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LETTERS

Unconventional superconductivity in $Ba_{0.6}K_{0.4}Fe_2As_2$ from inelastic neutron scattering

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A new family of superconductors containing layers of iron arsenide[1–3] has attracted considerable interest because of their high transition temperatures (T_c), some of which are >50 K, and because of similarities with the high-T_c copper oxide superconductors. In both the iron arsenides and the copper oxides, superconductivity arises when an antiferromagnetically ordered phase has been suppressed by chemical doping[4]. A universal feature of the copper oxide superconductors is the existence of a resonant magnetic excitation, localized in both energy and wavevector, within the superconducting phase[5–9]. This resonance, which has also been observed in several heavy-fermion superconductors[10–12], is predicted to occur when the sign of the superconducting energy gap takes opposite values on different parts of the Fermi surface[13], an unusual gap symmetry which implies that the electron pairing interaction is repulsive at short range[14]. Angle-resolved photoelectron spectroscopy shows no evidence of gap anisotropy in the iron arsenides, but such measurements are insensitive to the phase of the gap on separate parts of the Fermi surface[15]. Here we report inelastic neutron scattering observations of a magnetic resonance below T_c in $Ba_{0.6}K_{0.4}Fe_2As_2$, a phase-sensitive measurement demonstrating that the superconducting energy gap has unconventional symmetry in the iron arsenide superconductors.

Although the first iron arsenide superconductors were based on doped variants of *R*FeAsO, where *R* is a rare-earth element, there has been considerable interest in a new series of tetragonal compounds based on AFe_2As_2 (where *A* is barium, strontium or calcium), in which superconductivity is induced either by doping the *A* site with potassium or sodium[16,17] or by applying pressure[18]. These contain the same tetrahedrally coordinated Fe_2As_2 planes as the LaFeAsO compounds (Fig. 1), separated by planes of the doped *A* site, which act as a charge reservoir. So far, the maximum T_c observed is 38 K (ref. 16), which was seen in $Ba_{0.6}K_{0.4}Fe_2As_2$, the compound we are investigating in this Letter. The antiferromagnetic structure of the undoped parent compound, $BaFe_2As_2$, is illustrated in Fig. 1 (ref. 19).

Polycrystalline samples of $Ba_{0.6}K_{0.4}Fe_2As_2$ were prepared using solid-state synthesis techniques (see Supplementary Information for details). A sharp superconducting transition was observed in magnetic susceptibility at the previously reported temperature of 38 K (ref. 16), and neutron powder diffraction showed no evidence of any impurity phase within the statistical precision of the Rietveld refinement. The inelastic neutron scattering experiments were performed on the recently commissioned time-of-flight MERLIN spectrometer[20] at the ISIS Pulsed Neutron and Muon Facility, UK, using incident energies of 15, 30, 60 and 100 meV. The data were placed on an absolute intensity scale by normalization to a vanadium standard.

Figure 2 shows colour plots of the measured inelastic neutron scattering intensity as a function of momentum transfer, Q (expressed in units of reciprocal length), and energy transfer, ω, below ($T = 7$ K) and above ($T = 50$ K) the superconducting transition temperature. The most striking difference between the two temperatures is seen at $Q \approx 1.15$ Å^{-1} and an energy transfer of ~14 meV. At 7 K, there is clearly a peak that is well defined in both Q and ω and that is not present at 50 K. The value of Q characterizing this contribution to the magnetic response corresponds to the periodicity of the antiferromagnetic order within each plane of iron spins observed in the undoped parent compound, $BaFe_2As_2$ (ref. 19; see also Fig. 1).

To elucidate the evolution of the magnetic response, we combine measurements below T_c at three incident energies, in each case using a low-energy cut-off that excludes the tail of strong elastic nuclear scattering. The resulting data are shown in Fig. 3a, where the resonant excitation is seen to peak sharply at $\omega \approx 14$ meV. We cannot rule out the possibility that there could be a small phononic contribution to the scattering within this energy window, but the strong temperature dependence indicates that it is predominantly magnetic. Figure 3b

