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Nematic magnetoelastic effect contrasted between $Ba(Fe_{1-x}Co_x)_2As_2$ and FeSe

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To elucidate the origin of nematic order in Fe-based superconductors, we report a Raman scattering study of lattice dynamics, which quantify the extent of C_4-symmetry breaking, in $BaFe_2As_2$ and FeSe. FeSe possesses a nematic ordering temperature T_s and orbital-related band-energy split below T_s that are similar to those in $BaFe_2As_2$, but unlike $BaFe_2As_2$ it has no long-range magnetic order. We find that the E_g phonon-energy split in FeSe becomes substantial only well below T_s, and its saturated value is much smaller than that in $BaFe_2As_2$. Together with reported results for the $Ba(Fe_{1-x}Co_x)_2As_2$ family, the data suggest that magnetism exerts a major influence on the lattice.

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In copper- and iron-based high-temperature superconductors, as well as in heavy-fermion and organic superconductors, the superconducting phase is commonly found in close proximity to an antiferromagnetic phase. Not only does this important commonality suggest that the mechanism for unconventional superconductivity builds upon electronic correlations that give rise to the magnetism [1–8], but it also implies that intriguing intertwined phases, which have been a subject of intense study [9,10], may arise from the same electronic correlations [11]. In the Fe-based superconductors [12], the most prominent intertwined phase is the so-called nematic phase [13–15], in which the discrete C_4 rotational symmetry is broken but the lattice translational symmetry is not. Because electronic properties exhibit pronounced C_2 (rather than C_4) symmetry in the nematic phase while the crystal structure is only weakly orthorhombic [16–18], there has been a general consensus that the nematic phase is electronically driven [19]. The possible existence of a nematic quantum critical point has been intensively explored in this context [20,21], as it might explain some of the most unusual properties of these materials including the superconductivity itself.

Consistent with the notion that all essential intertwined phases in unconventional superconductors arise from a common magnetic origin [11], the tendency towards formation of stripe antiferromagnetic order in the Fe-based superconductors is considered a likely driving force for the nematic order. Such theoretical ideas have been explored in contexts both with [22–24] and without [25–28] stripe antiferromagnetic order as the system's low-temperature ground state. The latter theories are motivated by the case of bulk FeSe [29], which exhibits a nematic transition at $T_s \approx 90$ K but no long-range magnetic order down to the lowest temperature.

However, photoemission studies [30–34] have revealed below T_s a dramatic electronic reconstruction, which leads to an uneven occupation of the Fe d_{xz} and d_{yz} orbitals. When the magnetic ordering temperature T_{mag} is well below T_s, the reconstruction has been reported to be seen already above T_{mag} [31,32], although the effect of detwinning uniaxial pressure on T_{mag} [35,36] remains yet be considered. The electronic reconstruction in the pnictides improves the quality of Fermi-surface nesting [32], which can in turn help stabilize the stripe antiferromagnetic order. Together with the absence of long-range magnetic order and anomaly in the low-energy spin fluctuations near T_s [37,38], yet similarly pronounced electronic reconstruction in FeSe [33,34,39–41] as in other systems, these results support the alternative scenario that the nematic order is driven by orbital interactions [42–46] or by a related Pomeranchuk instability [47]. To what extent some of the most recent results can be thought of as refuting spin-driven and/or ferro-orbital nematic order is currently under heated debate [48–51].

To experimentally determine whether the nematic order is spin or orbital driven, in principle one would need to measure the susceptibility of spin-correlation anisotropy to orbital polarization, or vice versa, much in the fashion of what has been achieved between the electronic and lattice degrees of freedom [52], but this is obviously difficult. Here we take an alternative approach by using lattice dynamics to detect the so-called strength of nematicity in $BaFe_2As_2$ and FeSe. Since the lattice is linearly coupled to the electronic nematicity [53], and because the lattice (as we will show) and orbital-related [30,33] characteristic energies are respectively similar between the two systems, our measurement can determine how spin structures substantiate the nematic order. We find that the lattice-dynamics signature of C_4-symmetry breaking in FeSe only becomes substantial below $T^* \sim 60$ K rather than immediately below T_s, and that its saturated value is much smaller than that in $BaFe_2As_2$. Our results suggest that spin supersedes orbital in causing nematic lattice deformations.

