Spin-wave excitations and superconducting resonant mode in Cs$_x$Fe$_{2-y}$Se$_2$

A. E. Taylor,$^1$ R. A. Ewings,$^2$ T. G. Perring,$^{2,3}$ J. S. White,$^{4,5}$ P. Babkevich,$^5$
A. Krzton-Maziopa,$^{6,7}$ E. Pomjakushina,$^6$ K. Conder,$^6$ and A. T. Boothroyd$^1$

$^1$Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford, OX1 3PU, United Kingdom
$^2$ISIS Facility, Rutherford Appleton Laboratory, STFC, Chilton, Didcot, Oxford, OX11 0QX, United Kingdom
$^3$Department of Physics and Astronomy, University College London, Gower Street, London, WC1E 6BT, United Kingdom
$^4$Laboratory for Neutron Scattering, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
$^5$Laboratory for Quantum Magnetism, Ecole Polytechnique Fédérale de Lausanne (EPFL), 1015 Lausanne, Switzerland
$^6$Laboratory for Developments and Methods, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
$^7$Faculty of Chemistry, Warsaw University of Technology, PL-02006 Warsaw, Poland

We report neutron inelastic scattering measurements on the normal and superconducting states of single-crystalline Cs$_{0.8}$Fe$_1.0$Se$_2$. Consistent with previous measurements on Rb$_x$Fe$_{2-y}$Se$_2$, we observe two distinct spin excitation signals: (i) spin-wave excitations characteristic of the block antiferromagnetic order found in insulating $A_x$Fe$_{2-y}$Se$_2$ compounds, and (ii) a resonance-like magnetic peak localized in energy at 11 meV and at an in-plane wave vector of $(0.25, 0.5)$. The resonance peak increases below $T_c = 27$ K, and has a similar absolute intensity to the resonance peaks observed in other Fe-based superconductors. The existence of a magnetic resonance in the spectrum of Rb$_x$Fe$_{2-y}$Se$_2$ and now of Cs$_x$Fe$_{2-y}$Se$_2$ suggests that this is a common feature of superconductivity in this family. The low energy spin-wave excitations in Cs$_{0.8}$Fe$_{1.0}$Se$_2$ show no measurable response to superconductivity, consistent with the notion of spatially separate magnetic and superconducting phases.

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I. INTRODUCTION

The $A_x$Fe$_{2-y}$Se$_2$ compounds ($A = $ K, Rb, Cs and Tl) present an interesting new twist in the field of iron-based superconductors. The discovery of superconductivity with transition temperatures $T_c \approx 30$ K in this series in conjunction with antiferromagnetism with an unusually high ordering temperature $T_N$ of up to 559 K and large ordered moment of about 3.3 $\mu_B$ per Fe$^{2+}$ naturally raises the question: can superconductivity coexist microscopically with such a robust magnetic state? Although there are regions in the phase diagrams of the iron pnictide superconductors in which magnetism and superconductivity are believed to coexist microscopically, the highest $T_c$s and bulk superconductivity are found when the magnetic state has been suppressed. Another distinct feature of the $A_x$Fe$_{2-y}$Se$_2$ systems is their band structure. The Fermi surface lacks the large hole pocket at the zone center that features prominently in theories of superconductivity and magnetism in other iron-based superconductors.

