

Dynamic multiferroicity of a ferroelectric quantum critical point

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Quantum matter hosts a large variety of phases, some coexisting, some competing; when two or more orders occur together, they are often entangled and cannot be separated. Dynamical multiferroicity, where fluctuations of electric dipoles lead to magnetisation, is an example where the two orders are impossible to disentangle. Here we demonstrate elevated magnetic response of a ferroelectric near the ferroelectric quantum critical point (FE QCP) since magnetic fluctuations are entangled with ferroelectric fluctuations. We thus suggest that any ferroelectric quantum critical point is an *inherent* multiferroic quantum critical point. We calculate the magnetic susceptibility near the FE QCP and find a region with enhanced magnetic signatures that appears near the FE QCP, and controlled by the tuning parameter of the ferroelectric phase. The effect is small but observable - we propose quantum paraelectric strontium titanate as a candidate material where the magnitude of the induced magnetic moments can be $\sim 5 \times 10^{-7} \mu_B$ per unit cell near the FE QCP.

Quantum matter exhibits a plethora of novel phases and effects upon driving [1], one of which is the strong connection between the quantum critical point (QCP) of one order parameter and the presence of another phase. The discussion has often focussed on the relation between superconductivity and one or more magnetic phases [2–4]. However, other fluctuation-driven phase transitions, for example nematic phases in iron-based superconductors [3, 5], have also received significant attention. We focus here on the ferroelectric (FE) QCP which is a key part of the discussion of FE behaviour, particularly in displacive quantum paraelectrics [6, 7]. The behaviours that may occur near or as a result of such an FE QCP have been explored in various contexts [6–13], and the list of systems where the effects of quantum fluctuations can be observed is expanding, with temperatures up to $\sim 60\text{K}$ in some organic charge-transfer complexes [10, 14].

The concept of dynamical multiferroicity was introduced recently as the dynamical counterpart of the Dzyaloshinskii-Moriya mechanism, reflecting the symmetry between electric and magnetic properties [15]. In the Dzyaloshinskii-Moriya mechanism [16–18], ferroelectric polarisation is caused by a spatially varying magnetic structure, leading to strong coupling between ferroelectricity and magnetism [19–21]. In the related phenomenon of dynamical multiferroicity, magnetic moments \mathbf{m} can be induced by time-dependent oscillations of electric dipole moments \mathbf{p} :

$$\mathbf{m} = \lambda \mathbf{p} \times \partial_t \mathbf{p} = C \mathbf{n} \times \partial_t \mathbf{n}. \quad (1)$$

For magnetism to occur, \mathbf{p} has to exhibit transverse fluctuations; we therefore focus on rotational degrees of freedom of electric dipole moments [22]. The unit direction vector of the constant amplitude electric dipole moment is $\mathbf{n} \equiv \mathbf{n}(\mathbf{r}, t)$, with time derivative $\partial_t \mathbf{n}$, and $C = \lambda |\mathbf{p}|^2$

in terms of the polarization per unit volume \mathbf{p} (we use estimates from uniform polarisation $P_0 = |\mathbf{P}|V$ in FE phases), and coupling $\lambda = \pi/e$. Generally, we expect that orders entangled with the underlying static order can be excited dynamically. One possibility is to use external driving mechanisms such as light, magnetic field or lattice strain to induce transient excitations of the entangled orders [2]. The present work addresses the complementary case where inherent FE quantum fluctuations induce entangled ferromagnetic order fluctuations without any external drive.

In this Letter, we demonstrate that i) the fluctuating dipoles can induce magnetic fluctuations that surround the FE QCP, as shown in Fig. 1a. The mechanism for this effect is the induction of magnetic moments by fluctuating electric dipoles, described by Eq. (1), near the FE QCP and therefore describes *inherent* dynamic multiferroicity. We support this scenario by calculating the magnetic susceptibility that, as we show, diverges in the paraelectric phase (Figs. 2 and 3), indicating a transition to a new regime, labelled ‘Multiferroic PE’ in Fig. 1a. We thus surmise that any FE QCP is a multicritical multiferroic (MF) QCP with elevated magnetic fluctuations. While the proposed effect is general, we will consider the specific implications for magnetism in strontium titanate (STO) and provide estimates relevant to STO. ii) Within the approximations used, the effective action for \mathbf{p} does not acquire a static, \mathbf{B} -dependent mass term, and the FE QCP is independent of \mathbf{B} . The Zeeman splitting of the FE active phonon modes [15, 22] meanwhile does affect the magnetic susceptibility χ_m and, in higher order approximations, will lead to a \mathbf{B}^2 term in the free energy, affecting the FE QCP. iii) We estimate the typical induced magnetic moment from a single rotating electric dipole to be $|\mathbf{m}| \approx 8.0 \times 10^{-4} \mu_N$, where μ_N is the nu-

