

Equilibrium nonlinear phononics by electric field fluctuations of terahertz cavities

Emil Viñas Boström^{1,2,*}, Marios H. Michael^{1,*}, Christian Eckhardt¹ and Angel Rubio^{1,2,3,§}

¹*Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany*

²*Nano-Bio Spectroscopy Group, Departamento de Física de Materiales, Universidad del País Vasco, 20018 San Sebastian, Spain*

³*Center for Computational Quantum Physics, Flatiron Institute, Simons Foundation, New York City, New York 10010, USA*



(Received 27 September 2024; revised 2 April 2025; accepted 13 July 2025; published 15 August 2025)

Selective excitation of vibrational modes using strong laser pulses has emerged as a powerful material engineering paradigm. However, to have deterministic control over material properties for device applications, an analogous scheme without drive, operating in thermal equilibrium, is desirable. We here propose such an equilibrium analog of the light-driven paradigm, leveraging the strong coupling between lattice degrees of freedom and the quantum fluctuations of the electric field of a THz microcavity. We demonstrate this approach by showing, using *ab initio* data, how electric field fluctuations can induce a subdominant ferromagnetic order, on top of the dominant zigzag antiferromagnet order, in FePS₃ close to its Néel temperature.

DOI: [10.1103/fqq1-9f52](https://doi.org/10.1103/fqq1-9f52)

Dramatic changes to a material's electronic and magnetic orders can be achieved by driving selected phonon modes far out of equilibrium. This scheme, known as nonlinear phononics [1,2], exploits the nonlinear coupling between various phonon modes to resonantly drive and selectively break symmetries of the equilibrium system, to favor a nonequilibrium state with qualitatively different properties. For phenomena, where the underlying physics is driven by phonon fluctuations, such as ferroelectricity and superconductivity, this can lead to large modifications in the transition temperature [2–8]. Similarly, magnetic orders in magnetic insulators are sensitive to small changes in ionic positions [9–13], because spin interactions typically arise from an interplay of local correlations and virtual kinetic processes, which depend strongly on the bond angles and bond lengths of the crystal [14].

Although nonlinear phononics has been established as a powerful paradigm to manipulate matter, a general drawback of the scheme is that it operates within a nonequilibrium setting. The effect is therefore short-lived, and typically lasts on the order of 10–100 ps. Although in some cases, the effects induced by nonlinearly driven phonons have been found to be quasistationary [11,12,15], with a lifetime extending orders of magnitude beyond the microscopic timescales, the true equilibrium state remains unchanged. To fully unlock the engineering potential of controlling material structures

with light, nonlinear phononics must be extended to thermal equilibrium.

In this work, we make progress in this direction by using that the nonlinear mechanism of shifting the position of a Raman-active phonon through laser pumping is an incoherent process [16]. As a result, the same result can be achieved through electric field noise of the same magnitude. We show that a realistic route toward replacing intense laser light is to use thermal or quantum electric field fluctuations of THz cavities, and provide a concrete example for the antiferromagnet (AFM) FePS₃.

Our work is part of a larger effort to use optical microcavities to effect material control in thermal equilibrium [17–26], resting on the idea that changing a material's dielectric environment can alter its properties [27]. Recent experiments have demonstrated this effect and shown that it is highly off-resonant, and therefore sensitive to changes in the electromagnetic mode structure at all frequencies [20,23]. This type of ground-state reconfiguration is therefore qualitatively different from polaritonic cavity physics, where selected cavity modes are driven and resonantly coupled to excitations of matter [22,28–33]. To capture such off-resonant effects, it is necessary to develop a theoretical framework that accounts for the full-mode structure of the electromagnetic field, as well as their thermal population.

We here propose a scheme to control the properties of two-dimensional quantum materials through their coupling to the fluctuations of a confined electromagnetic field, which we term equilibrium nonlinear phononics. This field can arise from surface phonon polaritons (SPPs) close to a paraelectric insulating substrate [7], from surface plasmon polaritons [34,35] close to a plasmonic substrate, or from an optical microcavity. In contrast to standard nonlinear phononics, this scheme operates in global thermal equilibrium. More specifically, by coupling a Raman-active phonon mode to the vacuum and thermal fluctuations of a confined electric field, we can effect deterministic control over the phonon

*These authors contributed equally to this work.

†Contact author: emil.bostrom@mpsd.mpg.de

‡Contact author: marios.michael@mpsd.mpg.de

§Contact author: angel.rubio@mpsd.mpg.de

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI. Open access publication funded by Max Planck Society.

displacement. To demonstrate the power of this approach, we show how a subdominant ferromagnetic (FM) magnetization appears in the van der Waals antiferromagnet FePS₃, when tuned close to its Néel temperature, due to the collective coupling between spins and phonons, and phonons and SPPs. This magnetization is due to a cavity-mediated asymmetry of the free energy landscape of FePS₃, which is amplified by critical magnetic fluctuations appearing close to the antiferromagnetic transition [15].

The nonlinear phononics scheme proceeds by identifying a material property M that is sensitive to the displacement of a phonon mode Q , such that $M = 0$ for $Q = 0$ but acquires a finite value for $Q \neq 0$. A net displacement of Q is achieved through a nonlinear coupling to some other (typically infrared active) phonon mode Q_{IR} , of the general form gQQ_{IR} . When the infrared (IR) mode is driven by an external electric field, Q_{IR} displays an oscillatory motion that persists during the phonon lifetime τ . The oscillatory motion of Q_{IR} generates a net force on the coordinate Q , leading to a net displacement whose magnitude and direction are set by the coupling constant g . Unless M or Q is trapped in a metastable state, the effect will decay after a time $t \sim \tau$, typically on the order of 10–100 ps.

