Laser-induced coherent control of an electronic nematic quantum phase transition

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In recent years, nonequilibrium studies of strongly-correlated electron systems have undergone significant progress, due to both theoretical advances [1–3] and outstanding developments in ultrafast pump-probe techniques [4–7]. A major goal in this field is to obtain coherent control of quantum materials, using laser pulses to switch between different electronic states of their complex phase diagram. A natural route to realizing such control is to exploit the electron-phonon coupling and coherently excite appropriate optical phonon modes [7–11]. In this work, we demonstrate that the nonequilibrium excitation of $E_u$ phonons in tetragonal systems can be employed to quench, via nonlinear effects, an electronic nematic state across a quantum phase transition and at the same time avoid excessive heating. While this result is general, as it relies only on the symmetry properties of the system, we illustrate its applicability by considering the microscopic parameters of the nematic unconventional superconductor FeSe. Because electronic nematicity is present in several correlated systems, including the known families of unconventional superconductors [12–15], our result provides an efficient and feasible route to control quantum phenomena in correlated materials.

The basis for light control of quantum materials via electron-phonon interactions is to precisely excite infrared-active optical phonon modes via strong terahertz laser pulses [16–19]. Because infrared phonon modes do not couple directly to the electronic charge density, the changes in electronic structure and interactions are mediated by nonlinear effects [20, 21]. Importantly, the ordered states that can be most efficiently controlled by this approach are those that couple strongly to the lattice, such as superconductivity or metal-to-insulator transitions [20–23].

Another correlated state known to be intimately coupled to the lattice is electronic nematicity [24–26]. In this state, electronic degrees of freedom spontaneously break the rotational symmetry of the system [27], inevitably triggering a structural distortion. Nematic phases have been widely observed in quantum materials, from unconventional superconductors such as cuprates, pnictides, and heavy fermions, to ruthenates and semiconductors displaying the Quantum Hall effect [12]. The driving mechanism of nematicity in these compounds and its relationship with other phenomena such as superconductivity, magnetism, and charge-order, are all hotly debated [14, 28, 29]. Most suggestively, in several of these materials it is widely believed that the nematic phase ends at a putative quantum critical point (QCP) [30], which may host an exotic non-Fermi-liquid phase. Controlling the nematic degrees of freedom would therefore provide a high-precision tool for studying the competing and intertwined orders in these systems. The fact that nematic order couples strongly with the lattice suggests that light control provides a feasible route, which has hitherto remained largely unexplored.

In this paper, we provide a theory of nonequilibrium optical control of a generic nematic phase on the square lattice. The onset of nematic order breaks the tetragonal symmetry either by making the horizontal and vertical bonds inequivalent (called $d_{x^2−y^2}$-wave, or $B_{1g}$, nematic order) or the diagonals inequivalent (called $d_{xy}$-wave, or $B_{2g}$, nematic order). For concreteness, we consider hereafter the system to be close to an instability in the $B_{1g}$ nematic channel — similar results hold for an instability in the $B_{2g}$ channel. The orthorhombic lattice distortion that accompanies nematic order is due to the linear coupling between the electrons and a transverse acoustic phonon mode that propagates along the [110] direction. The phonon velocity is strongly renormalized by nematic fluctuations, and vanishes at the nematic transition. Although this linear nemato-elastic coupling has been widely employed to investigate the nematic properties of many materials [15, 30], the acoustic nature of the phonon makes it inconvenient for optical control [31]. Instead of acoustic phonons, we suggest to control the nematic phase by optically exciting the $E_u$ optical phonon mode, corresponding to two degenerate planar stretching lattice vibrations. This infrared-active mode, ideal for laser manipulation, is present in any system with a tetragonal point group. Furthermore, symmetry forbids both incoming photons and the $E_u$ phonon from coupling linearly to the electronic density. However, the $E_u$ vibrations do couple quadratically to the electronic nematic degrees of freedom. Therefore, in contrast to traditional pump-probe setups, the lattice is excited directly, whereas the electronic density in the nematic channel is excited only indirectly, via the nonlinear coupling to the phonons.