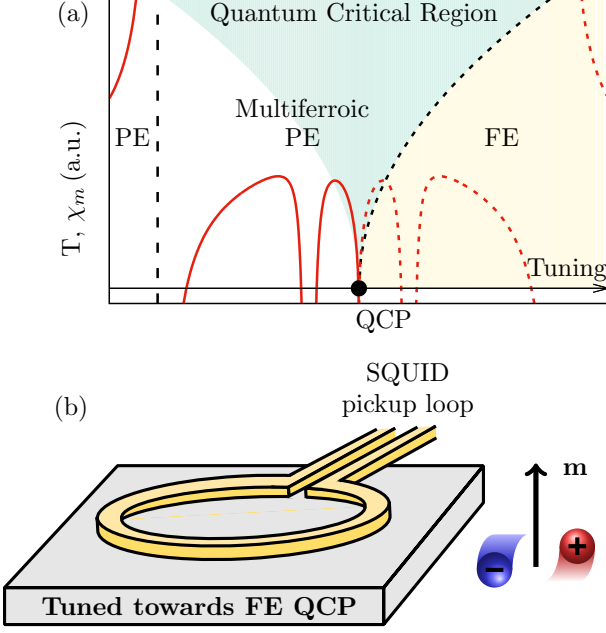


FIG. 1. (a) Phase diagram near a ferroelectric quantum critical point with the magnetic susceptibility (red line, dashed in FE phase) at $\omega = 0.5\omega_0$ which diverges at the vertical dashed line, leading to a new ‘Multiferroic PE’ phase. The ferroelectric quantum critical region (pale green) is now a dynamical multiferroic quantum critical region. In both the PE and FE phases, qualitatively similar behaviour of χ_m is expected, despite the different underlying orders. (b) A simple experiment using a SQUID could detect magnetic signatures resulting from rotating electric dipoles in a system towards its ferroelectric phase transition. Here, the electric dipoles are constrained to the horizontal plane and lead to an out-of-plane magnetic moment \mathbf{m} and susceptibility.

clear magneton, and the coupling $\lambda = \pi/e$ of charge $4e$ and length $1 \times 10^{-2}\text{\AA}$, rotating with frequency 0.5 THz , typical of the titanium atom displacements [23–26] and the ferroelectric phonon modes in STO [27–29] (supplemental material [30] §I).

Model: The system considered consists of fluctuating electric dipoles close to the PE-FE phase transition, inducing a magnetic moment via Eq. (1). In the absence of external fields, the generic description of the system of rotating electric dipoles consists of the paraelectric phase: $L_{PE} = (\omega^2 - \omega_q^2) \mathbf{p}_{\omega,q} \mathbf{p}_{-\omega,-q}$ which has negligible intrinsic magnetic contribution and we therefore ignore intrinsic magnetisation altogether. However, magnetic susceptibility of the paraelectric will occur near the FE QCP due to the dynamic induction of \mathbf{m} , Eq. (1).

The interaction between induced magnetic moments can be neglected in the PE phase since the lowest order contribution $|\mathbf{m}|^2 \propto |\mathbf{p}|^4$. We assume optical phonons, relevant for the PE-FE transition in STO [27], with dis-

person ω_q given by:

$$\omega_q^2 = \omega_0^2 \left(1 - \frac{x}{x_{cr}} \right) + bq^2 = \omega_0^2 \delta_x + bq^2, \quad (2)$$

where δ_x describes the distance to the *ferroelectric* QCP at x_{cr} . If the system is very close to the FE QCP, the momentum dependence is negligible and a flat dispersion with $b = 0$ can be used. The system is paraelectric for $\delta_x > 0$ and ferroelectric when $\delta_x < 0$.

Although in reality both amplitude and directional fluctuations of \mathbf{p} are present near the FE QCP, we will ignore the amplitude fluctuations, so the time dependence is contained entirely in the unit direction vector \mathbf{n} . At the boundary between the PE and FE phases instead of $|\mathbf{p}| \rightarrow 0$, the dipoles rotate. In the PE phase, finite-sized electric dipoles are present, but not aligned so the net polarisation is zero; in the FE phase the dipoles align. \mathbf{n} is linearised as: $\mathbf{n} = \mathbf{n}_0 + \tilde{\mathbf{n}}(t)$ with $\partial_t \mathbf{n}_0 = 0$ and $\langle \tilde{\mathbf{n}} \rangle = 0$. The zero-temperature Green’s function of the \mathbf{n} field in the Matsubara frequency-momentum space reads

$$\langle \tilde{n}_\omega^j \tilde{n}_{-\omega}^m \rangle = A_j \delta_{jm} G(i\omega, q), \quad (3)$$

with A_j as a constant factor. To find dynamic susceptibilities, we use the retarded Green’s function, obtained by analytical continuation to real frequencies ($i\omega \rightarrow \omega + i\eta$) [30]:

$$G^R(\omega, q) = \text{Re} \left(\frac{1}{\omega_q^2 - \omega^2} \right) + \frac{i\pi}{2\omega_q} [\delta(\omega_q - \omega) - \delta(\omega_q + \omega)]. \quad (4)$$

We now calculate the magnetic susceptibility in the PE phase:

$$\chi_m = \langle \mathbf{m}(r_1, t_1) \mathbf{m}(r_2, t_2) \rangle \equiv \chi^{(1)} + \chi^{(2)} \quad (5)$$

and \mathbf{m} is given by Eq. (1). The two contributions are $\chi^{(1)} \propto \langle \tilde{n}^k \tilde{n}^n \rangle$ and $\chi^{(2)} \propto \langle \tilde{n}^j \tilde{n}^k \tilde{n}^m \tilde{n}^n \rangle$.

