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Investigation of the modulated magnetic phase of superconducting $HoMo_6S_8$

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Small-angle neutron scattering has been used to study the temperature and magnetic field dependence of the magnetic scattering in the superconducting phase of $HoMo_6S_8$. Just above the transition $T_m = 0.732$ K to the modulated phase, precursor scattering which peaks at $Q_0(0.030\ \text{Å}^{-1})$ has been observed. This peak is suppressed by modest ($\sim$20-Oe) fields, whereas below T_m the intensity of the Bragg peak at Q_0 is initially enhanced, achieving a maximum at $H \sim 200$ Oe.

The properties of Chevrel-phase superconductors containing magnetic rare-earth elements ($R\,Mo_6S_8$, R = rare earth) have received considerable attention recently because of the interesting interplay between magnetism and superconductivity these compounds display.[1] In cases where interactions between the magnetic ions favor antiferromagnetic alignment, the spatially averaged magnetization is zero and thus the electromagnetic coupling to superconductivity is weak. This leads to coexistence of long-range antiferromagnetic order with superconductivity.[2] Ferromagnetism, on the other hand, generates an internal magnetic field which couples strongly to the superconducting order parameter, leading to a competition between these cooperative states. In the only Chevrel phase where ferromagnetism has been observed, $HoMo_6S_8$, this competition results in an ordered state in which a long wavelength ($\lambda = 200$ Å) magnetic modulation coexists with superconductivity.[3,4] At lower temperatures pure ferromagnetic alignment is favored and the superconducting state is destroyed. Similar effects have been observed[5,6] in the related material $ErRh_4B_4$.

One of the puzzling features of these "ferromagnetic-superconductor" systems is that no precursor (critical) scattering associated with the transition to the modulated phase has been observed. Theoretically[7-10] a peak in the scattering at finite Q would be expected in the paramagnetic state because ferromagnetism favors long-wavelength (small-Q) fluctuations which are energetically the most costly magnetic fluctuations for superconductivity. The resulting compromise is expected to give rise to a peak at a characteristic wave vector Q_0. We have succeeded for the first time in observing precursor scattering which peaks at Q_0, but only in a very narrow temperature regime ($\sim$5 mK) just above T_m. The effect of a magnetic field on this scattering is particularly interesting. For $T > T_m$ the peak at Q_0 is suppressed isotropically (i.e., without regard for the relative orientation of $\vec{Q}_0$ and the magnetic field $\vec{H}$) by small fields (e.g., 20 Oe). In contrast, below T_m the intensity of the Bragg peak at Q_0 varies anisotropically with field. At low fields the intensity increases, reaching a maximum at $H \sim 200$ Oe, while fields in excess of 500 Oe suppress the peak at Q_0 in favor of ferromagnetic spin alignment.

Our experiments were carried out on the D-11 small-angle scattering spectrometer at the Institut Laue Langevin reactor. The powder sample of

