Expansion of the tetragonal magnetic phase with pressure in the iron arsenide superconductor Ba_{1−x}K_{x}Fe_{2}As_{2}

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Expansion of the tetragonal magnetic phase with pressure in the iron-arsenide superconductor $Ba_{1-x}K_xFe_2As_2$

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In the temperature-concentration phase diagram of most iron-based superconductors, antiferromagnetic order is gradually suppressed to zero at a critical point, and a dome of superconductivity forms around that point. The nature of the magnetic phase and its fluctuations is of fundamental importance for elucidating the pairing mechanism. In $Ba_{1-x}K_xFe_2As_2$ and $Ba_{1-x}Na_xFe_2As_2$, it has recently become clear that the usual stripe-like magnetic phase, of orthorhombic symmetry, gives way to a second magnetic phase, of tetragonal symmetry, near the critical point, in the range of $x = 0.24$ and $x = 0.28$ for $Ba_{1-x}K_xFe_2As_2$. In a prior study, an unidentified phase was discovered for $x < 0.24$ but under applied pressure, whose onset was detected as a sharp anomaly in the resistivity. Here we report measurements of the electrical resistivity of $Ba_{1-x}K_xFe_2As_2$ under applied hydrostatic pressures up to 2.75 GPa, for $x = 0.22$, 0.24 and 0.28. The critical pressure above which the unidentified phase appears is seen to decrease with increasing x and vanish at $x = 0.24$, thereby linking the pressure-induced phase to the tetragonal magnetic phase observed subsequently at ambient pressure. In the temperature-concentration phase diagram of $Ba_{1-x}K_xFe_2As_2$, we find that pressure greatly expands the tetragonal magnetic phase, while the stripe-like phase shrinks. This reveals that pressure may be a powerful tuning parameter with which to explore the interplay between magnetism and superconductivity in this material.

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The phase diagram of iron-based superconductors of the $BaFe_2As_2$ family is characterized by competing antiferromagnetic (AF) order and superconductivity. Usually, the AF order decreases with concentration (doping) and a dome of superconductivity surrounds the critical point.[1] The AF order is a stripe-like spin-density wave, with a wavevector $\mathbf{Q} = (\pi, 0)$ and the magnetic moments lie in the plane. At the magnetic transition temperature, or slightly above it, the lattice changes from tetragonal at high temperature to orthorhombic at low temperature.[2,3]

In $Ba_{1-x}X_xFe_2As_2$, where X = K or Na, the phase diagram was recently found to be richer than this simple picture. Resistivity measurements under pressure revealed the existence of an internal transition inside the AF phase of $Ba_{1-x}K_xFe_2As_2$.[4] As the onset temperature T_N of the orthorhombic AF phase (o-AF) is suppressed with hydrostatic pressure P, an additional phase transition to a "new phase" appears below a transition temperature $T_0 < T_N$, for $0.16 < x < 0.21$, when $P > 0.9$ GPa.[4] A tetragonal magnetic phase (t-AF) was then discovered in the closely related compound $Ba_{1-x}Na_xFe_2As_2$, by neutron and x-ray diffraction on powder samples.[5] Subsequent neutron scattering on single crystals showed that in this t-AF phase the spins are aligned parallel to the c axis.[6] A similar phase of tetragonal symmetry was then found in $Ba_{1-x}K_xFe_2As_2$ at ambient pressure, for $0.24 < x < 0.28$.[7] The magnetic moments in the t-AF phase of $Ba_{1-x}K_xFe_2As_2$ are also oriented along the c axis.[8,9] Infrared spectroscopy showed that the t-AF phase has a double-Q magnetic structure,[8] as opposed to the single-Q structure of the o-AF phase. A pressure study of a $Ba_{1-x}K_xFe_2As_2$ sample with $x = 0.15$ by specific heat, transport and especially the Nernst effect confirms the bulk nature of the sequence of phase transitions previously detected only in resistivity.[10] Additionally the authors show that the pressure induced "new phase" suppresses the large Nernst signal of the o-AF phase, indicating the suppression of the nematicity as in the t-AF phase at ambient pressure. Several theoretical studies have investigated the properties of the tetragonal magnetic phase in iron-based superconductors.[5,11–20]

In this Article, we extend our prior study of $Ba_{1-x}K_xFe_2As_2$ under pressure, performed up to $x = 0.21$,[4] by studying three further samples, with $x = 0.22$, 0.24 and 0.28. We are able to connect the additional phase induced by pressure with the tetragonal phase seen at ambient pressure. Pressure is seen to cause a dramatic expansion of the tetragonal magnetic phase, on the backdrop of a shrinking orthorhombic phase.

