Spin-wave excitations and superconducting resonant mode in $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ 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, Oxon, 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-00664 Warsaw, Poland*

(Received 17 August 2012; published 28 September 2012)

We report neutron inelastic scattering measurements on the normal and superconducting states of single-crystalline $\text{Cs}_{0.8}\text{Fe}_{1.9}\text{Se}_2$. Consistent with previous measurements on $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$, we observe two distinct spin excitation signals: (i) spin-wave excitations characteristic of the block antiferromagnetic order found in insulating $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$ 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 $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ and now of $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ suggests that this is a common feature of superconductivity in this family. The low-energy spin-wave excitations in $\text{Cs}_{0.8}\text{Fe}_{1.9}\text{Se}_2$ show no measurable response to superconductivity, consistent with the notion of spatially separate magnetic and superconducting phases.

DOI: [10.1103/PhysRevB.86.094528](https://doi.org/10.1103/PhysRevB.86.094528)

PACS number(s): 74.25.Ha, 74.70.Xa, 78.70.Nx, 75.30.Ds

I. INTRODUCTION

The $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$ compounds ($A = \text{K}, \text{Rb}, \text{Cs}$, 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,^{1–4} 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,⁵ naturally raises the following 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.^{6–8} Another distinct feature of the $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$ systems is their band structure.^{9,10} 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 $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$ 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 $\text{A}_{0.8}\text{Fe}_{1.6}\text{Se}_2$.^{5,11–14} 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.^{13,15,16}

Initial experimental investigations of $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$ supported a picture of microscopic coexistence of the superconducting and antiferromagnetic states.^{1,3,5,17–21} These studies were backed up by calculations based on the antiferromagnetic state.^{22–25} Further work, however, has found evidence for a

spatial separation of superconducting (metallic) and antiferromagnetic (insulating or semiconducting) phases.^{13,26–30} The most recent results, from NMR,³¹ scanning electron microscopy (SEM),³² optical spectroscopy,³³ Raman scattering and optical microscopy,³⁴ and low-energy muon spin rotation³⁵ may help to explain the apparent discrepancies in the earlier work. They indicate that phase separation occurs with a complex platelike morphology on a submicron 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 $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$ family. Another important property is the magnetic dynamics, which are widely thought to play a role in mediating superconductivity in the iron-based superconductors.^{6–8} Up to now, investigations of the magnetic dynamics have focused on $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$. The spin-wave spectrum of the insulating parent antiferromagnetic phase has been measured by inelastic neutron scattering and the results were successfully modeled in terms of a local moment Heisenberg Hamiltonian.³⁶ Superconducting samples of $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ have also been studied, and a spin resonance has been discovered.³⁷ 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 $\mathbf{Q} = (0.25, 0.5)$ and equivalent positions, which correspond to $(\pi/2, \pi)$ in square-lattice notation.³⁸ This wave-vector is not the same as the usual resonance wave-vector of the iron-based superconductors, which is $\mathbf{Q} = (0.5, 0)$, etc. In Ref. 37, based on the theory of Maier *et al.*,³⁹ it was suggested that the position of the resonance in $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$

FIG. 1. (Color online) Magnetic susceptibility of one of the $\text{Cs}_{0.8}\text{Fe}_{1.9}\text{Se}_2$ single crystals used here, measured with a field of 1 Oe applied along the c axis after cooling in zero field. A susceptibility of -1 corresponds to full Meissner flux exclusion, but as no demagnetization corrections have been applied, -1 is not a rigorous lower bound.

can be traced to the nesting of electron-like Fermi surface pockets together with a d -wave superconducting pairing state, unlike the $s_{\pm}$ pairing generally thought to be present in other iron-based superconductors. In another study of $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$,

a magnetic signal was reported close to $\mathbf{Q} = (0.5, 0)$ in addition to spin-wave excitations from the block antiferromagnetic order and the magnetic resonance at $\mathbf{Q} = (0.5, 0.25)$.⁴⁰

In this work, we studied the spin excitations in superconducting $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$, 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 nonsuperconducting $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ up to the highest energy probed (~ 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 .^{5,34} Finally, we observe a spin resonance below T_c at $(0.25, 0.5)$, establishing that this feature is not confined to $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$, but is present in other members of the $\text{A}_x\text{Fe}_{2-y}\text{Se}_2$ family. The results suggest that the sample consists of distinct magnetically ordered and superconducting phases.