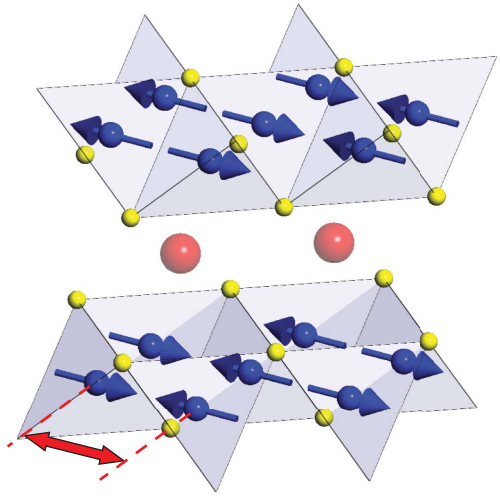

Figure 1 | The crystal structure of $Ba_{0.6}K_{0.4}Fe_2As_2$. The unit cell of $Ba_{0.6}K_{0.4}Fe_2As_2$ contains two layers of Fe_2As_2 tetrahedra (Fe, blue spheres; As, yellow spheres), separated by planes of barium or potassium atoms (red spheres). The blue arrows show the ordering of the iron spins observed in the undoped parent compound $BaFe_2As_2$ (ref. 19). The atomic distance of 2.77 Å that characterizes both the antiferromagnetic modulation and the newly observed resonant excitation is indicated by the red double-headed arrow.

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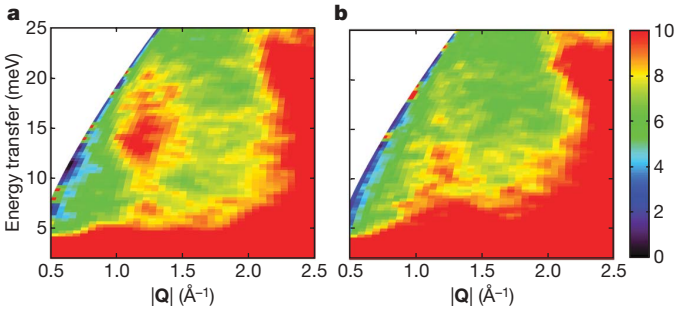

Figure 2 | Resonant spin excitation in $Ba_{0.6}K_{0.4}Fe_2As_2$. Inelastic neutron scattering, measured using an incident neutron energy of 60 meV at temperatures below (**a**, 7 K) and above (**b**, 50 K) T_c, shows the development of a magnetic excitation in the superconducting phase at an energy transfer of 14 meV and a momentum transfer of 1.15 Å^{-1}. The strong scattering at low energy transfers arises from the tail of strong elastic nuclear scattering, and the strong increase in scattering at higher values of Q is due to inelastic phonon scattering. The colour scale indicates scattering intensity in units of millibarns per steradian per millielectronvolt per mole.

shows that spectral weight in the resonant excitation seen below T_c is transferred to lower energy above T_c.

We performed a series of shorter measurements to determine the temperature dependence of this resonant excitation. Figure 4 shows data integrated over the (Q, ω) region of maximum intensity in the resonant excitation. As also observed in the copper oxide superconductors, the intensity of the resonance falls to zero at T_c, confirming the strong coupling of this excitation to the superconducting order parameter.

Similar resonant excitations have been observed in other strongly correlated superconductors, such as high-T_c copper oxide[5–9] and heavy-fermion superconductors[10–12], where they are commonly taken as evidence of an unconventional symmetry of the superconducting order parameter[13]. Below the superconducting transition temperature, the dynamic magnetic susceptibility is predicted to be enhanced at certain values of $\mathbf{Q}$ by a coherence factor, provided that the energy gap has the form $\Delta_{\mathbf{k}+\mathbf{Q}} = -\Delta_{\mathbf{k}}$ (here $\mathbf{k}$ and $\mathbf{k}+\mathbf{Q}$ are wavevectors on different parts of the Fermi surface).

In the copper oxide and heavy-fermion superconductors, this form results from $d_{x^2-y^2}$ symmetry, which has nodes in the energy gap within a single Fermi surface. In these cases, $\mathbf{Q}$ spans sections of the same Fermi surface that are gapped with opposite phases; in the copper oxide superconductors, $\mathbf{Q} = (\pi, \pi)$ is such a wavevector. However, this mechanism seems to be ruled out by results of angle-resolved photoemission spectroscopy (ARPES) of $Ba_{0.6}K_{0.4}Fe_2As_2$ that show no evidence of any anisotropy of the energy gap[15]. According to band structure calculations, the Fermi surfaces of the iron arsenide superconductors are predominantly derived from the iron d electrons, and comprise two small hole pockets centred at the centre of the Brillouin zone and two small electron pockets at the zone boundary[21,22]. ARPES shows there to be isotropic gaps around each of the measured surfaces, apparently ruling out a d-wave gap symmetry[15].