Methods.– Single crystals of $Ba_{1-x}K_xFe_2As_2$ were grown from self flux.[21] Three underdoped samples were measured, with a superconducting transition temperature $T_c = 20.8 \pm 0.5$ K, 25.4 ± 0.5 K, and 30.1 ± 0.5 K, respectively. Using the relation between T_c and the nominal K concentration x reported in ref. 3 and wavelength-dispersive x-ray spectroscopy,[22] we obtain $x = 0.22$, 0.24

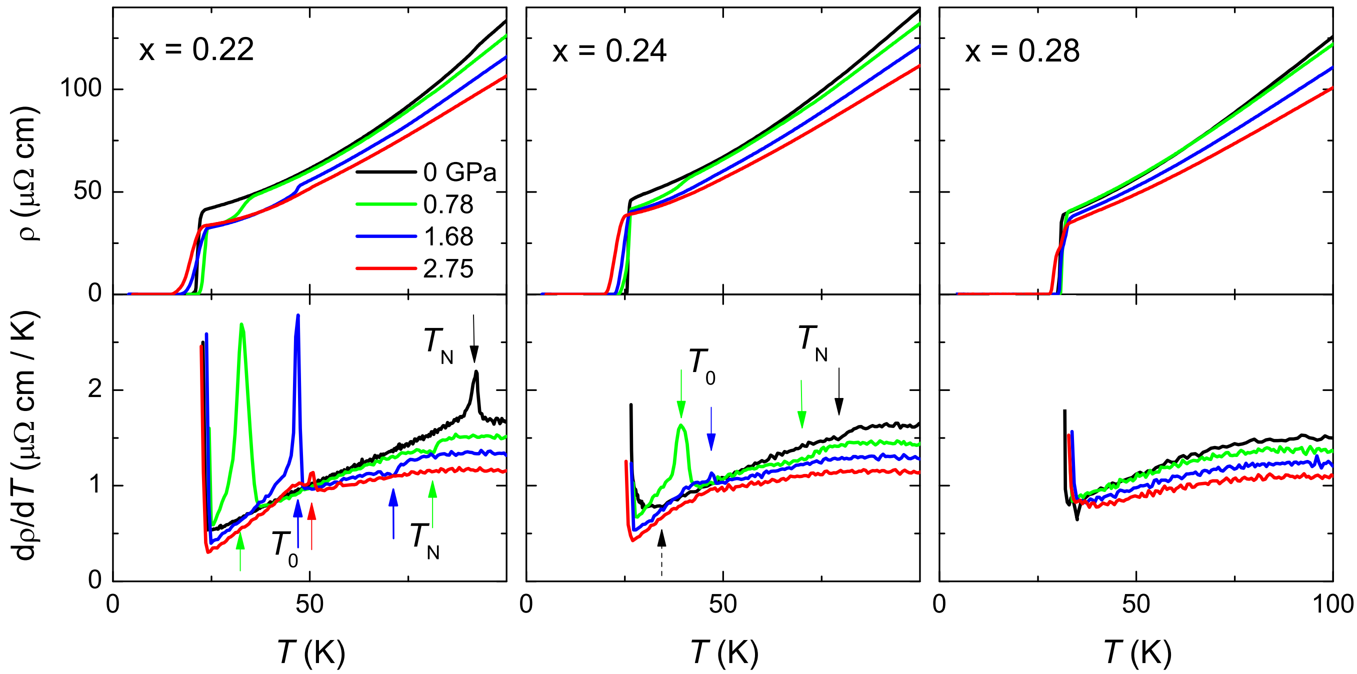

FIG. 1: *Top*: In-plane electrical resistivity of $Ba_{1-x}K_xFe_2As_2$ for $x = 0.22$, $x = 0.24$ and $x = 0.28$ (different columns) for four different pressures, as indicated. *Bottom*: Temperature derivative of the data in the top panels. The peak (dip) between 60 K and 100 K signals the onset of stripe-like antiferromagnetic order at T_N (arrows). The peak at lower temperature signals the onset of the tetragonal magnetic phase at T_0 (arrows).