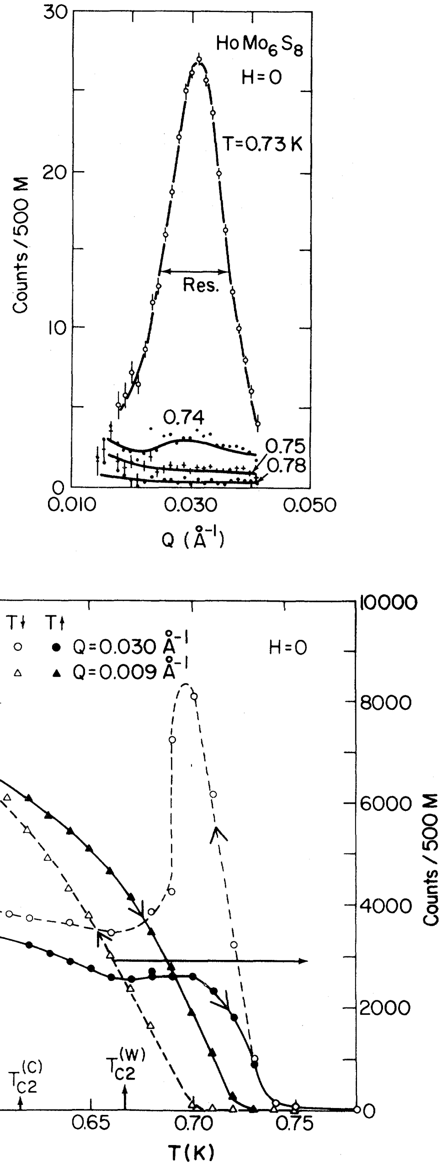

FIG. 1. Temperature dependence of the scattering with no applied field. (a) Wave-vector dependence. For $T > 0.740$ K there is no peak in the scattering at finite Q. For $T = 0.730$ K there is a Bragg peak at $Q_0 = 0.030$ Å^{-1}. (b) Temperature dependence of the scattering for the modulated component and the ferromagnetic (0.009 Å^{-1}) component. Note that the hysteresis is in the opposite sense for these two components.

$HoMo_6S_8$ has been well characterized[3,4]; it has an upper superconducting transition $T_{c1} = 1.82$ K, and a (first-order) reentrant transition T_{c2} at about 0.64 K. The sample was mounted in a dilution refrigerator equipped with a superconducting Helmholtz pair capable of producing a field of 2000 Oe. The spectrometer has a two-dimensional, position-sensitive detector which measures the scattering about the incident beam direction. Thus simultaneous measurements were made for all relative orientations of the magnetic field $\vec{H}$ and the wave vector $\vec{Q}$. We first summarize the results we have obtained in zero field, and then present the field-dependent data.

Above 0.750 K (Ref. 11) magnetic critical scattering which peaks at zero (or very small) wave vector is observed as shown in Fig. 1(a). In this figure the intensity is plotted as a function of the magnitude Q of the wave vector since the scattering from a powder is isotropic in the absence of an applied magnetic field. At 0.740 K there is a slight indication of a maximum at $Q_0 = 0.030$ Å^{-1}, which at 0.730 K has grown into a pronounced peak whose width is resolution limited under all experimental conditions.[4] Figure 1(b), which shows the temperature dependence of the intensity at Q_0, indicates that a magnetically modulated phase coexists with bulk superconductivity (MM-S) for $0.700 < T \leq 0.732$ K. Figure 1(b) also demonstrates that no ferromagnetic scattering is observed in this temperature interval (on cooling). The presence of a pure modulated magnetic phase contrasts with the results for $ErRh_4B_4$, where ferromagnetic and modulated magnetic components occur together over the entire temperature regime in which the modulated component is observed.[6]

For temperatures between 0.700 and T_{c2} a first-order transition proceeds from the MM-S state to a ferromagnetic, normal conducting (FN) state. We presume that in this temperature interval a mixed phase exists where some regions of the sample are in the MM-S state and others are in the FN state. The presence of the ferromagnetic component is manifested in these measurements by magnetic intensity at small Q, for example at 0.009 Å^{-1} as shown in Fig. 1(b). Both the neutron scattering and ac susceptibility measurement indicate that the system takes several hours to equilibrate between T_{c2} and 0.700 K. In this same temperature interval hysteresis is observed in all three order parameters of the system—ferromagnetic, magnetic modulation, and superconductivity. Below T_{c2} superconductivity disappears and the system is a pure ferromagnet.

Figure 2 shows the scattering at 0.735 K, just above T_m. A peak at Q_0 is observed, with a width which is considerably larger than the instrumental resolution. The development of this peak in the scattering as a function of temperature is much too rapid to be explained by conventional critical scattering theory. As a function of applied magnetic field we find that the intensity changes isotropically. For $H = 10$ Oe we observe no change in intensity, whereas for larger fields the intensity is reduced steadily with increasing H. The isotropy of the scattering indicates that the field-induced change of intensity orginates in the energetics of the system and

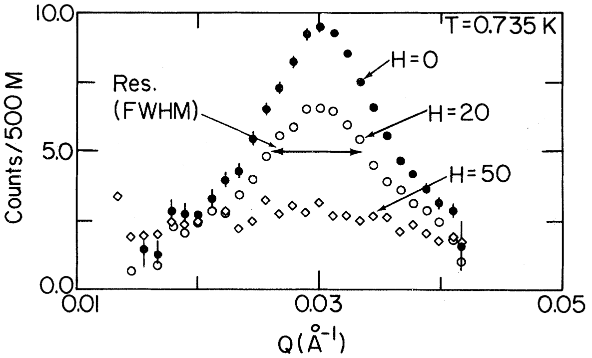

FIG. 2. Wave-vector dependence of the precursor scattering at 0.735 K at several applied fields. The intensity of the scattering is isotropic and shows no hysteresis.

is not due to alignment of the spins by the external field.

