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FAST TRACK COMMUNICATION

Superconductivity up to 29 K in $SrFe_2As_2$ and $BaFe_2As_2$ at high pressures

Patricia L Alireza, Y T Chris Ko, Jack Gillett, Chiara M Petrone, Jacqueline M Cole, Gilbert G Lonzarich and Suchitra E Sebastian[1]

Department of Physics, University of Cambridge, Cavendish Laboratory, J J Thomson Avenue,
Cambridge CB3 0HE, UK

E-mail: suchitra@phy.cam.ac.uk

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Abstract
We report the discovery of superconductivity at high pressure in $SrFe_2As_2$ and $BaFe_2As_2$. The superconducting transition temperatures are up to 27 K in $SrFe_2As_2$ and 29 K in $BaFe_2As_2$, the highest obtained for materials with pressure-induced superconductivity thus far.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recently a new class of high-temperature superconductors has been discovered based on iron arsenide layered structures. As in the cuprates and other examples of unconventional superconductivity, the parent compounds tend to be antiferromagnetic and superconductivity emerges under chemical doping or in some cases at high pressure once antiferromagnetism is suppressed.

The magnetic parent compounds of the iron arsenide family of superconductors crystallize in tetragonal structures, with LaFeAsO [1] forming in the tetragonal ZrCuSiAs structure, and AFe_2As_2 (A = Ba, Sr, Ca) forming in the more familiar tetragonal $ThCr_2Si_2$ structure (figure 1). The spin and charge transitions exhibited by the AFe_2As_2 materials at ambient pressure are suppressed by hole doping, leading to superconductivity with a transition temperature T_{sc} as high as 38 K [2]. The application of pressure has also been shown to suppress the spin and charge transitions in these materials [3–5]. Here we report high pressure measurements that reveal a superconducting dome in $SrFe_2As_2$ and $BaFe_2As_2$ with maximum T_{sc} of approximately 30 K. This constitutes the highest pressure-induced observation of superconductivity in any material thus far, to the best of our knowledge.

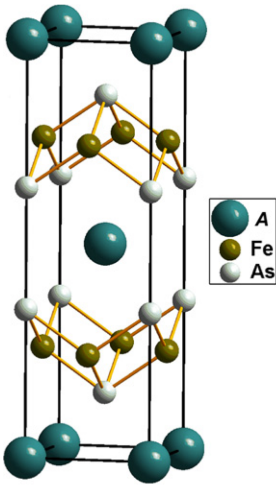

Figure 1. Nominal parent crystal structure of AFe_2As_2 (A = Ba, Sr, Ca), of the $ThCr_2Si_2$ type.

2. Experimental details and results

Single crystals of $BaFe_2As_2$ and $SrFe_2As_2$ were prepared by the flux growth technique [6, 7] and starting elements of greater than 99.99% purity. $SrFe_2As_2$ crystals were grown using Fe:As:Sn flux, and $BaFe_2As_2$ using FeAs flux. Crystals were characterized by x-ray diffraction, electron beam microprobe analysis and measurement of the temperature dependences of

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[1] Author to whom any correspondence should be addressed.

Table 1. Unit cell parameters for alternative structural possibilities of $SrFe_2As_2$ above and below the Neel temperature. The least-squares fit figures of merit represent the extent to which crystallographic systematic absences are observed in the data given each crystal system model; a value of zero corresponds to a perfect fit.

Bravais lattice	a (Å)	b (Å)	c (Å)	α (deg)	β (deg)	γ (deg)	Least-squares fit of data to unit cell
$T = 220$ K							
Tetragonal (I)	3.86	3.86	12.10	90	90	90	2.18
Orthorhombic (I)	3.84	3.97	12.16	90	90	90	1.51
Orthorhombic (F)	5.43	5.54	12.11	90	90	90	1.79
Monoclinic (C)	12.69	3.90	3.90	90	106.42	90	0.72
$T = 155$ K							
Tetragonal (I)	3.83	3.82	12.14	90	90	90	0.29
Orthorhombic (I)	3.81	3.84	12.14	90	90	90	0.09
Orthorhombic (F)	5.40	5.41	12.14	90	90	90	0.27
Monoclinic (C)	12.71	3.83	3.81	90	107.35	90	0.09

the magnetic susceptibility and electrical resistivity. Notably, while all single crystals were stoichiometric within limits of resolution of electron probe microanalysis (Sn inclusion in $SrFe_2As_2$ single crystals was no higher than 0.3%), not all samples show superconductivity in the reported pressure range. Superconductivity was favoured in $SrFe_2As_2$ single crystals grown out of an Fe-rich flux, although there was no detectable deviation from stoichiometry.

