Elastoresistance measurements on CaKFe$_4$As$_4$ and KCa$_2$Fe$_4$As$_4$F$_2$ with the Fe site of C$_{2v}$ symmetry

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We report resistance and elastoresistance measurements on (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$, CaKFe$_4$As$_4$, and KCa$_2$Fe$_4$As$_4$F$_2$. The Fe-site symmetry is $D_{2d}$ in the first compound, while $C_{2v}$ in the latter two, which lifts the degeneracy of the Fe $d_{xz}$ and $d_{yz}$ orbitals. The temperature dependence of the resistance and elastoresistance is similar between the three compounds. Especially, the [110] elastoresistance is enhanced with decreasing temperature irrespective of the Fe-site symmetry. This appears in conflict with recent Raman scattering studies on CaKFe$_4$As$_4$, which suggest absence of nematic fluctuations. We consider possible ways of reconciliation and suggest that the present result is important in elucidating the origin of in-plane resistivity anisotropy in iron-based superconductors.

Parent compounds of iron-based superconductors typically exhibit a tetragonal-to-orthorhombic structural phase transition at $T_s$ and a stripe-type antiferromagnetic transition at $T_N \leq T_s$ [1,13]. In spite of the tiny orthorhombicity $\delta = (a-b)/(a+b) = 2 - 4 \times 10^{-3}$ [2,13], noticeable in-plane anisotropy appears in various electronic properties below $T_s$, as revealed by e.g. resistivity [4], optical conductivity [5,6], inelastic neutron scattering (INS) [7,8], and NMR measurements [9]. We especially note that angle-resolved photoemission spectroscopy (ARPES) measurements show that the degenerate $d_{xz}$ and $d_{yz}$ levels of Fe in the tetragonal phase split considerably below $T_s$ [10]. Those observations suggest that the transition at $T_s$ is electronically driven, and hence it is regarded as an electronic nematic transition (see e.g. [11,14] for a review). However, which electronic degrees of freedom, spin or orbital, is responsible for the nematic transition is still highly debated [11,17]. The primary order parameter of the nematic phase would be $\epsilon = \langle\cos(\theta)\rangle$, where $\theta$ is the angle between the strain direction and the perpendicular direction induced by the strain. The elastoresistance with strain applied along the tetragonal [110] direction, which becomes the $a$ or $b$ axis in the orthorhombic phase below $T_s$, is assumed to be a proxy for the nematic susceptibility [15]. Raman scattering in $B_{1g}$ symmetry (1-Fe unit cell) can detect nematic fluctuations and hence can be used to derive the nematic susceptibility [16,23]. The shear modulus can also be related to the nematic susceptibility [25]. Nematic susceptibilities in 1111-, 122-, and 11-type iron-based superconductors (or parent compounds) estimated by these different techniques are broadly consistent (when comparison can be made), exhibiting strong enhancement as $T \to T_s$ (or $T \to 0$ in the case of moderately overdoped compounds where the structural transition does not occur down to absolute zero) [15,16,18,19,20].

In this context, it is intriguing that recent Raman scattering measurements on CaKFe$_4$As$_4$ [11] fail to observe nematic fluctuations [22,25], despite the fact that the [110] elastoresistance is enhanced with decreasing temperature [14]. The authors of [13] ascribe the absence of nematicity in CaKFe$_4$As$_4$ to the lowered symmetry of the Fe site (see Fig. 1). The Fe site symmetry in 1111-, 122-, and 11-type iron-based superconductors (or parent compounds) is $D_{2d}$, where the $d_{xz}$ and $d_{yz}$ orbitals are degenerate. On the other hand, the Fe site symmetry in CaKFe$_4$As$_4$ is $C_{2v}$ as explained in Fig. 1, where the $d_{xz}$ and $d_{yz}$ orbitals are no longer degenerate. The authors argue that this would result in a static antiferro-
were prepared in Tsukuba using KAs and FeAs as flux, and the height of the As layers above and below are different. The symmetry of the Fe site is lowered to $C_{2v}$, where the $d_{zx}$ and $d_{yz}$ orbitals are no longer degenerate.