The magnetic structure observed in superconducting $A_x$Fe$_{2-y}$Se$_2$ samples consists of blocks of four ferromagnetically aligned Fe spins, with antiferromagnetic alignment between these blocks. This magnetic state forms on a $\sqrt{5} \times \sqrt{5}$ superstructure of ordered Fe vacancies that has optimal composition $A_0.8$Fe$_{1.0}$Se$_2$. A $\sqrt{2} \times \sqrt{2}$ ordered phase has also been observed in some samples and is thought to be closely related to the superconducting phase. Initial experimental investigations of $A_x$Fe$_{2-y}$Se$_2$ supported a picture of microscopic coexistence of the superconducting and antiferromagnetic states. These studies were backed up by calculations based on the antiferromagnetic state. Further work, however, has found evidence for a spatial separation of superconducting (metallic) and antiferromagnetic (insulating or semiconducting) phases. The most recent results, from NMR scanning electron microscopy (SEM), optical spectroscopy, Raman scattering and optical microscopy, and low energy muon spin rotation, may help to explain the apparent discrepancies in the earlier work. They indicate that phase separation occurs with a complex plate-like morphology on a sub-micron scale. Proximity effects between nanodomains could therefore allow an interplay between superconducting and magnetic regions, and may explain the apparent bulk superconductivity despite estimates of a genuine superconducting phase fraction of only 5–10%.

The interplay between superconductivity and static magnetic order remains a key issue in the $A_x$Fe$_{2-y}$Se$_2$ family. Another important property is the magnetic dynamics, which are widely thought to play a role in mediating superconductivity in the iron-based superconductors. Up to now, investigations of the magnetic dynamics have focussed on Rb$_x$Fe$_{2-y}$Se$_2$. The spin-wave spectrum of the insulating parent antiferromagnetic phase has been measured by inelastic neutron scattering and the results were successfully modelled in terms of a local moment Heisenberg Hamiltonian.

Su-
perconducting samples of Rb$_x$Fe$_{2-y}$Se$_2$ have also been studied, and a spin resonance has been discovered. The resonance is quasi-two-dimensional and characterized by an increase in scattering intensity below $T_c$ at an energy of approximately 14 meV and at the wave vector $Q = (0.25, 0.5)$ and equivalent positions, which corresponds to $(\pi/2, \pi)$ in square lattice notation. This wave vector is not the same as the usual resonance wave vector of the iron based superconductors, which is $Q = (0.5, 0)$ etc. In Ref. [8] it was suggested that the position of the resonance in Rb$_x$Fe$_{2-y}$Se$_2$ can be traced to the nesting of electron-like Fermi surface pockets together with a $d$-wave superconducting pairing state based on the theory of Maier et al. [9] unlike the $s_\pm$ pairing generally thought to be present in other iron-based superconductors. In another study of Rb$_x$Fe$_{2-y}$Se$_2$, a magnetic signal was reported close to $Q = (0.5, 0)$ in addition to spin-wave excitations from the block antiferromagnetic order and the magnetic resonance at $Q = (0.5, 0.25)$ [9].

In this work we studied the spin excitations in superconducting Cs$_x$Fe$_{2-y}$Se$_2$, with particular focus on the low energy magnetic features and their response to superconductivity. We find that the spin excitations associated with the block antiferromagnetic order have a very similar spectrum to those observed in non-superconducting Rb$_x$Fe$_{2-y}$Se$_2$ up to the highest energy probed ($\sim 150$ meV). To within experimental error, we find no influence of superconductivity on the low energy magnetic excitations from the block antiferromagnetic order, in contrast to the response of the magnetic Bragg peak and a two-magnon Raman peak which both show a small anomaly in intensity on cooling below $T_c$. Finally, we observe a spin resonance below $T_c$ at $(0.25, 0.5)$, establishing that this feature is not confined to Rb$_x$Fe$_{2-y}$Se$_2$ but is present in other members of the A$_x$Fe$_{2-y}$Se$_2$ family. The results suggest that the sample consists of distinct magnetically ordered and superconducting phases.

II. EXPERIMENTAL METHODS

The Cs$_x$Fe$_{2-y}$Se$_2$ single crystals were grown by the Bridgman process as described in Ref. [8]. The nominal composition of the crystals used in this study is Cs$_{0.8}$Fe$_{1.9}$Se$_2$, and their superconducting and magnetic properties have been reported previously [31,32]. The crystals were coated in Cytop varnish before handling in air, and then checked for crystalline quality prior to the experiment. Magnetic susceptibility measurements shown in Fig. 1 established that the onset of bulk superconductivity occurs at $T_c = 27$ K. By ‘bulk’ we mean that full flux exclusion is achieved after cooling in zero field. However, this does not necessarily imply 100% superconducting volume fraction, since non-superconducting regions can be screened by surface currents in a zero-field-cooled measurement. A crystal from the neutron scattering sample was re-measured after the experiment and found to have an unchanged $T_c$.

