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SMALL-ANGLE NEUTRON SCATTERING OF $DyMo_6S_8$ AT LOW TEMPERATURE

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Abstract

The Chevrel phase compound $DyMo_6S_8$ becomes superconducting at 2.05 K and is antiferromagnetic (AFM) below $T_M = 0.4$ K. Experiments with an applied magnetic field have shown that the AFM state is gradually suppressed and a ferromagnetic component seems to be induced before H_{c2} (1200 Oe) is reached. The present experiments have been performed at the small-angle scattering facility D11 at the Institut Laue Langevin with a dilution refrigerator and applied fields of up to 2000 Oe. Neutron wavelengths of 6.25 and 10Å have been used allowing a minimum wavevector $Q_{min} \simeq 0.005$ Å^{-1} to be achieved. For increasing H we find a sudden increase in the small-angle scattering at H = 180 Oe, which we identify as H_{c1}. With a position sensitive detector we have also investigated the scattering as a function of the relative orientation of $\vec{Q}$ and $\vec{H}$. For a simple paramagnet we would expect more magnetic intensity with $\vec{Q} \perp \vec{H}$ than with $\vec{Q} \parallel \vec{H}$, however, we find the exact reverse with somewhat more intensity for $\vec{Q} \parallel \vec{H}$. This is not understood, but we propose that the small-angle scattering is predominantly inelastic. Similar anisotropic effects have been found in other Chevrel phase systems.

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Introduction

The ternary Chevrel compound $DyMo_6S_8$ becomes superconducting at $T_c = 2.05$ K and is antiferromagnetic (AFM) below $T_M = 0.40$ K.[1] The magnetic field required to destroy superconductivity H_{c2} has been measured[2] on the sample used in the present experiments as 1200 Oe. Recent neutron diffraction experiments[3] have shown that fields $H < H_{c2}$ applied in the magnetic state ($T < T_M$) cause a partial suppression of antiferromagnetism and the appearance of ferromagnetic (FM) "Bragg peaks", in spite of the persistence of superconductivity.

A considerable amount of work has recently been reported on the coexistence of ferromagnetic order and superconductivity. In particular, neutron experiments on $HoMo_6S_8$ (Ref. 4 and 5) and $ErRh_4B_4$ (Ref. 6) have addressed this problem. It appears that, as expected, ferromagnetism (i.e. $q = 0$ magnetic order) and superconductivity probably do not exist in the same spatial region of the crystal. In $DyMo_6S_8$ at zero field this problem does not arise because the ordering is antiferromagnetic and the coupling between the magnetic and superconducting electrons is predominantly electromagnetic. Since the AFM period ($= 2\pi/q_m$ with $q_m = 0.40$ Å^{-1}) is less than the London penetration depth, AFM order and superconductivity may coexist.[7] However, FM ordering gives rise to a macroscopic dipolar field which would destroy superconductivity. In $HoMo_6S_8$ the ordering in the intermediate regime at temperatures just above that at which the FM state destroys superconductivity displays[4,5] a modulated magnetic phase with a wavevector of $Q = 0.030$ Å^{-1}; this state is a result of the competition between the FM and superconducting order parameters[7]. With the instrumental arrangements as in Ref. 3 the Q resolution was ~ 0.022 Å^{-1}, so that modulations with smaller Q values than this would appear as FM correlations. One principle aim of the present experiment was to investigate the possible occurence of a magnetic modulation in $DyMo_6S_8$ in an applied field with a resolution of ~ 0.005 Å^{-1}.

Experimental Details and Results

The experiments were performed at the D11 small-angle diffractometer at the Institut Laue Langevin, Grenoble, France. Neutrons of wavelengths 6.25 Å or 10 Å from a cold source were incident on the sample. The sample (~ 3g) was mounted in a special cell[4] with single-crystal sapphire windows and placed in a dilution refrigerator, which was equipped with a superconducting Helmholtz pair capable of producing a field of 2000 Oe in a direction perpendicular to the incident neutron beam. The neutron detector, which was 10 m from the sample, was planar and position sensitive, thus covering a Q range about the origin of reciprocal space from ~ 0.002 $Å^{-1}$ (depending on the beam stop configuration) to ~ 0.165 $Å^{-1}$. With this configuration simultaneous measurements were made for all relative orientations of the magnetic field $\vec{H}$ and the scattering wavevector $\vec{Q}$. Note, however, that the first AFM reflection appears at Q = 0.400 $Å^{-1}$, which is outside the range studied in this experiment.

