Local nematic susceptibility in stressed $BaFe_2As_2$ from NMR electric field gradient measurements

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The electric field gradient (EFG) tensor at the ^{75}As site couples to the orbital occupations of the As p-orbitals and is a sensitive probe of local nematicity in $BaFe_2As_2$. We use nuclear magnetic resonance to measure the nuclear quadrupolar splittings and find that the EFG asymmetry responds linearly to the presence of a strain field in the paramagnetic phase. We extract the nematic susceptibility as a function of temperature and find that it diverges near the structural transition in agreement with other measures of the global susceptibility.

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The iron-based superconductors exhibit a complex interplay between orbital, electronic and lattice degrees of freedom. In $BaFe_2As_2$ a ferro-orbital instability is accompanied by an orthorhombic distortion and long-range antiferromagnetic order [1, 2]. This nematic phase breaks the C_4 tetragonal symmetry of the lattice, and is preceded by critical nematic fluctuations and divergent nematic susceptibility in the disordered phase [3, 4]. In the nematic phase, the Fe d_{xz} and d_{yz} orbitals become non-degenerate, with an energy splitting on the order of 40 meV, and different occupation levels [5]. This phase also stabilizes antiferromagnetic ordering of the Fe spins, which order either concomitantly with the nematic phase transition, or at a temperature T_N only a few Kelvins below. As a result, many low energy experimental probes actually sense a complex interplay of the orbital, lattice, and magnetic degrees of freedom simultaneously, precluding quantitative analyses.

Several techniques have been developed to probe the nematic degrees of freedom. Anisotropic resistivity [6, 7], elastoresistance [3], electronic Raman scattering [8], elastic constants [9–11], thermopower, polarized light image color analysis [12, 13] and optical conductivity [14] probe global, macroscopic anisotropies. NMR and neutron scattering, on the other hand, are microscopic probes, and have been used to investigate the effect of nematicity on the spin fluctuations [15–19]. The nuclear quadrupolar interaction, however, can probe the microscopic orbital occupations directly [20]. The ^{75}As ($I = 3/2$) quadrupolar moment couples to the local electric field gradient (EFG), which is dominated by the on-site occupations of the As 4p electrons. These orbitals are hybridized with the Fe 3d orbitals, and thus the EFG is a sensitive probe of the d-orbital occupations. Indeed, the EFG tensor exhibits a dramatic lowering from axial symmetry at the nematic phase transition in the absence of applied strain [21]. In this Rapid Communication we present new data on the EFG under uniaxial strain. We find that the EFG asymmetry parameter is linearly proportional to the in-plane strain applied to the crystal, and is a direct measure of the nematic susceptibility. This approach enables one to probe the *local*, rather than global, nematic susceptibility.

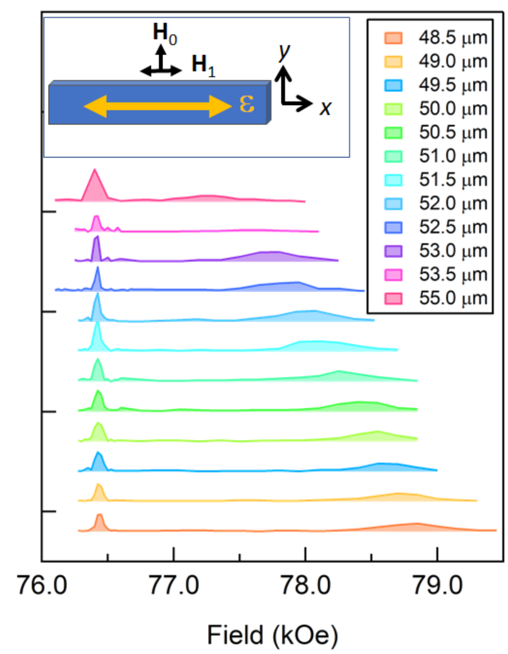

FIG. 1. (color online) Field-swept spectra of $BaFe_2As_2$ at constant frequency $f = 55.924$ MHz at 138 K for several different displacements of the piezoelectric device, showing the central and upper satellite transitions. INSET: Orientation of the crystal with respect to the external field, $\mathbf{H}_0$, the strain axis, and the rf field $\mathbf{H}_1$.