The key feature of this scheme is the net force on Q effected via the IR-active phonon Q_{IR} . A similar force can be generated through the quadratic coupling to an electric field, λQE^2 , which, however, only persists during the action of the pulse. The situation changes drastically when the electric field of the laser is replaced by the fluctuating electric field of a confined electromagnetic mode. We can therefore imagine replacing the Hamiltonian of a typical nonlinear phononics setup:

$$H = H_{\text{e-p}} + \sum_i g_i QQ_{\text{IR},i}^2 + \sum_i Z_{ij} Q_{\text{IR},i} E_j, \quad (1)$$

where $H_{\text{e-p}}$ is the electron-phonon Hamiltonian, g_i is the nonlinear phonon-phonon coupling, and Z_{ij} is the Born effective charge tensor for the IR mode i , with the Hamiltonian

$$H = H_{\text{e-p}} + \sum_i \lambda_i Q \hat{E}_i^2 + \sum_i \hbar \omega_i a_i^\dagger a_i. \quad (2)$$

Here, $\hat{E}_i$ is the electric field of a quantized electromagnetic mode i , with frequency ω_i , whose corresponding creation and annihilation operators are $a_i^\dagger$ and a_i , respectively. The parameter λ_i is the quadratic light-matter coupling, related to the Raman tensor of mode Q , and quantifies the interaction between the phonon mode and the electric field fluctuations.

For a given phonon mode Q , the quantized electric field effects a dispersive force $\sum_i \lambda_i \langle \hat{E}_i^2 \rangle$ stretching the mode away from its equilibrium configuration. This force is proportional to the mode confinement $1/V_{\text{eff}}$, which for a microcavity is set by the effective mode volume and for SPPs by the cube of the substrate-material distance (see Fig. 1). The above equation illustrates that if the effective force is appreciable, the fluctuations of the confined electromagnetic mode will have the same effect on the material as the nonlinear phonon coupling, but in thermal equilibrium. Such fluctuations are expected to be rather insensitive to the precise cavity geometry, and could be realized, e.g., with a distributed Bragg reflector [22,31], a split-ring resonator [20], or other types of cavities

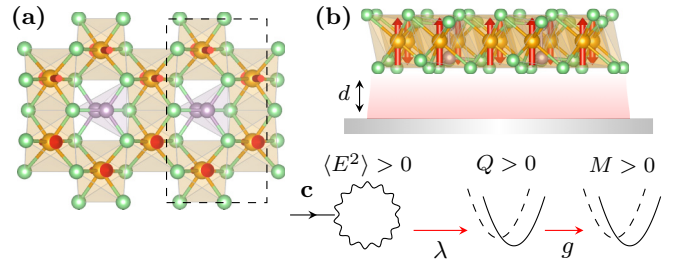


FIG. 1. (a) Top view of a monolayer of the van der Waals antiferromagnet FePS₃, showing the Fe (orange), P (purple), and S (green) ions, as well as the zigzag antiferromagnetic order of the magnetic moments (red). The black dashed lines indicate the magnetic unit cell. (b) Illustration of FePS₃ deposited on a plasmonic substrate, with the substrate-material distance d indicated. For a plasmonic cavity, the effective mode volume is related to the substrate-material distance by $V_{\text{eff}} = d^3$. (c) Illustration of how electric field fluctuations generate a net phonon displacement, which in turn generates a net magnetization.

[36]. For concreteness, however, we focus in the remainder of this work on the electric field coming from SPPs, and show how their coupling to a Raman phonon leads to a finite magnetization in the van der Waals magnet FePS₃.

To calculate the fluctuations of the phonon-polariton electric field, we assume a paraelectric substrate hosting a phonon polariton whose normal to the surface is along the z direction. The components of the electric field operator parallel ($\parallel$) and perpendicular ($\perp$) to the surface can then be expanded in terms of polaritonic mode functions as

$$\mathbf{E}(\mathbf{r}, t) = \sum_{\mathbf{q}} \mathbf{f}_{\mathbf{q}} e^{-qz} e^{i\mathbf{q} \cdot \mathbf{x} - i\omega_s t} (a_{\mathbf{q}}^\dagger + a_{-\mathbf{q}}). \quad (3)$$

Here, ω_s is the SPP frequency, and $\mathbf{f}_{\mathbf{q}}$ is a mode function depending on the transverse optical (TO) and longitudinal optical (LO) phonon frequencies ω_{TO} and ω_{LO} , as well as on the relative permittivities ϵ_{sub} and ϵ_{mat} of the substrate and the material. Since the SPP dispersion is approximately flat as a function of $\mathbf{q}$ [see Fig. 2(a)], we have replaced the frequency $\omega_{\mathbf{q}}$ with ω_s , and note that due to the longitudinal polarization of the phonon-polariton modes the electric field $\mathbf{E} \parallel \mathbf{q}$ for the in-plane components. A detailed discussion of the quantization and orthonormalization of the phonon-polariton modes is found in Notes A–C in the Supplemental Material [37].

For our present purposes, there are two important benefits with SPPs. The first is the geometry of their confinement, localizing the SPPs to a region of height $d = q^{-1}$ above the paraelectric surface. For the interaction with a two-dimensional (2D) system, this confinement gives a close to optimal overlap between the modes and the material. The second benefit is the dispersion of the modes, with its almost flat $\omega_{\mathbf{q}} = \omega_s$ for $|\mathbf{q}| > q_c$ [see Fig. 2(a)]. This gives a large density of states at $\omega = \omega_s$ [see Fig. 2(b)], whose effect is to produce large electric field fluctuations.