II. EXPERIMENTAL METHODS

The $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ single crystals were grown by the Bridgman process as described in Ref. 3. The nominal composition of the crystals used in this study is $\text{Cs}_{0.8}\text{Fe}_{1.9}\text{Se}_2$, and their superconducting and magnetic properties have been reported previously.^{3,11,17} The crystals were coated in Cytop varnish

FIG. 2. (Color online) (a) Map of two-dimensional reciprocal space for $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$, showing positions of the magnetic ordering vectors and the magnetic resonance signals. Wave-vectors are indexed in reciprocal lattice units (r.l.u.) 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 (Ref. 5). The paths along which cuts shown in later figures were taken are represented by thick solid lines. (b) Neutron scattering intensity map of $\text{Cs}_{0.8}\text{Fe}_{1.9}\text{Se}_2$ in the same area of reciprocal space as shown in (a). The data were recorded with an incident neutron energy of 100 meV and averaged over the energy range from 10 to 20 meV. The areas of missing data are due to the beam stop and masked detectors.

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 nonsuperconducting regions can be screened by surface currents in a zero-field-cooled measurement. A crystal from the neutron scattering sample was remeasured 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.⁴¹ Three single crystals were coaligned 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 $[110]$ direction 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 $(\mathbf{Q}, E)$ 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 $\text{mb sr}^{-1} \text{meV}^{-1} \text{f.u.}^{-1}$, where $1 \text{ mb} = 10^{-31} \text{ m}^2$ and f.u. stands for formula unit of $\text{Cs}_{0.8}\text{Fe}_{1.9}\text{Se}_2$.

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. 37. We index positions in reciprocal space with respect to the one-Fe sublattice, lattice parameters $a = b = 2.8 \text{ \AA}$, $c = 7.7 \text{ \AA}$. Figure 2(b) is a map of the neutron scattering intensity averaged over the energy range 10 to 20 meV and projected onto the same region of the (H, K) plane as shown in Fig. 2(a). The strong scattering signal localized at $\mathbf{Q}_{\text{AFM}} = (0.1, 0.3)$ and equivalent positions is due to magnetic fluctuations associated with the block antiferromagnetic order on the $\sqrt{5} \times \sqrt{5}$ Fe vacancy superstructure. The eightfold symmetry of the magnetic spectrum, which derives from the superposition of two fourfold patterns from left-handed and right-handed magnetic structures, respectively, is apparent from this figure. All spectra presented hereafter have been folded into one octant to improve statistics.

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 wave-vectors of $(0.1, 0.5)$ 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,

FIG. 3. (Color online) Spin-wave spectrum of $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$. (a) Energy cuts showing the band of magnetic scattering around 100 meV. The data are from a run with incident neutron energy $E_i = 180$ meV at 4 K. The orange triangles were recorded at the wave-vector $(0.1, 0.5)$, where there is a clear magnetic signal with maximum intensity near 105 meV. The green diamonds are a similar cut from the nearby position $(0.1, 1)$, which is away from any magnetic scattering. (b) Spin-wave dispersion of the low-energy band, measured at 4 K. The method used to obtain the data points is described in the text. The different colored symbols indicate data obtained with different incident neutron energies E_i . The dashed lines mark the magnetic Brillouin zone boundaries. For energies below 60 meV, the vertical error bars represent the width of the cut in energy and the horizontal error bars represent the error in the fitted peak position. For the two points at 63 meV, the horizontal error bar is the width of the cut in wave-vector and the vertical error bar is the error from the fit. The point at 7 meV is the minimum anisotropy gap and has an out-of-plane wave-vector component $L \approx 0.5$ r.l.u.