The difference between the linear coupling to the acoustic phonon mode and the quadratic coupling to the $E_u$ optical mode is sketched in Fig. 1. For illustration purposes, consider a square lattice consisting of balls and springs, with electrons that can hop from site to site and may have some charge profile. When electrons and phonons are linearly coupled, the main effect of electronic...
FIG. 1. Distinct types of nemato-elastic coupling. The lattice here is depicted by a springs-and-balls caricature, whereas the local electronic charge distributions are illustrated by charge quadrupoles on the lattice sites. In the case of linear nemato-elastic coupling, via the lattice \( B_{1g} \) mode, the breaking of rotational symmetry is manifested as a lattice deformation, as seen on the left. In contrast, if the coupling is quadratic, via the degenerate \( E_u \) phonons, the breaking of rotational symmetry manifests itself through a change of the resonant frequencies of the two planar modes, as seen on the right. Consequently, exciting the lattice \( B_{1g} \) mode induces a nonzero nematic order parameter, while exciting the \( E_u \) modes induces nematic fluctuations, and thus an effective attraction in the electronic nematic channel.

nematic order is the modification of the rest positions of the balls. However, when the coupling is quadratic, the effect is the modification of the spring constants, causing a shift in the resonance frequencies of the previously degenerate phonon modes \([21, 32]\). When these modes are excited out of equilibrium, this resonance-shift oscillates rapidly, and the background of oscillations enhances the much slower electronic quadrupole fluctuations, pushing the system towards the nematic phase. The shift is proportional to the number of excited phonons, and by extension to the laser beam intensity. As we show below, the nematic susceptibility \( \chi_{\text{nem}} \) is enhanced according to

\[
\chi_{\text{nem}}^{-1} \rightarrow \chi_{\text{nem}}^{-1} - \frac{\lambda^2}{2\hbar \Omega_{\text{ph}}} n_{\text{ph}}
\]  

where \( \Omega_{\text{ph}} \) is the \( E_u \) phonon resonance frequency, \( \lambda \) is the (quadratic) coupling constant, and \( n_{\text{ph}} \propto |\varepsilon_0|^2 \) is the laser-induced phonon occupation number, where \( \varepsilon_0 \) is the electric field strength.

Crucial to our result is the fact that, near the nematic instability, due to the phenomenon of critical slowing down, the phonons are much faster than the nematic fluctuations. The reason for this is the aforementioned difference between the impact of a nematic transition on the acoustic mode and the optical \( E_u \) modes. Whereas the acoustic mode softens at the transition, quadratic coupling leads to a small splitting of the \( E_u \) phonons, breaking the degeneracy but leaving both modes gapped. This separation of lattice and nematic time scales ensures that the change in the effective nematic interaction is quasi-instantaneous. Furthermore, the absence of a direct coupling between light and electrons ensures long electronic heating times, maintaining the quantum nature of the system. As we illustrate below using experimental parameters of the material FeSe, a posterchild of nematic order, these features support a feasible protocol for an optically-induced quantum quench of the electronic nematic transition.

To derive these results, we start from a general, material-independent model for coupled nematic degrees of freedom and \( E_u \) phonons. We are not concerned with the microscopic mechanism of the nematic transition, and merely describe it by a bosonic field \( \phi \) that transforms as the \( B_{1g} \) irreducible representation of the tetragonal point group. In terms of the electronic field operators \( \psi(q, \omega) \), the nematic bosonic field is proportional to the quadrupolar charge density, given by

\[
\phi(q, \Omega) \sim \sum_{k,\omega}(k_x^2 - k_y^2)\psi(k + q/2, \omega + \Omega/2)\psi(k - q/2, \omega - \Omega/2);
\]

alternatively, it could also be written as a composite order from e.g. spin fluctuations. In any case, close enough to the nematic transition, the nematic degrees of freedom are described by the effective Hamiltonian:

\[
H_{\text{nem}} = \sum_{x_i} \phi(x_i, t) \chi_0^{-1} r \phi(x_i, t),
\]
where we assumed a typical Landau form for the nematic dynamical susceptibility:

$$\chi^{-1}_{\text{nem}}(\Omega) = \chi_0^{-1}(r - \Omega^2/c^2 + \cdots). \quad (3)$$