Single-crystal x-ray diffraction was used to obtain possible clues as to the sample dependence of superconductivity under pressure. A 300 μm × 275 μm × 60 μm crystal of $SrFe_2As_2$ that showed superconductivity under pressure was mounted onto a Rigaku SCXmini diffractometer, equipped with an Oxford Cryosystems Nitrogen cryostream. Unit cell parameters were determined above and below the Neel temperature, at $T = 220$ K and 155 K, respectively. Results, given in table 1, show that the structure is consistent with previous reports where there is much discussion as to the exact nature of its body-centred tetragonal, face-centred orthorhombic or possibly C-centred monoclinic characteristics [8–11]. Significant structural features appear in the diffraction patterns—of particular interest is the recurrent characteristic v-shaped diffuse scattering signature throughout reciprocal-space, illustrated in figure 2. This reveals that two-dimensional disorder is present in the three-dimensional crystal structure in some form. The two most likely origins of this disorder are (i) substantial defects in the *ab* crystallographic plane, or (ii) twinning. Indeed, table 1 reveals tell-tale signs of a classical form of twinning: the fact that $a \approx b$ in any of the possible options listed, and that a and b in the body-centred tetragonal and face-centred orthorhombic options are related by a factor of $\sqrt{2}$. The exact nature of the three-dimensional crystal structure of $SrFe_2As_2$ with relation to the appearance of superconductivity under pressure is the subject of on-going work.

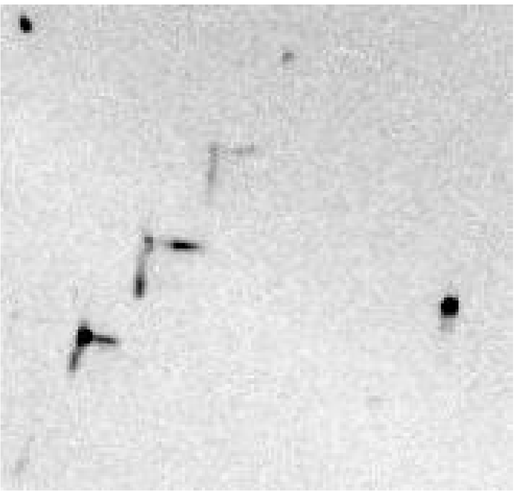

Figure 2. An x-ray diffraction pattern showing characteristic diffuse scattering signatures in $SrFe_2As_2$, in reciprocal space, indicating two-dimensional disorder in the *ab* plane.

Superconducting transitions were detected by means of a miniature diamond anvil cell (the L-A cell- [12]) with ultra-low background magnetic susceptibility designed for use with a SQUID magnetometer, i.e., the magnetic properties measurement system made by Quantum Design (details of experimental technique are in [12]). The pressure transmitting medium is Daphne Oil 7373 and the pressure is measured at room temperature by means of ruby fluorescence before and after each cool down. The change in pressure on cooling to 5 K, the base temperature of our study, has been checked using the known pressure dependence of T_{sc} of a Pb sample and is typically less than 3 kbar in our experiments. A superconducting anomaly is clearly visible upon cooling as a change in the magnetic moment of the sample as a function of decreasing temperature in the presence of an applied magnetic field of 50 G (shown in figure 3). Superconducting anomalies have also been measured using an ac field modulation technique with a detection microcoil mounted around the sample inside the sample space of a moissanite anvil cell (details of experimental technique are in [13]). The pressure transmitting medium and the method to determine the pressure is the same as for DC magnetization measurements. In a narrow pressure range close to 40 kbar, the size of magnetic moment screening below T_{sc} is close to that expected if superconductivity exists throughout the sample (figures 3 and 4). Bulk superconductivity is inferred

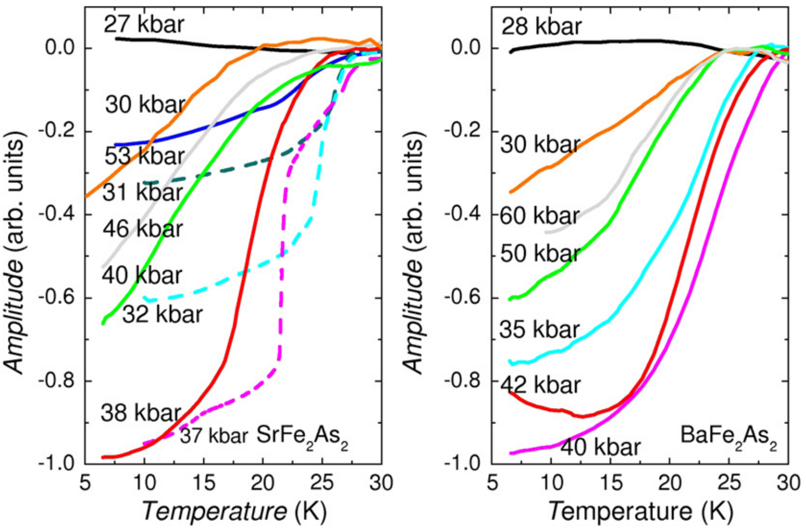

Figure 3. SQUID magnetization data (solid lines) and AC susceptibility data (dashed lines), measured on plate-like samples approximately 200 μm × 200 μm × 60 μm with the c-axis normal to the plate face. Shielding is observed in the pressure range ~28–60 kbar, consistent with bulk superconductivity within a more narrow pressure range around 40 kbar in both materials. Data shown for (a) SrFe$_2$As$_2$ and (b) BaFe$_2$As$_2$, with the magnetic field (50 G in the case of DC magnetization, and 10 G in the case of AC susceptibility) applied parallel to the c-axis. Details of the experimental technique are in [12, 13].