the low-energy spin-wave scattering appears as a ring of intensity centered on the $\mathbf{Q}_{\text{AFM}}$ positions. The points in Fig. 3(b) were obtained as follows. Gaussian fits were made to peaks in constant-energy cuts along the line $(0.1, K)$ passing through the magnetic wave-vectors $\mathbf{Q}_{\text{AFM}} = (0.1, 0.3)$ and $(0.1, 0.7)$ [see Fig. 2(a)]. Gaussian functions fitted to pairs of peaks symmetrically displaced either side of each $\mathbf{Q}_{\text{AFM}}$ 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 ΔH of the cuts. Where appropriate, a nonmagnetic background was

estimated from cuts taken along nearby lines in reciprocal space. The two peaks could not be resolved for energies below 20 meV. 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 minimum point on the dispersion was determined from an energy cut at $\mathbf{Q}_{\text{AFM}}$ through the $E_i = 60$ meV data.

The results shown in Figs. 3(a) and 3(b) bear a very close resemblance to the magnetic spectrum of nonsuperconducting $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ 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 $\mathbf{Q}_{\text{AFM}}$ at different L values are consistent with a minimum anisotropy gap of 7 ± 1 meV and a maximum of about 20 meV, somewhat lower than the maximum of 30 meV reported³⁶ for $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$. In Ref. 36, a third spin-wave band was observed in $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$, 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 $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$.

In Fig. 4, we show the temperature dependence of the spin-wave peak at $\mathbf{Q}_{\text{AFM}} = (0.1, 0.3)$ averaged over the energy range 13 to 15 meV. Figure 4(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 normalized the data by the Bose population factor. Figure 4(b) plots the areas of the fitted peaks as a function of temperature. To within the experimental error (about 3%), there is no change upon crossing the superconducting transition temperature.

Finally, we consider the magnetic dynamics at wave-vectors away from the $\mathbf{Q}_{\text{AFM}}$ points in reciprocal space. Figure 5(a) shows an intensity map recorded at 4 K and averaged over the energy range 9 to 13 meV. The data have been folded onto an octant of reciprocal space to improve statistics. To within the experimental error, there is no evidence for the excitations observed near $(0,0.5)$ and equivalent positions by Wang *et al.* in similar measurements on superconducting $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$.⁴⁰ However, our data do reveal a weak signal centered on $(0.25, 0.5)$ with a maximum at an energy of about 11 meV [see inset to Fig. 5(a)]. Wave-vector cuts through this peak in the $(H, 0)$ direction averaged over 9–15 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 (dotted line). 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, consistent with a magnetic resonance peak. To determine the absolute strength of the peak, we have converted its integrated intensity into the $\mathbf{Q}$ -averaged or local susceptibility $\chi''(\omega)$.⁴² We assumed the peak to be 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. Figure 6 shows

FIG. 4. (Color online) (a) Constant-energy cuts through $\mathbf{Q}_{\text{AFM}} = (0.1, 0.3)$ showing the temperature dependence of the spin-wave scattering averaged over the energy range 13 to 15 meV. Data were recorded with an incident energy $E_i = 33$ meV, giving an out-of-plane wave-vector component $L = 1.28$ r.l.u. Successive cuts are displaced vertically by 40 units for clarity. Dashed lines are fits to Gaussian peaks on a linear background, as described in the main text. (b) The integrated intensity (in $\text{mb sr}^{-1} \text{meV}^{-1} \text{f.u.}^{-1} \text{r.l.u.}$) given by the area of the fitted Gaussian peaks as a function of temperature. The data have been normalized by the Bose factor $[1 - \exp(-E/k_B T)]^{-1}$.

cuts through the $(0.25, 0.5)$ peak parallel to $(H, 0)$ and $(0, K)$ together with Gaussian fits. The full widths at half maximum are found to be 0.066 ± 0.010 r.l.u. and 0.070 ± 0.016 r.l.u., respectively.

