Competition between Ferromagnetism and Superconductivity in $HoMo_6S_8$

J. W. Lynn
Department of Physics, University of Maryland, College Park, Maryland 20742

and

G. Shirane and W. Thomlinson
Department of Physics, Brookhaven National Laboratory, Upton, New York 11973

and

R. N. Shelton
Ames Laboratory—U. S. Department of Energy, Ames, Iowa 50011, and Department of Physics, Iowa State University, Ames, Iowa 50011

(Received 19 August 1980)

Neutron scattering has been used to investigate the development of magnetic order in the region of the reentrant superconducting transition in $HoMo_6S_8$. On cooling, an ordered oscillatory magnetic state develops in the superconducting phase, with a wavelength which increases with the application of a magnetic field. On further cooling the sample reverts to the normal conducting state as ferromagnetism sets in. No oscillatory magnetic phase is observed, however, when the superconducting phase is approached from low temperatures.

PACS numbers: 75.25.+z, 61.12.Dw, 74.40.Rv

The relationship between magnetism and superconductivity has received renewed attention since the discovery of ternary superconducting materials which also achieve long-range magnetic order at low temperatures.[1] For materials which develop antiferromagnetic order[2] the superconducting state has been found to be preserved so that there is true coexistence of long-range magnetic order and superconductivity. Ferromagnetic alignment, on the other hand, can be expected to be strongly opposed by superconductivity. In the two examples discovered so far, $HoMo_6S_8$[3,4] and $ErRh_4B_4$,[5-7] the ferromagnetic state destroys the superconductivity at sufficiently low temperatures. At intermediate temperatures, however, the competition between these two cooperative states may give rise to a compromise oscillatory magnetic behavior, and considerable theoretical effort has been devoted to elucidating various possibilities and their origins.[8-15] It is the purpose of this Letter to report the direct observation of an ordered oscillatory magnetic state in $HoMo_6S_8$ which coexists with superconductivity.

The measurements were carried out at the Brookhaven National Laboratory high-flux beam reactor with use of a triple-axis neutron spectrometer, with a 4-g polycrystalline sample mounted in a ^{3}He cryostat. Bulk susceptibility measurements on this same sample yield a superconducting transition temperature of 1.82 K, and a reentrant superconducting transition of $T_S^{(C)} = 0.612$ K on cooling and $T_S^{(W)} = 0.668$ K on warming.

Figure 1 shows the observed magnetic scattering, with background measured at 2.0 K subtracted, as a function of wave vector for several tem-

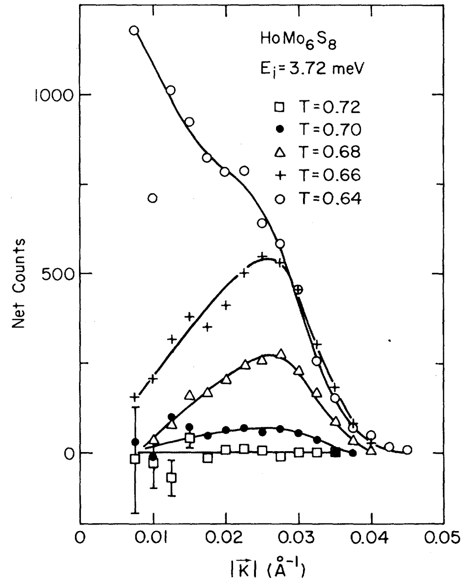

FIG. 1. Net scattering intensity, after subtraction of background at 2.0 K, as a function of wave vector for various temperatures. The peak at finite wave vector, which is limited in width by the instrumental resolution, indicates that an oscillatory magnetic state has developed on cooling.

peratures. The incident energy was 3.72 meV, with 10′ full width at half maximum collimation before and after the sample. The vertical divergence was also restricted in these measurements. At $T = 0.70$ K we begin to detect scattering which has a peak at a finite wave vector of $|\vec{K}| \simeq 0.025$ Å^{-1}, which corresponds to a wavelength in real space of ~250 Å. The strength of this scattering increases with decreasing temperature with little change in the wave vector down to 0.66 K. The observed width of the peak is limited by the instrumental resolution, which provides a lower limit to the coherence length of the oscillation of 1500 Å. Since the reentrant superconducting transition temperature for this sample is 0.612 K on cooling, the sample is most definitely still superconducting. At lower temperatures additional scattering develops at smaller wave vectors, and for $T < 0.64$ there is no longer a peak in the scattering at a finite wave vector. This additional small-angle scattering persists to low temperatures, and is likely due to scattering from domain walls since the overall intensity is proportional to the square of the magnetization. To establish the origin of this low-temperature scattering higher-resolution measurements capable of probing smaller wave vectors will be needed.