FIG. 1. Crystal structures of (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$, CaKFe$_4$As$_4$, and KCa$_2$Fe$_4$As$_4$F$_2$ (from left to right, prepared using the software VESTA [47]). In (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$, the Fe layers are located at $z = 1/4$ (and equivalent), and the heights (distances) $h$ measured from the Fe layers of the As layers above and below are the same. The symmetry of the Fe site is $D_{2d}$, where the $d_{xz}$ and $d_{yz}$ orbitals are degenerate. In CaKFe$_4$As$_4$ and KCa$_2$Fe$_4$As$_4$F$_2$, the Fe layers are displaced from $z = 1/4$, and the height $h_1$ of the As layers above and $h_2$ of the As layers below are different. The symmetry of the Fe site is lowered to $C_{2v}$, where the $d_{zx}$ and $d_{yz}$ orbitals are no longer degenerate.

quadrupolar (AFQ) order and preclude “Pomeranchuk-like fluctuations” [43]. A sample and a strain gage were glued on the surface of a piezostack, and the sample resistance $R$ was recorded as a function of the strain $\epsilon$ as the operating voltage of the piezostack was ramped up and down. Instead of holding a constant temperature for each measurement, we collected $R$ vs $\epsilon$ data continuously, slowly cooling or warming the sample (typically 0.3 K/min), and corrected the data for the resistance variation due to the temperature variation to determine the elastoresistance [51]. Results reported below are for the longitudinal configuration, i.e., the current $I$ was applied parallel to the strain $\epsilon$. The elastoresistance coefficients $m_{[110]}$ and $m_{[100]}$ were measured with $I \parallel [110]$ and [010], respectively. Nematic fluctuations under consideration enhance $m_{[110]}$ but do not $m_{[100]}$. In some previous studies, using both longitudinal and transverse configurations, values of $2m_{66}$ and $(m_{11} - m_{12})$ were determined, where $m_{ij}$’s are components of the elastoresistance tensor [20]. $m_{[110]}$ and $m_{[100]}$ are practically proportional to $2m_{66}$ and $(m_{11} - m_{12})$ [15, 51, 52].

Figure 2 shows the temperature dependence of the resistance and elastoresistance for (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$, CaKFe$_4$As$_4$, and KCa$_2$Fe$_4$As$_4$F$_2$. For each compound, the same sample was used to measure the temperature dependence of the resistance and [110] elastoresistance, while a different sample was used for the [100] elastoresistance (it is impossible to remove epoxy without damaging samples). In addition, at least one more sample for each compound was measured to confirm the reproducibility of the [110] elastoresistance. For the [110] samples used in the figure, the residual resistivity ratio defined as $R(300 \text{ K})/R(36 \text{ K})$ and superconducting transition temperature $T_c$ determined from the midpoint of transition are summarized in Table 1. The large residual resistivity ratios and transition temperatures indicate the high quality of our samples. Although the actual resistivity $\rho$ is not easy to accurately determine because of uncertainty in the sample dimensions and inhomogeneous current distribution, especially along the interlayer direction, resistivities at $T = 300 \text{ K}$ of (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$ and CaKFe$_4$As$_4$ were
FIG. 2. Temperature dependence of resistance (a, c, e) and elastoresistance (b, d, f) for (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$, CaKFe$_4$As$_4$, and KCa$_2$Fe$_4$As$_2$F$_2$ (from left to right). The broken lines in (a, c, e) are the temperature derivatives. In the elastoresistance measurements (b, d, f), the electrical current $I$ and the strain $\epsilon$ are parallel and applied parallel to [110] and [100]. The broken lines show Curie-Weiss fits $m_{[110]} = C/(T - \theta) + m_0$ to the [110] data in the region between 100 and 185 K (see text).