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 $A_x\text{Fe}_{2-y}\text{Se}_2$ 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 $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ and have a similar spectrum to that of insulating $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$.³⁶ We find the top of the

FIG. 5. (Color online) (a) Neutron scattering intensity map of $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ averaged over energies between 9 and 13 meV. The data were recorded at 4 K with an incident energy $E_i = 33$ meV. The weak signal centered on (0.25,0.5) is identified with the resonance peak. The inset shows the resonance peak intensity as a function of energy, where the intensity is given as the imaginary part of the local susceptibility. (b) Constant-energy cuts showing the temperature dependence of the resonance peak. The out-of-plane wave-vector at the peak is $L = 1.3$ r.l.u. Successive cuts are displaced vertically by 5 units for clarity. The dotted lines are fits to a Gaussian peak on a linear background. The inset in (b) shows the integrated intensity (in $\text{mb sr}^{-1} \text{meV}^{-1} \text{f.u.}^{-1}$ r.l.u.) given by the Gaussian fits as a function of temperature. The data have been normalized by the Bose factor $[1 - \exp(-E/k_B T)]^{-1}$.

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 $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$.³⁶

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 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 greater than 3%–4% at low energies. By contrast, previous studies on superconducting $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ 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 .^{5,34} 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 (nonsuperconducting) regions which only interact at the interfaces.^{31–33} 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 the spectrum of superconducting $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$.^{37,40,43} As shown in Fig. 5(b), we find that the magnetic signal at (0.25,0.5) increases in intensity on cooling below T_c , and the peak persists at temperatures

above T_c in agreement with the observations of Friemel *et al.*⁴³ However, one notable difference is the shape of the resonance peak in momentum space, which for $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ was found to be highly elliptical with an aspect ratio of 2.1.⁴³ For $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$, we have observed that the resonance peak is more isotropic, with an aspect ratio of 1.1 ± 0.3 (see Fig. 6).

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.⁴⁴ Within this framework, and with a realistic band-structure model, Friemel *et al.*⁴³ were able to reproduce the position of the magnetic resonance in $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ assuming a $d_{x^2-y^2}$ superconducting gap. Further theoretical

FIG. 6. (Color online) Constant-energy cuts through the resonance peak parallel to the (a) $(H, 0)$ and (b) $(0, K)$ directions. The data were recorded at 4 K with an incident energy $E_i = 33$ meV. The solid lines are fits to a Gaussian peak on a linear [panel(a)] or quadratic [panel(b)] background.

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 $A_x\text{Fe}_{2-y}\text{Se}_2$ superconductor. This suggests that the resonance could be a characteristic feature of superconductivity in this family.

By contrast, we do not observe the magnetic signal reported by Wang *et al.* at (0,0.5) and equivalent wave-vectors in similar measurements on superconducting $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$.⁴⁰ This affirms that the $A_x\text{Fe}_{2-y}\text{Se}_2$ compounds are distinct from other families of Fe-based superconductors.