This peak in the scattering is fundamentally different from that expected for a conventional second-order ferromagnetic phase transition, where the temperature evolution of the critical scattering as one approaches the ordered state can be described to a good approximation by a wave vector dependent susceptibility $\chi(|\vec{K}|)$ of the Ornstein-Zernike form

$$\chi(|\vec{K}|) \propto \frac{1}{|\vec{K}|^2 + (1/\xi)^2}.$$

Here ξ is the range in real space of the correlations between spins. Typically $\xi(T)$ increases as the transition temperature T_m is approached from above and diverges at T_m. The scattering at fixed temperature (ξ fixed) is maximum at $|\vec{K}| = 0$ and monotonically falls to zero with increasing $|\vec{K}|$. In addition $\chi(\vec{K} = 0)$ diverges at T_m, which is the characteristic feature of a ferromagnetic transition. For a coupled ferromagnetic superconductor, on the other hand, one can argue on rather general grounds[8-15] that the long-wavelength (small-$|\vec{K}|$) magnetic fluctuations (either static or dynamic) will be energetically costly for the superconductor. The basic reason for this is that the most favorable configuration for ferromagnetism is parallel alignment of the spins. The associated macroscopic magnetic field, however, will have an effect similar to an applied field, which is of course energetically unfavorable for the superconductor. The system may then compromise by forming an oscillation in the magnetization, the criterion being that the wavelength be as long as possible for the ferromagnetism, but not long compared to the superconducting penetration depth. This oscillatory tendency will give rise to a scattering function with a maximum at some finite wave vector Q_c, which would correspond to critical fluctuations, or to a long-range-ordered state with a Bragg peak at Q_c as in the present case.

Figure 2 shows the temperature dependence of the intensity at a wave vector of 0.027 Å^{-1} near the maximum. With no magnetic field applied, the oscillatory magnetic state rapidly develops

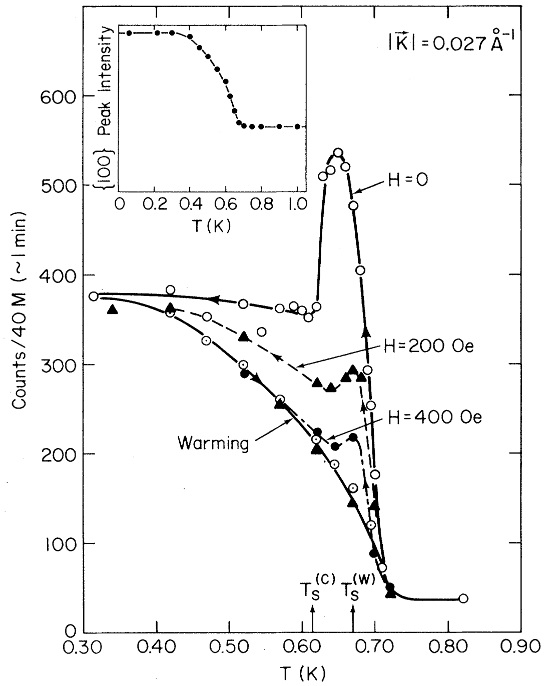

FIG. 2. Temperature dependence of the observed scattering intensity at a wave vector near the maximum. On cooling, the intensity rapidly rises as the oscillatory state develops, then decreases at lower temperature as ferromagnetism sets in. The application of a magnetic field is seen to suppress the scattering at this wave vector. On warming, no oscillatory state is observed, and the field has little effect on the scattering. The inset shows the temperature dependence of the {100} Bragg peak, which under higher resolution shows a small intensity difference on heating vs cooling (Ref. 4).

on cooling and reaches a maximum at $T \sim 0.65$ K. The reduction in intensity below $T_S^{(C)}$ is apparently associated with the reentrance to the normal conducting state as determined by bulk susceptibility measurements. Warming from low temperatures follows quite a different curve. There is no evidence of the oscillatory state in these warming data, and in fact we find no peak in the scattering as a function of wave vector at any temperature on warming. We remark that the bulk susceptibility measurements show the superconducting transition to have hysteresis, with the midpoint at $T_S^{(W)} = 0.668$ K on warming as indicated in Fig. 2. The general temperature behavior of the magnetic transition is shown in the inset to Fig. 2. Here the peak intensity of the $\{100\}$ Bragg position is plotted versus temperature, and consists of a temperature-independent nuclear contribution and the magnetic contribution.