For a slightly lower temperature of 0.730 K, in the ordered MM-S phase, the Bragg peak intensity is anisotropically affected by a field as shown in Fig. 3. Here the intensity for $\vec{H} \parallel \vec{Q}$ and $\vec{H} \perp \vec{Q}$ is plotted for Q_0. The intensity initially increases with increasing field, and then decreases at larger fields. Upon returning to smaller fields there is hysteresis, and the data suggest that there may be a remanent field in the system. Note that the intensity after cycling the field is larger than that observed in the virgin state; this is consistent with the initial field-increasing data and the assumption of a remanent field. In addition to an increased modulated component we observe some intensity in the small-Q ferromagnetic component after field cycling. These data were obtained by cooling in zero field, and then isothermally increasing H; cooling in a finite field gives qualitatively similar results, but quantitatively does not yield the same intensity distribution for any (H,T). Hysteretic effects were observed below T_m as a function of field *and* temperature, including $H=0$ as shown in Fig. 1(b). Careful zero-field measurements close to T_m demonstrate that this behavior persists up to the transition, suggesting that the transition from the paramagnetic to the magnetically modulated state is also discontinuous.

It is tempting to relate the temperature interval $0.732 < T \leq 0.74$ K in which we observe precursor scattering to the theoretical interval $T_m < T < T_F^*$, where T_F^* is the ferromagnetic transition temperature which would apply if superconductivity were absent.[7-10] Ferrell and Bhattacharjee[8] have proposed that enhanced precursor scattering should occur below T_F^* as a result of vortices induced by an externally applied field. These vortices provide a driving field which couples to the susceptibility of the $4f$ holmium electrons and enhances their response around Q_0. The fact that we observe enhanced scattering in zero field suggests that vortices may form spontaneously,[7,9,10] below T_F^*. The scattering from such vortices, however, would be expected to occur at wave vectors smaller than those accessible in the present experiments. Our observations of the asymmetry of the scattering in a magnetic field demonstrate that the scattering in the MM-S state cannot originate *directly* from a vortex lattice. The scattering could result from a spiral magnetic state,[7,8] or from the laminar state proposed by Tachiki.[9]

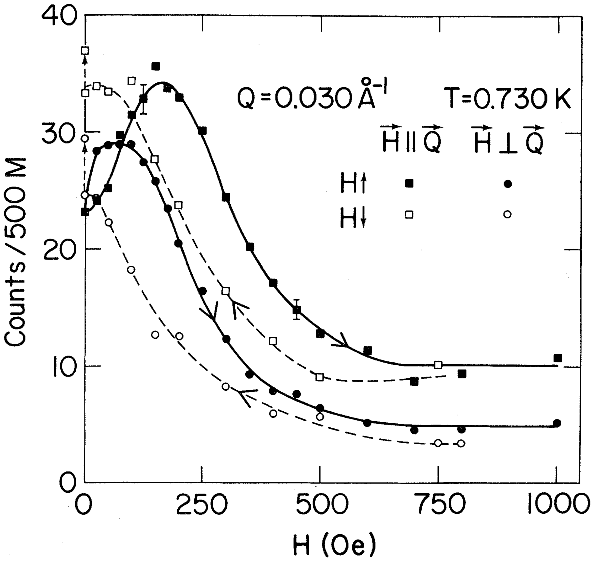

FIG. 3. Magnetic field dependence of the Bragg peak intensity in the modulated phase. Note that the intensity of the scattering is anisotropic and shows strong hysteretic effects.

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[11] The relative temperature accuracy is ±1 mK. The absolute temperature accuracy, however, is only ±20 mK.