Finally, we make some remarks about the absolute intensities of the magnetic features. The scattering intensities in our measurements and those of Ref. 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 $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ used in Ref. 40 with those from the sample of $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ used here. The amplitude of the spin-wave peak at 14 meV for $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ (Fig. 4 above) is about $55 \text{ mb sr}^{-1} \text{ meV}^{-1} \text{ f.u.}^{-1}$, which is similar to the amplitude of $40 \text{ mb sr}^{-1} \text{ meV}^{-1} \text{ f.u.}^{-1}$ at 10 meV for $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ [Fig. 5(b) of Ref. 40]. However, the amplitude of the resonance peak in $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$, about $2.5 \text{ mb sr}^{-1} \text{ meV}^{-1} \text{ f.u.}^{-1}$, is about five times larger than that reported for $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ [compare Fig. 5(b) above with Fig. 6 of Ref. 40]. One should of course be cautious when comparing peak amplitudes. Nevertheless, it does appear that the resonance peak is more prominent in $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ than in $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$. This could indicate an intrinsic difference between the two materials, or that the crystal used here has a higher volume fraction of superconducting phase than that used in Ref. 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 $\text{BaFe}_{1.85}\text{Co}_{0.15}\text{As}_2$, $\text{BaFe}_{1.87}\text{Co}_{0.13}\text{As}_2$, and $\text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$ (Refs. 45, 42, and 46, respectively). With the latter two materials, the resonance peak enhancement (i.e., the increase on cooling below T_c) estimated from the presented spectra is $3\text{--}4\mu_B^2 \text{ eV}^{-1} \text{ f.u.}^{-1}$ and the enhancement in the energy-integrated signal is $\sim 0.015\mu_B^2 \text{ f.u.}^{-1}$. The values for $\text{BaFe}_{1.85}\text{Co}_{0.15}\text{As}_2$ are about a factor of 2 larger.⁴⁷ In the case of $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$, if we assume the $T > T_c$ signal to be about 40% of that at $T = 4 \text{ K}$ based on the ratio of 44 to 4 K data shown in the inset to Fig. 5(b), then the enhancements in the peak and energy-integrated local susceptibilities are $(1.8 \pm 0.5)\mu_B^2 \text{ eV}^{-1} \text{ f.u.}^{-1}$ and $(0.009 \pm 0.003)\mu_B^2 \text{ f.u.}^{-1}$, which are comparable with the values found for the arsenide superconductors. Since the arsenides were studied near optimal doping, they are

expected to be bulk superconductors with close to 100% superconducting volume fraction. It is tempting, therefore, to infer that the resonance peak, and hence superconductivity, in $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ is associated with most or all of the sample volume. However, there are several other factors that could influence 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 $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$ is similar in strength to that in other Fe-based superconductors despite the apparent small volume fraction of superconducting phase.

V. CONCLUSIONS

The magnetic spectrum of $\text{Cs}_{0.8}\text{Fe}_{1.9}\text{Se}_2$ 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 $\mathbf{Q}_{\text{AFM}} = (0.1, 0.3)$, which do not respond to superconductivity to within the experimental sensitivity. The spin-wave component closely resembles that of nonsuperconducting (insulating) $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$. Together with other recent studies, these results are consistent with a microstructure composed of spatially separate superconducting and nonsuperconducting domains, with the $\sqrt{5} \times \sqrt{5}$ Fe vacancy superstructure and block antiferromagnetism confined to the nonsuperconducting phase. It remains a materials challenge to try to maximize the volume fraction of the superconducting phase.

Note added. Recently, a paper appeared describing the observation of a magnetic resonant mode in the neutron scattering spectrum of superconducting $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$.⁴⁸ The wave-vector, energy, and integrated spectral weight of the resonant mode of $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ reported in Ref. 48 are very similar to those found here for $\text{Cs}_x\text{Fe}_{2-y}\text{Se}_2$.

ACKNOWLEDGMENTS

This work was supported by the UK Engineering & Physical Sciences Research Council and the Science & Technology Facilities Council. Work in Switzerland was supported by the Swiss National Science Foundation and its NCCR programme MaNEP. We thank M. Kenzelmann, A. Podlesnyak, L.-P. Regnault, and F. Bourdarot for assistance with the experimental work, and D. S. Inosov for helpful comments on the manuscript.

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¹J. Guo, S. Jin, G. Wang, S. Wang, K. Zhu, T. Zhou, M. He, and X. Chen, *Phys. Rev. B* **82**, 180520 (2010).