A single crystal of $BaFe_2As_2$ was synthesized via a self-flux method and cut to dimensions of approximately 1.5

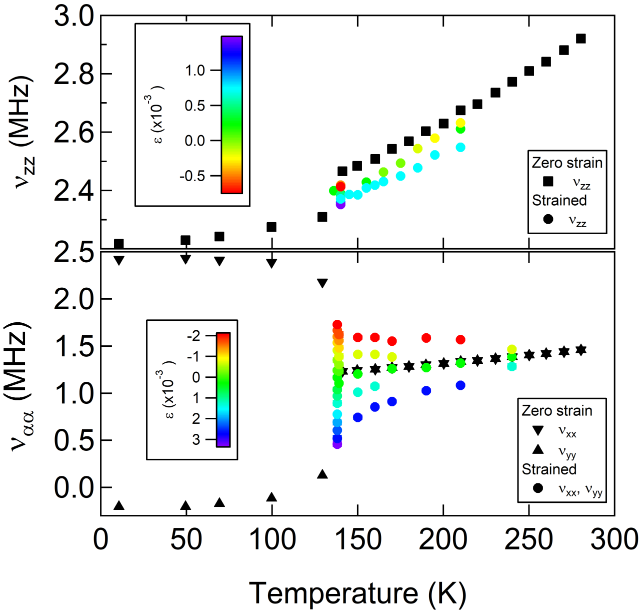

FIG. 2. (color online) The electric field gradient components (ν_{xx}, ν_{yy}, ν_{zz}) for the As versus temperature for $BaFe_2As_2$ both in zero strain (reproduced from [21]) and under uniaxial strain.

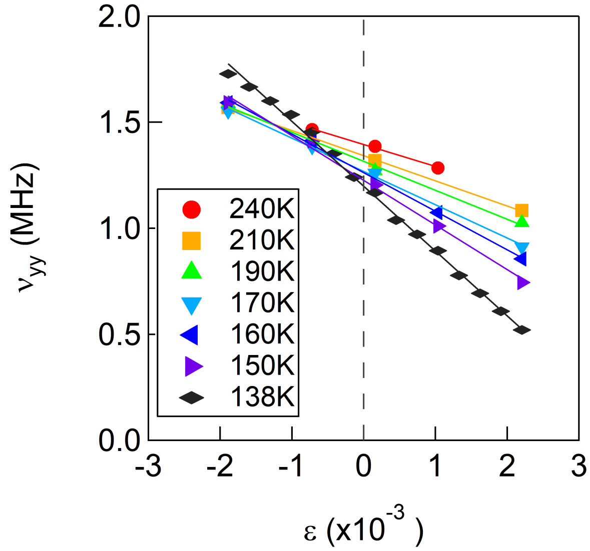

FIG. 3. (color online) The quadrupolar splitting ν_{yy} as a function of strain at several fixed temperatures. The solid lines are linear fits to the data.

mm×0.5 mm with the long axis parallel to the $(110)_T$ direction in the tetragonal basis along the Fe-Fe bond direction. The sample was mounted in a custom-built NMR probe incorporating a Razorbill cryogenic strain apparatus [22]. Uniaxial stress was applied to the crystal as described in [16] by piezoelectric stacks as illustrated in the inset of Fig. 1, and strain was measured by a capacitive dilatometer. A free-standing NMR coil was placed around the crystal, and spectra were measured by acquiring echoes while sweeping the magnetic field H_0 at fixed frequency, as shown in Fig. 1. ^{75}As has spin $I = 3/2$, with three separate resonances separated by the quadrupolar interaction. The higher quadrupolar satellite resonance occurs at field $H_{sat} = (f_0 + \nu_{\alpha\alpha})/\gamma(1 + K_{\alpha\alpha})$, where $f_0 = 55.924$ MHz is the rf frequency, $\gamma = 7.29019$ MHz/T is the gyromagnetic ratio, and $K_{\alpha\alpha}$ and $\nu_{\alpha\alpha}$ are the Knight shift and EFG tensor components in the $\alpha = (x, y, z)$ direction. The central transition field is given by: $H_{cen} = \frac{f_0}{\gamma(1+K_{\alpha\alpha})}\left(\frac{1}{2} + \sqrt{\frac{3f_0^2 - 2(\nu_{\beta\beta}+\nu_{\alpha\alpha})^2}{12}}\right)$, where $\beta = (y, x, z)$ for $\alpha = x, y, z$. The center of gravity of each peak was used to determine the resonance field, and hence $K_{\alpha\alpha}$ and $\nu_{\alpha\alpha}$ as a function of strain. The Knight shift shows essentially no change with strain [16], however, all components of the EFG tensor show strong variations, as shown in Fig. 2 and Fig. 3.