The quadratic contribution in $\omega - q$ space is:

$$\chi_{il}^{(1)} = C^2 n_0^j n_0^m A_k \epsilon_{ijk} \epsilon_{lmk} \omega^2 G^R(\omega), \quad (6)$$

with $G^R(\omega)$ given by Eq. (4). The factor ω^2 comes from the Fourier transform of $\langle \partial_t \tilde{n}^k \partial_t \tilde{n}^n \rangle$.

The quartic contribution to the magnetic corresponds to a one-loop diagram as discussed in the supplemental material §III [30]. The real part of the diagonal element is:

$$\text{Re}[\chi_{ii}^{(2)}] = \frac{C^2 A_j A_k \Lambda^3}{\pi \omega_0} \left[\frac{\omega_0^2 \sqrt{\delta_x}}{\pi (\omega^2 + 4\omega_0^2 \delta_x)} + \frac{f(\omega)}{8\sqrt{\delta_x}} \right] \quad (7)$$

where $f(\omega)$, given in full in the supplemental material §III [30], contains δ -functions at $2\omega_0 \sqrt{\delta_x} \pm \omega$ and ω with weights ω or $\omega_0 \sqrt{\delta_x}$. The imaginary part is:

$$\text{Im}[\chi_{ii}^{(2)}] = \frac{C^2 A_j A_k \Lambda^3}{\pi^2 \omega} \left[\frac{\omega^2 - 2\omega_0^2 \delta_x}{\omega^2 - 4\omega_0^2 \delta_x} \right]. \quad (8)$$

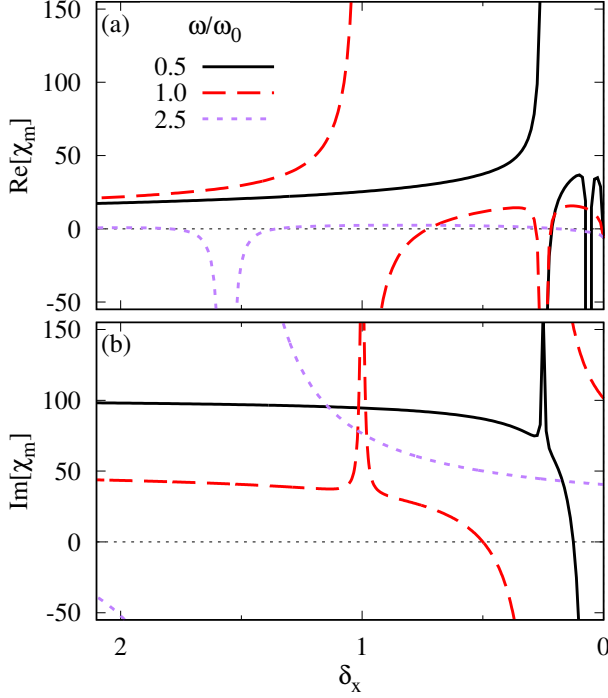


FIG. 2. The total magnetic susceptibility in units of the common prefactor $\lambda^2 V^4 P_0^4$, and with $\Lambda^3/\omega_0 = 1000$ for clarity of plots, as a function of δ_x at several energies. (a) the real part; (b) the imaginary part. The behaviour in the FE phase $\delta_x < 0$ is expected to share the main qualitative features despite the underlying order. The effects of changing the $\chi^{(2)}$ prefactor Λ^3/ω_0 and the individual contributions of $\chi^{(1)}$ and $\chi^{(2)}$ are discussed in the supplemental material §IV [30].

Λ is a momentum cut-off, see the supplemental material [30] §III for details. If ω is written in terms of ω_0 , the size of the $\chi^{(2)}$ contribution is determined by Λ^3/ω_0 . For $\Lambda = 2\pi/a$ where $a = 3.905\text{\AA}$ is the lattice parameter of strontium titanate, and $\omega_0 = 0.5\text{THz}$, we have $\Lambda^3/\omega_0 \sim 10^{18}$. Meanwhile, local regions of ordered fluctuations on the nanometre scale [31] lead to $\Lambda^3/\omega_0 \sim 10^{15}$ and coherence over sub-micrometre ferroelectric domains [32] gives $\Lambda^3/\omega_0 \sim \times 10^{11}$. Larger areas of coherent fluctuations are limited to tetragonal domains, $\sim 10\mu\text{m}$ [33], in which case $\Lambda^3/\omega_0 \sim 5 \times 10^5$, the distribution and size of tetragonal domains can be controlled by both applied electric fields [34, 35] and pressure [34].

Results: The total magnetic susceptibility χ_m from Eq. (5) is plotted in Figs. 2 and 3 with the overall scale given by the shared prefactor $C^2 = \lambda^2 V^4 P_0^4$ set to unity in all plots. In bulk STO samples, the value of C^2 can be estimated from experimental data of samples tuned through the FE phase transition by applied strain or ^{18}O isotope substitution, which indicates the possible size of the dipole moments in the PE phase: $C^2 \sim 2 \times 10^{-3}$, for bulk STO crystals, $C^2 \sim 3 \times 10^{-58}$ for sub-micrometre

sized ferroelectric domains and $C^2 \sim 4 \times 10^{-34}$ for $10\mu\text{m}$ tetragonal domains, all in units of C^2m^4 [23–25].

We consider tuning towards the FE QCP at a constant energy (fixed ω/ω_0) first. In Fig. 2a, far from the FE QCP, the system is dielectric with $\text{Re}[\chi_m] > 0$ but not large. On moving towards the FE QCP, χ_m diverges and changes sign at $\delta_x = \omega^2/\omega_0^2$; this indicates a phase transition into a region where magnetic signatures can be expected. As the energy is decreased, the divergence moves towards the FE QCP and the features are compressed into a narrower range of the tuning parameter. The height of the positive peak at small δ_x also increases at lower energies.