and 0.28, respectively. These x values are also consistent with the measured antiferromagnetic ordering temperature T_N (which coincides with the structural transition from tetragonal to orthorhombic),[3] equal to 91 ± 2 K and 79 ± 5 K, respectively for the two lower dopings. The sample with $x = 0.28$ shows no magnetic or structural transition. The resistivity at room temperature of all samples lies between 250 and 350 $\mu\Omega$ cm, in agreement with previous studies.[23] As before,[4] we have normalized the resistivity at $T = 300$ K to 300 $\mu\Omega$cm. Hydrostatic pressures up to 2.75 GPa were applied with a hybrid piston-cylinder cell,[24] using a 50:50 mixture of n-pentane:isopentane. This pressure transmitting medium has been shown to present the best hydrostatic conditions, i.e., the smallest uniaxial pressure component, in the pressure range up to 3 GPa.[25] The pressure was measured via the superconducting transition of a lead wire inside the pressure cell. The electrical resistivity ρ was measured for a current in the basal plane of the orthorhombic crystal structure, with a standard four-point technique using a Lakeshore ac-resistance bridge. The transition temperatures are defined as follows: T_c is where $\rho = 0$; T_N and T_0 are detected as extrema in the derivative $d\rho/dT$.

Resistivity.– Fig. 1 shows the in-plane resistivity (top panels) and its temperature derivative (bottom panels) of each sample, for a selection of pressures. T_N is detected as a peak in the derivative for the first sample at ambient pressure, and then as a dip for higher pressures or doping. The transition at T_0 shows up as a sharp peak, below T_N. For those concentrations and applied pressures where both T_N and T_0 are detected, the resistivity curves and their temperature derivatives resemble the curves of a sample with $x = 0.25$ at ambient pressure, where the t-AF phase is present (see the supplemental online material of ref. 7.) In that publication, resistivity is identified as a good probe of T_0 via a comparison with thermodynamic probes such as the thermal expansion or specific heat. In Fig. 2, the full set of derivative curves is displayed for $x = 0.22$ and $x = 0.24$, allowing to track the anomalies at T_N and T_0 as a function of pressure.

As previously reported for samples with lower doping,[4] T_N decreases linearly with pressure. For $x = 0.22$, the peak in the derivative at T_N evolves into a dip at 0.48 GPa. We are able to follow this dip up to $P = 2$ GPa, above which it disappears. The evolution of the peak at T_0 is different. At 0.48 GPa, the peak at T_0 appears. T_0 goes up with pressure until it stays almost constant above 2.3 GPa. The height of the sharp peak at T_0 increases slightly at first, and then decreases above $P \simeq 1.5$ GPa. The behavior for $x = 0.24$ is similar, but shifted to lower pressures. T_N can be followed only up to 0.94 GPa. The transition at T_0 appears as a peak as soon as we apply pressure. In fact, a slight upturn of the derivative with decreasing T, indicative of an onset of the transition at T_0, can be seen even at ambient pressure. The onset is marked by an up-pointing dashed arrow in the lower middle panel of Fig. 1. We see that the new phase is present in this sample at $P = 0$. This provides a direct link between what was initially called the “new

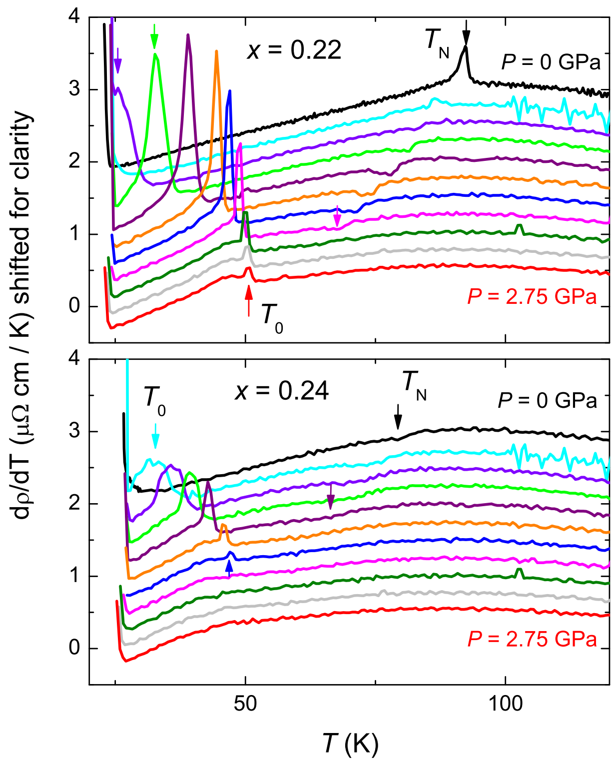

FIG. 2: *Top*: Temperature derivative of the resistivity of $Ba_{1-x}K_xFe_2As_2$ with $x = 0.22$, for 11 different pressures, from ambient pressure ($P = 0$) at the top (black) to $P = 2.75$ GPa at the bottom (red), with the following intermediate values: $P = 0.28$, 0.48, 0.78, 0.94, 1.37, 1.68, 2.0, 2.31, and 2.4 GPa. The curves are shifted for clarity. The black down-pointing arrow marks T_N at $P = 0$. The next down-pointing arrow marks T_N at the highest pressure where it can still be detected. T_0 shows up as a peak at low temperature. The up-pointing arrows mark T_0 at the highest pressure where the peak can still be detected. *Bottom*: The same for $x = 0.24$.