At a given temperature T , the electric field fluctuations of SPPs stretching a Raman phonon mode are given by

$$\epsilon_0 \sum_{ij} \langle E_i E_j \rangle_T = \frac{\omega_s^2 - \omega_{\text{TO}}^2}{16\pi \omega_s (\epsilon_{\text{sub}} + \epsilon_{\text{mat}}) d^3} n(\omega_s), \quad (4)$$

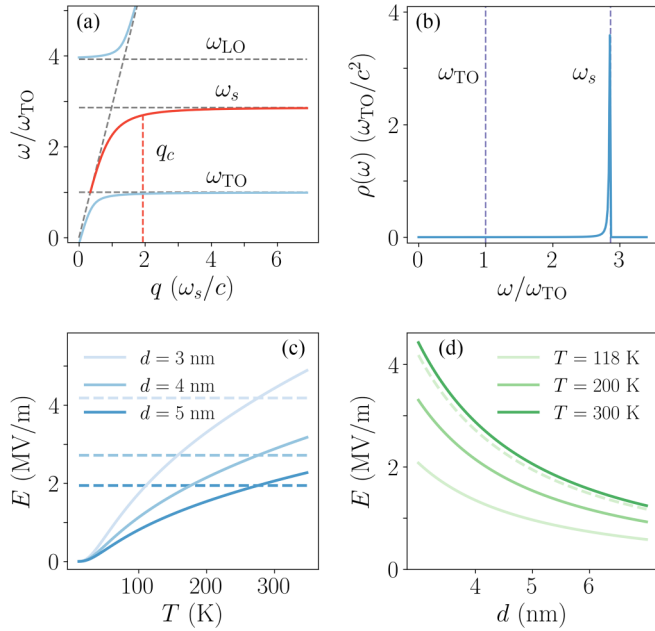


FIG. 2. (a) SPP dispersion (red) as a function of momentum. (b) SPP density of states as a function of frequency. (c) Temperature dependence of the SPP electric field fluctuations for distances $d = 3, 4$, and 5 nm (light blue to dark blue). The dashed lines indicate the vacuum contribution to $E = \sqrt{\langle E^2 \rangle}$, assuming the ultraviolet contribution is unchanged by the paraelectric substrate. (d) Distance dependence of the SPP electric field fluctuations for temperatures $T = 118$ K, 200 K and 300 K (light green to dark green). The dashed line indicates the vacuum contribution. The electric field E is evaluated for the parameters $\omega_s/2\pi = 2.9$ THz, $\omega_{\text{TO}}/2\pi = 1.3$ THz, $\epsilon_{\text{sub}} = 6.3$, and $\epsilon_{\text{mat}} = 1$.

where d is the distance between the substrate and the material, and $n(\omega)$ is the Bose-Einstein distribution. The dependence of the fluctuations $E = \sqrt{\langle E^2 \rangle}$ on d and T is shown in Fig. 2. We note that this coupling is highly off-resonant, and involves a summation over the complete set of SPP modes contributing to the local fluctuations.

To derive the above expression, we calculated the thermal fluctuations using the expression: $E^2 = a^2 + (a^\dagger)^2 + 2a^\dagger a = E^2 - \langle E_{\text{vac}}^2 \rangle$, normal ordered with respect to the phonon-polariton vacuum. It remains a subtle question whether the difference in vacuum fluctuations between free space and a phonon-polariton surface contributes to the stretching of the Raman phonon, as both terms in $\langle E_{\text{vac}}^2 \rangle - \langle E_{\text{vac},0}^2 \rangle$ formally diverge with the ultraviolet cutoff Λ . While we expect these divergences to cancel, yielding a finite positive contribution, the challenge of regularizing the theory in a physically consistent way remains an open question. In Fig. 2, we show an estimate of the vacuum contribution, calculated by assuming that the mode spectrum above $\omega \sim \omega_s$ is unchanged by the paraelectric substrate. We note that the effective light-phonon coupling is even larger if quantum fluctuations also contribute to this coupling.

Interestingly, a qualitatively similar result is obtained for the noise of the electric field parallel to the mirrors of an idealized Fabry-Perot cavity (see Note D in the Supplemental Material [37]). Indeed, in this case, we find $\epsilon_0 \langle E^2 \rangle =$

$7\pi^2\omega_c/(1080h^3)$, where ω_c is the fundamental cavity frequency and h is the height of the cavity. For both the SPPs and the Fabry-Perot cavity, we thus find that the electric field fluctuations are given by the ratio of a characteristic frequency (ω_s or ω_c) to a characteristic volume (d^3 or h^3). Given the disparate nature of these cavities, we believe this to be a general result, such that the fluctuations of a cavity can be estimated from its characteristic frequency and length scales. We note that this is a nontrivial result, since the volume of each independent mode is infinite due to the macroscopic in-plane extent of the cavities. However, when all modes are summed, their contribution is finite, highlighting the importance of a multimode treatment.

We now discuss the effect of SPP fluctuations on FePS₃, whose magnetoelastic properties are described by the Hamiltonian [9,38]

$$H = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \Delta \sum_{ij} S_i^z S_j^z + \sum_{\alpha} \frac{\Omega_{\alpha}}{2} [P_{\alpha}^2 + Q_{\alpha}^2]. \quad (5)$$

Here, the parameters J_{ij} encode the coupling of spins $\mathbf{S}_i$ and $\mathbf{S}_j$, Δ is an Ising anisotropy, P_{α} and Q_{α} are the momentum and coordinate of the phonon mode α , and Ω_{α} is the corresponding phonon energy. The magnetic interactions depend on the ionic positions, which generates an interaction between the spins and lattice displacements. For small displacements Q , the exchange interaction can be expanded as $J_{ij}(Q) \approx J_{ij} - \alpha_{ij}Q$, where α_{ij} quantifies the phonon modulation of the magnetic interactions. All parameters of this Hamiltonian have been calculated from first principles within the frozen phonon approximation [15] (see Note K in the Supplemental Material [37]). The elements of α_{ij} were found to decay rapidly with interatomic distance, and to be significant only for nearest-neighbor spins.

