CONDENSED MATTER

Magnetic scattering and vortex lattice in an iron-based ferromagnetic superconductor

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The magnetic properties of superconductors have a rich and interesting history. Early work showed that even tiny concentrations of magnetic impurities destroyed the superconducting pairing through the exchange-driven spin depairing mechanism, prohibiting any possibility of cooperative magnetic behavior [1]. The first exception to this rule was provided by the cubic rare-earth substituted $CeRu_2$ alloys, followed by the ternary Chevrel-phase (RMo_6S_8, R = rare earth) and related superconductors, where the first demonstrations of long-range magnetic order coexisting with superconductivity were obtained. The majority of these Chevrel-phase materials order antiferromagnetically where coexistence of long-range order with superconductivity is common, but these materials also provided the first examples of the rare occurrence of ferromagnetism and consequent electromagnetic competition with superconductivity. Antiferromagnetic order is also found for the Cu spins and all the rare earths in the cuprates, and for all the RNi_2B_2C borocarbides with the singular exception of $ErNi_2B_2C$ at low temperature (below 2.3 K) where a net magnetization developed that resulted in the spontaneous formation of flux quanta (vortices).

For the high-T_C iron-based superconductors of direct interest here, all the iron and rare-earth orderings have been antiferromagnetic in nature until very recently, with the apparent development of 3d iron ferromagnetism around 10 K in the form of spontaneous vortex creation in $(Li_{1-x}Fe_xOH)FeSe$, well below the superconducting transition temperature T_C which can be as high as 43 K. To investigate both the magnetic and superconducting properties of this system, we have carried out neutron diffraction and small-angle neutron scattering (SANS) measurements on a polycrystalline sample $(^7Li_{0.82}Fe_{0.18}OD)FeSe$, where the ^7Li isotope has been employed to avoid the neutron absorption of ^6Li, and H has been replaced by D to avoid the huge nuclear incoherent cross section [2].

High intensity powder diffraction measurements were carried out to search for magnetic Bragg peaks. For the present system no evidence for magnetic ordering was found. This perhaps is not surprising given that the ferromagnetism is expected to arise in the Li-Fe layer; with an expected moment of less than 1 μ_B/Fe and only 18 % of the sites occupied, the site-averaged ferromagnetic moment will be very difficult to observe in powder diffraction.

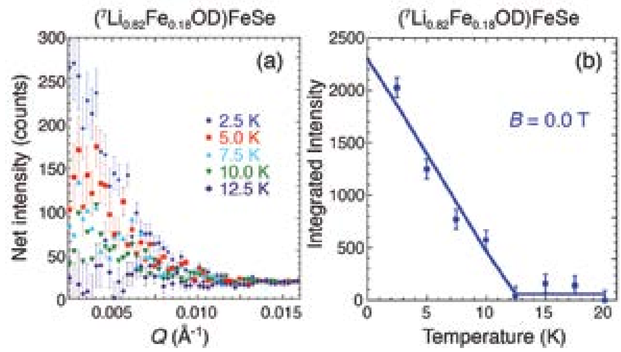

FIGURE 1: (a) Magnetic scattering as a function of wave vector Q for several temperatures. No magnetic field is applied. At 12.5 K very little magnetic scattering is observed, as is the case at higher temperatures (see text). Below the ferromagnetic transition magnetic scattering intensity develops, which increases monotonically with both decreasing Q and T. No peak in this Q range is observed, ruling out the formation of a long range ordered oscillatory magnetic state. (b) Integrated intensity as a function of temperature, revealing a magnetic transition temperature of ≈ 12.5 K. Uncertainties are statistical in origin and represent one standard deviation.

To detect long wavelength oscillatory magnetic order, pure ferromagnetism, and study the vortex scattering, small-angle neutron scattering (SANS) is the technique of choice. We have observed two separate components of magnetic scattering, one in zero applied field due to an inhomogeneous mixed state originating from either the spontaneous formation of vortices or ferromagnetic domains, while in an applied field we observe the scattering from a well-developed vortex lattice that arises below the superconducting transition temperature for this sample of T_C = 18 K [2]. Figure 1a shows the wave vector dependence of the difference scattering in this regime at several temperatures of interest, with data being taken between 2.5 K and 37.5 K in steps of 2.5 K. At and above 12.5 K no magnetic signal is observed, while for lower

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temperatures we observe a rapid increase in intensity with decreasing T. At each temperature this scattering increases monotonically with decreasing Q but otherwise does not appear to change its shape. We see that there is a well-defined onset of scattering at the onset of ferromagnetism at ≈ 12 K, which could originate from the spontaneous formation of vortices. They will be oriented randomly in the powder and most won't coherently Bragg diffract, but individual vortices will scatter and is likely the origin of the SANS scattering shown in Fig. 1.

