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Signatures of filamentary superconductivity in antiferromagnetic $BaFe_2As_2$ single crystals

D MOSELEY[1], K A YATES [1], W R BRANFORD[1], A S SEFAT[2], D MANDRUS[2,3], S J STUARD[4], S SALEM-SUGUI[4], L GHIVELDER[3] AND L F COHEN[1(a)]

[1] *Blackett Laboratory, Imperial College, London, SW7 2AZ, UK*
[2] *Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA*
[3] *Department of Materials Science and Engineering,University of Tennessee, Knoxville, TN 37996, USA*
[4] *Physics Department, Federal University of Rio de Janeiro, Rio de Janeiro, Brazil*

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Abstract – In this paper, we present ac susceptibility and magnetotransport measurements on aged single crystals of the ferropnictide parent compound, $BaFe_2As_2$ with a paramagnetic to antiferromagnetic transition temperature of 134 K. The ac susceptibility shows the clear onset of a partial diamagnetic response with an onset temperature, commensurate with a subtle downturn in resistivity at approximately 20K. Below 20K the magnetotransport shows in plane anisotropy, magnetic field history dependence and a hysteretic signature. Above 20K the crystals show the widely reported high field linear magnetoresistance. Between 20K and 40K an enhanced noise signature in ac susceptibility is observed for large ac amplitudes, reminiscent of Barkhausen noise associated with domain wall movement in magnetic materials. The hysteresis in magnetoresistance and the observed sensitivity of the superconducting phase to the amplitude of the ac signal are indicative characteristics of granular or weakly linked filamentary superconductivity. These features taken together with the observed noise signature above T_c suggests a link between the formation of the superconducting filamentary phase and the freezing of antiphase domain walls, known to exist in these materials.

Introduction. – The ferropnictides are a family of materials in which electrical doping or applied pressure can induce a superconducting state [1,2,3]. The parent ferropnictides are antiferromagnetic with a structural transition (from high temperature tetragonal to low temperature orthorhombic) occurring close to the Néel temperature, T_N. For $BaFe_2As_2$ this coupled structural /antiferromagnetic transition occurs at around 134-138 K [2]. At the magnetic transition, a spin-density wave (SDW) striped magnetic state is produced, with the spins aligned antiparallel along the *a*-axis direction and parallel in *b*-axis direction[4].

Although the parent compounds do not develop a fully superconducting (SC) state, early transport studies reported a reduced resistance response under ambient conditions in stoichiometric crystals indicative of SC at low temperatures [5]. Initially ascribed to internal crystallographic strain, it has now been shown that for the 122 [6,7,8,9], 111 [10,11] and 11 [12,13] families, air exposure can lead to the creation of a superconducting phase. The SC derived from this mechanism is reported to be inhomogeneous, with a superconducting fraction reported to be of the order of 10% [5]. The precise geometric arrangement of the SC fraction has not been established, though it has been suggested that the distribution is filamentary.

It has been widely discussed that two types of magnetic domain walls exist in the pnictides below the Néel temperature, twin boundaries and antiphase domain walls (APDWs) [7]. APDWs are known to exist in the undoped ferropnictides [14] and unlike twin boundaries, APDWs are considered mobile at high temperatures [4] and freeze in position as the temperature is reduced [8]. It has been suggested that the filamentary superconductivity nucleates on these immobile APDWs although evidence is limited, [7,8] or that a spin-glass like AFM state coexists with superconductivity along domain walls [15].

The fundamental relationship in the pnictides between magnetism and superconductivity is certainly worthy of further exploration and it is possible that the superconductivity that arises in aged crystals might hold some