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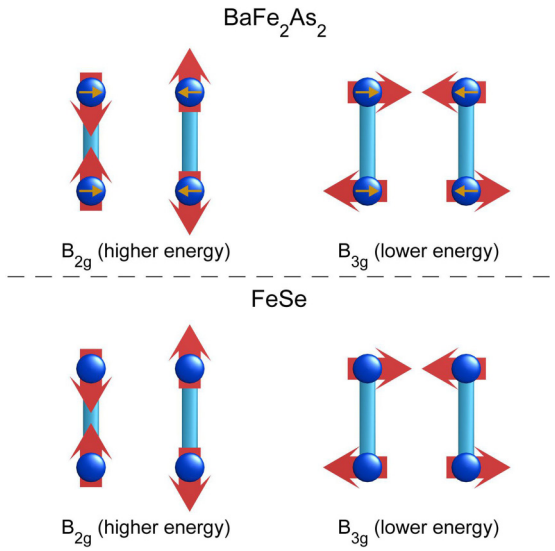

FIG. 1. Thick arrows indicate displacement of Fe atoms in the B_{2g} and B_{3g} phonon modes in $BaFe_2As_2$ and FeSe. As and Se atoms are omitted for clarity. The vertical Fe-Fe bonds are highlighted as being more rigid than the horizontal ones.

$BaFe_2As_2$ is the parent compound of the so-called 122 family of Fe-based superconductors, exhibiting an orthorhombic stripe antiferromagnetic phase below $T_{mag} \approx T_s = 138$ K [54]. FeSe is structurally the simplest Fe-based superconductor with an orthorhombic structural transition at $T_s \approx 90$ K but no long-range magnetic order [29]. At high temperatures, $BaFe_2As_2$ and FeSe belong to the $I4/mmm$ and $P4/nmm$ space groups, respectively, and the Fe and As-Se atoms contribute two two-fold degenerate E_g phonon modes at the Brillouin zone center. When the C_4 rotational symmetry is lowered into C_2 in the nematic phase, each of the E_g modes splits into B_{2g} and B_{3g} modes that are of slightly different energies. Since all these phonons are Raman active, we can utilize the high-energy resolution and sensitivity of Raman scattering to detect the energy split, which provides information about the *ab* anisotropy of the lattice spring constants arising from the spin and/or orbital interactions.

We performed our variable-temperature Raman scattering experiment in a confocal backscattering geometry, using a Horiba Jobin Yvon LabRAM HR Evolution spectrometer equipped with 1800 g/mm gratings and a liquid-nitrogen-cooled CCD detector. Long-wavelength $\lambda = 785$ nm and 633 nm lasers were used as excitations to achieve high energy resolution ($\approx$0.7 cm^{-1}). We kept our laser power low ($\sim$1 mW) to reduce heating [55], which led to very long exposure time (>4 h per spectrum) in order to obtain satisfactory statistics in the photon counts. Samples were kept in a cryostat under better than 5×10^{-8} Torr vacuum to ensure surface stability over the entire measurements. High-quality single crystals of $BaFe_2As_2$ and FeSe were grown by self-flux and chemical vapor transport methods, respectively. The Raman measurements were performed on surfaces that are perpendicular to the easy-cleavage *ab* plane, which allowed us to use perpendicular linear polarizations of incoming and scattered photons to detect the E_g, B_{2g}, and B_{3g} phonons. Such