Below its Néel temperature $T_N = 118$ K, FePS₃ orders into a zigzag antiferromagnet as illustrated in Fig. 1(a). It was recently observed that when subjected to intense THz pulses, at temperatures close to T_N , the system develops a subdominant ferromagnetic order on top of the antiferromagnetic order [15]. The THz-induced magnetization is linked to the nonlinear excitation of a Raman-active phonon mode at $\Omega = 3.27$ THz, which interacts strongly with the magnetic structure.

To describe the interaction between this Raman phonon mode and the magnetic orders, the Hamiltonian in Eq. (5) can be mapped onto an effective theory in terms of the phonon coordinate Q , and the macroscopic magnetic variables L and M corresponding to the AFM and FM orders (related to the microscopic spins as discussed in Ref. [15], Appendix B, and Note F in the Supplemental Material [37]). In the effective theory, the system is described by a Ginzburg-Landau free energy

$$F = \frac{a_L(T)}{2} L^2 + \frac{b_L}{4} L^4 + \frac{a_M(T)}{2} M^2 + \frac{b_M}{4} M^4 + \frac{\Omega}{2} Q^2 + g(L - \langle L \rangle)MQ. \quad (6)$$

The general form of the coupling follows from symmetry, by noting that both L and Q are finite momentum orders (Q is a zone-folded phonon), and that L and M are both odd

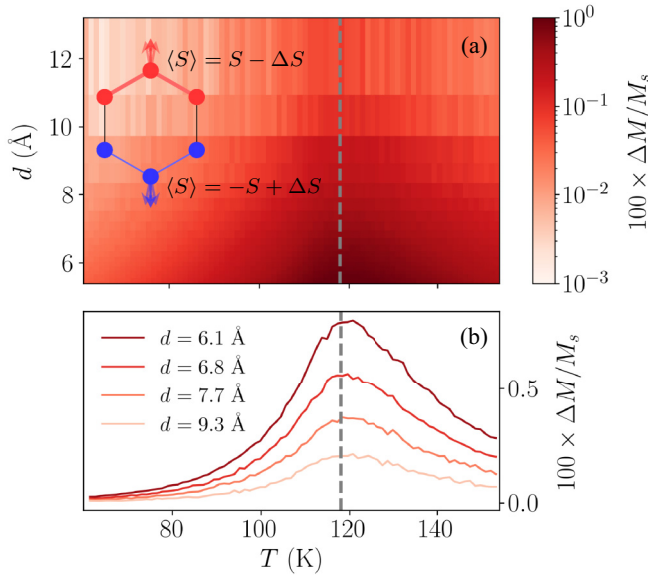


FIG. 3. (a) Equilibrium magnetization induced by the coupling between surface phonon polariton fluctuations and the phonon mode Q , as a function of temperature T and substrate-material distance d . The phonon energy is $\Omega = 13.5$ meV (3.27 THz) and the spin-phonon coupling is $g = 0.5$ meV. The results were obtained from classical Monte Carlo simulations of the spin-phonon Hamiltonian in Eq. (5) on a spin lattice with 20×20 unit cells (800 spins), at fixed coupling λ . The value for λ was then translated into a distance d , as discussed in the text. Inset: The Raman phonon increases and decreases magnetic interactions within every other zigzag chain, indicated by thick red lines and thin blue lines. This leads to a suppression of the fluctuations ΔS in the red chains, and an increase of ΔS in the blue chains, and thereby to a net magnetization. (b) Line cuts of the data in panel (a) for selected values of the substrate-material distance.

under inversion. The specific form of the coupling has been corroborated by spin Monte Carlo simulations of the magnetic structure, with the microscopic magnetic interactions modulated according to the phonon displacement pattern [15] (see Notes G–I in the Supplemental Material [37]). The real space motion of the atoms corresponding to this phonon mode modifies the bond lengths between the Fe atoms, which enhances the exchange interaction within every other FM zigzag chain, while weakening it within adjacent chains (see Fig. 3). The effect of the modulation is to decrease or increase the magnetic fluctuations ΔS in every other chain, leading to a net magnetization from the fluctuating moments. Far above or far below the Néel temperature, where there is no magnetic order or it is completely frozen in, the effect of the phonon is negligible.

The phonon mode Q is Raman active, and therefore couples to the fluctuations of the cavity electric field via the Raman tensor. The coupling is of the form (see Appendix A)

$$H_R \approx \frac{\epsilon_0 V_c}{2} \hat{\mathbf{E}} \frac{\partial \epsilon}{\partial Q_i} \hat{\mathbf{E}} \hat{Q}_i, \quad (7)$$

where V_c is the unit cell volume, ϵ is the relative permittivity matrix, and all operators are evaluated in a given unit cell i . We note that when transitioning from the local

picture to a picture in terms of the macroscopic variable Q (the $q = 0$ component of the phonon mode), the Raman tensor is suppressed by a factor $1/\sqrt{N}$, while $Q_i = \sqrt{N}Q$. The coupling to the macroscopic mode therefore remains finite in the thermodynamic limit. Introducing the phonon operator $\hat{Q} = (l_0/\sqrt{2})(b^\dagger + b)$, where $l_0 = \sqrt{\hbar/(M\Omega)}$ is the phonon oscillator length and M is its effective mass, the coupling can be written as $H_R = \lambda(L/L_s)(b^\dagger + b)$ with the effective Raman coupling $\lambda = (\epsilon_0 V_c l_0)/(2\sqrt{2})(\hat{\mathbf{E}} \partial_Q \epsilon \hat{\mathbf{E}})$. Here, the factor L/L_s , with L_s the saturation magnetization, is included to account for the fact that the sign of Q is defined relative to the sign of L [15].