(a) E-mail: l.cohen@imperial.ac.uk

interesting clues. Accordingly, in this paper, we present resistivity vs. temperature (ρ_n vs. T), magnetoresistance (MR) and ac susceptibility ($\chi(T)$) measurements on micromechanically cleaved single crystals of $BaFe_2As_2$ which have been aged and exposed to air repeatedly over a period of two years. As with other studies [7] we observe sub-20 K variations in ρ_n vs. T. In this temperature range the $\chi(T)$ also shows a partial diamagnetic response which we ascribe to the development of an inhomogeneous SC state. In addition, for each crystal, but in only one of the two possible MR Van der Pauw (VDP) configurations, we observe a clear hysteresis in magnetic field. The hysteresis displays a dependence on magnetic field history. This behaviour shows similarities to previous studies of the magnetotransport properties of granular superconductors [16]. We also observe a reproducible noise signature in $\chi(T)$ at $20 \leq T \leq 40K$ which is reminiscent of Barkhausen noise. We propose that the hysteresis in magnetoresistance, the sensitivity of the superconducting phase to the amplitude of the ac signal combined with the noise signature in the ac susceptibility above T_c support the scenario that filamentary superconductivity is indeed forming at antiphase domain walls as they freeze.

Experimental Details. – Large single crystals of $BaFe_2As_2$ grown employing the self-flux method [17], were cleaved using a micromechanical exfoliation technique. We consider four crystals derived from the same batch of crystals but from different single crystals within the batch. The single crystals were stored in a desiccator for approximately 2 years. Sample sizes were of the order of millimetres in the *ab*-plane and tens of micrometers along the *c*-axis. All measurements were taken using the VDP technique [18] with the magnetic field applied along the *c*-axis and current within the *ab*-plane, as shown in the inset of Fig. 1 b). Only one of the MR configurations displays hysteretic magnetotransport signature and this configuration is labelled R_1 in all crystals. The MR Ratio is defined as $[R(\mu_0 H) - R(0T)]/R(0T)$, where $R(0T)$ is the zero field state after magnetic ramping at the previous temperature. The thickness of the samples was determined from the mass of the crystals by measuring the surface area of the crystal faces and using the experimentally determined mass density [19]. The real (in phase) and imaginary (out of phase) parts of the ac susceptibility were measured with a commercial magnetometer (Quantum Design PPMS). The ac field could be varied from 0.005 to 1.5 mT; lower ac fields require a higher ac frequency, up to 10 kHz, because of the small signal to noise ratio. The measurements were performed with or without a superimposed dc field, up to 9 T after cooling to the set temperature in a zero field.

Results and Discussion. – In Fig. 1 a, the ρ_n vs. T response for several of the crystals is shown. The temperature dependence of the resistivity is typical for Ba122 crystals [3]. The location of the T_N has been considered a hallmark of the quality of a sample, with values of T_N = 138 K observed in annealed crystals [20]. We observe T_N = 134 K suggesting that the crystal quality maybe somewhat impaired due to aging.

In Fig. 1 b, we show the low temperature ρ_n data in more detail. The results in this study are compared with data from a number of other studies on aged crystals by plotting the resistivity normalized to the resistivity at 30 K. A variety of behaviours are observed at low temperatures in the ferropnictides ranging from an upturn in resistance to a down turn or even a zero resistance state in some cases [9]. The crystals considered in this work, show either a slight downturn, or a small upturn at 20K. The features are subtle, and as with many similar reported features in the literature, not obviously associated with superconductivity.