²A. F. Wang, J. J. Ying, Y. J. Yan, R. H. Liu, X. G. Luo, Z. Y. Li, X. F. Wang, M. Zhang, G. J. Ye, P. Cheng, Z. J. Xiang, and X. H. Chen, *Phys. Rev. B* **83**, 060512 (2011).

³A. Krzton-Maziopa, Z. Shermadini, E. Pomjakushina, V. Pomjakushin, M. Bendele, A. Amato, R. Khasanov,

H. Luetkens, and K. Conder, *J. Phys.: Condens. Matter* **23**, 052203 (2011).

⁴M.-H. Fang, H.-D. Wang, C.-H. Dong, Z.-J. Li, C.-M. Feng, J. Chen, and H. Q. Yuan, *Europhys. Lett.* **94**, 27009 (2011).

⁵W. Bao, Q. Huang, G. Chen, D. Wang, J. He, and Y. Qiu, *Chin. Phys. Lett.* **28**, 086104 (2011).

⁶D. C. Johnston, *Adv. Phys.* **59**, 803 (2010).

⁷G. R. Stewart, *Rev. Mod. Phys.* **83**, 1589 (2011).

- ⁸M. D. Lumsden and A. D. Christianson, *J. Phys.: Condens. Matter* **22**, 203203 (2010).
- ⁹Y. Zhang, L. X. Yang, M. Xu, Z. R. Ye, F. Chen, C. He, H. C. Xu, J. Jiang, B. P. Xie, J. J. Ying, X. F. Wang, X. H. Chen, J. P. Hu, M. Matsunami, S. Kimura, and D. L. Feng, *Nat. Mater.* **10**, 273 (2011).
- ¹⁰T. Qian, X. P. Wang, W. C. Jin, P. Zhang, P. Richard, G. Xu, X. Dai, Z. Fang, J. G. Guo, X. L. Chen, and H. Ding, *Phys. Rev. Lett.* **106**, 187001 (2011).
- ¹¹V. Y. Pomjakushin, D. V. Sheptyakov, E. V. Pomjakushina, A. Krzton-Maziopa, K. Conder, D. Chernyshov, V. Svitlyk, and Z. Shermadini, *Phys. Rev. B* **83**, 144410 (2011).
- ¹²V. Y. Pomjakushin, E. V. Pomjakushina, A. Krzton-Maziopa, K. Conder, and Z. Shermadini, *J. Phys.: Condens. Matter* **23**, 156003 (2011).
- ¹³M. Wang, M. Wang, G. N. Li, Q. Huang, C. H. Li, G. T. Tan, C. L. Zhang, H. Cao, W. Tian, Y. Zhao, Y. C. Chen, X. Y. Lu, B. Sheng, H. Q. Luo, S. L. Li, M. H. Fang, J. L. Zarestky, W. Ratcliff, M. D. Lumsden, J. W. Lynn, and P. Dai, *Phys. Rev. B* **84**, 094504 (2011).
- ¹⁴F. Ye, S. Chi, W. Bao, X. F. Wang, J. J. Ying, X. H. Chen, H. D. Wang, C. H. Dong, and M. Fang, *Phys. Rev. Lett.* **107**, 137003 (2011).
- ¹⁵A. Ricci, N. Poccia, B. Joseph, G. Arrighetti, L. Barba, J. Plaisier, G. Campi, Y. Mizuguchi, H. Takeya, Y. Takano, N. L. Saini, and A. Bianconi, *Supercond. Sci. Technol.* **24**, 082002 (2011).
- ¹⁶W. Li, H. Ding, Z. Li, P. Deng, K. Chang, K. He, S. Ji, L. Wang, X. Ma, J.-P. Hu, X. Chen, and Q.-K. Xue, *Phys. Rev. Lett.* **109**, 057003 (2012).