This expression for λ holds irrespective of the origin of the electric field, but for a numerical estimate, we evaluate the local electric field fluctuations arising from the SPPs of SrTiO₃. For a substrate-material distance $d = 1$ nm and a temperature $T = T_N = 118$ K, the electric field fluctuations of the vacuum are found to be $E = \sqrt{\langle E^2 \rangle} \approx 21.7$ MV m⁻¹, while the full temperature-dependent field is $E(T) \approx 30.0$ MV m⁻¹. We note that this field strength is comparable to the peak value used in recent experiments [15], such that a finite effect on the system is expected. With the Raman tensor $\partial \epsilon / \partial Q_i = 4.2a_0^{-1}$, unit cell volume $V_c \approx 10^3$ Å³, and oscillator length $l_0 = 0.35a_0$, as obtained from first-principles simulations (see Note K in Supplemental Material [37]), this gives an effective coupling of $\lambda \approx 26$ μeV stretching the phonon by $\Delta Q = 0.34$ mÅ.

Having calculated the phonon-cavity coupling, the Ginzburg-Landau theory can now be used to investigate the effect of the SPPs on the magnetic order. Minimizing the phonon energy gives a displaced equilibrium position $Q_0 = \lambda L/(\Omega L_s) - g(L - \langle L \rangle)M/\Omega$, which will influence the magnetization. To calculate the induced magnetization, the Ginzburg-Landau equation for M is solved to lowest order in λ/Ω . Introducing for simplicity the coefficient $c_M(T) = g\lambda(\langle L^2 \rangle - \langle L \rangle^2)/(\Omega L_s)$, the location of the free energy minimum is given by $M_0 = c_M/\bar{a}_M$. Here, $\bar{a}_M(T) = a_M(T) - (g^2/\Omega)(\langle L^2 \rangle - \langle L \rangle^2)$ is a renormalized quadratic term for M , arising from the coupling between M and L by the phonon. Since L is the dominant order parameter, the coefficient $a_L(T) = a_{L0}(T - T_N)^\beta$ will change sign at the Néel temperature, leading to a finite value $\langle L \rangle \sim (T_N - T)^\beta$. Similarly, the fluctuations of L will show a critical enhancement at the Néel temperature, such that $\chi_L = V(\langle L^2 \rangle - \langle L \rangle^2) \sim |T - T_N|^{-\gamma}$.

To obtain the induced magnetization ΔM , we can write $\Delta M = M_0 P_0$, where P_0 is the probability distribution for M evaluated at M_0 . Assuming the probability distribution is localized at the minimum, we can take $P_0 = 1$ to obtain $\Delta M = M_0 = c_M/\bar{a}_M$. Close to the critical temperature, there is a finite magnetization

$$\Delta M = \left(\frac{g\lambda}{\Omega L_s} \right) \frac{\chi_L}{V a_M - (g^2/\Omega) \chi_L} \approx \left(\frac{g\lambda}{\Omega a_M} \right) \frac{\chi_L}{V L_s}, \quad (8)$$

where we have assumed $V a_M \gg \chi_L$ in the macroscopic limit. This result is in good agreement with spin Monte Carlo simulations (see Fig. 3), showing a finite magnetization developing for $T \approx T_N$. A similar magnetization was recently observed when driving the system with intense THz radiation [15].

From a finite-size scaling analysis (see Note J in the Supplemental Material [37]), it is expected that $\chi_L \sim L^{\gamma/\nu} = L^{2-\eta}$ for temperatures close to T_N , where η is the anomalous dimension. Therefore, the magnetization is expected to scale like $\chi_L/L^d \sim L^{-1/4}$ in two dimensions and $\chi_L/L^d \sim L^{-1.036}$ in three dimensions. However, these scaling laws assume a cubic system with volume $V = L^d$. In the present case, the system is highly asymmetric, with the vertical extent $L_z \ll L$. For such a system, the critical exponents are expected to follow the three-dimensional universality class, while the scaling of the susceptibility gives $\chi_L/V = (\chi_L/L^2)L_z^{-1} \sim L^{-0.036}$ in the macroscopic limit.

These scaling arguments combine a mean-field analysis of F with renormalization group exponents for χ_L and ξ , and predict a magnetization weakly decaying with system size. This should be compared with our Monte Carlo simulations, which predict a size-independent magnetization density (see Fig. S2 [37]). Since the Monte Carlo simulations include higher-order terms in the Ginzburg-Landau theory, the discrepancy is likely due to higher-order effects neglected in the analytical analysis. In particular, we note that using the mean-field exponents for χ_L gives an anomalous dimension $\eta = 0$, such that the magnetization becomes independent of volume close to T_N , in line with the Monte Carlo results.