phase" and what is now known to be the t-AF phase. (In our previous study, a similar situation was found for $x = 0.19$ at $P = 1.08$ GPa. At zero magnetic field, a slight onset of the transition at T_0 was seen above T_c, which was completely uncovered by a magnetic field of $H = 15$ T shifting the T_c far below T_0, which is unaffected by the field.[4]) This $x = 0.24$ sample is apparently right at the border of the t-AF phase, as a very tiny amount of either pressure or additional K content is enough to clearly induce the t-AF phase. The peak at T_0 stays sharp but its height decreases above $P \simeq 1$ GPa, and the last pressure where it is observed is 1.68 GPa. The curve at this pressure looks very much like the one at the highest pressure in the $x = 0.22$ sample.

Temperature-pressure phase diagram.– Fig. 3 presents the temperature-pressure phase diagram for the three samples. T_N decreases linearly with P, with a slightly steeper slope at $x = 0.24$. By contrast, T_0 rises rapidly, at least initially. At $x = 0.22$, T_0 saturates above $P = 2.3$ GPa. At $x = 0.24$, we can no longer detect T_0 above $P = 1.68$ GPa (Fig. 2), the pressure at which it merges with the T_0 line at $x = 0.22$ (Fig. 3).

At $x = 0.24$, the phase diagram is such that if the T_0 line (blue) saturates at high pressure as it does in the case of $x = 0.22$ (red T_0 line), then a linear extension of the T_N line (blue) will hit that T_0 line, implying that the t-AF phase would persist to pressures beyond the end of the o-AF phase.

As for superconductivity, note that T_c decreases as soon as the tetragonal phase appears (Fig. 3), as found in prior studies of $Ba_{1-x}K_xFe_2As_2$ (refs. 4,27) and $Ba_{1-x}Na_xFe_2As_2$,[26,27] in agreement with the negative dT_c/dP expected from the Ehrenfest relation applied to the thermodynamic data.[7]

Temperature-concentration phase diagram.– Combining our present results with those of our previous study,[4] we plot the temperature-concentration phase diagram of $Ba_{1-x}K_xFe_2As_2$ in Fig. 4. For comparison, we also reproduce the phase diagram at zero pressure reported in ref. 7; the agreement with our own ambient-pressure data is excellent. We see that the T_N line moves down with pressure, in parallel fashion. This suggests that the critical concentration x_N where T_N goes to zero shifts down with pressure.

On the backdrop of this shrinking o-AF phase, the tetragonal magnetic phase undergoes a major expansion with pressure (Fig. 4). While the t-AF phase occupies a small area below T_N at ambient pressure, its area grows by an order of magnitude at $P = 2.4$ GPa. In other words, at high pressure the tetragonal phase becomes the dominant magnetic phase in the temperature-concentration phase diagram of $Ba_{1-x}K_xFe_2As_2$. A recent study of thermal expansion and specific heat revealed a complex phase diagram in $Ba_{1-x}Na_xFe_2As_2$ with an expanded tetragonal phase. There, in agreement with our results, chemical pressure might lead to the expansion of the tetragonal phase.[28] In the context of recent calculations, it may be that pressure favours the t-AF phase because it changes the ellipticity of the electron pockets in the Fermi surface of $Ba_{1-x}K_xFe_2As_2$.[16]

Summary.– In summary, we have shown that the new phase discovered in $Ba_{1-x}K_xFe_2As_2$ from sharp signatures in the resistivity under pressure [4] is the tetragonal antiferromagnetic phase observed and identified subsequently by various probes in both $Ba_{1-x}Na_xFe_2As_2$ (refs. 5,6) and $Ba_{1-x}K_xFe_2As_2$.[7–9] Under pressure, this t-AF phase expands enormously, by an order of magnitude for 2.4 GPa in terms of the area it occupies in the temperature-concentration phase diagram, relative to the orthorhombic stripe-like AF phase that dominates at ambient pressure. As a result, at high pressure superconductivity exists on the border of a dominant tetragonal magnetic phase. It is then likely that fluctuations of that double-Q phase play a role in the pairing. Recent calculations suggest that such fluctuations could actually enhance T_c.[19]