There are two contributions to the peaks in real part of susceptibility: one is from the poles in $\text{Re}[\chi^{(1)}]$ resulting in the large derivative feature, at $\delta_x = (\omega/\omega_0)^2$, the other comes from the δ -functions in $\text{Im}[G^R]$ that result in poles in $\text{Re}[\chi^{(2)}]$ and negative dips in the total susceptibility at $\omega = 2\omega_0\sqrt{\delta_x}$. On moving closer to the FE QCP, after the initial divergent transition, χ_m becomes positive again without any divergence, and there is a further sharp dip from the $\delta(2\omega_0\sqrt{\delta_x} - \omega)$ term in $\chi^{(2)}$, before a final decrease to an energy independent value at $\delta_x = 0$, again from $\chi^{(2)}$.

The imaginary part of χ_m , plotted in Fig. 2b, also diverges as expected at the border of the magnetic region. This is followed by a smooth decrease to a negative divergence with $\text{Im}[\chi_m] = 0$ at $2\delta_x = \omega^2/\omega_0^2$, followed by a sign change via a divergence corresponding to the negative peaks in $\text{Re}[\chi_m]$.

Changing energy while at a fixed distance from the FE QCP is considered in Fig. 3. The divergences and sign changes at finite ω are exactly those seen in Fig. 2, with an extra, artificial, divergence of both the real and imaginary parts at $\omega = 0$, originating from calculating $\chi^{(2)}$ in the continuum limit. We note that upon increasing δ_x to move away from the QCP, the onset of the magnetic transition moves to higher energy. The energy dependence of the size of the imaginary part is seen particularly clearly in Fig. 3b. The two positive peaks in χ_m below the magnetic transition, and that, in the bulk, $\langle \mathbf{M} \rangle = 0$, indicate that the instability may be towards several different types of magnetic fluctuations perhaps occurring sequentially as the system is tuned towards the FE QCP. In the FE phase, we expect qualitatively similar features, despite the underlying FE order.

An applied magnetic field will have two effects. Firstly, the phonon Zeeman effect splits the phonon modes with a linear dependence on the applied magnetic field [15] and moves the divergence of χ_m (which occurs at $\delta = \omega^2/\omega_0^2$) linearly with \mathbf{B} applied perpendicular to the plane of the rotating dipoles. Secondly, an additional term in the Lagrangian for the interaction of magnetic moments with an applied magnetic field: $\mathbf{B} \cdot \mathbf{m} = \lambda \mathbf{B} \cdot (\mathbf{p} \times \partial_t \mathbf{p})$ [22] can be treated as a perturbation to the paraelectric system. Calculating the corresponding second order diagram (Sup-

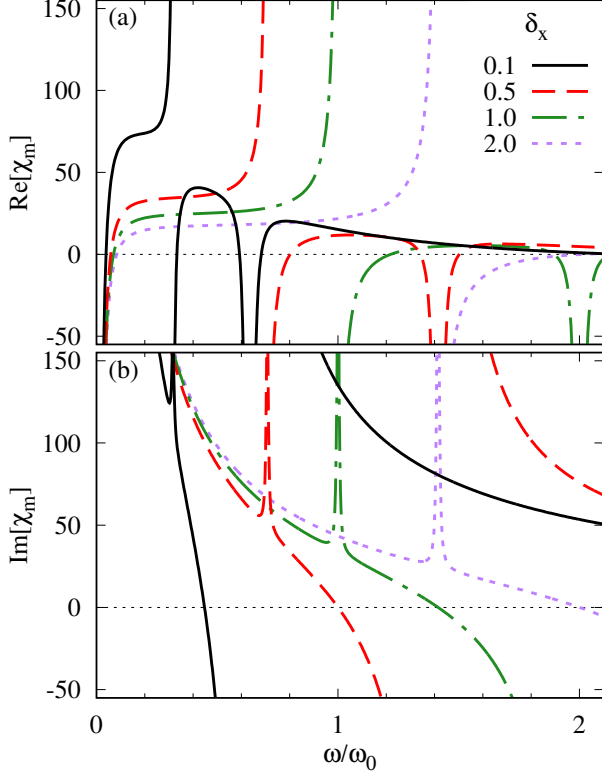


FIG. 3. Total magnetic susceptibility, in units of $\lambda^2 V^4 P_0^4$, and with $\Lambda^3/\omega_0 = 1000$ for clarity, as a function of ω/ω_0 at several distances from the FE QCP. (a) real part; (b) imaginary part.

plemental material, §V [30]) does not introduce a static, \mathbf{B} -dependent mass term, but may do so at higher orders.

Experimental proposal: We suggest strontium titanate (STO) as a suitable candidate material for the observation of magnetic signatures on tuning towards the FE QCP because of its incipient ferroelectric nature below c. 35 K and its quantum paraelectric nature below 4 K [36] where the zero-point motion of the soft transverse optical phonon mode is high enough to prevent ferroelectricity even at zero temperature [37]. In ^{18}O substituted STO, $\omega_{q=0}(T)$ becomes constant below 4 – 10 K depending on the distance from the FE QCP [38–42]. Thus, rotating electric dipoles can be present over an appreciable temperature range. There is additional flexibility because there are several methods for tuning STO towards the FE QCP (Ca doping [43], ^{18}O substitution [24, 25, 44], strain or applied pressure [23, 28]).