- ¹⁷Z. Shermadini, A. Krzton-Maziopa, M. Bendele, R. Khasanov, H. Luetkens, K. Conder, E. Pomjakushina, S. Weyeneth, V. Pomjakushin, O. Bossen, and A. Amato, *Phys. Rev. Lett.* **106**, 117602 (2011).
- ¹⁸D. A. Torchetti, M. Fu, D. C. Christensen, K. J. Nelson, T. Imai, H. C. Lei, and C. Petrovic, *Phys. Rev. B* **83**, 104508 (2011).
- ¹⁹W. Yu, L. Ma, J. B. He, D. M. Wang, T. L. Xia, G. F. Chen, and W. Bao, *Phys. Rev. Lett.* **106**, 197001 (2011).
- ²⁰L. Ma, G. F. Ji, J. Zhang, J. B. He, D. M. Wang, G. F. Chen, W. Bao, and W. Yu, *Phys. Rev. B* **83**, 174510 (2011).
- ²¹H. Kotegawa, Y. Hara, H. Nohara, H. Tou, Y. Mizuguchi, H. Takeya, and Y. Takano, *J. Phys. Soc. Jpn.* **80**, 043708 (2011).
- ²²T. Das and A. V. Balatsky, *Phys. Rev. B* **84**, 115117 (2011).
- ²³X. W. Yan, M. Gao, Z. Y. Lu, and T. Xiang, *Phys. Rev. Lett.* **106**, 087005 (2011).
- ²⁴C. Cao and J. Dai, *Chin. Phys. Lett.* **28**, 057402 (2011).
- ²⁵G. M. Zhang, Z. Y. Lu, and T. Xiang, *Phys. Rev. B* **84**, 052502 (2011).
- ²⁶W. Li, H. Ding, P. Deng, K. Chang, C. Song, K. He, L. Wang, X. Ma, J. Hu, X. Chen, and Q. Xue, *Nat. Phys.* **8**, 126 (2011).
- ²⁷A. Charnukha, J. Deisenhofer, D. Pröpper, M. Schmidt, Z. Wang, Y. Goncharov, A. N. Yaresko, V. Tsurkan, B. Keimer, A. Loidl, and A. V. Boris, *Phys. Rev. B* **85**, 100504(R) (2012).
- ²⁸A. Ricci, N. Poccia, G. Campi, B. Joseph, G. Arrighetti, L. Barba, M. Reynolds, M. Burghammer, H. Takeya, Y. Mizuguchi, Y. Takano, M. Colapietro, N. L. Saini, and A. Bianconi, *Phys. Rev. B* **84**, 060511 (2011).
- ²⁹F. Chen, M. Xu, Q. Q. Ge, Y. Zhang, Z. R. Ye, L. X. Yang, J. Jiang, B. P. Xie, R. C. Che, M. Zhang, A. F. Wang, X. H. Chen, D. W. Shen, J. P. Hu, and D. L. Feng, *Phys. Rev. X* **1**, 021020 (2011).
- ³⁰V. Ksenofontov, G. Wortmann, S. A. Medvedev, V. Tsurkan, J. Deisenhofer, A. Loidl, and C. Felser, *Phys. Rev. B* **84**, 180508 (2011).
- ³¹Y. Texier, J. Deisenhofer, V. Tsurkan, A. Loidl, D. S. Inosov, G. Friemel, and J. Bobroff, *Phys. Rev. Lett.* **108**, 237002 (2012).
- ³²S. C. Speller, T. B. Britton, G. M. Hughes, A. Krzton-Maziopa, E. Pomjakushina, K. Conder, A. T. Boothroyd, and C. R. M. Grovenor, *Supercond. Sci. Technol.* **25**, 084023 (2012).
- ³³R. H. Yuan, T. Dong, Y. J. Song, P. Zheng, G. F. Chen, J. P. Hu, J. Q. Li, and N. L. Wang, *Sci. Rep.* **2**, 221 (2012).