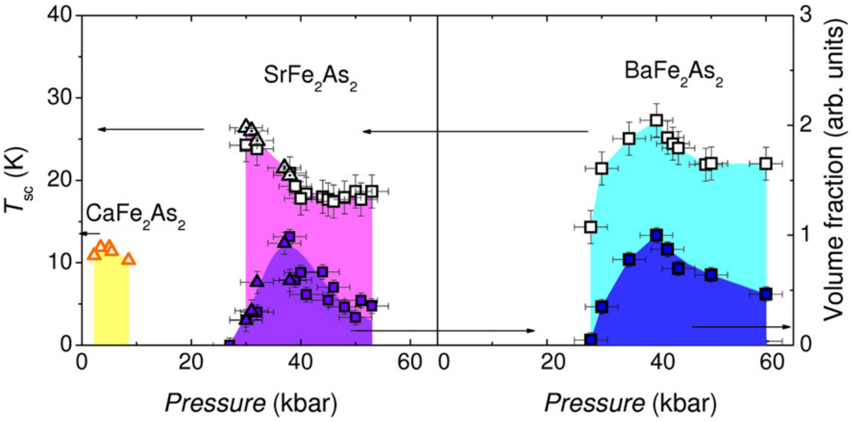

Figure 4. The superconducting transition temperature and superconducting volume fraction of AFe$_2$As$_2$ (A = Sr, Ba) as a function of pressure. The white squares (triangles) show the critical temperature measured by SQUID magnetization and (AC susceptometry). The filled squares (triangles) show the volume fraction measured by SQUID magnetization (AC susceptometry). The superconducting dome for CaFe$_2$As$_2$ is taken from [3]. SrFe$_2$As$_2$ reveals a maximum critical temperature of ~27 K, while that of BaFe$_2$As$_2$ is ~29 K. Curiously, while superconductivity in BaFe$_2$As$_2$ appears more gradually with increasing pressure, the onset of superconductivity in SrFe$_2$As$_2$ occurs abruptly, accompanied by a maximum in superconducting temperature. Surprising also is the narrow pressure range at which bulk superconductivity is almost complete in both materials.

by comparing the measured signal with that of a Pb sample of similar size and shape.

Figure 4 shows the measured pressure dependences of T_{sc} in BaFe$_2$As$_2$ and SrFe$_2$As$_2$. T_{sc} denotes the temperature at which 25% of the total drop in signal from shielding is reached. We see that the superconducting dome in BaFe$_2$As$_2$ is relatively broad, extending between about 28 to 60 kbar and has a peak of $T_{sc} \cong 29$ K near 40 kbar. In contrast, the superconducting dome is narrower in SrFe$_2$As$_2$, showing a peak of $T_{sc} \cong 27$ K near 30 kbar and a curiously sharp onset around 28 kbar. Interestingly, in both materials, the volume fraction of superconductivity is near complete only over a very narrow pressure range, with the superconducting dome bounded by a decrease in superconducting volume fraction.

3. Discussion

An interesting finding is the decrease in the peak of T_{sc} on reducing the ionic size of A in AFe$_2$As$_2$ from Ba and Sr to the isoelectric element Ca (figure 4). Also striking is the appreciably broader dome of superconductivity in BaFe$_2$As$_2$ as compared to the Ca and Sr analogues. This increase in the height of the superconducting dome in going from CaFe$_2$As$_2$ to SrFe$_2$As$_2$ and BaFe$_2$As$_2$ may be connected in part with the degree of abruptness with which the spin and charge transitions are suppressed with pressure. Evidence suggests that these transitions are more strongly discontinuous in CaFe$_2$As$_2$ than in BaFe$_2$As$_2$ [14]. The sharper discontinuity may lead effectively to a truncation of the superconducting dome and potentially therefore a reduction in the peak value of T_{sc} in $CaFe_2As_2$ compared with $BaFe_2As_2$. While pressure-induced superconductivity is now ubiquitous in various families of materials, the AFe_2As_2 class of materials are unique in manifestation of a strongly varying volume fraction of bulk superconductivity within the superconducting dome, while the superconducting temperature remains fairly high. Further experiments will assist in revealing whether the variation in volume fraction reflects either a macroscopic or microscopic inhomogeneity, perhaps reflecting structural twinning effects or coexisting order parameters. The peak superconducting transition temperatures in $BaFe_2As_2$ and $SrFe_2As_2$ are the highest to be induced thus far in a non-superconducting material by the application of pressure. Pressure is thus seen to be a powerful tuning parameter in engineering materials properties.

Acknowledgments

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