A simple experimental set up, consisting of a superconducting quantum interference device (SQUID) above an STO sample, that may permit the observation of the region of pronounced magnetic fluctuations is sketched in Fig. 1b. Strain is a particularly flexible means of tuning STO samples towards the FE QCP, and biaxial strain in STO thin films can confine polarisation to the

plane perpendicular to the tetragonal c -axis, but does not unambiguously determine the polarisation direction [45–47]. Strain could therefore lead to polarisation with no strongly preferred direction in the 2D plane perpendicular to the tetragonal c -axis [48], a favourable condition for the observation of the magnetic signatures proposed here. Although strained STO is considered here, other FE QCPs and tuning mechanisms could be studied, e.g.: $\text{Ca}_{1-x}\text{Pb}_x\text{TiO}_3$ [49], strained KTaO_3 [50] or bromide substituted tris-sarcosine calcium chloride [10]. The quantum dipole phase of the triangular lattice Mott insulator $\kappa - (\text{BEDT} - \text{TTF})_2\text{Hg}(\text{SCN})_2\text{Br}$ [51] may also exhibit magnetic signatures of inherent dynamical multiferroicity.

Discussion: Including the long range interactions between electric dipoles, such as those resulting from twin boundaries between tetragonal domains with differently oriented c -axes [52, 53], would introduce off-diagonal terms to the Green’s function [48]. The immediate effect is a non-zero average magnetisation $\langle \mathbf{M} \rangle \propto \langle \mathbf{n} \times \partial_t \mathbf{n} \rangle$. Alongside this, the off-diagonal components of the dielectric susceptibility $\chi_e^{ij} = \langle \mathbf{pp} \rangle_{ij} \propto \langle \tilde{n}^i \tilde{n}^j \rangle$ would also be non-zero at the twin boundaries, leading to a finite Kerr effect [54]. Further, the motion of twin boundaries may be a means to induce relevant fluctuations of the electric dipoles [55]. Scanning SQUID measurements able to resolve the individual tetragonal domains would be required to investigate the effects of domain structures on the magnetic signals. Again, STO is a promising material since tetragonal domains form naturally on cooling through the antiferrodistortive structural phase transition at 105 K and their distribution can be controlled by applied pressure [34].

The situation examined here is distinct from that recently considered in the context of multiferroic criticality [12] and other systems where the quantum critical points of two or more types of order can be tuned by the same or different parameters leading to a fan where the quantum fluctuations of both orders are important [12, 56]. In our model, the magnetic order does not exist independently of the ferroelectric order, leading to an FE quantum critical region that is surrounded by a region of strong magnetic fluctuations. While distinct from the nematic phase transitions seen in iron pnictides [5, 57], the multiferroic paraelectric region is another realization of competing orders near a QCP. The interaction between the induced magnetic moments and an external magnetic field is expected to mostly affect the nature of the FE phase transition, as discussed for magnetic phase transitions [58–60].

Conclusions: We have expanded the framework of dynamic multiferroicity [15], and predict strongly enhanced ferromagnetic (FM) susceptibility in a paraelectric material near its FE QCP. The induced magnetic susceptibility diverges at a finite distance from the FE QCP. The effect we predict points to another way for entan-

gled quantum order to appear. On the approach to the FE QCP, the fluctuations of the entangled (FM) order are enhanced as the static FE order develops quantum fluctuations. We thus propose that any FE QCP is in fact an *inherent* multiferroic QCP that contains entangled ferroelectric and (much weaker but present) ferromagnetic fluctuations. We expect magnetic signatures of fluctuating dipoles to be observable experimentally, e.g. in SQUID measurements. We also expect that magnetic effects near an FE QCP can be detected in optical Kerr and Faraday effects. Our results are applicable to any ferroelectric-paraelectric transition including classical transitions at finite temperatures, where the fluctuations will be confined to a narrow Ginzburg-Levanyuk region near the transition. The effect will become pronounced near the $T = 0$ QCP. To illustrate the physics we have considered STO as a concrete example of a system that can be tuned towards its FE QCP.

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SUPPLEMENTAL MATERIAL

I: Coupling strength

To calculate the strength of the coupling in the dynamical multiferroic set up, we consider a current flowing round a loop of radius a_B (the Bohr radius) and a period τ_B such that the energy \hbar/τ_B is one Rydberg $R_y = \hbar^2/2m_e a_B^2$. The magnetic dipole moment is perpendicular to the plane of the loop and has magnitude [61]

$$m_{dip} = I\pi r^2 = \frac{e\pi a_B^2}{\tau_B} = \frac{e\hbar\pi}{2m_e} = \pi\mu_B, \quad (9)$$

where $\mu_B = e\hbar/2m_e$ is the Bohr magneton.