The inelastic neutron scattering experiments were performed on the MERLIN time-of-flight (TOF) chopper spectrometer at the ISIS Facility [33]. Three single crystals were co-aligned to give a sample of total mass 0.42 g, with a uniform mosaic of 2.5° (full width at half maximum). The sample was mounted with the c axis parallel to the incident neutron beam, and the a axis horizontal. Spectra were recorded in the large position-sensitive detector array with neutrons of incident energy $E_i = 33, 40, 50, 60, 100$ and 180 meV at $T = 4$ K, and $E_i = 33$ meV at $T = 4, 20, 34$ and 44 K. For a fixed sample orientation only three of the four $(Q, E)$ components are independent. We will use $E$ and the two in-plane wave vector components $(H, K)$. This means that the out-of-plane wave vector component varies with $E$. The scattering from a standard vanadium sample was used to normalize the spectra and place them on an absolute intensity scale, with units mb sr$^{-1}$ meV$^{-1}$ f.u.$^{-1}$, where 1 mb = 10$^{-31}$ m$^2$ and f.u. stands for formula unit of Cs$_{0.8}$Fe$_{1.9}$Se$_2$.

III. RESULTS

Figure 2(a) is a map of the $(H, K)$ plane in two-dimensional reciprocal space, showing the positions of the antiferromagnetic Bragg peaks and the magnetic resonance signal reported in Ref. [9]. We index positions in reciprocal space with respect to the one-Fe sub-lattice, lattice parameters $a = b = 2.8$ Å. Figure 2(b) is a map of the neutron scattering intensity averaged over the en-
Figure 2. (Color online) (a) Map of two-dimensional reciprocal space for Cs$_x$Fe$_{2-y}$Se$_2$, showing positions of the magnetic ordering vectors and the magnetic resonance signals. Wave vectors are indexed with respect to the one-Fe unit cell (unfolded Brillouin zone). The open circles are reciprocal lattice points, and the dashed square is the first Brillouin zone for this unit cell. The two small tilted squares are the first Brillouin zones for the unit cells of the left-handed (LH) and right-handed (RH) magnetic domains. The paths along which cuts shown in later figures were taken are represented by thick solid lines. (b) Neutron scattering intensity map of Cs$_{0.8}$Fe$_{1.9}$Se$_2$ in the same area of reciprocal space as shown in (a). The data were recorded with an incident neutron energy of 100 meV. The areas of missing data are due to the beam stop and masked detectors.

The magnetic spectrum is revealed in more detail in Fig. 3, which shows a strongly dispersive spin-wave band extending from below 20 meV up to 63 meV, and a second band between 85 and 120 meV. The existence of the latter is demonstrated in Fig. 3(a) via two energy scans recorded at fixed wavevectors of $Q_{AFM} = (0.1, 0.5)$ and $(0.1, 1)$. These positions were chosen after inspection of an intensity map like that in Fig. 2(b) but at an energy of 100 meV, which showed a regular pattern of diffuse magnetic scattering with maximum intensity at $(0.1, 0.5)$ and minimum at $(0.1, 1)$.

