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Twisted phonons produce giant magnetic moments

Terahertz electric-field-driven dynamical multiferroicity in $SrTiO_3$
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An archetypical perovskite compound, $SrTiO_3$ (STO), never ceases to amaze. Originally viewed as a potential synthetic replacement of diamond in the '50s,[*] STO was the first doped insulator in which superconductivity was discovered in 1967, [1] but the mechanism of superconductivity still remains the subject of depbate. In the '70s and '80s, STO received a new wave of attention after Müller and Burkard [2] identified it as a quantum paraelectric: a material balancing on the verge of ferroelectricity but never quite making it due to disruptive zero-point motion of light oxygen atoms (later on, it was realized that a structural antiferrodistortive transition at 105 K also plays a role in arresting ferroelectricity [3, 4]). From 2015 and on, STO is again on the stage thanks to finely crafted transport measurements, which unveiled a T^2 resistivity scaling in doped STO [5][†] and the strongest thermal Hall effect among all non-magnetic insulators [6].

Paraelectricity is STO (and related compounds, such as $KTaO_3$, $EuTiO_3$, etc.) is linked to the softening of transverse optical phonon modes. In STO, there are two such modes at the Γ point of the Brillouin zone, with E_u and A_{2u} symmetries respectively. The frequencies of these modes drops precipitously as temperature decreases; namely, the frequency of the softest mode, A_{2u}, drops from about 12 meV at room temperature to the saturation value of 0.8 meV (0.2 THz) at liquid Helium temperatures. Concomitantly, the dielectric constant increases from about 300 at room temperature to 25,000 at Helium, making STO the highest-K insulator known. The soft mode had long been suspected to be responsible for superconductivity in STO [7–11] and has been recently proposed to be the reason for the T^2 scaling of its resistivity [12]. It is this soft mode that is the main subject of recent experimental study by Basini et al. [13].

Common wisdom says that STO may be many things, but magnetic it is not. Basini et al. defy this notion by demonstrating that intense circularly polarized THz pulses induce

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[*] It did not work out.

[†] If you think that this is just the usual Fermi-liquid T^2 scaling, note that in STO it extends well beyond the Fermi temperature.

dynamical magnetization in undoped STO. The mechanism, known as "dynamical multiferroicity" (DMF), was theoretically proposed in Ref. [14]. [‡] The idea of DMF is based on a symmetry argument: the cross-product of a circularly-polarized electric polarization, $\mathbf{P}(t) \propto \cos\omega t\hat{x} + \sin\omega t\hat{y}$, and its time derivative, $\partial_t\mathbf{P}(t)$, is a time-reversal-odd pseudovector; hence the magnetization $\mathbf{M}(t)$–another time-reversal-odd pseudovector–can be expressed as $\mathbf{M}(t) \propto \mathbf{P}(t) \times \partial_t\mathbf{P}(t)$. To test this idea, Basini et al. [13] used circularly-polarized 3 THz pulses with 0.5 THz bandwidth to pump the 0.2 THz soft phonon mode, thereby forcing the ions to move in circles and inducing the magnetization along the normal to the plane of ionic motion. The induced magnetization was detected via Kerr rotation of 400 nm (≈ 750 THz) probe pulses; the maximum value of the Kerr angle amounted to about 50 μrad. To prove that the effect indeed comes from the electric polarization induced by the soft phonon mode, Basini et al. followed the dependence of the Kerr signal on temperature, which was varied from 160 to 360 K. According to neutron spectroscopy [15], the soft phonon frequency should vary in this temperature interval by almost a factor of 2, which should be enough to detune the mode from the resonance with THz pulses. Indeed, the Kerr effect was found to depend in T non-monotonically, taking a maximum value at 280 K and decreasing towards both ends of the temperature interval.

The observation of an induced magnetization, linked to a soft phonon mode, alone confirms the proposal of Ref. [14]. Nevertheless, a number of experimental observations do not fit the theoretical predictions. Some of them are related to the fact that a response to intense (up 250 kV/cm) THz pulses is strongly non-linear. In fact, the Kerr signal contains two peaks of comparable amplitude: one at twice the frequency of the THz pulse and another one is near zero. On the other hand, the original theory [14] predicted peaks at $\omega = 0$ and twice the phonon frequency, with the second peak being much smaller than the first. Basini et al. (see also a subsequent paper [16]) convincingly explain this discrepancy by invoking a non-linear response and anharmonicity of the soft mode. There is, however, a much more tantalizing and yet unexplained discrepancy. As the magnetization is induced by ionic motion, one would expect the induced magnetic moment to be on the order of the nuclear magneton, $\mu_{\mathrm{N}} = e\hbar/2m_{\mathrm{p}}$ with m_{p} being the proton mass, divided by the average atomic mass (in units of m_{p}). Taking the latter to be ~ 100, one obtains $\mu \sim 10^{-2}\mu_{\mathrm{N}} \sim 10^{-5}\mu_{\mathrm{B}}$ per unit cell, where μ_{B} is the Bohr magneton. Instead, the analysis of Kerr data yields a whopping value of $\mu \sim 0.1\mu_{\mathrm{B}}$, which is 10^4 times larger than expected. Apparently, an (almost) electronic magnitude of the magnetic moment hints at that the electric polarization of bound electrons must be involved. It is not clear however, why the effect is observed only if ionic motion is also driven by the electric field of a THz pulse.

It would be only natural to wonder if there is a link between the giant phonon magnetic moment in STO and its large Hall thermal conductivity. Experimentally, it is known that a large Hall thermal conductivity does not necessarily go hand-by-hand with paraelectricity: for example, $KTaO_3$–a cousin of STO–exhibits much smaller Hall thermal conductivity [6]. It would be quite instructive to repeat the experiment of Ref. [13] on a number of quantum paraelectrics, including $KTaO_3$.

STO is not the only material in which giant magnetic moments associated with phonons have been observed. Other examples include a Dirac semimetal Cd_3As_2 [17] and another

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[‡] In general terms, DMF can be viewed as a particular realization of the inverse Faraday effect.

quantum paraelectric PbTe [18]. There are also theoretical proposals for giant phonon magnetic moments in topological [19] and antiferromagnetic [20] insulators, but is STO clearly none of the above. Time and again, a carefully collected experimental data on this relatively simple material sends the ball to the theoretical side of the court.

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