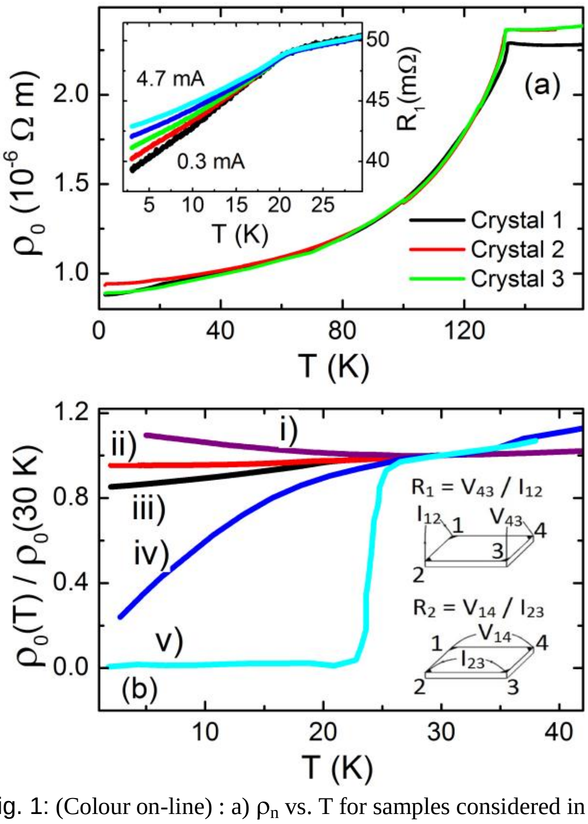

Fig. 1: (Colour on-line) : a) ρ_n vs. T for samples considered in this work. Inset: Current dependence of the low temperature resistance for Sample 1, from top to bottom at 4.7, 2.4, 1.2, 0.6, 0.3 mA. b) Low temperature $\rho_n(T)/\rho_n(30K)$ comparison with literature data: i) polycrystalline LaFeAsO [21], ii) Crystal 3 this work iii) Crystal 1 this work, iv) single crystal $SrFe_2As_2$ after 6 months in desiccator [9], and v) single crystal $SrFe_2As_2$ after 9 months in desiccator [9]. Inset: Schematic of the Van der Pauw measurement configuration.

To further explore the low temperature behaviour, the current dependence of the low temperature R_1 (T) was considered in Crystal 1, (inset of Fig. 1 a). Below 20 K, the low temperature resistance downturn is suppressed with increasing current while above 20K no current dependence is observed. Such a current dependence has been previously tied to the inhomogeneous SC model in granular superconductors [22].

Fig. 2 shows the temperature dependence of the real (χ') and imaginary (χ") parts of the ac mass magnetic susceptibility, X, for crystal 4 using an ac frequency of 10 kHz, zero DC magnetic field and an ac amplitude of 0.1mT. At around 20 K, the onset of diamagnetism is observed. Figure 3 shows the dependence of χ' on the amplitude of the ac field up to 1.5mT (in figure (a)) and on the dc field up to 9T (in figure 3b). Although sensitivity of the diamagnetic response to the amplitude of the ac field has been reported for granular superconductors [23], the inconsistency between the dc and ac response suggests a more complex scenario here. It is clear that the diamagnetic screening response is much less sensitive to the application of a dc field, with a dc field of greater than 100mT needed to produce equivalent results to a 0.3 mT ac field. The value of dc field required to suppress the diamagnetic signal is consistent with the upper critical field observed in the filamentary superconducting state found in Ca122 crystals [7] suggesting that it is not directly related to the suppression of inter-filamentary screening.

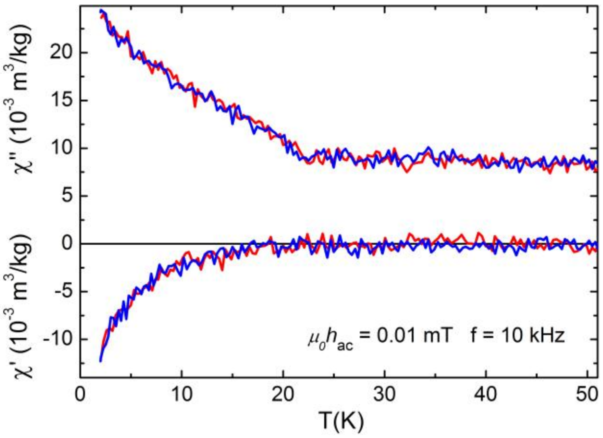

Fig. 2: (Colour on-line): Real and imaginary parts of the AC mass magnetic susceptibility on Crystal 4. Two separate sets of measurements are shown indicated by the blue and red curves.

Taken together, the resistivity and susceptibility measurements are suggestive of a form of inhomogeneous superconductivity developing at low temperatures in aged parent Ba122 crystals. Moreover, the current dependence of ρ(T) and ac field dependence of χ(T) are indicative of granular superconductivity. In granular superconductors, a matrix of superconducting grains are connected by Josephson junction type weak links. With increasing measurement current, the critical current of some of the superconducting grains may be exceeded, decreasing the superconducting fraction and effectively increasing the measured resistance. It is also possible that the observed current dependence of the resistivity could be due to the self-field produced by the current through micron sized superconducting filaments.