The experiment consisted of cooling the sample and then applying a magnetic field. The total counts registered on the detector per second are plotted in Fig. 1 as a function of applied field. We identify the break in the total intensity curve as H_{c1}, the field at which the field begins to penetrate the superconducting sample and additional magnetic scattering is induced in the forward direction. In the non-magnetic state ($T > 0.40$ K) the increase in signal is still present, although less marked, at the same value of H_{c1} as found for $T < 0.40$ K. The difference in the approach to saturation of the curves for T = 0.16 and 0.6 K may be understood by recalling the very rapid onset of saturation of the Dy moments below T_M (see Fig. 5 of Ref. 3). Thus at higher temperatures the full moment is more difficult to align. Presumably this is due to a series of low lying crystal-field states although details of such states are as yet unavailable. We note from Ref. 3 that the Dy antiferromagnet moment at 0.1 K is ~ 9 μ_B, just below the free ion value of 10 μ_B. No abrupt change in the induced moment is observed at the value of H_{c2} given by Ref. 2.

The next point to consider is the wavevector distribution of the small-angle scattering. From a series of difference patterns $I(H > 0) - I(H = 0)$ we can conclude that there is no appreciable difference between the spatial aspect of the additional magnetic scattering above or below T_m and that there is no apparent peak at finite values of Q. These runs are with the counts radially averaged over the whole detector to give good statistics. Given the limitations of the beam stop size, and the very high absorption (by Dy) of neutrons with long wavelength (10 - 15 Å), our minimum Q value is ~ 0.005 Å^{-1}. It thus appears that the range of the induced ferromagnetic correlations exceeds 1200 Å in the superconducting state. We should, however, note the caution of Ref. 3, that this does not necessarily imply that ferromagnetism is coexisting with superconductivity in the same manner as the AFM state. Induced FM order may occur in domains or along flux lines. The neutron experiments cannot give a detailed microscopic picture of <u>where</u> the FM order is developing, since they integrate over the whole sample.

Finally, we have studied the dependence of the induced FM scattering on the relative orientation of $\vec{Q}$ and $\vec{H}$. For a simple induced moment system we would expect $\vec{\mu}_{ind} \parallel \vec{H}$, where $\vec{\mu}_{ind}$ is the induced magnetic moment. Since only those components of $\vec{\mu}_{ind}$ which are perpendicular to $\vec{Q}$ scatter neutrons we would expect to observe more scattered intensity for $\vec{Q} \perp \vec{H}$ if the scattering were elastic. Indeed, in the absence of crystal field or exchange anisotropy we would expect no elastic scattering for $\vec{Q} \parallel \vec{H}$.

We show in Fig. 2 a series of plots of intensity (I) versus Q for different conditions of T and applied field H. Note that these are already difference counts (i.e. the background, taken with H = 0, has been subtracted) and that we have treated only those portions of the detector in angular sectors of ± 15° near the $Q_\perp$ and $Q_\parallel$ positions. A number of statements can immediately be made.

(1) At low T $I_\parallel$ (intensity with $\vec{Q} \parallel \vec{H}$) > $I_\perp$ (intensity with $\vec{Q} \perp \vec{H}$).

(2) This difference is approximately proportional to H and does not disappear when $H > H_{c2}$, i.e. when the material is no longer superconducting [2(b)].

(3) Consistent with Fig. 1 the total counts decrease significantly with increasing temperature [2(d)].

(4) The difference counts also decrease above T_M, but the anisotropy remains [2(d)].

(5) The counts are much less for $\lambda = 10$ Å because of the high absorption by Dy, but the anisotropy and approximate Q dependence are similar for the two incident neutron wavelengths [2(c) and 2(e)]. This is a good check on the instrument and data reduction technique.