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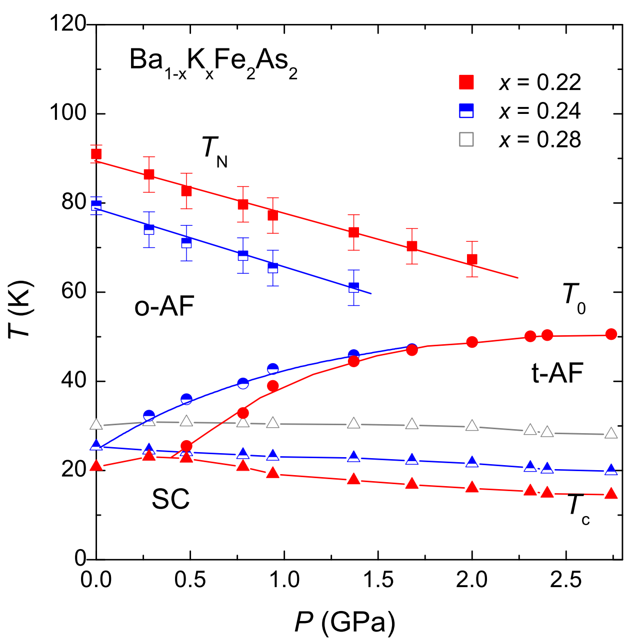

FIG. 3: Temperature-pressure phase diagram of $Ba_{1-x}K_xFe_2As_2$, for $x = 0.22, 0.24$ and 0.28 (full, half-full and empty symbols, respectively), showing the orthorhombic antiferromagnetic (o-AF) transition temperature T_N (squares), the superconducting (SC) transition temperature T_c (triangles), and the tetragonal antiferromagnetic (t-AF) transition temperature T_0 (circles).

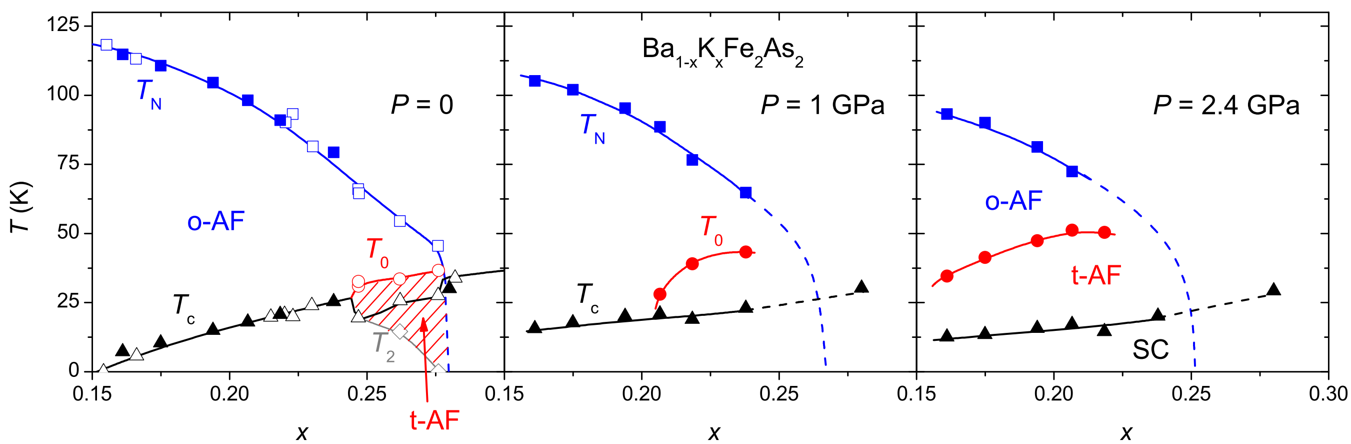

FIG. 4: Temperature-concentration phase diagram of $Ba_{1-x}K_xFe_2As_2$, showing T_N (blue squares), T_0 (red circles) and T_c (black triangles), for three different values of the applied pressure: $P = 0$ (left panel), 1.0 GPa (middle panel) and 2.4 GPa (right panel). This includes data from our previous study.[4] Ambient-pressure data from ref. 7 are also shown in the left panel (open symbols), including a transition back to the o-AF phase, below T_2 (diamonds). All lines are a guide to the eye. The evolution from left to right, with increasing pressure, reveals a major expansion of the tetragonal magnetic phase (t-AF), on the backdrop of a shrinking stripe phase (o-AF). Extrapolating to higher pressure, we expect the former to become the dominant magnetic phase coexisting with superconductivity in $Ba_{1-x}K_xFe_2As_2$.

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