TABLE I. The first column shows resistivities at $T = 300$ K of (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$ and CaKFe$_4$As$_4$ estimated from measurements on four and five samples, respectively. Resistivity was not determined for KCa$_2$Fe$_4$As$_2$F$_2$. The rest of the columns show values determined for the [110] samples of Fig. 2 residual resistivity ratio defined as $R(300 \text{ K})/R(36 \text{ K})$, superconducting transition temperature $T_c$ determined from the midpoint of transition, the coefficients $C$, $\theta$, and $m_0$ determined from the Curie-Weiss fit to the [110] elastoresistance data (see text).

|                  | $\rho$(300 K) ($\mu\Omega$ cm) | $R$(300 K)/$R$(36 K) | $T_c$ (K) | $C$ (K) | $\theta$ (K) | $m_0$ |
|------------------|-------------------------------|----------------------|-----------|---------|--------------|-------|
| (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$ | 430(180)                     | 14                   | 34.8      | 1280(20) | 44.3(5)      | -1.8(1) |
| CaKFe$_4$As$_4$  | 350(190)                     | 17                   | 35.9      | 890(20) | 49(1)        | -3.7(2) |
| KCa$_2$Fe$_4$As$_2$F$_2$ | -                             | 12                   | 33.7      | 750(20) | 45(1)        | -1.8(1) |

Estimated from measurements on four and five samples, respectively, and shown in the table as well. The resistivity was not determined for KCa$_2$Fe$_4$As$_2$F$_2$ because of technical reasons. The dashed lines in Figs. 2(b), (d), and (f) are Curie-Weiss fits $m_{[110]} = C/(T - \theta) + m_0$ to the [110] data in the temperature range between 100 and 185 K (the upper bound 185 K was set because it was the highest temperature for the elastoresistance measurements on KCa$_2$Fe$_4$As$_2$F$_2$). The estimated parameters $C$, $\theta$, and $m_0$ are also listed in Table I.

The three compounds show a similar temperature dependence of resistance [Figs. 2(a), (c), and (e)]: the $R(T)$ curves are slightly concave at high temperatures and become convex at low temperatures. The inflection point is 86, 93, and 69 K for (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$, CaKFe$_4$As$_4$, and KCa$_2$Fe$_4$As$_2$F$_2$, respectively (see the derivative curves). Furthermore, the derivative curves (dashed lines) of the former two compounds emphasize a particularly close similarity between them.

The measured [110] elastoresistances are enhanced considerably with decreasing temperature irrespective of the Fe-site symmetry, while the [100] ones are much smaller and much less temperature dependent [Figs. 2(b), (d), and (f)]. The [110] elastoresistances can be described well by the Curie-Weiss formula down to $\sim 100$ K (dashed lines) but deviate downward at lower temperatures. Although the coefficient $C$ in (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$ is considerably larger than those in the other two (Table I), its significance is unclear. A quantitative analysis of $C$ would require an elaborate theory dealing with changes in the electronic structure and electron scattering due to an applied strain. The [110] elastoresistance data for (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$ is qualitatively consistent with the $2m_{66}$ data for (Ba$_{0.6}$K$_{0.4}$)Fe$_2$As$_2$ [28]. The present fit parameter $\theta = 44.3(5)$ K (Table I) is close to $\theta = 46.1(2.4)$ K reported in [28]. The [110] elastoresistance data for CaKFe$_4$As$_4$ is also qualitatively consistent with the $2m_{66}$ data reported in [41].

In order to compare the [110] elastoresistance data between the three compounds, we prepared two types of plots (Fig. 3). Figure 3(a) shows $C(m_{[110]} - m_0)^{-1}$ as a function of temperature. The [110] elastoresistance data of the three compounds show similar behavior: a Curie-Weiss behavior (i.e. straight line) in a limited temperature range, upward deviation at lower temperatures, and downward deviation at higher temperatures. This is ac-
FIG. 3. (a) $C(m_{[110]} - m_0)^{-1}$ and (b) $m_{[110]}T$ as a function of temperature for (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$, CaKFe$_4$As$_4$, and KCa$_2$Fe$_4$As$_4$F$_2$. $m_{[110]}$ is the [110] elastoresistance shown in Figs. 2(b), (d), and (f), while $C$ and $m_0$ are the Curie-Weiss fit parameters listed in Table I. The dashed line in (a) corresponds to the Curie-Weiss to the CaKFe$_4$As$_4$F$_2$ itself is also highly two-dimensional [40, 50]. These might be related to the difference. 