The EFG tensor is given by $\nu_{\alpha\beta} = (eQ/12h)\partial^2 V/\partial x_\alpha \partial x_\beta$, where $Q = 3.14 \times 10^{-29}\text{m}^2$ is the quadrupolar moment of the ^{75}As and V is the electrostatic potential at the As site. This quantity is dominated by the occupation of the As $4p$ orbitals, which in turn are hybridized with the $d_{xz,yz}$-orbitals of the neighboring Fe atoms [20]. In the tetragonal phase the EFG asymmetry parameter $\eta = (\nu_{yy} - \nu_{xx})/(\nu_{xx} + \nu_{yy})$ vanishes, as seen in Fig. 2. In the presence of finite nematicity, the C_4 symmetry of the EFG tensor is broken and $\nu_{xx} \neq \nu_{yy}$ [23]. In-plane anisotropic strain fields, $\varepsilon_{ani} = \frac{1}{2}(\varepsilon_{xx} - \varepsilon_{yy})$, with B_{2g} symmetry (in the coordinate system of the tetragonal unit cell) couple bilinearly to nematicity, therefore η responds to strain in the same manner that the magnetization of a ferromagnet responds to a uniform magnetic field [3, 12, 24]. Due to a finite Poisson ratio, uniaxial stress induces strains $\varepsilon_{\alpha\alpha}$ ($\alpha = x, y, z$) along three different directions, but the dominant contribution is ε_{ani} that couples to η.In our configuration we can only apply $\mathbf{H}_0$ perpendicular to the stress axis. We measure both $\nu_{zz} = \nu_{cc}$ along the $\hat{c}$ axis of the crystal, and ν_{yy} for $\mathbf{H}_0$ in the basal plane. For the latter case, $\nu_{yy} = \nu_{aa}$ for compressive strain ($\varepsilon_{ani} < 0$) and $\nu_{yy} = \nu_{bb}$ for tensile strain ($\varepsilon_{ani} > 0$), and $\nu_{xx}(\varepsilon_{ani}) = \nu_{yy}(-\varepsilon_{ani})$. The EFG thus enables us to identify the zero-strain displacement, x_0, by the condition $|\nu_{xx}| = |\nu_{yy}| = |\nu_{zz}|/2$. Note that η can exceed unity, since $\nu_{xx} + \nu_{yy} + \nu_{zz} = 0$. Furthermore, in the absence of strain a global order parameter in a twinned sample would average to zero, whereas the local order measured by NMR reveals all domains simultaneously [21].

As seen in Fig. 2, the applied strain significantly alters the local EFG. Just above the structural transition $T_s = 135$ K, the strained EFG values approach those in the spontaneously ordered phase in the absence of strain. Furthermore, the maximum strain levels as measured by the dilatometer reach approximately 60% of the spontaneous values of the orthorhombicity in the ordered phase [25]. Nevertheless, ν_{yy} remains linear over this range as shown in Fig. 3. The slope of this response is therefore a measure of the static nematic susceptibility, $\chi_{\rm nem}$. Similar behavior was observed in elastoresistance [3], shear modulus [11], and electronic Raman scattering [26]. However, the NMR probes the local nematicity in terms of the different orbital occupations obtained from calculated EFGs, rather than the global response due to different nematic domains.

Figure 4 shows the temperature dependence of $d\eta/d\varepsilon_{ani}$ and compares the response to elastoresistance measurements [3]. The NMR data exhibit a similar behavior with a divergence at T_s. We fit the EFG data to the sum of a Curie-Weiss term plus a background susceptibility: $\chi_{\rm nem} = C/(T - T_0) + \chi_0$, and find $C_0 = 4700 \pm 700$ K, $T_0 = 116 \pm 3$ K, and $\chi_0 = 54 \pm 8$. The background term reflects the intrinsic response of the lattice, whereas the Curie-Weiss term represents the nematic instability. Our observed value of T_0 is consistent with elastoresistance and shear modulus measurements, but differs from that observed by Raman scattering [11, 26, 27]. The difference between T_0 and T_s arises due to the coupling between the electronic nematic and the lattice, so that the free energy instability occurs before the divergence [28].

In order to understand the relationship between the EFG asymmetry and the splitting between the Fe d_{xz} and d_{yz} orbitals, we have performed GGA-based DFT calculations [? ?] for the tetragonal structure at 300 K and 0.2 GPa [?] under anisotropic in-plane strain ε_{ani}. Our values of the EFG are consistent with previous calculations in the absence of strain, but underestimate the experimental values by approximately a factor of three [29, 30]. We confirm that the EFG is dominated by the occupation of the As p orbitals [20], which are hybridized with the neighboring d_{xz} and d_{yz} orbitals. We calculate that $d\eta/d\varepsilon_{ani} = 33$, which is close to the experimental value of the background susceptibility, χ_0. The strong temperature-dependent divergence at T_s is a collective phenomenon driven by the electronic system and cannot be captured by the DFT calculations which are valid only at $T = 0$. Under strain, the two bands with dominant d_{yz} and d_{xz} character become non-degenerate, and develop a finite splitting, Δ_{xz-yz}, at the X point in **k**-space. We find that $\eta = A\Delta_{xz-yz}$, where $A = 5.7$/eV. These values are consistent with angle-resolved photoemission experiments that indicate a splitting $\Delta_{xz-yz} \sim 40$ meV in the nematic phase [5], whereas NMR studies reveal a value of $\eta \sim 1.2$ [21].