The coupling strength λ is obtained by equating the magnitude of the magnetic dipole moment of the current loop with that of the dynamical multiferroic set up [Eq. (1) of the main text] for electric dipole moments of charge e and length a_B rotating with period τ_B :

$$m_{dyn} = \frac{\lambda e^2 a_B^2}{\tau_B}. \quad (10)$$

Requiring that these magnetic moments are equal, $m_{dip} = m_{dyn} = m_B$, gives the coupling strength:

$$\lambda = \pi\mu_B \frac{\tau_B}{e^2 a_B^2} = \frac{\pi e a_B^2}{\tau_B} \frac{\tau_B}{e^2 a_B^2} = \frac{\pi}{e}. \quad (11)$$

The ratio of the induced magnetic moment of any rotating electric dipole to the Bohr model is:

$$\frac{m_{FE}}{m_B} = \frac{(n_q)^2 (n_d)^2 \tau_B}{\tau_F} \quad (12)$$

where τ_F is the rotation period of the electric dipole(s), n_q and n_d are the charge and size of the dipoles in units of the electron charge and Bohr radius respectively.

II: Model

The analytical continuation from the Matsubara frequencies to real frequencies consists of replacing $i\omega$ in the Green's function by $\omega + i\eta$ [62]:

$$\begin{aligned} G(i\omega, q) &= \frac{1}{\omega_q^2 - (i\omega)^2} \rightarrow G^R(\omega, q) \\ G^R(\omega, q) &= \frac{1}{\omega_q^2 - (\omega + i\eta)^2} \\ &= \frac{1}{2\omega_q} \left[\frac{1}{\omega_q + \omega + i\eta} + \frac{1}{\omega_q - \omega - i\eta} \right] \end{aligned} \quad (13)$$

This is then evaluated using the principal value integrals to give:

$$\begin{aligned} G^R(\omega, q) &= \frac{1}{2\omega_q} \left\{ \frac{1}{\omega_q + \omega} + \frac{1}{\omega_q - \omega} \right. \\ &\quad \left. + i\pi [\delta(\omega_q - \omega) - \delta(\omega_q + \omega)] \right\} \\ &= \frac{1}{\omega_q^2 - \omega^2} + \frac{i\pi}{2\omega_q} [\delta(\omega_q - \omega) - \delta(\omega_q + \omega)], \end{aligned} \quad (14)$$

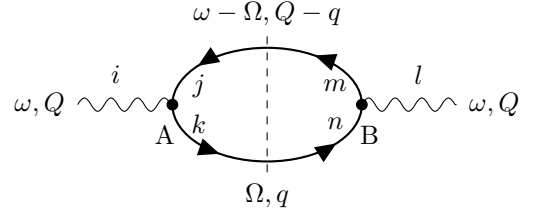
where ω_q is the dispersion of the ferroelectric (FE) phonons: $\omega_q = \sqrt{\omega_0^2 \delta_x + bq^2}$. Near the ferroelectric quantum critical point (FE QCP), the momentum dependence can be neglected [48].

III: Calculation of $\chi^{(2)}$

The second contribution to the magnetic susceptibility is:

$$\chi_{m,il}^{(2)} = C^2 \epsilon_{ijk} \epsilon_{lmn} \langle \tilde{n}^j(t_1) \partial_{t_1} \tilde{n}^k(t_1) \tilde{n}^n(t_2) \partial_{t_2} \tilde{n}^m(t_2) \rangle \quad (15)$$

where the temporal (and spatial) arguments have been included explicitly ($(t_1) \Rightarrow (r_1, t_1)$), and $C^2 = \lambda^2 V^4 P_0^4$. The easiest way to evaluate this is to recognise that the angular bracket corresponds to the loop of the diagram:



The equivalence of the internal lines means δ_{jm} and δ_{kn} and the $\epsilon_{ijk} \epsilon_{lmn}$ prefactor becomes $\epsilon_{ijk} \epsilon_{ljk} = +\delta_{il} d!$. Integration over the internal degrees of freedom is the integral $\int d\Omega d^d q / (2\pi)^{d+1}$ which is to be computed. The coupling constant λ has already been factored out so the factor at A is $i\Omega$, and that at B is $i(\omega - \Omega)$ which we assume are independent of momentum. The internal lines give contributions of the (FE phonon) Green's functions: $A_k G^R(\Omega)$ for the lower and $A_j G^R(\omega - \Omega)$ for the upper parts of the loop respectively.

Evaluating the diagram corresponds to calculating the integral:

$$D_{jk} = -A_j A_k \int \frac{d^d q}{(2\pi)^d} \frac{d\Omega}{2\pi} G^R(\Omega) G^R(\omega - \Omega) \Omega(\omega - \Omega), \quad (16)$$

with $\chi_{il}^{(2)} = C^2 d! \delta_{il} D_{jk}$.

The Green's function given by Eq. (14) has both real and imaginary parts, so $G^R(\Omega) G^R(\omega - \Omega)$ leads to several terms:

$$\begin{aligned}
G^R(\Omega)G^R(\omega - \Omega) &= \frac{1}{\omega_q^2 - \Omega^2} \frac{1}{\left(\omega_{Q-q}^2 - (\omega - \Omega)^2\right)} \\
&- \frac{\pi^2}{4\omega_q\omega_{Q-q}} \left[\delta(\omega_q - \Omega)\delta(\omega_{Q-q} - \omega + \Omega) - \delta(\omega_q + \Omega)\delta(\omega_{Q-q} - \omega + \Omega) \right. \\
&\quad \left. - \delta(\omega_q - \Omega)\delta(\omega_{Q-q} + \omega - \Omega) + \delta(\omega_q + \Omega)\delta(\omega_{Q-q} + \omega - \Omega) \right] \\
&+ \frac{i\pi}{2} \left\{ \frac{\delta(\omega_{Q-q} - \omega + \Omega) - \delta(\omega_{Q-q} + \omega - \Omega)}{\omega_{Q-q}(\omega_q^2 - \Omega^2)} + \frac{\delta(\omega_q - \Omega) - \delta(\omega_q + \Omega)}{\omega_q(\omega_{Q-q}^2 - (\omega - \Omega)^2)} \right\}.
\end{aligned}$$