(6) Because of the rapid loss of magnetic response with increasing T very little moment can be induced with 2000 Oe at $T > T_c$, and any anisotropy is certainly outside statistics [2(f)].

Conclusions

In this experiment we have confirmed the results of Ref. 3 but with improved resolution. Indeed, an apparent $\vec{q} = 0$ component of magnetization is found for $H < H_{c2}$.

In Fig. 2 we show that the scattering is anisotropic with $I_{\parallel} > I_{\perp}$. This is exactly the reverse of what we expect for elastic scattering from a simple induced magnet system. That this anisotropy persists for $H > H_{c2}$ excludes the possibility of our having seen complicated effects from a spin-flopped vortex lattice, and also suggests the effects have little directly to do with superconductivity. Similar anisotropic effects have been seen [5] in $HoMo_6S_8$, although these are mainly associated with the long-wavelength scattering at $Q \gtrsim 0.03$ $Å^{-1}$. The anisotropy in neither system is understood. One explanation, which may pertain in the case of $DyMo_6S_8$, is that the neutron scattering is predominantly inelastic in origin. Since the Chevrel phase compounds order with almost the full free-ion moment, there may be a series of almost free-ion levels very closely spaced. Transverse ($\Delta M = \pm 1$) matrix elements would then dominate both the crystal-field and spin-wave responses. Inelastic scattering from such components does indeed have the correct symmetry to produce $I_{\parallel} > I_{\perp}$. Such a hypothesis, which still needs further investigation, opens up new possibilities for understanding the interplay between superconductivity and magnetism in these compounds.

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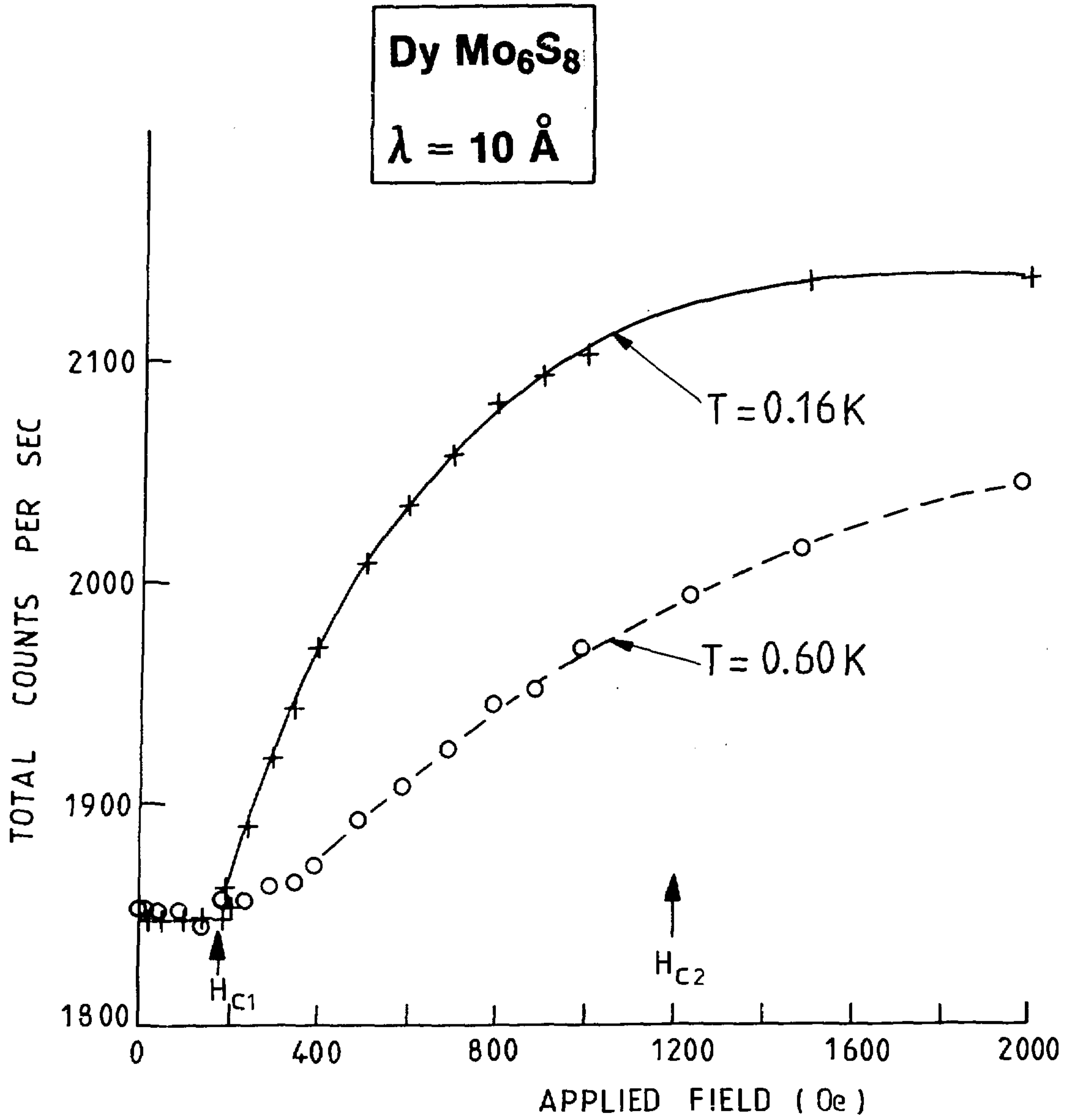
Figure Captions