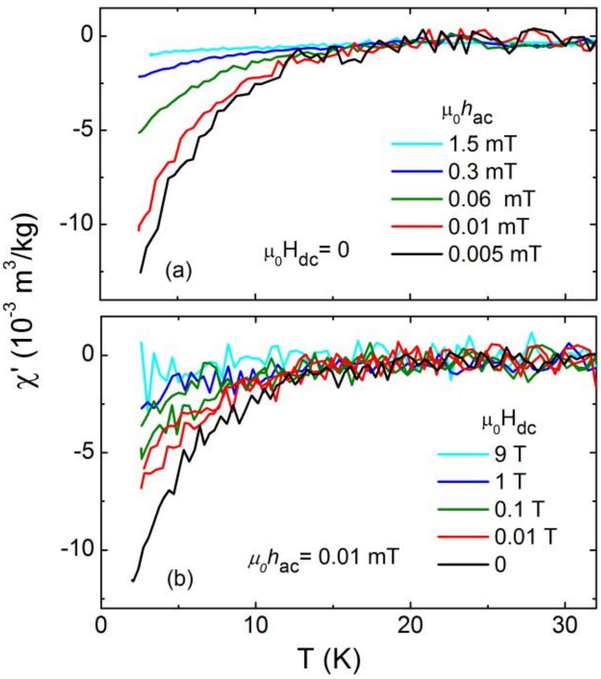

Fig 3: a) Dependence of χ'(T) on increasing h_{ac}. b) Dependence of χ'(T) on increasing H_{dc}.

An explanation based on granular superconductivity is also consistent with the magnetic history dependence observed for the magnetoresistance shown in figure 4. To study the magnetic history dependence, the sample was warmed to 25 K then cooled to the required temperature followed by magnetic sweeping. The initial field sweep for crystal 1 shows a sharp positive MR followed by a more gradual increase to the highest field measured. Crystal 3 by contrast shows a decrease to a minimum MR at ~0.3 T followed by an increasing MR until the highest fields measured. Both crystals show a different resistance on decreasing magnetic field (path 2). On increasing the field again, the resistance does not re-trace path 1 but forms a new path (path 3) until a field (B_{rev}) greater than approximately 1.5 T whereupon the magnetoresistance appears to once more become reversible.

The magnetoresistance appears to be defined by at least two characteristic fields. A field of maximum resistance difference (B_{hys}) and a field above which the behaviour once more becomes reversible, (B_{rev}). If the virgin field sweep data is attributable to a form of filamentary superconductivity in the crystals, it suggests that either the filamentary distribution of the superconducting regions are irreversibly altered by the application of a magnetic field or that there is irreversible flux pinning produced by minor hysteresis loop field cycling.

The location of B_{hys} has been described in terms of the magnetization of a granular material by Balaaev et al., [16]. It is shown more clearly in the inset to figure 4a which shows the full MR curve for crystal 3 at 2K. Hysteresis in R(H) has previously been observed in grain-aligned bulk cuprate superconductors [16,23,24] and an explanation in terms of grain boundary weak links [21,26] or intragranular flux pinning [16] both appear plausible. We find that B_{hys} is extremely similar for all samples and persists to around 15 K (inset to fig 4b). This demonstrates that a similar process occurs in all crystals studied and links the magnetic history and the hysteresis to the same phenomenon.

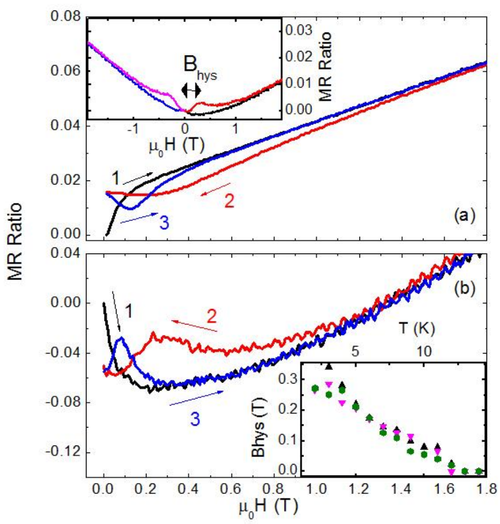

Fig 4: a) Unidirectional sweeping at 2 K for crystal 1 showing magnetic history dependence. Inset shows the full magnetic hysteresis data for crystal 3 at 2K defining B_{hys}. (b) Unidirectional sweeping for crystal 3 at 2K, inset shows B_{hys} as a function of temperature for samples 1 (▲), 2 (▼), 3 (●).