- ³⁴A. M. Zhang, J. H. Xiao, Y. S. Li, J. B. He, D. M. Wang, G. F. Chen, B. Normand, Q. M. Zhang, and T. Xiang, *Phys. Rev. B* **85**, 214508 (2012).
- ³⁵A. Charnukha, A. Cvitkovic, T. Prokscha, D. Pröpper, N. Ocelic, A. Suter, Z. Salman, E. Morenzoni, J. Deisenhofer, V. Tsurkan, A. Loidl, B. Keimer, and A. V. Boris, *Phys. Rev. Lett.* **109**, 017003 (2012).
- ³⁶M. Wang, C. Fang, D. Yao, G. Tan, L. W. Harriger, Y. Song, T. Netherton, C. Zhang, M. Wang, M. B. Stone, W. Tian, J. Hu, and P. Dai, *Nat. Commun.* **2**, 580 (2011).
- ³⁷J. T. Park, G. Friemel, Y. Li, J. H. Kim, V. Tsurkan, J. Deisenhofer, H. A. Krug von Nidda, A. Loidl, A. Ivanov, B. Keimer, and D. S. Inosov, *Phys. Rev. Lett.* **107**, 177005 (2011).
- ³⁸J. T. Park, D. S. Inosov, A. Yaresko, S. Graser, D. L. Sun, P. Bourges, Y. Sidis, Y. Li, J. H. Kim, D. Haug, A. Ivanov, K. Hradil, A. Schneidewind, P. Link, E. Faulhaber, I. Glavatsky, C. T. Lin, B. Keimer, and V. Hinkov, *Phys. Rev. B* **82**, 134503 (2010).
- ³⁹T. A. Maier, S. Graser, P. J. Hirschfeld, and D. J. Scalapino, *Phys. Rev. B* **83**, 100515 (2011).
- ⁴⁰M. Wang, C. Li, D. L. Abernathy, Y. Song, S. V. Carr, X. Lu, S. Li, Z. Yamani, J. Hu, T. Xiang, and P. Dai, *Phys. Rev. B* **86**, 024502 (2012).
- ⁴¹R. Bewley, R. Eccleston, K. McEwen, S. Hayden, M. Dove, S. Bennington, J. Treadgold, and R. Coleman, *Phys. B: Condens. Matter* **385-386**, 1029 (2006).
- ⁴²C. Lester, J.-H. Chu, J. G. Analytis, T. G. Perring, I. R. Fisher, and S. M. Hayden, *Phys. Rev. B* **81**, 064505 (2010).
- ⁴³G. Friemel, J. T. Park, T. A. Maier, V. Tsurkan, Y. Li, J. Deisenhofer, H. A. Krug von Nidda, A. Loidl, A. Ivanov, B. Keimer, and D. S. Inosov, *Phys. Rev. B* **85**, 140511 (2012).
- ⁴⁴T. A. Maier and D. J. Scalapino, *Phys. Rev. B* **78**, 020514 (2008).
- ⁴⁵D. S. Inosov, J. T. Park, P. Bourges, D. L. Sun, Y. Sidis, A. Schneidewind, K. Hradil, D. Haug, C. T. Lin, B. Keimer, and V. Hinkov, *Nat. Phys.* **6**, 178 (2010).
- ⁴⁶M. Liu, L. W. Harriger, H. Luo, M. Wang, R. A. Ewings, T. Guidi, H. Park, K. Haule, G. Kotliar, S. M. Hayden, and P. Dai, *Nat. Phys.* **8**, 376 (2012).
- ⁴⁷D. S. Inosov (private communication).
- ⁴⁸G. Friemel, W. P. Liu, E. A. Goremychkin, Y. Liu, J. T. Park, O. Sobolev, C. T. Lin, B. Keimer, and D. S. Inosov, *Europhys. Lett.* **99**, 67004 (2012).