The results in Fig. 3 show that close to the Néel temperature, electric field fluctuations of the SPP modes induce a finite magnetization in FePS₃. In contrast to standard nonlinear phononics, this magnetization appears in thermal equilibrium and thereby amounts to a true reconfiguration of the equilibrium magnetic state. Due to the strong links between nonlinear phononics experiments and the present protocol, we expect that quite generally, nonequilibrium states of 2D materials induced by resonant nonlinear driving have an equilibrium analog realized through the coupling to SPPs. In particular, we note that the Ginzburg-Landau free energy discussed above is rather general, and appears across a wide range of systems with intertwined dominant and subdominant orders [39]. Provided that a coupling of the form $gLMQ$ is realized for some phonon mode, we therefore expect a similar phenomenology for the dominant and subdominant orders L and M , irrespective of their physical nature. This observation opens the door to stabilizing exotic electronic phases in quantum materials in thermal equilibrium.

More broadly, we emphasize that while the particular effect of a phonon-induced magnetization is relatively moderate, the electric field fluctuations generated by the SrTiO₃ substrate are very large in amplitude and rival the amplitude of state-of-the-art THz lasers. This implies that equilibrium nonlinear phononics could rival experiments performed by coherent lasers in a noise-driven context.

ACKNOWLEDGMENTS

We thank Daniel A. Bustamante Lopez and Dominik M. Juraschek for insightful discussions. E.V.B. acknowledges funding from the European Union's Horizon Europe research and innovation program under the Marie Skłodowska-Curie Grant Agreement No. 101106809 (CavityMag), and M.H.M. acknowledges support from the Alexander von Humboldt Foundation. We further acknowledge support from the

Cluster of Excellence “CUI: Advanced Imaging of Matter”—EXC 2056—project ID 390715994 and SFB-925 “Light induced dynamics and control of correlated quantum systems”—project 170620586 of the Deutsche Forschungsgemeinschaft (DFG), from Grupos Consolidados (IT1453-22), and from the Max Planck-New York City Center for NonEquilibrium Quantum Phenomena at the Flatiron Institute. The Flatiron Institute is a division of the Simons Foundation.

APPENDIX A: RAMAN COUPLING AND SURFACE PHONON POLARITON FLUCTUATIONS

The phonon mode at 3.27 THz is Raman active and folded from the M -point of the paramagnetic unit cell. It therefore couples to the fluctuations of the SPP modes via a Raman-like term of the form

$$H_R = \int d^3x \frac{\epsilon_0 \hat{E}_l(x) \hat{E}_m(x)}{2} \frac{\partial \epsilon_{lm}(\mathbf{Q}_i)}{\partial Q_i} u_i(x) \hat{Q}_i \approx V_{\text{cell}} \frac{\partial \epsilon_{lm}}{\partial Q_i} \frac{\hat{E}_l \hat{E}_m}{2} \hat{Q}_i, \quad (\text{A1})$$

where in the last, line all operators are evaluated in unit cell i . Here, $u_i(x)$ corresponds to the wave function of the Raman mode at position i , which we take to be localized on a single unit cell so that $\int d^3x u_i(x) = V_{\text{cell}}$. In terms of phonon operators, this coupling can be written as $H_R = \lambda(L/L_s)(b^\dagger + b)$, with the effective Raman coupling $\lambda = (\epsilon_0 V_{\text{cell}} l_0)/(2\sqrt{2})(\hat{\mathbf{E}} \partial_Q \epsilon \hat{\mathbf{E}})$. Here, the factor L/L_s , with L_s the saturation magnetization, is included to account for the fact that the sign of Q is defined relative to the sign of L [15].

To estimate the value of g , we use SPPs to compute the local fluctuations $\langle E^2 \rangle$. Expanding the modes as

$$E_{\parallel}(\mathbf{x}, z, t) = \sum_q \frac{\mathbf{q}(-i\omega_s) f_q}{|q|} e^{-qz} e^{i\mathbf{q} \cdot \mathbf{x} - i\omega_s t} (a_q^\dagger - a_{-q}),$$

$$E_{\perp}(\mathbf{x}, z, t) = \sum_q \omega_s f_q e^{-qz} e^{i\mathbf{q} \cdot \mathbf{x} - i\omega_s t} (a_q^\dagger + a_{-q}),$$

$$f_q = \sqrt{\frac{q(\omega_s^2 - \omega_{\text{TO}}^2)}{4A\epsilon_0(\epsilon_{\text{sub}} + \epsilon_{\text{mat}})\omega_s^3}}, \quad (\text{A2})$$

where ω_s is the SPP frequency, A is the surface area, ω_{TO} is the frequency of the TO phonon, ϵ_{sub} and ϵ_{mat} are the relative dielectric constants of the phonon-polariton substrate and the material, respectively, and d is the substrate-material distance. The electric field fluctuations can be evaluated as

$$\epsilon_0 \sum_{lm} \langle E_l E_m \rangle = \sum_{lm} \delta_{lm} \left(\frac{\delta_{lx}}{2} + \frac{\delta_{ly}}{2} + \delta_{lz} \right) \int \frac{d^2q}{(2\pi)^2} \frac{(\omega_s^2 - \omega_{\text{TO}}^2) q e^{-2qd}}{4(\epsilon_{\text{sub}} + \epsilon_{\text{mat}})\omega_s} = \frac{\omega_s^2 - \omega_{\text{TO}}^2}{16\pi\omega_s(\epsilon_{\text{sub}} + \epsilon_{\text{mat}})d^3}. \quad (\text{A3})$$

For an SrTiO₃ substrate a distance 5 nm away from the magnetic material, this gives electric field fluctuations of the order $\langle E^2 \rangle \sim 1 \text{ MV m}^{-1}$. We note that this has the general form of a characteristic frequency (ω_s) over a characteristic volume (d^3).

