl} \hat{e}^{\text{in}}_j \hat{e}^{\text{in}}_k P_{0l}|^2 \tag{1.16}$$

with $\hat{e}^{\text{in}}_i||x$ and $P_{0l}||y$, we have

$$\Delta I_{\text{SHG}} \propto 2\hat{e}^{\text{out}}_i \hat{e}^{\text{out}}_j \chi_{ixxy}\chi_{jxxx} P_{0y}\delta P_x + \hat{e}^{\text{out}}_i \hat{e}^{\text{out}}_j \chi_{ixxx}\chi_{jxxx}\delta P_x \delta P_x \tag{1.17}$$

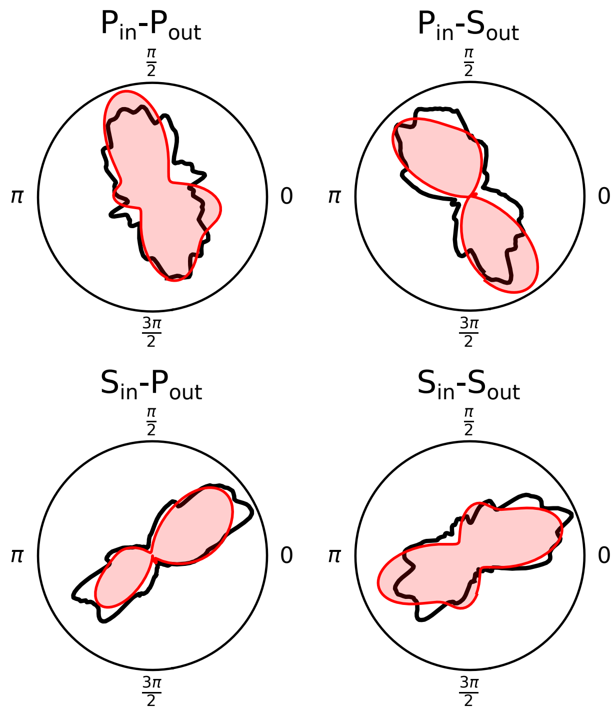

Figure 1.8: Fits (red) to static RA-SHG data (black) depicted in Fig. 1.3.

Since we are in $S_{\text{in}}P_{\text{out}}$, $\hat{e}_i^{\text{out}} \perp x$; thus, Eq. 1.17 involves the tensor elements χ_{yxxx} and χ_{zxxx}. Both of these elements are zero due to the $x \to -x$ mirror symmetry. Thus, the $\delta\vec{P}||\hat{x}$ oscillation is not visible in our experiment.

Additionally, since the $\delta\vec{P}||\hat{x}$ mode does not couple to the spin order in this compound[17], its frequency should be far above the frequencies observed in our experiment (which are determined by the energy scales of the spin Hamiltonian).

1.8.6.2 Zone-folded acoustic phonons

Phonon band structure calculations were carried out using the finite displacement method[30] with a distance of $0.01\,\text{Å}$ within a $3 \times 3 \times 3$ supercell. Forces were calculated via the DFT-D2 method[12] and LDA+U method[7] ($U_{\text{Cu}} = 3\,\text{eV}$) using a $7 \times 7 \times 5$ k-mesh with 122 irreducible k-points and a plane-wave cutoff energy of $100\,\text{eV}$. The result is shown in Fig. 1.9. The acoustic phonons in Fig. 1.9 all disperse too rapidly for the