Figure 3(b) plots the in-plane dispersion of the lower spin-wave band. In constant-energy maps in the $(H, K)$ plane, the low-energy spin-wave scattering appears as a ring of intensity centered on the $Q_{AFM}$ positions. The points in Fig. 3(b) were obtained as follows. Gaussian functions fitted to pairs of peaks symmetrically displaced either side of each $Q_{AFM}$ were constrained to have the same area and width. The peak positions were corrected for the systematic shift caused by the curvature of the dispersion surface over the width $\Delta H$ of the cuts. Where appropriate, a non-magnetic background was estimated from cuts taken along nearby lines in reciprocal space. The points at the magnetic Brillouin zone (BZ) boundaries (marked in Fig. 3(b) by dashed lines) were obtained from a Gaussian fit to the peak in a background-corrected energy cut.

The results shown in Figs. 3(a) and (b) bear a very close resemblance to the magnetic spectrum of non-superconducting Rb$_x$Fe$_{2-y}$Se$_2$ reported in Ref. 36. Our data are not sufficient to determine the detailed dispersion in the out-of-plane direction $(0, 0, L)$, but the spectra measured with different $E_i$ to probe $Q_{AFM}$ at different $L$ values are consistent with a minimum anisotropy gap of $7 \pm 1$ meV and a maximum of about 20 meV, somewhat lower than the maximum of 30 meV reported for
Rb$_2$Fe$_{2-y}$Se$_2$. In Ref. [37], a third spin-wave band was observed in Rb$_2$Fe$_{2-y}$Se$_2$, with a dispersion from 180 and 230 meV. Our data do not extend high enough in energy to confirm the existence of this band in Cs$_2$Fe$_{2-y}$Se$_2$.

In Fig. 3 we show the temperature dependence of the spin-wave peak at $Q_{\text{AFM}} = (0.1, 0.3)$ averaged over the energy range 13 to 15 meV. Figure 3(a) shows wave vector scans recorded at four different temperatures, two below $T_c$ and two above $T_c$. The peaks show no discernible change within this temperature range. To check this quantitatively we fitted the data to a Gaussian function on a linear background, allowing the width, center and area of the Gaussian, and the slope and intercept of the background to vary. To correct for the increase in signal due to the thermal population of spin-waves we have considered the background to vary. To correct for the increase in signal due to the thermal population of spin-waves we have considered the background to vary. To correct for the increase in signal due to the thermal population of spin-waves we have considered the background to vary. 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Figure 5. (Color online) (a) Neutron scattering intensity map of Cs$_x$Fe$_{2-y}$Se$_2$ averaged over energies between 9 and 13 meV. The data were recorded at 4 K with an incident energy $E_i = 33$ meV. The inset shows the peak intensity as a function of energy, where the intensity is given as the imaginary part of the local susceptibility. (b) Constant-energy cuts through the map in (a) showing the temperature dependence of the magnetic signal at ($0.25, 0.5$). The out-of-plane wave vector at the peak is $L = 2.4$. Successive cuts are displaced vertically by 5 units for clarity. The dotted line through the 4 K cut is a fit to a Gaussian peak on a linear background. The inset in (b) shows the integrated intensity of the 4 K Gaussian peak and of constrained Gaussian fits to the higher temperature data (see text) as a function of temperature. The integrated intensities (in mb sr$^{-1}$ meV$^{-1}$ f.u.$^{-1}$ Å$^{-1}$) have been normalized by the Bose factor $[1 - \exp(-E/k_B T)]^{-1}$.

to Fig. 5(a)). Wave vector cuts through this peak in the ($H, H$) direction averaged over 9–13 meV are shown in Fig. 5(b) at a series of temperatures. The cut at 4 K, well below $T_c$, shows a well defined peak which has been fitted with a Gaussian function on a linear background (dashed line). Above $T_c$ the peak is either strongly suppressed or absent. Fits were made to the cuts at higher temperatures with the width and center of the Gaussian fixed to the values found at 4 K. The inset to Fig. 5(b) shows the integrated intensity of the fitted Gaussian peaks as a function of temperature. The signal clearly increases as the temperature decreases. To determine the absolute strength of the peak we have converted its integrated intensity into the $Q$-averaged or local susceptibility $\chi''(\omega)$. We assumed the peak is two-dimensional and used the dipole form factor of Fe$^{2+}$. The inset to Fig. 5(a) shows the energy dependence of $\chi''(\omega)$ at $T = 4$ K.