The integral over the internal energy is calculated first, then one considers that the momentum dependence of the phonon spectrum is irrelevant near the FE QCP so $\omega_{Q-q} = \omega_q = \omega_0\sqrt{\delta_x}$. Assuming spherical symmetry, the integral over $d^d q$ becomes $\int_0^\infty dq q^2$ in $d = 3$; evaluating up to some cut-off value Λ introduces a Λ^3 weight to $\chi^{(2)}$.

The real part is:

$$\begin{aligned}
\text{Re}[\chi_{ii}^{(2)}] &= C^2 A_j A_k \Lambda^3 \left[\frac{\omega_0 \sqrt{\delta_x}}{\pi^2 (\omega^2 + 4\omega_0^2 \delta_x)} - \frac{f(\omega)}{8\pi\omega_0 \sqrt{\delta_x}} \right] \\
f(\omega) &= \omega \left[\delta(2\omega_0 \sqrt{\delta_x} - \omega) - \delta(2\omega_0 \sqrt{\delta_x} + \omega) \right] + \omega_0 \sqrt{\delta_x} \left[2\delta(\omega) - \delta(2\omega_0 \sqrt{\delta_x} + \omega) - \delta(2\omega_0 \sqrt{\delta_x} - \omega) \right], \quad (17)
\end{aligned}$$

and the imaginary part is given in the main text [Eq. (8)]. Dimensional analysis gives the dimensions of the A_k factors in $\chi^{(2)}$, and the n_0^2 factor in $\chi^{(1)}$ contains an implicit integral over $d\omega d^d q$ to ensure dimensional consistency. For simplicity, $|A_k| = 1$ and $|n_0^2| = 1$ are used.

In the limit of static dipoles ($\omega = 0$), both the real and imaginary parts of $\chi^{(1)}$ are zero due to the ω^2 factor. Meanwhile, $\text{Re}[\chi^{(2)}]$ diverges with an overall negative factor, thus determining the static behaviour. The imaginary part of $\chi^{(2)}$ also modifies χ_m significantly by providing an additional positive contribution independent of the distance from the FE QCP. At the FE QCP where $\delta_x = 0$, the real part of $\chi^{(1)}$ is a constant ($d!\lambda^2 V^4 P_0^4 A_k n_0^2$) and the imaginary part is zero. In contrast, the real part of $\chi^{(2)}$ is zero at the FE QCP and the imaginary part provides a positive contribution that depends on energy ($d!\lambda^2 V^4 P_0^4 A_k A_j \Lambda^3 / \pi^2 \omega$).

At the other extreme, of $\omega \rightarrow \infty$, the real part of $\chi^{(1)}$ is a negative constant, while $\text{Re}[\chi^{(2)}]$ is zero. The imaginary parts are $\text{Im}[\chi^{(1)}] = 0$ unless $\delta_x = \infty$ too and $\text{Im}[\chi^{(2)}] = 0$ as $1/\omega$. The system is well behaved in that the rate of energy absorption, as quantified by $\text{Im}[\chi_m]$ is finite, even in the limit of infinite energies [63].

IV: $\chi^{(1)}$ and $\chi^{(2)}$ contributions

The contribution from $\chi^{(2)}$ depends on a momentum-dependent factor Λ^3/ω_0 . As seen in Fig. 4, the regions of positive χ_m after the initial divergence at $\delta_x = \omega^2/\omega_0^2$ are suppressed for small Λ^3/ω_0 , corresponding to large distances (or sample size) for a given phonon frequency ω_0 . The observation of these features close to the FE QCP will depend strongly on the distances and energies

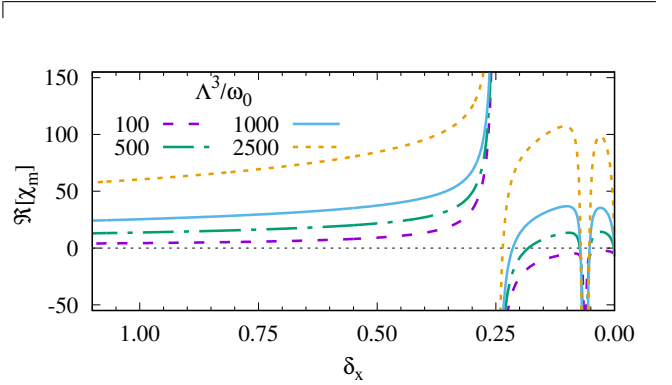


FIG. 4. Real part of χ_m at $\omega/\omega_0 = 0.5$ for several values of Λ^3/ω_0 . Reducing the significance of the $\chi^{(2)}$ contribution can remove the second region of $\chi_m > 0$ near the FE QCP.

considered.