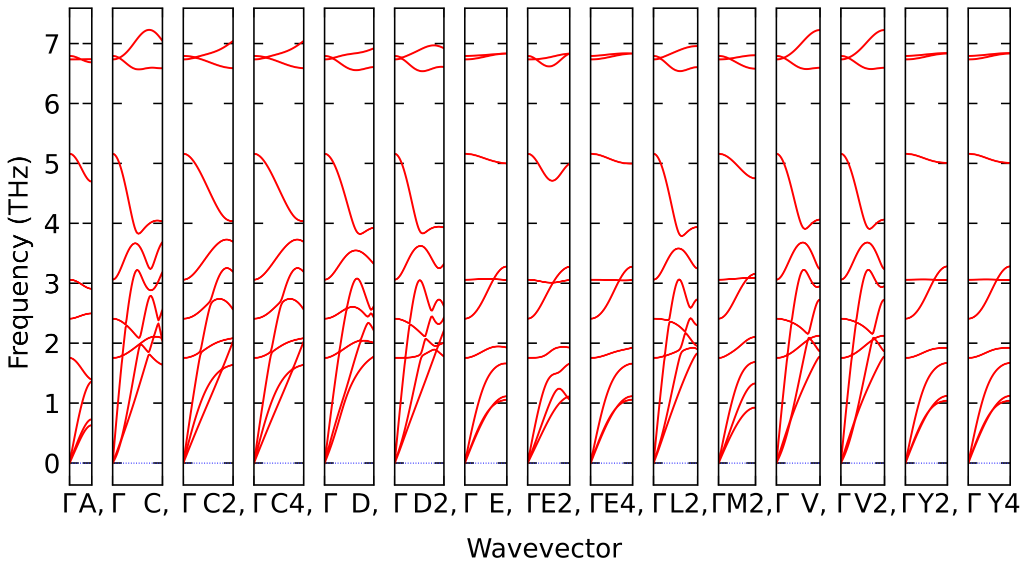

Figure 1.9: Phonon band structure of $CuBr_2$ within a finite displacement calculation.

$0.05\,\mathrm{THz}$ oscillation in the tr-SHG to be consistent with a zone-folded (at $k = (0, 0.235, 0.5)$) acoustic phonon.

1.8.6.3 Magnetic SHG

In principle, magnetic systems with broken inversion symmetry may generate electric-dipole SHG with or without a static electric dipole moment. In this section, we wish to show that this is not the case in $CuBr_2$; i.e., in $CuBr_2$, the SHG intensity is a direct measure of the macroscopic electric dipole moment.

Indeed, in the presence of such a static electric dipole moment $\vec{P}_0$, we typically expect the SHG response to be directly proportional to it; i.e.

$$I(2\omega) \propto |\hat{e}_i^{\mathrm{out}} \chi_{ijk} \hat{e}_j^{\mathrm{in}} \hat{e}_k^{\mathrm{in}}|^2, \tag{1.18}$$

where

$$\chi_{ijk} = \chi_{ijkl} P_{0l}, \tag{1.19}$$

and $\hat{e}^{\mathrm{in}}$, $\hat{e}^{\mathrm{out}}$ are unit vectors in the direction of the incoming and outgoing electric fields, respectively. In $CuBr_2$, we have

$$\vec{P}_0 = \sum_{\langle i,j \rangle} \hat{x} \times \vec{S}_i \times \vec{S}_j, \tag{1.20}$$

i.e., the static polarization is quadratic in the spin degree of freedom.

The question, then, is whether there exists some additional term

$$\chi'_{ijk} = \chi_{ijkl} G_{0l}, \tag{1.21}$$

where $\vec{G}_0$ is either (a) linear in spin, or (b) quadratic in the spins but not of the form $\sum_{\langle ij \rangle} \vec{S}_i \times \vec{S}_j$. For case (b), note that the term $\sum_{\langle ij \rangle} \vec{S}_i \times \vec{S}_j$ is the only quadratic form which is simultaneously antisymmetric in the bond direction and $\vec{q} = 0$ (i.e. each bond has the same coefficient).

For case (a), we argue here that any such term is weak due to the approximate time-reversal symmetry of the spiral magnetic order. Consider first a four-site commensurate approximant of the incommensurate spin spiral. This phase has a symmetry element consisting of the time-reversal operation followed by a translation by half of the magnetic supercell. Thus, the point group contains time-reversal symmetry. Since $\vec{G}_0$ is linear in spin, time-reversal takes $\vec{G}_0 \to -\vec{G}_0$; but since time-reversal is a symmetry, it must also take $\chi'_{ijk} \to \chi'_{ijk}$ and $\chi_{ijkl} \to \chi_{ijkl}$. Thus, $\chi'_{ijk} = 0$ in the commensurate approximation.

In the incommensurate case, note that the magnetic point group of an incommensurate magnetic phase is defined as the set of point-group operations present in the operations belonging to the superspace group. Thus, for a single-k incommensurate magnetic structure, time-reversal is always an element of the magnetic point group. This is due to the fact that the lattice constant in the chain direction is 3.51 Å, so lengthscales associated with translations in the space group are much smaller than the probe wavelength ($\sim 800\,\mathrm{nm}$). The symmetry group "seen" by the probe thus contains time reversal to a very good approximation.

1.8.6.4 Multi-phason excitation

While the amplitude mode of the spin spiral in $CuBr_2$ is the only single-particle excitation which couples to δP_y (see Eq. 1.7), in principle multiparticle excitations consisting of, e.g., two phasons with opposite momenta are also allowed. However, note that the relevant energy scale which defines the phason sound velocity depends on the intra-chain coupling terms, which are $\mathcal{O}(10\,\mathrm{meV})$; the peak in the phason joint density of states thus occurs at this high energy scale, which is much larger than the $0.2\,\mathrm{meV}$ energy of our low-frequency oscillation. Multi-phason excitations are thus not consistent with the long-lived oscillation observed in our experiment.

Chapter Two

Concluding remarks

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