**IV. DISCUSSION**

One of the goals of this work was to determine whether the spin dynamics of the block antiferromagnetic phase in superconducting samples of $A_x$Fe$_{2-y}$Se$_2$ are different to those in insulating samples, and whether they respond to superconductivity. Figure 3 presents a clear demonstration that the antiferromagnetic spin-waves persist in superconducting Cs$_x$Fe$_{2-y}$Se$_2$ and have a similar spectrum to that of insulating Rb$_x$Fe$_{2-y}$Se$_2$. We find the top of the low energy acoustic spin-wave branch to be 63 ± 1 meV, and the center of the medium energy band to be 105 ± 5 meV, compared with ∼67 meV and ∼115 meV, respectively, found in Rb$_x$Fe$_{2-y}$Se$_2$. We find no evidence for a coupling between the low energy spin-waves and superconductivity. This is illustrated in Fig. 4 for an energy near 14 meV where the scattering is strongest. However, we also examined the data from 8 meV up to 27 meV and found no change in the spin-wave scattering on cooling through $T_c$ at any energy in this range. From our results we can rule out any superconductivity-induced change at low energies greater than 3–4%. By contrast, previous studies on superconducting K$_x$Fe$_{2-y}$Se$_2$ reported systematic reductions of 5% or more in the intensities of a magnetic Bragg peak and a two-magnon Raman peak at ∼200 meV on cooling below $T_c$. One possibility is that the size of the effect depends on the energy probed, however a more plausible explanation is based on the notion that these samples
are phase-separated on a nanoscale into superconducting and magnetically ordered (non-superconducting) regions which only interact at the interfaces.\textsuperscript{31,32} Below $T_c$, the superconducting proximity effect could suppress magnetic order near the phase boundaries, so that samples with different interfacial surface areas would respond to superconductivity by different amounts.

Although we find no effect of superconductivity on the magnetic excitations associated with the block antiferromagnetic order, we do observe the magnetic resonance peak at $(0.25, 0.5)$ previously reported in superconducting Rb$_2$Fe$_{2\text{−}y}$Se$_2$\textsuperscript{27,42,43}. As shown in Fig. 5, we find that the magnetic signal at $(0.25, 0.5)$ increases in intensity on cooling below $T_c$, and there is tentative evidence that the peak persists at temperatures above $T_c$ in agreement with the observations of Friemel \textit{et al.}\textsuperscript{44} The existence of resonance peaks in the iron pnictides has been explained in terms of nesting features in the Fermi surface enhanced by electronic correlations and superconducting coherence effects.\textsuperscript{34} Within this framework, and with a realistic band structure model, Friemel \textit{et al.}\textsuperscript{44} were able to reproduce the position of the magnetic resonance in Rb$_2$Fe$_{2\text{−}y}$Se$_2$ assuming a $d_{x^2\text{−}y^2}$ superconducting gap. Further theoretical work is needed to understand the magnetic resonance in detail, but our results at least establish that the $(0.25, 0.5)$ resonance is present in another A$_x$Fe$_{2\text{−}y}$Se$_2$ superconductor. This suggests that the resonance could be a characteristic feature of superconductivity in this family.