SANS measurements in a horizontal applied magnetic field are shown in Fig. 2. Figure 2a shows the net intensity at 5 K upon cooling in a field of 0.4 T. In comparison with the data in Fig. 1a, we see that the smallest Q ferromagnetic scattering has reduced in intensity as expected since in this layered superconductor only some of the spontaneous vortices will align with the field. We also see a well-defined peak at larger Q. A least-squares fit to a (resolution-limited) Gaussian peak yields a position $Q = 0.0078(3)$ Å^{-1}. The expected position for a triangular vortex lattice is given by

$$Q_{10} = 2\pi\sqrt{\frac{2B}{(\sqrt{3})\phi_0}}$$

where $\phi_0 = 2.068\times10^5$ T Å^2 is the flux quantum and B is the internal field. For an applied field of 0.4 T the calculated position is $Q = 0.0077$ Å^{-1}, which is in excellent agreement with the measurement.

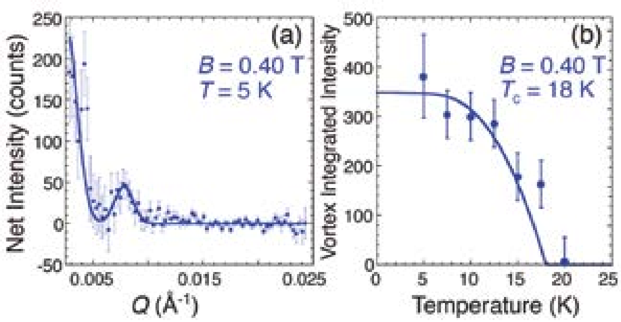

FIGURE 2: (a) Magnetic intensity at 5 K after cooling from 25 K in an applied field of 0.4 T. The ferromagnetic scattering has shifted to smaller Q, indicating that the length scale has increased or the strength for this scattering has decreased as would be expected when a field is applied. The peak at $Q = 0.0077$ Å^{-1} is due to the vortex lattice. (b) Integrated intensity of the vortex scattering as a function of temperature. The onset of scattering occurs at $T_C = 18$ K. No evidence of the ferromagnetic ordering is observed, indicating that the ordered moment is small (see text).

The temperature dependence of the vortex scattering, determined by integrating the net scattering over the peak is shown in Fig. 2b, develops a signal below T_C as expected. In the ferromagnetic state we expect an additional contribution from the internally generated magnetic flux, which would shift the vortex peak to larger Q, and increase its intensity. Neither trend is observed in these data, again indicating that the ferromagnetic moment is quite small.

For ferromagnetic superconductors like $ErRh_4B_4$, $HoMo_6S_8$ and $HoMo_6Se_8$ the magnetization that develops in the superconducting state competes with the Meissner screening through the London penetration depth. This competition results in a long wavelength oscillatory magnetic order that coexists with superconductivity, with a wavelength that is either strongly temperature dependent, or results in a strongly first-order transition to pure ferromagnetism where the superconductivity is destroyed. The present system certainly behaves differently in that we do not see any oscillatory magnetic order, thus leaving the other possibility that vortices spontaneously form. In a polycrystalline sample the vortices will be randomly oriented and it is very unlikely that a spontaneous vortex lattice could ever be observed, and indeed a true spontaneous vortex lattice—formed in the absence of an applied field—still remains to be observed in any ferromagnetic superconductor. Efforts to observe this will require single crystal samples, and such measurements are underway. What is clear is that $Li_{1-x}Fe_xOHFeSe$ is a fascinating ferromagnetic superconductor, and further measurements to investigate the magnetic order, spin fluctuations, lattice dynamics, and vortex structure in greater detail should prove very interesting.

References

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