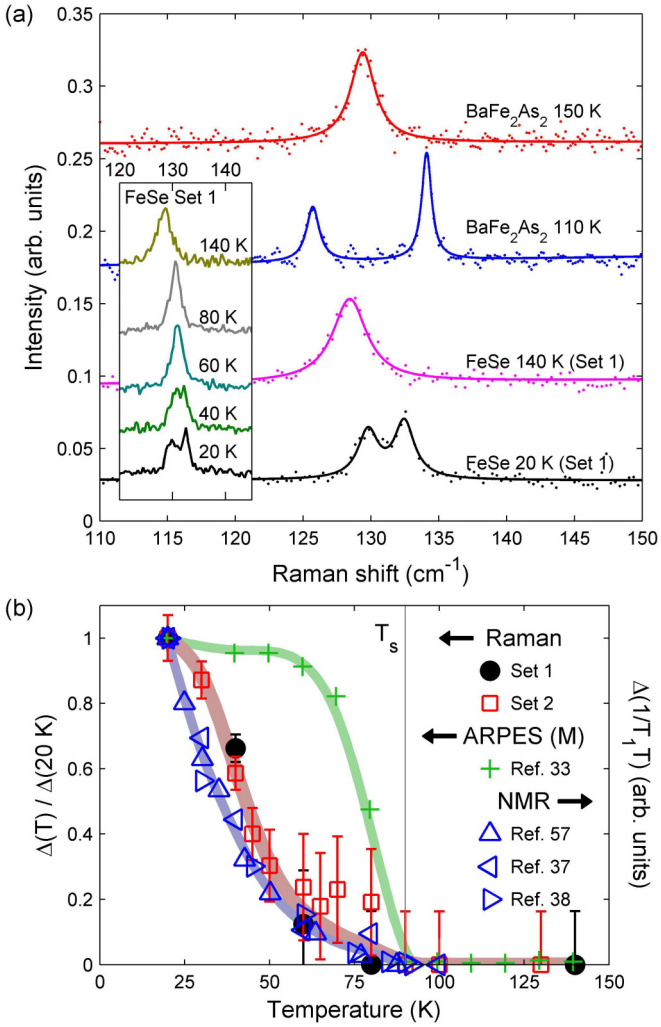

FIG. 2. (a) Raman spectra measured on $BaFe_2As_2$ and FeSe single crystals. The inset shows temperature dependence of the spectrum of FeSe near 130 cm^{-1}, vertically offset for clarity. (b) Temperature dependence of phonon-energy split in FeSe, plotted together with band-energy split observed with photoemission at the Brillouin-zone M point and spin-lattice relaxation rate increase below 90 K observed with NMR. Data are normalized at 20 K for comparison.

sample surfaces were prepared by cleaving the crystals after freezing in liquid nitrogen.

Figure 1 illustrates the vibrational patterns of Fe atoms in B_{2g} and B_{3g} modes that derive from the same E_g mode in the high-temperature phase. Because of the uneven d_{xz} and d_{yz} orbital occupation, bonds along one of the Fe-Fe directions are expected to be stronger, and atomic vibrations along that direction are expected to occur at slightly higher frequency (or energy). The difference between $BaFe_2As_2$ and FeSe is that the former also exhibits a stripe antiferromagnetic order, which is expected to further influence the lattice dynamics via magnetoelastic coupling [56]. The question is how large such effects are compared to the influence of the orbital and/or Fermi-surface anisotropy in the nematic phase. Importantly, photoemission experiments have found comparable magnitudes of orbital-related band-energy split

in the two systems [30,33,34], so any substantial difference we identify has to arise from the difference in the magnetism.

We present our key result in Fig. 2. At high temperatures, $T = 150$ K $> T_s$ in $BaFe_2As_2$ and $T = 140$ K $> T_s$ in FeSe, the E_g phonon peaks of both systems are observed at very similar energies. This shows that the two systems possess similar lattice dynamics in the tetragonal phase, which is not an unexpected result given the similar atomic masses of As and Se and the structural similarity between the FeAs and FeSe layers. As we have recently reported [57], at $T = 110$ K $< T_s$ in $BaFe_2As_2$, the E_g peak splits into B_{2g} and B_{3g} peaks that differ in energy by 9.4 cm^{-1}, consistent with a previous report [56]. In contrast, although a splitting of the E_g peak is also observed at $T = 20$ K $\ll T_s$ in FeSe, the B_{2g} and B_{3g} peaks only differ in energy by 2.6 cm^{-1}. We attribute the much smaller energy split in FeSe to the lack of magnetic order as discussed above.

A further unexpected observation is that, unlike in $BaFe_2As_2$, where the phonon-energy split rapidly increases below T_s and reaches its saturated value about 30 K below T_s [56], the split in FeSe remains small (albeit nonzero) over a considerable temperature range below T_s, and only becomes greater than 20% of its value at the lowest temperature below $T^* \approx 60$ K, as shown in the inset of Figs. 2(a) and 2(b). While the precise value of T^* depends on its definition and exhibits a slight variation among different sample spots [55], the overall behavior is consistent with the temperature dependence of the spin-lattice relaxation rate [37,38,58], in that a rapid increase is found only below a temperature somewhat lower than T_s. A comparison among the normalized T dependence of the phonon split, the increase of spin-lattice relaxation rate below T_s, and the orbital-related band splitting (at the Brillouin-zone M point), clearly shows that the phonon split is related more directly to magnetic than to orbital degree of freedom. Thus the split in FeSe, albeit small and in the absence of static magnetic order, might nevertheless be caused by low-energy spin fluctuations which are presumably nematic in nature.

In the $Ba(Fe_{1-x}Co_x)_2As_2$ family, the phonon-energy split is found to decrease with increasing Co doping [56], which simultaneously suppresses the tetragonal-to-orthorhombic structural transition temperature T_s, the stripe antiferromagnetic ordering temperature T_{mag} [59], the orbital-related band-energy split Δ_{orb} [30], and the transport anisotropy [16]. It is therefore difficult to decipher the relationship among these quantities by studying this material family alone. To this end, we have attempted to empirically relate the phonon-energy split to the magnetic and orbital characteristic energies, accommodating both $Ba(Fe_{1-x}Co_x)_2As_2$ and FeSe.