Here, $r > 0$ describes the distance to the nematic critical point, $\Omega$ is the frequency, and $c$ is a characteristic velocity. We do not need to consider spatial variations of the nematic mode, as the laser excitation has a wavelength much longer than any other scale in the system. The $E_u$ phonon corresponds to two degenerate in-plane stretching modes, and can be thus represented by the operators $(X_1, X_2)$ corresponding to the two in-plane orthogonal directions $x_1$ and $x_2$. For our purposes, it is sufficient to consider a dispersionless Einstein-like mode with frequency $\Omega_{\text{ph}}$:

$$H_{\text{ph}} = \sum_{x_i} \sum_{j=1,2} \left[ \frac{1}{2M} P_j^2(x_i,t) + \frac{M\Omega_{\text{ph}}^2}{2} X_j^2(x_i,t) \right]. \quad (4)$$

The $E_u$ mode cannot couple linearly to $\phi$, which transforms as $B_{1g}$. It does couple quadratically though, because $E_u \otimes E_u = A_{1g} \oplus A_{2g} \oplus B_{1g} \oplus B_{2g}$. Note that, although the $E_u$ mode couples to all quadrupole channels equally, near the transition only one channel is relevant. The coupling between nematic and phononic degrees of freedom is then

$$H_{\text{nem}-\text{ph}} = \frac{\lambda}{a^2} \sum_{x_i} \phi(x_i, t) (X_1^2(x_i, t) - X_2^2(x_i, t)). \quad (5)$$

Here, $a = \sqrt{\hbar/M\Omega_{\text{ph}}}$ is the quantized oscillator length and $\lambda$ is the coupling constant. We now excite the system with an external monochromatic electric field at a frequency $\sim \Omega_{\text{ph}}$, close to resonance with the $E_u$ modes. At very low temperatures, which is relevant for quantum quenches, the thermal occupation of the phonons is negligible. The main effect of the external field is to excite the phonon modes $X_j$. After the excitation, the phonon modes decay according to a decay rate $\Gamma_{\text{ph}}$. For narrow phonon lines, which is usually the case in the materials of interest, $\Gamma_{\text{ph}} \ll \Omega_{\text{ph}}$. Typically, the coupling between the lattice and the electronic background is weak, and we assume that the electronic heating time $\tau_{\text{el}}$ is much longer than the typical decay time of the phonons, i.e. $\tau_{\text{el}}^{-1} \ll \Gamma_{\text{ph}}$. We will discuss this assumption in more detail below.

To understand how such a resonant excitation of the $E_u$ phonon mode affects the nematic degrees of freedom, it is sufficient to treat the system classically and focus on a single site $x_0$. According to Eq. (3), the typical frequency of the nematic mode is $\Omega_{\text{nem}} = c\sqrt{r}$. Since $r \to 0$ at the nematic transition, the phonons are typically much faster than the nematic mode near the transition, $\Omega_{\text{ph}} \gg \Omega_{\text{nem}}$. We can therefore treat the system in a Born-Oppenheimer-type approximation, except now with fast phonons and slow electrons. The coupling term in Eq. (5) gives a quasi-static splitting of the resonance frequencies of the two planar stretch modes,

$$\Omega_{1,2}(t) = \Omega_{\text{ph}} \sqrt{1 \pm \frac{2\lambda}{\hbar\Omega_{\text{ph}}}} \phi(t), \quad (6)$$

yielding the effective phonon Hamiltonian,

$$\hat{H}_{\text{ph}}^{(2)} = -\frac{\lambda^2}{2\hbar\Omega_{\text{ph}}} n_{\text{ph}} \phi^2(t), \quad (7)$$

where $n_{\text{ph}} = n_1 + n_2$. This term corresponds to an effective attractive interaction in the nematic channel, which enhances the uniform static susceptibility of the nematic degrees of freedom as shown in Eq. (1). This is the main result of our work. Clearly, the magnitude of the enhancement depends on the occupation number $n_{\text{ph}}$, which in turn depends on the strength of the external field $\varepsilon_0$. The relationship between these quantities can be obtained via the solution of the classical damped harmonic oscillator, $(X_j^2) \approx \frac{q_j^2\varepsilon_0^2}{8M\Omega_{\text{ph}}^2\Gamma_{\text{ph}}}$, where $\varepsilon_j$ are the components of the electric field, combined with the usual classical-quantum correspondence:

$$n_{\text{ph}} \approx \frac{X_1^2 + X_2^2}{a^2} \approx \frac{1}{2} \left( \frac{\varepsilon_0 a}{2\hbar\Gamma_{\text{ph}}} \right)^2 \quad (9)$$

Although here we motivated the results using a semiclassical approach, in the Methods section we derive them in the Keldysh formalism, and provide a more general form appropriate for analyzing arbitrary pulse shapes.