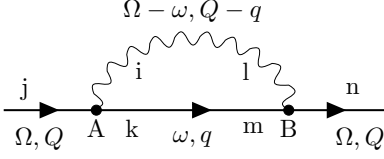
The contributions of $\chi^{(1)}$, given by the Green's function, Eq. (14), and $\chi^{(2)}$, Eq. (17) here and Eq. (8) of the main text, are plotted as functions of δ_x in Fig. 5 (a) and (b), and as functions of energy in Fig. 5 (c) and (d). This highlights the origin of the features seen in Figs. 2 and 3 of the main text, particularly the divergences and $\delta_x = 0$ limit. In all plots, both here and the main text, the δ -functions from $\text{Im}[G^R(\omega)]$ have been replaced by Lorentzian functions.

V: Behaviour in a magnetic field

The additional term in the Lagrangian describing the interaction between a magnetic moment and an external field is:

$$L_B = \mathbf{B} \cdot \mathbf{m} = \lambda \mathbf{B} \cdot (\mathbf{p} \times \partial_t \mathbf{p}). \quad (18)$$

This can be treated as a perturbative term in the full Lagrangian, the second order expansion of which gives the following diagram:



which, using the standard diagrammatic rules, corresponds to the integral:

$$I_B = -A_k \lambda^2 \Omega \int \frac{d^d q}{(2\pi)^d} \frac{d\omega}{2\pi} \omega G^R(\omega, q) G_B(\Omega - \omega, Q - q). \quad (19)$$

G^R is the usual Green's function for the ferroelectric propagator, Eq. (14); G_B is the magnetic propagator:

$$G_B(\omega', q') = \langle B_i(-\omega, -q) B_l(\omega', q') \rangle = B^2 \delta_{il} \delta(\omega - \omega') \delta(q - q'). \quad (20)$$

Energy and momentum conservation at the vertices with zero energy and momentum transfer with the magnetic field gives $\omega' = \Omega - \omega$, $q' = Q - q$. Thus, the integral over the internal energy and momenta evaluates to:

$$I_B = -\frac{A_k \lambda^2 B^2 d! \delta_{jn}}{(2\pi)^{d+1}} \Omega^2 G^R(\Omega, Q) \propto \frac{B^2 \Omega^2}{\omega_Q^2 - \Omega^2}. \quad (21)$$

A magnetic field therefore affects the energy of the FE phonons, but does not, at this level of approximation move the FE QCP.

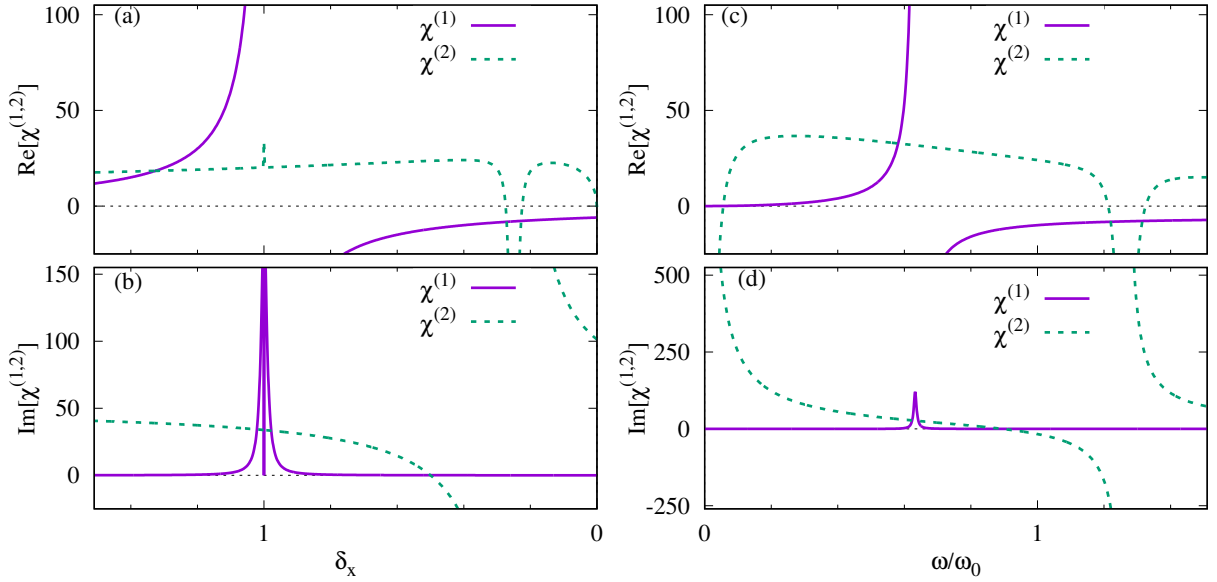


FIG. 5. Real and imaginary parts of $\chi^{(1)}$ (solid purple lines) and $\chi^{(2)}$ (dashed green lines) contributions to χ_m . (a), (b) as a function of distance from the FE QCP at $\delta_x = 0$ at fixed $\omega = \omega_0$; (c), (d) as a function of energy at fixed $\delta_x = 0.4$. In all cases, the scale is in terms of the common size $\lambda^2 V^4 P_0^4$ and the scale of $\chi^{(2)}$ is given by $\Lambda^3/\omega_0 = 1000$. In (a), (c) and (d), the finite width and height of the peaks in $\text{Re}[\chi^{(2)}]$ and $\text{Im}[\chi^{(1)}]$ are the result of replacing $\delta(\omega_0 \sqrt{\delta_x} - \omega)$ by a Lorentzian function.