Finally, we make some remarks about the absolute intensities of the magnetic features. The scattering intensities in our measurements and those of Ref. \textsuperscript{40} are calibrated and given in absolute units of cross section. This allows us to compare the strengths of the magnetic signal from the sample of Rb$_2$Fe$_{2\text{−}y}$Se$_2$ used in Ref. \textsuperscript{40} with those from the sample of Cs$_8$Fe$_{2\text{−}y}$Se$_2$ used here. The amplitude of the spin-wave peak at 14 meV for Cs$_8$Fe$_{2\text{−}y}$Se$_2$ (Fig. 4 above) is about 55 mb sr$^{-1}$ meV$^{-1}$ f.u.$^{-1}$, which is similar to the amplitude of 40 mb sr$^{-1}$ meV$^{-1}$ f.u.$^{-1}$ at 10 meV for Rb$_2$Fe$_{2\text{−}y}$Se$_2$ (Fig. 5(b) of Ref. \textsuperscript{40}). However, the amplitude of the resonance peak in Cs$_8$Fe$_{2\text{−}y}$Se$_2$, about 2.5 mb sr$^{-1}$ meV$^{-1}$ f.u.$^{-1}$, is about five times larger than that reported for Rb$_2$Fe$_{2\text{−}y}$Se$_2$ — compare Fig. 5(b) above with Fig. 6 of Ref. \textsuperscript{40}. One should of course be cautious when comparing peak amplitudes. Nevertheless, it does appear that the resonance peak is more prominent in Cs$_8$Fe$_{2\text{−}y}$Se$_2$ than in Rb$_2$Fe$_{2\text{−}y}$Se$_2$. This could indicate that the crystal used here has a higher volume fraction of superconducting phase than that used in Ref. \textsuperscript{40}.

It is also interesting to compare the strength of the resonance peak with that in other Fe-based superconductors. Results for $\chi''(\omega)$ have been reported previously for BaFe$_{1.87}$Co$_{0.13}$As$_2$ and BaFe$_{1.9}$Ni$_{0.1}$As$_2$.\textsuperscript{27,42,43} In both cases the resonance peak amplitudes (i.e. the increase on cooling below $T_c$) are $3\text{−}4 \mu_B^2$ eV$^{-1}$ f.u.$^{-1}$ and the energy-integrated signal is $\sim 0.015 \mu_B^2$ f.u.$^{-1}$. From the inset to Fig. 7(a) the corresponding values for Cs$_8$Fe$_{2\text{−}y}$Se$_2$ are $(3.0 \pm 0.5) \mu_B^2$ eV$^{-1}$ f.u.$^{-1}$ and $(0.015 \pm 0.003) \mu_B^2$ f.u.$^{-1}$, remarkably similar to the values for the two arsenide superconductors. Since the latter were near optimal doping they are expected to be bulk superconductors with close to 100% superconducting volume fraction. It is tempting, therefore, to conclude that the resonance peak and hence superconductivity in Cs$_8$Fe$_{2\text{−}y}$Se$_2$ is associated with most or all of the sample volume. However, there are many other factors that could control the size of the resonance peak, e.g. the degree of nesting, strength of magnetic correlations, etc. and these may differ from one material to another. We simply note that the resonance peak in Cs$_8$Fe$_{2\text{−}y}$Se$_2$ is similar in strength to that in other Fe-based superconductors.

V. CONCLUSIONS

The magnetic spectrum of Cs$_{0.8}$Fe$_1.9$Se$_2$ studied in this work comprises two components: a low energy resonance-like excitation with wave vector $(0.25, 0.5)$ which responds to superconductivity and is similar in strength to the corresponding feature found in other Fe-based superconductors, and spin-wave excitations of the block antiferromagnetic order with wave vector $Q_{\text{AFM}} = (0.1, 0.3)$ which do not respond to superconductivity to within the experimental sensitivity. The spin-wave component closely resembles that of non-superconducting (insulating) Rb$_2$Fe$_{2\text{−}y}$Se$_2$. Together with other recent studies, these results are consistent with a microstructure composed of spatially separate superconducting and non-superconducting domains, with the $\sqrt{5} \times \sqrt{5}$ Fe vacancy superstructure and block antiferromagnetism confined to the non-superconducting phase. It remains a materials challenge to try to maximize the volume fraction of the superconducting phase.

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\textsuperscript{a}a.taylor1@physics.ox.ac.uk
\textsuperscript{b}a.boothroyd@physics.ox.ac.uk
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