The phase diagram of iron-based superconductors of the BaFe$_2$As$_2$ 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. The AF order is a stripe-like spin-density wave, with a wavevector $Q = (\pi, 0)$ 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.

In Ba$_{1-x}$K$_x$Fe$_2$As$_2$, 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$_x$Fe$_2$As$_2$. 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. A tetragonal magnetic phase (t-AF) was then discovered in the closely related compound Ba$_{1-x}$Na$_x$Fe$_2$As$_2$, for $0.24 < x < 0.28$, by neutron and x-ray diffraction on powder samples. Subsequent neutron scattering on single crystals showed that in this t-AF phase the spins are aligned parallel to the c axis. A similar phase of tetragonal symmetry was then found in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ at ambient pressure, for $0.24 < x < 0.28$. The magnetic moments in the t-AF phase of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ are also oriented along the c axis. Infrared spectroscopy showed that the t-AF phase has a double-$Q$ magnetic structure as opposed to the single-$Q$ structure of the o-AF phase.

Several theoretical studies have investigated the properties of the tetragonal magnetic phase in iron-based superconductors. In this Article, we extend our prior study of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ under pressure, performed up to $x = 0.21$ 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$_x$Fe$_2$As$_2$ were grown from self flux. Three underdoped samples were measured, with a superconducting transition temperature $T_c = 20.8 \pm 0.5$ K, 25.4 $\pm$ 0.5 K, and 30.1 $\pm$ 0.5 K, respectively. Using the relation between $T_c$ and the nominal K concentration $x$ reported in ref. and wavelength-dispersive x-ray spectroscopy, we obtain $x = 0.22$, 0.24 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) equal to 91 $\pm$ 5 K and 79 $\pm$ 2 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. As before, 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 using a 50:50 mixture of n-pentane:isopentane. The pressure was measured via the superconducting transition of a lead wire inside the pressure cell. The electrical resistivity $\rho$ 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: $T_N$ 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 $T_N$. 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, $T_N$ 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 $P \approx 1.5$ 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 peak at $T_0$ stays sharp but its height decreases above $P \approx 1$ 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$_x$Fe$_2$As$_2$ (refs. [24] and Ba$_{1-x}$Na$_x$Fe$_2$As$_2$ [25,26] in agreement with the negative $dT_c/dP$ expected from the Ehrenfest relation applied to the thermodynamic data.

Temperature-concentration phase diagram.– Combining our present results with those of our previous study, we plot the temperature-concentration phase diagram of
Baticity of the electron pockets in the Fermi surface of \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) favours the t-AF phase because it changes the ellipsoidal concentration phase diagram of \( \text{Ba}\text{Fe}_2\text{As}_2 \). In other words, at high pressure the tetragonal phase grows by an order of magnitude at a small area below 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 \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \). 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 \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \).

**Summary.** In summary, we have shown that the new phase discovered in \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) from sharp signatures in the resistivity under pressure is the tetragonal antiferromagnetic phase observed and identified subsequently by various probes in both \( \text{Ba}_{1-x}\text{Na}_x\text{Fe}_2\text{As}_2 \) (refs. 5,6) and \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \). 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 \( T_c \).

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**FIG. 2:** Top: Temperature derivative of the resistivity of \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) 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 \( x = 0.24 \).

**FIG. 3:** Temperature-pressure phase diagram of \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) 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$_x$Fe$_2$As$_2$, 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. 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$_x$Fe$_2$As$_2$. 

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1 P. C. Canfield and S. L. Bud’ko, Annu. Rev. Condens. Matter Phys. 1, 27 (2010).
2 D. K. Pratt et al., Phys. Rev. Lett. 103, 087001 (2009).
3 S. Avci et al., Phys. Rev. B 85, 184507 (2012).
4 E. Hassinger et al., Phys. Rev. B 86, 140502(R) (2012).
5 S. Avci et al., Nat. Commun. 5, 3845 (2014).
6 F. Waßer et al., Phys. Rev. B 91, 060505 (2015).
7 A. Boehmer et al., Nat. Commun. 6, 7911 (2015).
8 B. P. Mallet et al., Phys. Rev. Lett. 115, 027003 (2015).
9 J. M. Allred et al., Phys. Rev. B 92, 094515 (2015).
10 J. Lorenzana et al., Phys. Rev. Lett. 101, 186402 (2008).
11 I. Eremin and A. V. Chubukov, Phys. Rev. B 81, 024511 (2010).
12 E. Berg et al., Phys. Rev. B 81, 172504 (2010).
13 G. Giovannetti et al., Nat. Commun. 2, 398 (2011).
14 P. M. R. Brydon et al., Phys. Rev. B 84, 214510 (2011).
15 J. Wang et al., Phys. Rev. B 91, 121104 (2015).
16 X. Kang et al., Phys. Rev. B 91, 024401 (2015).
17 M. N. Gastiasoro et al., arXiv:1502.05859 (2015).
18 R. M. Fernandes et al., arXiv:1504.03656 (2015).
19 J. M. Allred et al., arXiv:1505.06175 (2015).
20 H.-Q. Luo et al., Supercond. Sci. Technol. 21, 125014 (2008).
21 M. Tanatar et al., Phys. Rev. B 89, 144514 (2014).
22 Y. Liu et al. Phys. Rev. B 89, 134504 (2014).
23 I. R. Walker, Rev. Sci. Instrum. 70, 3402 (1999).
24 W. J. Duncan et al., J. Phys.: Condens. Matter 22, 052201 (2010).
25 S. Bud’ko et al., Phys. Rev. B 89, 014510 (2014).
26 S. Bud’ko et al., Phys. Rev. B 87, 100509(R) (2013).