These results support a feasible protocol for a quantum quench near a putative nematic QCP. A sketch of the proposed experimental protocol is depicted in Fig. 2, consisting of a pulse of length $\tau_{\text{pulse}}$ and frequency $\Omega_{\text{ph}}$ resonant with the $E_u$ phonon mode. The other relevant time scales are: the phonon decay time $\Gamma_{\text{ph}}^{-1} \gg \Omega_{\text{ph}}$, which sets the time scale needed to excite the phonons from rest to the amplitude $\langle X_j^2 \rangle$; the time scale of the nematic fluctuations, $\Omega_{\text{nem}}^{-1} \gg \Omega_{\text{ph}}^{-1}$; and $\tau_{\text{el}}$, the electronic heating time. To establish coherent dynamics, the latter must be the longest time scale of the problem, i.e. $\tau_{\text{el}} \gg \Omega_{\text{nem}}^{-1}, \Gamma_{\text{ph}}^{-1}$. We expect this to be the case because, in contrast to traditional pump-probe experiments, the protocol in Fig. 2...
FIG. 2. Schematic setup of the nematic quantum quench experiment. The system is subjected to an external laser pulse with frequency $\Omega_{ph}$, matching the resonance frequency of the $E_u$ mode. The laser induces a change in the phonon occupation number, $n_{ph}$, that shifts the “mass” of the nematic mode, as seen in Eq. (1), leading to a coherently controlled nematic phase transition. Here $n_{ph} = 2\chi_{u}\hbar\Omega_{ph}/\lambda^2$ is the critical phonon occupation obtained from Eq. (1). As described in the text, there are two scenarios: If nematic timescales are shorter than the phonon width, $\Omega_{nem}^{-1} \ll \Gamma_{ph}^{-1}$, the laser controls the nematic phase adiabatically. In the opposite regime $\Omega_{nem}^{-1} \gg \Gamma_{ph}^{-1}$, which always occurs near enough to the transition, the laser induces a quantum quench of the nematic phase. In all cases the electronic heating timescale is very long, $\tau_{el} \gg \Omega_{nem}^{-1}, \Gamma_{ph}^{-1}$.

Excites the lattice directly while electronic heating occurs indirectly: First, the resonant $E_u$ modes relax to the phononic bath on a timescale of $\Gamma_{ph}^{-1}$. Then, the phonon bath exchanges energy with the electronic bath, and eventually excites the slow nematic modes. This stage is expected to be slow, and to become even slower close to the nematic transition. We still need to compare the two time scales $\Omega_{nem}^{-1}$ and $\Gamma_{ph}^{-1}$. If $\Gamma_{ph}^{-1} \gg \Omega_{nem}^{-1}$, which is expected to hold far from a nematic transition, the softening of the nematic mode, Eq. (1), may be considered adiabatic. On the other hand, if $\Omega_{nem}^{-1} \gg \Gamma_{ph}^{-1}$, which is expected to happen near the nematic transition, the softening is essentially instantaneous, resulting in a quantum quench of the nematic instability. Therefore, the condition on the model parameters for a quantum quench is:

$$\Gamma_{ph}^{-1} \ll \Omega_{nem}^{-1} \ll \tau_{el},$$

To show that these conditions can be realized in actual systems, we consider the case of FeSe [28, 33]. This iron-based superconductor ($T_{SC} \approx 8$ K) displays an electronic nematic transition at $T_{nem} \approx 90$ K, that is suppressed to zero upon $S$ doping, suggesting a putative metallic nematic QCP [34, 35]. Because FeSe$_{1-x}$S$_x$ does not display long-range magnetic order at ambient pressure, it is an ideal system for studying the interplay of nematicity and superconductivity. An illustration of the lattice structure and $E_u$ modes of FeSe is shown in Fig. 3. We estimate from existing measurements on FeSe and related compounds (see Methods) that $h\Omega_{ph} \approx 30$ meV, $h\Gamma_{ph} \approx 0.5 - 1.5$ meV, and $h\Omega_{nem} \approx 10$ meV away from the transition, softening further as one approaches it. In addition, we estimate $\tau_{el}^{-1} < 0.25$ meV away from the transition, which can get as small as 0.04 meV approaching the transition. These estimates show that the quantum quench regime is achievable in FeSe.