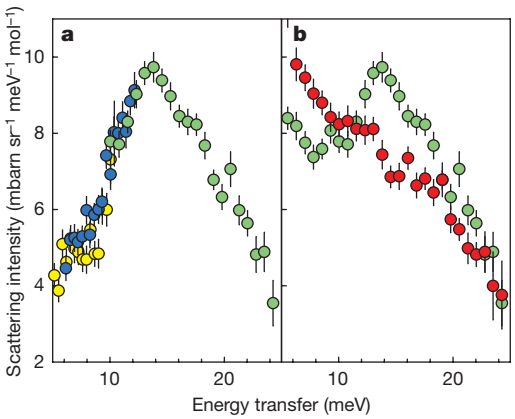

Figure 3 | Energy dependence of the resonant spin excitation. a, The inelastic neutron scattering intensity from $Ba_{0.6}K_{0.4}Fe_2As_2$ integrated over Q in the range 1.0–1.3 Å^{-1} at 7 K, measured using incident neutron energies of 15 meV (yellow circles), 30 meV (blue circles) and 60 meV (green circles). **b**, Same as in **a**, but at 7 K (green circles) and 50 K (red circles) using an incident neutron energy of 60 meV. The error bars are derived from the square root of the raw detector counts. The data show a resonant peak at 7 K and the transfer of spectral weight from this peak to lower energies at 50 K, that is, above T_c.

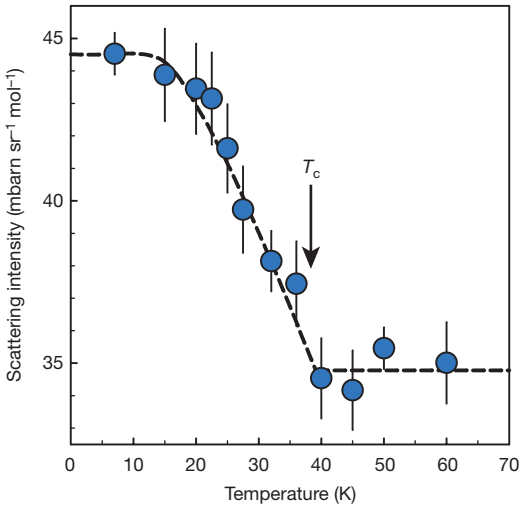

Figure 4 | Temperature dependence of the resonant spin excitation. The inelastic neutron scattering intensity from $Ba_{0.6}K_{0.4}Fe_2As_2$ integrated over Q in the range 1.0–1.3 Å^{-1} and over ω in the range 12.5–17.5 meV. The integration range corresponds to the region of maximum intensity of the resonant excitation observed below T_c (Fig. 2). The error bars are derived from the square root of the raw detector counts. The dashed line is a guide to the eye below T_c and shows the average value of the integrals above T_c.

A resolution of this apparent discrepancy has been provided by theoretical predictions that the symmetry is not d wave, but rather extended $s_{\pm}$ wave[14], in which the gaps at the hole pockets are isotropic and the gaps at the electron pockets are isotropic but are of opposite sign to those at the hole pockets. This means that magnetic fluctuations are amplified by the coherence factor at values of $\mathbf{Q}$ that couple the hole and electron pockets, as has been confirmed by explicit calculations of the neutron scattering intensities[23,24]. This is precisely where we have observed the resonant excitation, so our measurements, in conjunction with the ARPES data, provide phase-sensitive evidence for the validity of extended $s_{\pm}$-wave gap models.

The energy of this resonant excitation is $\omega_0 \approx 14$ meV. This is equivalent to $4.3T_c$, which is just less than the canonical value of $5T_c$ seen in the copper oxide superconductors[25]. However, it is more appropriate to consider the ratio $\omega_0/2\Delta_0$, where Δ_0 is the maximum value of the gap. This ratio has values ranging from 0.62 to 0.74 in a wide range of materials[11]. From ARPES data on $Ba_{0.6}K_{0.4}Fe_2As_2$, $\Delta_0 \approx 12$ meV (ref. 15), giving a ratio of $\omega_0/2\Delta_0 \approx 0.58$. It is remarkable that materials with such a divergent range of T_c (which varies over two orders of magnitude) can be unified by such a simple scaling relation.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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