Our results are presented in Fig. 3. First of all, we find that the phonon-energy split is not simply related to the structural transition temperature [Fig. 3(a)]. Despite its likely connection to the magnetism as discussed in the preceding paragraphs, the split is not simply linearly related to T_{mag} either, as in that case the split in FeSe would be expected to be nearly zero [Fig. 3(b)]. The split in FeSe appears to be bounded from below by another mechanism, which we assume here to be orbital interactions. By considering the reported values of phonon-energy split Δ_{Raman} [56] and Δ_{orb} at the Brillouin-zone M point [30] as functions of Co concentration x, which has a one-to-one correspondence to T_{mag} in $Ba(Fe_{1-x}Co_x)_2As_2$ [16,59], we find

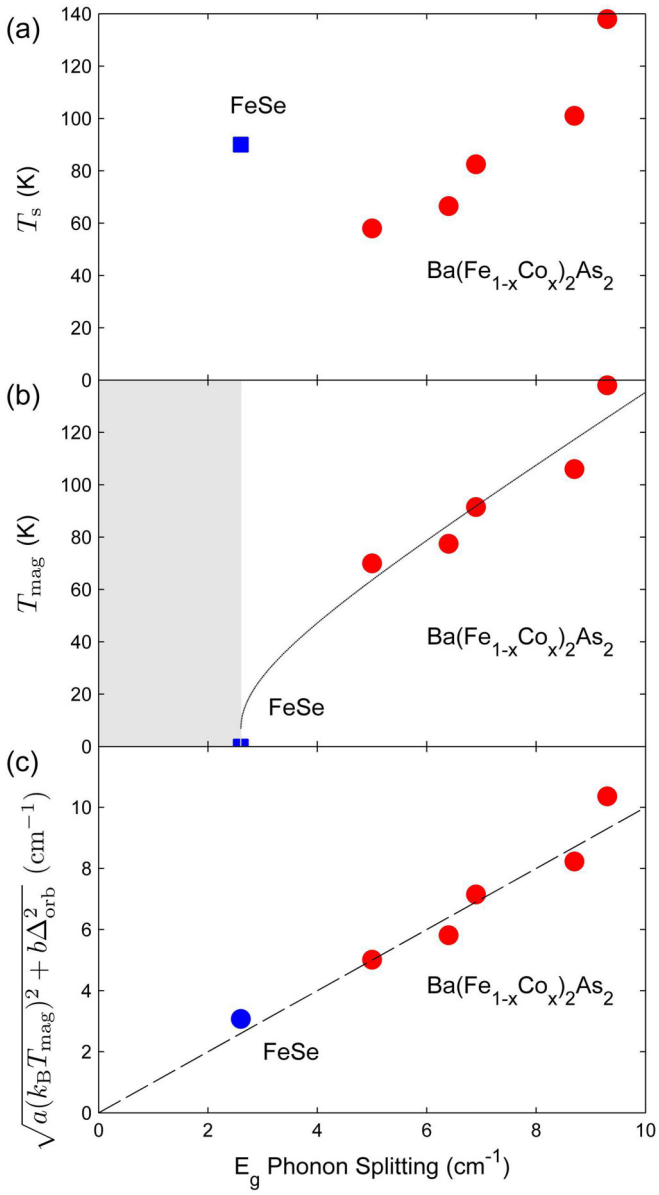

FIG. 3. Structural phase transition temperature (a), magnetic ordering temperature (b), and an empirical combination (see text) of magnetic and orbital energies (c) plotted vs E_g phonon-energy split for FeSe and $Ba(Fe_{1-x}Co_x)_2As_2$ [56].

that the empirical formula $\Delta_{Raman} = \sqrt{a(k_B T_{mag})^2 + b\Delta_{orb}^2}$, where a and b are dimensionless parameters, describes all the data very well [Fig. 3(c)]. The underlying assumption for this formula is that the spin-related energy $k_B T_{mag}$ and the orbital-related energy Δ_{orb} influence the lattice dynamics in an uncorrelated fashion. We find that $a = 1.0 \times 10^{-2}$, which is much greater than $b = 5.8 \times 10^{-5}$, i.e., the spin correlations exert a much stronger influence on the lattice dynamics than the orbital structure, as expected from the fact $\Delta_{orb} = 62$ and 50 meV in $BaFe_2As_2$ and FeSe [30,33], respectively, yet their phonon-energy splits differ by over a factor of three. Static orthorhombic lattice distortions are always weak in Fe-based superconductors, suggesting that they are only secondary once the nematic order is well developed, and hence their