We use these parameters to also estimate the coupling $\lambda$ and, thus, the expected maximum shift in the nematic susceptibility. As shown in Eq. (6), static nematic order $\langle \phi \rangle \neq 0$ splits the frequency of the $E_u$ mode by $h\Delta\Omega \approx 2\lambda\langle \phi \rangle$. The dimensionless nematic order parameter $\langle \phi \rangle$ is estimated by the elliptical distortion of the Fermi surface measured by ARPES, $\langle \phi \rangle \approx \Delta k_F/k_F$. This leads to $\lambda/h\Omega_{ph} \approx 0.04$. The maximum shift of the nematic transition temperature can then be obtained by a Lindemann criterion argument: the maximum possible occupation $n_{ph}$ will have $\langle \chi^2 \rangle \approx n_{ph} \approx c_L^2 \ell^2$, where $\ell = 3.7$ Å is the FeSe lattice constant, and $c_L$ is a fraction usually of the order of $c_L = 0.1 - 0.2$ [36]. Using these values we find a shift of $6.5 - 26$ K in the nematic transition temperature, indicating the experimental feasibility of a quantum quench.

In conclusion, we showed that the nonequilibrium excitation of the infrared-active $E_u$ phonon mode present in tetragonal systems mediates an attractive electronic interaction in the nematic channel. Besides establishing a robust protocol for light control of nematicity, this result unveils a promising and experimentally feasible avenue to induce a quantum quench across the transition of correlated materials that display nematic order. Because nematic fluctuations are intimately connected to other electronic instabilities, most notably superconductivity and magnetism, the nonequilibrium excitation of nematic fluctuations may also be used to drive transient states with different types of electronic orders.

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METHODS

Derivation of the results using the Keldysh formalism

We derive our main result, Eq. (8), by writing down the action for an \( E_n \) phonon mode \( X_j \) coupled linearly to an external electromagnetic field and quadratically to a nematic field \( \phi \). Then we integrate out the phonons to obtain an effective action for \( \phi \). Finally, we specialize to the limit where the electric field varies much faster than \( \phi \) to obtain a simple expression for the induced nematic attraction.

We write the action as

\[
S = S_{\text{ph}} + S_{\text{nem-ph}} + S_{\text{nem}}, \tag{11}
\]

where \( S_{\text{ph}} \) describes the phonon system coupled to the electromagnetic field:

\[
hS_{\text{ph}} = \int_{\mathcal{C}} dt \sum_{x_i} \left[ \frac{M}{2} |\dot{X}_j|^2(x) + q X_j(x) A_j(x) - \frac{M \Omega_{\text{ph}}^2}{2} |X_j|^2 \right]
\]

\[
= \int_{\mathcal{C}} dt \sum_{x_i} \left[ \frac{1}{2} X_j(x) D_0^{-1} X_j(x) + q X_j(x) E_j(x) \right], \tag{12}
\]

where \( \mathcal{C} \) denotes the Keldysh contour, \( x = (t, x_i) \) is shorthand for the position in spacetime on the lattice, and \( q \) is the electric charge. In Eq. (12) we employ the summation convention and choose the dynamic gauge \( E_j = -\partial_t A_j \).

The phonon propagator is:

\[
D_0^{-1} = M \left( -\partial_t^2 - \Omega_{\text{ph}}^2 \right). \tag{13}
\]

The coupling to the nematic field is given by

\[
hS_{\text{nem-ph}} = \lambda \int_{\mathcal{C}} dt \sum_{x_i} \left( X_i(x) \sigma_3^{ij} X_j(x) \right) \phi(x), \tag{14}
\]

where \( \sigma_n \) is the \( n \)-th Pauli matrix operating in the space of lattice directions. \( \phi(x) \) denotes the nematic field.