Fig. 1 Intensity (integrated over the whole detector) as a function of applied field at two different temperatures near $T_M = 0.4$ K.

Fig. 2 A series of I vs Q plots in the applied fields and temperatures noted in each panel. In each case the net counts are determined by subtracting the results with $H = 0$. To illustrate the anisotropy we have treated separately counts with an arc of ± 15° from $\vec{Q} \parallel \vec{H}$ (open circle) and $\vec{Q} \perp \vec{H}$ (closed circles).



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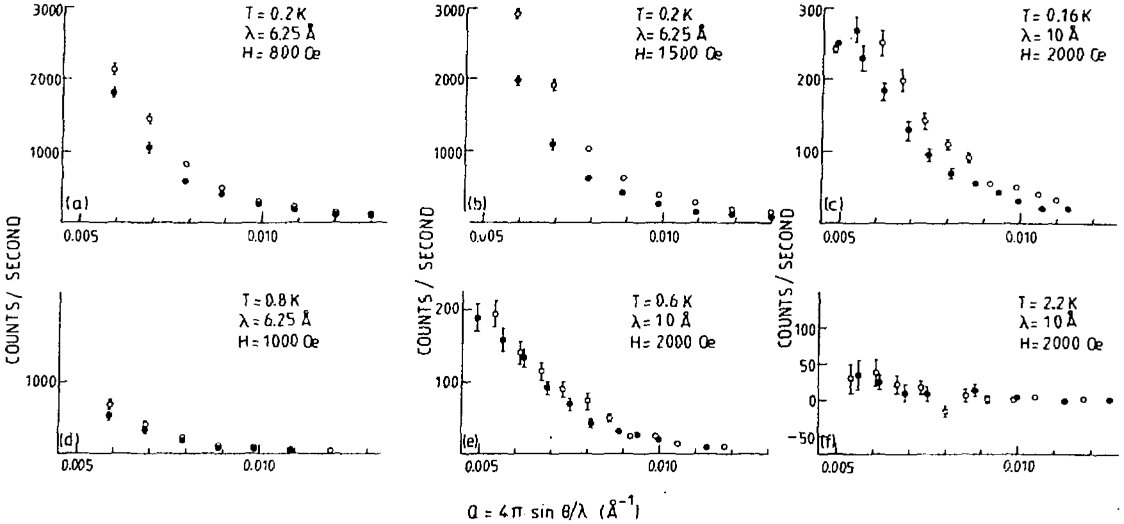

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