In contrast to B_{hys}, the field B_{rev}, could either be associated with the upper critical field of the superconducting filaments, or the critical field associated with the connectivity of the filaments. Alternatively, it could be associated with the field at which the antiphase domain walls become de-pinned. Certainly B_{rev} appears correlated with the dc fields required to quench the ac susceptibility diamagnetic screening signature (figure 3b). Interestingly, the explanation based on the movement of the APDWs is consistent with a curious observation above T_c in the ac susceptibility data. Figure 5 shows that we observe the surprising onset of a regime of noisy behaviour which starts rather abruptly at around 40K and stops at the temperature where the diamagnetic screening begins, around 20K. This signature is observed most clearly for the largest ac amplitude used (1.5mT) and is manifest in both the real and imaginary components of X. The observations are completely reproducible. The noise signature resembles that of Barkhausen noise found in magnetic media due to discontinuous jumps in magnetization due to domain wall movement. We rule out the suggestion that this feature is related to superconducting fluctuations above T_c. Such fluctuations may survive up to $1.4T_c$ when the superconductivity exists in reduced dimensions [25] and have been predicted to occur above the superconducting critical temperature in these materials [26]. However, we think this scenario is unlikely because of the extended temperature range of our observations and the relatively low frequency of our measurement. We suggest instead that this noise characteristic is a feature associated with the freezing of the APDW, which, at high ac amplitude, can be oscillated by the ac field. Once the APDW freezes, the noise in $\chi(T)$ decreases and superconductivity is formed. It has been suggested that the superconductivity forms along the APDW boundaries [8]. We speculate that the rapid suppression of the diamagnetic signature in our samples at high ac amplitude could be related to the onset of movement of the APDW.

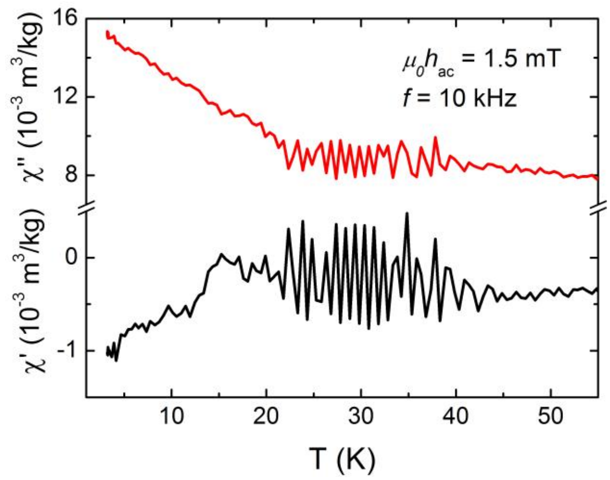

Fig 5: ac mass magnetic susceptibility using $\mu_0 h_{ac}$ = 1.5 mT as a function of temperature on warming from low temperatures, showing noise feature between 20 to 40 K.

In summary, we have studied single crystals of stoichiometric $BaFe_2As_2$ that have been stored in a desiccator for two years. The zero field resistivity and ac susceptibility taken together suggest that a form of inhomogeneous superconductivity develops at around 20K. The hysteresis in magnetoresistance, magnetic field history and current dependence of the zero field resistivity are all consistent with a granular type of inhomogeneity and possibly the filamentary sort that has been suggested for the pnictides [5,7]. Between 20K and 40K we observe a region of enhanced ac noise in $\chi(T)$ which is intriguing. We propose that this is associated with the freezing of antiphase domain boundaries. If the superconductivity is nucleated on frozen APDWs, moving them would cause the superconducting signature to be suppressed as the superconductivity itself is destroyed. The association of the freezing of APDW with the onset of superconductivity has been discussed extensively [7] using NMR and resistivity. Certainly the suppression of the enhanced noise signature in the ac χ, commensurate with the onset of superconducting screening which is the most significant new result here appears consistent with this scenario.

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