To integrate out the phonons we perform the Keldysh rotation for the bosonic fields \( \phi \) and \( X_j \) as well as for the external field \( E_j \),

\[
O^{\pm q}(t) = \frac{1}{2} \left( O^+(t) \pm O^-(t) \right), \tag{15}
\]

where \( O \) is some operator and \( \pm \) denote respectively the upper/lower branches of the contour. We then integrate out the \( X_j \)'s to obtain the effective action,

\[
S_n = S_0 + S_{\text{eff}}. \tag{16}
\]

Here,

\[
hS_{\text{eff}} = \text{Tr} \log D^{-1} \tag{17}
\]

describes the equilibrium contribution to the nematic fields that results from the coupling to \( X_j \). This term is already contained in \( S_{\text{nem}} \). However, the coupling to external fields also induces an effective action for \( \phi \), namely,

\[
hS_{\text{eff}} = -q^2 \int_{\mathcal{C}} dt \sum_{x_i} E_i^c(x) \pi_x^{\alpha\beta} D^{\alpha\beta}_{ij}(x) \tau_x^{\gamma\delta} E_j^\delta(x). \tag{18}
\]

Here \( \tau_n \) denotes Pauli matrices in Keldysh space. The full phonon propagator in Eqs. (17) and (18) is,

\[
D = (\tau_0 \sigma_0 - 2D_0 \lambda \sigma_3 \Phi)^{-1} \cdot D_0, \tag{19}
\]

where \( \Phi^{\delta\gamma} = \phi^\alpha \tau_0^{\delta\gamma} + \phi^{c\alpha} \tau_0^{\delta\gamma} \), and we suppressed indices for clarity. Here, the superscript \( cl \) (\( q \)) denotes the classical (quantum) Keldysh components. The propagator \( D_0 \) is the generalization of Eq. (13) to Keldysh space, with the addition of a damping term \( \Gamma_{\text{ph}} \), i.e.

\[
D_0^R(\omega) = M^{-1}(\omega^2 - \Omega_{\text{ph}}^2 + 2i\Gamma_{\text{ph}} \omega)^{-1}. \tag{20}
\]

Eqs. (18) and (19) are exact within our model, and work for any laser beam profile. However, to make things simpler, we can assume that the electromagnetic field is completely classical, \( E_j^c = 0 \), and furthermore that the field is essentially monochromatic with a frequency \( \Omega_{\text{ph}} \) and a width \( \ll \Gamma_{\text{ph}} \). In that case, expanding to second order in \( \lambda \) one finds, after some algebra,

\[
S_{\text{eff}}^{(2)} = -q^2 E_0^c |D_0(\Omega_{\text{ph}})|^2 \int d\omega \Re \left( D_0^R(\Omega_{\text{ph}} + \omega) + D_0^R(-\Omega_{\text{ph}} + \omega) \right) (\phi^c(\omega) + \phi^c(\omega)) \tag{21}
\]

In Eq. (21) we suppressed the summation over the lattice. We also ignored the first order contribution, since it vanishes for a pulse that does not explicitly break tetrag-
nematic degrees of freedom do vary slowly compared to the phonons, i.e. when \( \phi \) is peaked at frequencies \( \Omega_{\text{nem}} \ll \Omega_{\text{ph}} \), then the effective attraction in the nematic channel comes from the low-frequency tails of \( D_0 \), i.e. from the long-time averaged motion of the phonons. Indeed, we can show this by coarse-graining \( D_0 \), i.e. by averaging over some bandwidth \( \Gamma_{\text{ph}} \ll \Lambda \ll \Omega_{\text{ph}}, \)

\[
\mathbb{R} \int_{-\Lambda}^{\Lambda} \frac{d\omega}{2\Lambda} \left( D_0^R(\Omega_{\text{ph}} + \omega) + D_0^R(-\Omega_{\text{ph}} + \omega) \right) \approx \frac{-1}{2M\Omega_{\text{ph}}^2}. \tag{22}
\]

Thus, going back to Eq. (21), and transforming back to the Keldysh contour, we find,

\[
hS^{(2)}_{\text{eff}} \approx \frac{\chi^2}{4\hbar\Omega_{\text{ph}}} \left( \frac{qE_0a}{2\Gamma\Omega_{\text{ph}}M} \right)^2 \int dt \sum_{x_i} \phi^2(x). \tag{23}
\]

Eq. (23) is equivalent to Eq. (8) of the main text.