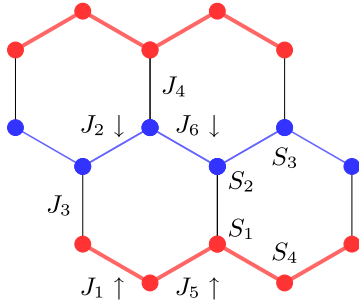


FIG. 4. Spatial modulation pattern of the magnetic interactions induced by the 3.27 THz Raman phonon mode. An increase in the magnetic interaction on a given bond is illustrated by a thick line and an upward-pointing arrow $\uparrow$, while a decrease is illustrated by a thin line and a downward-pointing arrow $\downarrow$.

APPENDIX B: DERIVATION OF THE LOCAL GINZBURG-LANDAU THEORY

The low-energy magnetoelastic properties of FePS₃ can be described by a Hamiltonian of the form [10,38]

$$H = \sum_{ij} \mathbf{S}_i \cdot (\mathbf{J}_{ij} \mathbf{S}_j) - \Delta \sum_i (S_i^z)^2 + \frac{1}{2} \sum_{\alpha} [P_{\alpha}^2 + \Omega_{\alpha}^2 Q_{\alpha}^2], \quad (\text{B1})$$

where the matrix $\mathbf{J}_{ij}$ encodes the (possibly anisotropic) couplings of spins $\mathbf{S}_i$ and $\mathbf{S}_j$, and Δ is a single ion anisotropy. P_{α} and Q_{α} are the momentum and displacement operators of phonon mode α , respectively, and Ω_{α} is the corresponding phonon frequency. In general, the magnetic interactions depend on the atomic positions, giving rise to an interaction

between the spins and the lattice displacements. For small displacements Q_{α} , the exchange tensor can be expanded as

$$\mathbf{J}_{ij}(Q) \approx \mathbf{J}_{ij} - \alpha_{ij} Q, \quad (\text{B2})$$

where the tensor α_{ij} quantifies the phonon modulation of the magnetic interactions.

The magnetic exchange interactions as well as the modulations α_{ij} for the 3.27 THz phonon mode were calculated from first principles within the frozen phonon approximation [15]. We find that α_{ij} decays rapidly with interatomic distance, and is significant only for nearest-neighbor exchange interactions. Due to the space group symmetries of the system, only six nearest-neighbor bonds of the magnetic unit cell are independent, and are henceforth labeled J_i in accordance with Fig. 4.

The spatial pattern in which the phonon modulates the magnetic interactions is shown in Fig. 4, and the strength of the couplings was found to be $|\alpha| = 17 \text{ meV}\text{\AA}^{-1}$. As the diagonal components of α are much larger than the off-diagonal components, α can be treated as a scalar.

Defining the zigzag antiferromagnetic and ferromagnetic order parameters $L = S_1 - S_2 - S_3 + S_4$ and $M = S_1 + S_2 + S_3 + S_4$ (within a given magnetic unit cell), the microscopic modulations of the magnetic parameters can be mapped onto a theory of macroscopic variables. These macroscopic variables are expected to govern the low-energy and long-wavelength behavior of the theory. More specifically, we can write the spin-phonon coupling as

$$H_{\text{int}}^{(i)} = \gamma Q_i (S_{2i} S_{3i} - S_{1i} S_{4i}) = -g Q_i L_i M_i, \quad (\text{B3})$$

where Q_i is the Raman phonon coordinate, L_i is the zigzag order, and M_i is the magnetization of cell i , coupled through the constant g .

-
- [1] R. Mankowsky, M. Först, and A. Cavalleri, Non-equilibrium control of complex solids by nonlinear phononics, *Rep. Prog. Phys.* **79**, 064503 (2016).
 - [2] A. S. Disa, T. F. Nova, and A. Cavalleri, Engineering crystal structures with light, *Nat. Phys.* **17**, 1087 (2021).
 - [3] T. F. Nova, A. S. Disa, M. Fechner, and A. Cavalleri, Metastable ferroelectricity in optically strained SrTiO₃, *Science* **364**, 1075 (2019).
 - [4] X. Li *et al.*, Terahertz field-induced ferroelectricity in quantum paraelectric SrTiO₃, *Science* **364**, 1079 (2019).
 - [5] M. Budden *et al.*, Evidence for metastable photo-induced superconductivity in K₃C₆₀, *Nat. Phys.* **17**, 611 (2021).
 - [6] E. Rowe *et al.*, Resonant enhancement of photo-induced superconductivity in K₃C₆₀, *Nat. Phys.* **19**, 1821 (2023).
 - [7] C. J. Eckhardt *et al.*, Theory of resonantly enhanced photo-induced superconductivity, *Nat. Commun.* **15**, 2300 (2024).
 - [8] S. Chattopadhyay *et al.*, Metastable photo-induced superconductivity far above T_c , *npj Quantum Mater.* **10**, 34 (2025).
 - [9] Q. Zhang *et al.*, Coherent strong-coupling of terahertz magnons and phonons in a van der Waals antiferromagnetic insulator, [arXiv:2108.11619](https://arxiv.org/abs/2108.11619).
 - [10] J. Cui *et al.*, Chirality selective magnon-phonon hybridization and magnon-induced chiral phonons in a layered zigzag antiferromagnet, *Nat. Commun.* **14**, 3396 (2023).
 - [11] B. Truc *et al.*, Light-induced metastable hidden skyrmion phase in the Mott insulator Cu₂OSeO₃, *Adv. Mater.* **35**, 2304197 (2023).
 - [12] A. S. Disa *et al.*, Photo-induced high-temperature ferromagnetism in YTiO₃, *Nature (London)* **617**, 73 (2023).
 - [13] E. Ergeçen *et al.*, Coherent detection of hidden spin-lattice coupling in a van der Waals antiferromagnet, *Proc. Natl. Acad. Sci. USA* **120**, e2208968120 (2023).
 - [14] G. Jackeli and G. Khaliullin, Mott insulators in the strong spin-orbit coupling limit: From Heisenberg to a quantum compass and Kitaev models, *Phys. Rev. Lett.* **102**, 017205 (2009).
 - [15] B. Ilyas *et al.*, Terahertz field-induced metastable magnetization near criticality in FePS₃, *Nature (London)* **636**, 609 (2024).
 - [16] M. Först *et al.*, Nonlinear phononics as an ultrafast route to lattice control, *Nat. Phys.* **7**, 854 (2011).
 - [17] S. Latini *et al.*, The ferroelectric photo ground state of SrTiO₃: Cavity materials engineering, *Proc. Natl. Acad. Sci.* **118**, e2105618118 (2021).