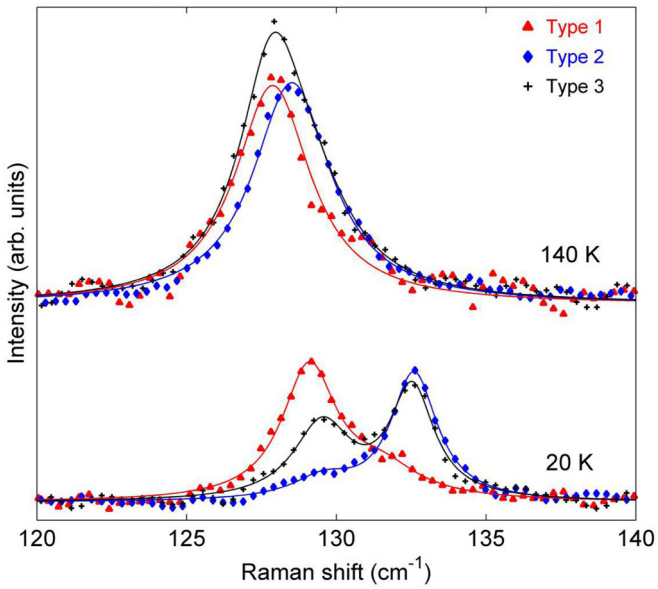

FIG. 4. Raman spectra measured on three representative surface spots of FeSe with different local stress.

comparable weakness in $BaFe_2As_2$ and FeSe [29,60] does not contradict our findings in the dynamic regime.

In the above analysis, we have used Δ_{orb} determined from photoemission experiments, some of which were performed on samples detwinned by uniaxial stress. Since we did not use a detwinned sample here, and because magnetic and transport properties are sensitive to uniaxial pressure especially near T_s, it is important to check the possible influence of local stress on our result. Indeed, we have identified three types of surface spots on our FeSe sample, as shown in Fig. 4. They correspond to local-stress environments that lead to different twin-domain distributions at 20 K under the laser spot. Importantly, the phonon energies change very little among the spots both well above and below T_s, in agreement with our recent finding for $BaFe_2As_2$ [57]. Together with consistent Δ_{orb} and its T dependence reported for twinned and detwinned FeSe [33,34], we believe that both the small value of Δ_{Raman} and the departure of T^* from T_s in FeSe are robust against local stress [55].

A conservative interpretation of our result is that the Fe-based superconductors exhibit strong nematic magnetoelastic coupling, consistent with recent transport and neutron Larmor diffraction measurements of the 122 family [61]. The fact that spin interactions appear dominant over orbital interactions in causing the C_2 lattice dynamics is consistent with recent inelastic neutron scattering experiments, in which the energy scale of spin anisotropy is found to be greater than that of the orbital ordering in optimally doped $BaFe_{2-x}Ni_xAs_2$ [62].

The pronounced magnetoelastic coupling does not prove by itself that the nematic order is driven by magnetism: Our data are consistent with the scenario that orbital-driven nematicity lifts the *ab* degeneracy for the spins and helps stabilize the stripe antiferromagnetic order in the pnictides, which in turn exerts a strong feedback on the lattice dynamics that is absent in FeSe (at least above T^*). However, it is not unlikely that both spin- and orbital-driven nematicity can only be stabilized in the presence of a deformable lattice [63], similar to the formation of charge density waves in metals [64]. Under such circumstances, the weakness of orbital's influence on the lattice, especially in the dynamic regime as demonstrated by the small phonon-energy split in FeSe and the lack of any substantial effect between T^* and T_s despite the nearly saturated value of Δ_{orb} at T^* [33,34], suggests that orbital interactions alone might not be able to cause the nematic order. In light of recent theoretical proposals for spin-driven nematicity in FeSe without long-range magnetic order [25–28], it will be interesting to compare anisotropic spin correlations, either derived from such theories [65] or in principle measurable by neutron scattering [66,67], to our measured phonon-energy splits, both in the zero-temperature limit and as functions of temperature.

To conclude, we have determined the E_g to $B_{2g} + B_{3g}$ phonon-energy split in FeSe and compared it to those in the $Ba(Fe_{1-x}Co_x)_2As_2$ system. A drastic difference is found both in the much-reduced energy split and in the lower characteristic temperature of the split in FeSe. Our result demonstrates that spin correlations have a much stronger influence on the lattice than orbital interactions. If the nematic order requires participation of lattice deformation to be fully stabilized, it is unlikely to be driven solely by orbital interactions.

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