**Estimates of the experimental parameters for FeSe**

In the main text, we invoked the iron-based superconductor \( \text{FeSe}_{1-x} \text{S}_x \) to demonstrate the feasibility of our quantum quench protocol. In this section, we briefly outline the sources and methods used to extract experimental parameters for this compound. FeSe has been widely studied, due at least partly to the fact that there are clean single crystals available, and that it does not suffer from the nematic transition, giving us the opportunity to experiment up to a prefactor (which can also be temperature dependent). In a Stoner-type theory for the nematic transition, \( \chi_0 \sim N_F \), where \( N_F \) is the density of states at the Fermi level. In FeSe, the Fermi surface evolves very strongly as a function of temperature [33, 38], indicating the importance of correlation effects. We therefore estimate in these systems that \( \chi_0^{-1}r = E_F(T - T_{\text{nem}})/T_{\text{nem}} \).

As mentioned in the main part of the paper, we took \( \Delta k_F/k_F \) as a proxy for \( \langle \phi \rangle \), which is again only correct up to an unknown prefactor. We stress that the uncertainty in our knowledge of \( \chi_0^{-1}r \) and \( \langle \phi \rangle \) means that our estimates for the effective coupling are only valid to within an order of magnitude.

For clarity, we have compiled our estimates of the various parameters for FeSe into Table I. We now briefly outline what sources we used to extract the experimental parameters in Table I and our estimate for the coupling, \( \lambda = \frac{\Delta \Omega}{2\langle \phi \rangle} \).

| \( \Omega_{\text{ph}} \) | \( \Gamma_{\text{ph}} \) | \( \Omega_{\text{nem}} \) | \( \tau_{\text{g}}^{-1} \) | \( E_F \) | \( k_F \) | \( \Delta k_F \) |
|---|---|---|---|---|---|---|
| 30-33 | 0.4 - 1.5 | 10 | 0.24 | 25 | 0.13 | 0.02 |

Table I. Estimated experimental parameters for the Fe-based superconductor FeSe. Energies are quoted in meV, and wavevectors in \( \text{Å}^{-1} \).

which, as noted in the paper, is

\[
\lambda = \frac{\Delta \Omega}{2\langle \phi \rangle}, \tag{24}
\]

where \( \Delta \Omega \) is the splitting of the \( E_u \) mode in the nematic phase.

The infrared phonon structure and dispersion relations in FeSe have been both calculated [39–41] and detected experimentally [37, 42–44]. We discuss data only for the tetragonal phase, above \( T_{\text{nem}} \approx 90 \text{K} \). Ref. 42 reported \( \Omega_{\text{ph}} = 30.9 \text{ meV} \) in a film of FeSe on CaF\(_2\), measured by optical reflectometry. Ref. 37 reported \( \Omega_{\text{ph}} = 32 \text{ meV} \) near the \( M \) point, measured by electron energy-loss spectroscopy on a single crystal. Ref. 44 reported \( \Omega_{\text{ph}} = 31.3 \text{ meV} \) in neutron scattering. This tallies with theoretical calculations [39, 41] predicting \( \Omega_{\text{ph}} = 30–35 \text{ meV} \), and only a weak dispersion for the \( E_u \) mode. We did not find a reported measurement of \( \Gamma_{\text{ph}} \) for FeSe. However, Ref. 45 reported \( \Gamma_{\text{ph}} \approx 1.2–1.5 \text{ meV} \) in the related chalcogenides \( \text{FeTe} \) and \( \text{FeTe}_{1-x}\text{Se}_x \). For the FeSe \( B_{1g} \) optical mode, Ref. 46 reported a decay rate \( \Gamma_{B_{1g}} \approx 0.4 \text{ meV} \) at the nematic transition.