- [18] J. B. Curtis, M. H. Michael, and E. Demler, Local fluctuations in cavity control of ferroelectricity, *Phys. Rev. Res.* **5**, 043118 (2023).
- [19] Ojeda H. P. Collado, M. H. Michael, J. Skulte, A. Rubio, and L. Mathey, Equilibrium parametric amplification in Raman-cavity hybrids, *Phys. Rev. Lett.* **133**, 116901 (2024).
- [20] F. Appugliese *et al.*, Breakdown of topological protection by cavity vacuum fields in the integer quantum Hall effect, *Science* **375**, 1030 (2022).
- [21] E. Viñas Boström, A. Sriram, M. Claassen, and A. Rubio, Controlling the magnetic state of the proximate quantum spin liquid α -RuCl₃ with an optical cavity, *npj Comput. Mater.* **9**, 202 (2023).
- [22] F. Dimberger *et al.*, Magneto-optics in a van der Waals magnet tuned by self-hybridized polaritons, *Nature (London)* **620**, 533 (2023).
- [23] G. Jarc *et al.*, Cavity-mediated thermal control of metal-to-insulator transition in IT-TaS₂, *Nature (London)* **622**, 487 (2023).
- [24] R. Riolo *et al.*, Tuning Fermi liquids with polaritonic cavities, *Proc. Natl. Acad. Sci. USA* **122**, e2407995122 (2025).
- [25] C. J. Eckhardt *et al.*, Cavity-mediated ultra-strong light-matter coupling of itinerant electrons, [arXiv:2409.10615](https://arxiv.org/abs/2409.10615).
- [26] J. Bloch, A. Cavalleri, V. Galitski, M. Hafezi, and A. Rubio, Strongly correlated electron-photon systems, *Nature (London)* **606**, 41 (2022).
- [27] H. Hübener, E. V. Boström, M. Claassen, S. Latini, and A. Rubio, Quantum materials engineering by structured cavity vacuum fluctuations, *Mater. Quantum Technol.* **4**, 023002 (2024).
- [28] J. Flick, M. Ruggenthaler, H. Appel, and A. Rubio, Atoms and molecules in cavities, from weak to strong coupling in quantum-electrodynamics (QED) chemistry, *Proc. Natl. Acad. Sci. USA* **114**, 3026 (2017).
- [29] M. A. Sentef, M. Ruggenthaler, and A. Rubio, Cavity quantum-electrodynamical polaritonically enhanced electron-phonon coupling and its influence on superconductivity, *Sci. Adv.* **4**, eaau6969 (2018).
- [30] A. Thomas *et al.*, Tilting a ground-state reactivity landscape by vibrational strong coupling, *Science* **363**, 615 (2019).
- [31] F. Dimberger *et al.*, Spin-correlated exciton-polaritons in a van der Waals magnet, *Nat. Nanotechnol.* **17**, 1060 (2022).
- [32] Y. J. Bae *et al.*, Exciton-coupled coherent magnons in a 2D semiconductor, *Nature (London)* **609**, 282 (2022).
- [33] M. K. Svendsen *et al.*, Theory of quantum light-matter interaction in cavities: Extended systems and the long wavelength approximation, [arXiv:2312.17374](https://arxiv.org/abs/2312.17374).
- [34] K. Lenk, J. Li, P. Werner, and M. Eckstein, Dynamical mean-field study of a photon-mediated ferroelectric phase transition, *Phys. Rev. B* **106**, 245124 (2022).
- [35] G. Kipp *et al.*, Cavity electrodynamics of van der Waals heterostructures, [arXiv:2403.19745](https://arxiv.org/abs/2403.19745).
- [36] H. Hübener *et al.*, Engineering quantum materials with chiral optical cavities, *Nat. Mater.* **20**, 438 (2020).
- [37] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/fqq1-9f52> for additional details on the quantization and orthogonalization of the surface phonon polariton (SPP) modes, the derivation of the SPP-phonon coupling, an explicit calculation of the SPP density of states, the derivation of the effective Ginzburg-Landau theory, additional details on the spin Monte Carlo simulations, a finite size analysis of the critical magnetic properties, and details on the first principles calculations.
- [38] S. Liu *et al.*, Direct observation of magnon-phonon strong coupling in two-dimensional antiferromagnet at high magnetic fields, *Phys. Rev. Lett.* **127**, 097401 (2021).
- [39] E. Fradkin, S. A. Kivelson, and J. M. Tranquada, *Colloquium: Theory of intertwined orders in high temperature superconductors*, *Rev. Mod. Phys.* **87**, 457 (2015).