Our experimental results show that (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$, CaKFe$_4$As$_4$, and KCa$_2$Fe$_4$As$_4$F$_2$ exhibit similar temperature dependence of resistance and elastoresistance. Especially, the similarity between the former two compounds is striking. This indicates that the electronic structure and scattering mechanisms, which dominate the electrical conduction, are similar between the three compounds. If the enhancement of the [110] elastoresistance in (Ba$_{0.5}$K$_{0.5}$)Fe$_2$As$_2$ is ascribed to nematic fluctuations, as widely believed, it therefore appears natural to ascribe the enhancement in the latter two compounds to nematic fluctuations as well. However, this assumption is in conflict with the Raman scattering studies on CaKFe$_4$As$_4$ [12, 13], which did not observe nematic fluctuations as already mentioned. We consider two possibilities to resolve this conflict in the following.

Firstly, one can argue that, although nematic fluctuations exist in CaKFe$_4$As$_4$, they were missed in the Raman studies. As suggested in [43], nematic fluctuations in the charge/orbital sector may be precluded by the AFQ-order-like arrangement of the $d_{xz}$ and $d_{yz}$ orbitals in CaKFe$_4$As$_4$. However, it does not necessarily mean that spin-driven nematicity, for which the degeneracy of the $d_{xz}$ and $d_{yz}$ orbitals is not a prerequisite, is precluded. We note that Raman scattering due to nematic fluctuations in hole-doped (Ba$_{1-x}$K$_x$)Fe$_2$As$_2$ [42, 43] is already weaker than that in electron-doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [17, 23]. It is necessary to theoretically examine if the lowered Fe-site symmetry or the absence of nematic fluctuations in the charge/orbital sector in CaKFe$_4$As$_4$ may further suppress Raman scattering intensity (to the extent that scattering due to nematic fluctuations is missed). Raman scattering measurements on KCa$_2$Fe$_4$As$_4$F$_2$ are also desirable.

Secondly, one can argue that enhanced elastoresistance does not necessarily indicate enhanced nematic fluctuations. Fluctuations of some order parameter other than nematic ones may also enhance the elastoresistance if it couples to the lattice, breaks tetragonal symmetry, and brings about a unidirectional in-plane anisotropy in the electronic structure and/or scattering. In the present case, mere stripe-type spin fluctuations without nematic correlations already suffice. The enhancement of spin fluctuations with decreasing temperature is observed in NMR and INS measurements on CaKFe$_4$As$_4$ [58, 61] and KCa$_2$Fe$_4$As$_4$F$_2$ [62]. As a strain is applied, spin fluctuations at $(\pi, 0)$ and $(0, \pi)$ become inequivalent, which can result in resistivity anisotropy. INS measurements on CaKFe$_4$As$_4$ and KCa$_2$Fe$_4$As$_4$F$_2$ under uniaxial pressure are desirable to access the validity of this scenario.

Finally, it is highly debated whether the in-plane re-
Further studies on the resistivity anisotropy in iron-based superconductors is due to anisotropic scattering or anisotropic Fermi surface [27, 30, 63–75]. If the AFQ-order-like arrangement of the $d_{xz}$ and $d_{yz}$ orbitals in $C_{2v}$ Fe-site compounds [43] suppresses Fermi surface deformation due to an applied strain, the present observation of enhanced elastoresistance in CaKFe$_4$As$_4$ and KCa$_2$Fe$_4$As$_4$F$_2$ may suggest the dominant role of anisotropic scattering.

In conclusion, we have observed that (Ba$_0.5$K$_{0.5}$)Fe$_2$As$_2$, CaKFe$_4$As$_4$, and KCa$_2$Fe$_4$As$_4$F$_2$ exhibit similar temperature dependence of resistance and elastoresistance. The [110] elastoresistance is enhanced irrespective of the Fe-site symmetry, which appears in conflict with the previous Raman studies [12, 43]. We suggested two possible ways of reconciliation: The first one assumes that nematic fluctuations were missed in the Raman studies, while the other assigns the enhanced elastoresistance to fluctuations other than nematic ones. We also suggested that the present result might shed new light on the issue of the origin of the resistivity anisotropy in iron-based superconductors. Further studies on $C_{2v}$ Fe-site compounds, not only experimental ones but also theoretical ones, are clearly called for.

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