The electronic structure and dynamics of FeSe have been extensively studied by (among others) ARPES and Raman techniques. To estimate the relevant timescale \( \Omega_{\text{nem}}^{-1} \), we considered reports of polarization-resolved Raman data measuring the dynamic response of the nematic mode in \( \text{FeSe}_{1-x}\text{S}_x \) [47, 48]. These measurements show a wide damped peak centered around 25 meV, and extending to about 50 meV before beginning to decay. Although there are interesting features in the entire region (for details see e.g. Ref. 48), the sharpest features show up at frequencies below about \( \Omega_{\text{nem}} \sim 10–12 \text{ meV} \), and soften as one approaches the nematic transition, giving us the estimate for \( \Omega_{\text{nem}} \) in Table I. To estimate the coupling constant \( \lambda \), we used Eq. (24). As a proxy for \( \langle \phi \rangle \) we took the elliptical distortion of the hole-like Fermi surface \( \Delta k_F = k_{F,x} - k_{F,y} \) at the \( Z \) point. We extracted the values for \( E_F = 25 \text{ meV} \), \( k_F = 0.13 \text{ Å}^{-1} \) and \( \Delta k_F = 0.02 \text{ Å}^{-1} \) from Ref. 33 (all at the \( Z \) point). We have not found a detailed study of the \( E_u \) mode splitting in FeSe. However, the \( E_g \) mode, which is Raman active but has almost the same resonant frequency as \( E_u \), has been measured. Ref. 49 reports a maximum split of \( \Delta \Omega = 0.4 \text{ meV} \) at 20 K.

Using these numbers, the shift of the nematic susceptibility, \( \chi_{\text{nem}}^{-1} \rightarrow \chi_{\text{nem}}^{-1} - n_{\text{ph}}\chi_0^{-1} \delta r \) (see Eq. 1 of the main
where
\[ \delta r \sim \frac{\lambda^2}{2\hbar \Omega_{ph} E_F} \approx 0.001 \] (25)

Using \( \delta r = \delta T_{nem}/T_{nem} \), where \( T_{nem} \) is the shift in the nematic transition temperature \( T_{nem} \approx 90 \) K, we find \( \delta T_{nem} = 0.1 \) K per phonon. To estimate the maximum possible phonon occupation number, it is simplest to consider what occupation number would melt the lattice. This can be found from the Lindemann criterion,
\[ n_{ph} \Delta^2 = c_L \ell^2, \] (26)

where \( \ell \) is the lattice constant, \( a = \sqrt{\hbar / M \Omega_0} \) is the quantized oscillator length, and \( c_L \) is some fraction (we picked the range 0.1–0.2, since 0.1 is the commonly used Lindemann value [36]). Since the \( E_u \) mode involves motion of both the Fe and Se atoms, we use the reduced mass, \( M = \sqrt{2M_{Fe}M_{Se}} / (M_{Fe} + M_{Se}) \approx 66u \). Then we find \( a = 0.05\,\text{Å} \), which in turn implies \( n_{ph} = 65–240 \) and \( \delta T_{nem} = 6.5–26 \) K.

To estimate the equilibrium time \( \tau_d \), we summed up the phonon decay time \( \Gamma_{ph}^{-1} \) with measured electronic decay times \( \Gamma_{el-ph}^{-1} \) from ultrafast optical reflectivity experiments. Typically, such measurements heat up the electronic subsystem, which then decays slowly into the lattice [50, 51]. This decay is characterized by two distinct timescales: a fast decay of the electrons into symmetry-preferred optical phonon modes (e.g. \( A_{1g} \)) and then a slow anharmonic decay of these modes to the lattice. We took as our estimate for the decay time \( \Gamma_{el-ph}^{-1} \), the decay constant of this slow anharmonic decay. We also considered temperatures not too close to the critical temperature \( T_{nem} \), out of the assumption that such a timescale roughly characterizes a generic electron-phonon decay. We obtained \( \Gamma_{el-ph} = 0.5–0.6 \) meV from Ref. [50]. We took as our estimate for \( \Gamma_{ph} \) the lower value quoted above in Table I (which is one that was measured for actual FeSe), implying a total decay \( \tau_d = \Gamma_{ph}^{-1} + \Gamma_{el-ph}^{-1} = 18.8–17.2 \) ps. However, this estimate does not take into account the expected slowing down of electronic heating rates near the nematic transition. Such slowing down has been measured in BaFe\(_2\)As\(_2 \) [52].
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