Fig. 2 also shows the quadrupolar splitting ν_{zz} along the c-axis to in-plane strain. This independent component of the EFG tensor does not couple to the nematic order, but nevertheless it is suppressed by the lattice distortion. We find that $|\nu_{zz}(\varepsilon_{ani})/\nu_{zz}(0)| = 1 - \beta\varepsilon_{ani}^2$, where $\beta \approx 9000$ is approximately temperature independent. Our DFT calculations reveal a small quadratic suppression with $\beta = 30$, due to changes in the relative occupations of the As p_z and $p_{x,y}$ orbitals. The difference between the experimental and theoretical values may reflect changes to the c-axis lattice parameters due to a finite Poisson ratio.

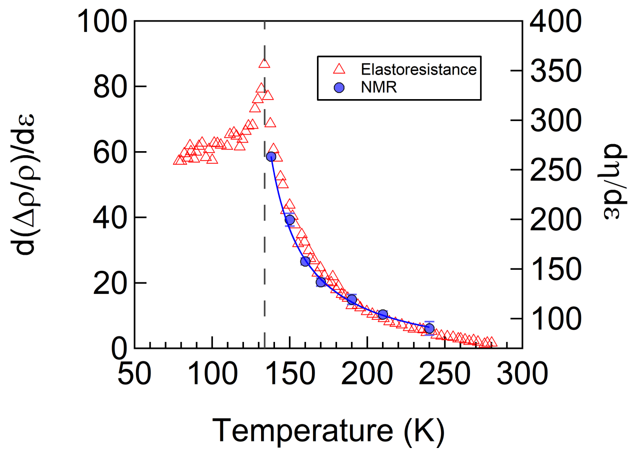

FIG. 4. (color online) The nematic susceptibility measured by the EFG asymmetry (•) and that measured by elastoresistance (△, reproduced from [3]). The solid line is a fit to the NMR data, as described in the text. The vertical dashed line indicates T_N.

Our measurements offer insight into the behavior of the EFG in electron-doped pnictides. In doped $Ba(Fe,M)_2As_2$ (M = Co, Ni), the quadrupolar satellite resonances are inhomogeneously broadened ($\sim 1.0 - 1.5$ MHz) relative to those in the parent compound (0.13 MHz) [31–33]. A large source of this broadening may arise from local strain distributions. Local strains at dopant atoms can reach up to 3% [34], which would correspond to a shift in the As EFG parameters of $\delta\eta \sim 10$ and $\delta\nu_{zz} \sim 2.9$ MHz at 140 K. The strain field relaxes with distance from the dopant giving rise to a distribution of local EFGs. Recently a finite EFG asymmetry $\eta \sim 0.1$ was reported in $BaFe_2(As_{1-x}P_x)_2$ in the tetragonal phase [20]. This value would be consistent with an average strain field on the order of 0.05%. We postulate, therefore, that the origin of the finite nematicity observed in this compound reflects inhomogeneous strain fields from the dopant atoms, rather than intrinsic nematicity above the structural transition [35]. Complex EFG distributions have also been reported in $RFeAsO_{1-x}F_x$ (R = La, Sm) that have been interpreted as nanoscale electronic order [36]. It is unclear whether these spatial variations arise due to ν_{zz} or η, although they may reflect a combination of both strain and/or orbital occupations.

In conclusion, we have conducted detailed measurements of the EFG under a uniform uniaxial stress, and observed a linear response that is strongly temperature dependent. The slope agrees well with other measurements of the nematic susceptibility, and demonstrates that C_4 symmetry is broken not only in the different Fe 3d orbital occupations, but also in the As 4p orbitals. Our results further demonstrate that ^{75}As NMR is sensitive to the charge degrees of freedom, and enable a quantitative measure of the local orbital occupations of the Fe d-orbitals. Measurements of the local nematicity by NMR provide an important microscopic complement to other techniques, and offer a unique opportunity to measure the response in the superconducting state. For example, in contrast to elasto-resistance and Raman scattering, NMR under strain can probe the nematic susceptibility below T_c. Such measurements may provide insight into the role of nematic degrees of freedom in the superconducting mechanism [37].

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