The application of a magnetic field has a rather marked effect on the oscillatory state. The scattering intensity at the peak wave vector (Fig. 2) is suppressed by the field, and above $H \sim 500$ Oe apparently the scattering intensity would follow a single reversible curve on both heating and cooling. Note that on heating the magnetic field has no discernible effect. The measured wave-vector dependence of the scattering at several applied magnetic fields is shown in Fig. 3. The intensity is seen to shift to *smaller* wave vectors with increasing field, which implies that the wavelength of the oscillatory state increases toward the ferromagnetic limit $\lambda \to \infty$.

These data clearly demonstrate that the competition between the ferromagnetism and superconductivity gives rise to an oscillatory magnetization on cooling, with the width of the observed oscillatory peak (Fig. 1) being limited by the instrumental resolution. We have also been able to observe the oscillatory nature of the scattering around the $\{100\}$ powder Bragg peak. In addition, we have observed a long time constant to approach equilibrium upon entering or exiting the oscillatory state on cooling. These data taken together show that the oscillatory state is long range in character, a possibility which has been predicted theoretically with use of a Ginzberg-Landau approach[9] to this coupled-order-parameter problem. Another possibility which has been suggested is the formation of a spontaneous vortex lattice.[11, 13-15] In this case, however, we would expect the scattering to shift to larger wave vector with increasing field rather than to smaller wave vector as observed.

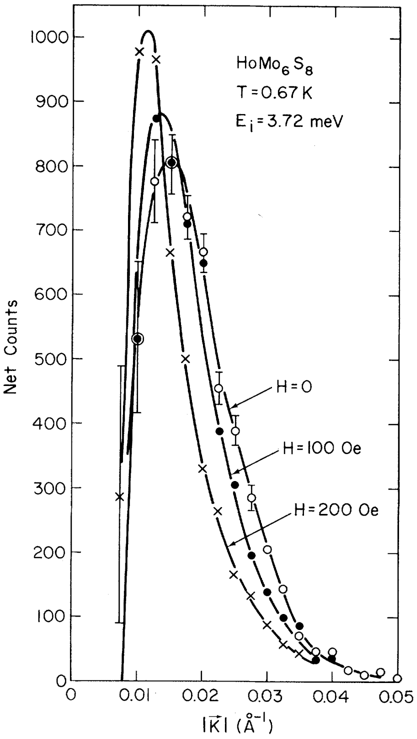

FIG. 3. Scattering as a function of wave vector at several values of the applied magnetic field, showing that the intensity shifts to smaller wave vectors with increasing field.

A peak in the small-angle neutron scattering has also been observed in the reentrant-superconductor system $ErRh_4B_4$ (Refs. 2 and 7). In contrast to the $HoMo_6S_8$, the intensity of the oscillatory peak in the $ErRh_4B_4$ case was found to be much weaker than expected for an ordered state, and it was therefore concluded that the peak (at 0.06 Å^{-1}) was most likely due to fluctuation effects resulting from the competition between the superconductivity and ferromagnetism. The complication of an additional magnetic component to the scattering in the same temperature region made it difficult to determine the width of the oscillatory peak, and hence left open the question of whether the oscillatory magnetization in that system is also long range in character.

We would like to thank H. C. Hamaker for carrying out some of the susceptibility measurements, and D. E. Moncton for helpful discussions. Work at the University of Maryland was supported by the National Science Foundation under Contract No. DMR 79-00908, at Brookhaven National Laboratory by the U. S. Department of Energy under Contract No. DE-AC02-76cH00016, and at Ames

Laboratory by the U. S. Department of Energy under Contract No. W-7405-Eng-82/WPAS-KC-02-02-02.

[1] See, for example, Ø. Fischer, Appl. Phys. 16, 1 (1978).

[2] See, for example, the review article by D. E. Moncton, G. Shirane, and W. Thomlinson, J. Magn. Magn. Mater. 14, 172 (1979); D. E. Moncton, J. Appl. Phys. 50, 1880 (1978).

[3] M. Ishikawa and Ø. Fischer, Solid State Commun. 23, 37 (1977); L. D. Woolf, M. Tovar, H. C. Hamaker, and M. B. Maple, Phys. Lett. 74A, 363 (1979).

[4] J. W. Lynn, D. E. Moncton, W. Thomlinson, G. Shirane, and R. N. Shelton, Solid State Commun. 26, 493 (1978).

[5] W. A. Fertig, D. C. Johnston, L. E. DeLong, R. W. McCallum, M. B. Maple, and B. T. Matthias, Phys. Rev. Lett. 38, 987 (1977).

[6] D. E. Moncton, D. B. McWhan, J. Eckert, G. Shirane, and W. Thomlinson, Phys. Rev. Lett. 39, 1164 (1977).

[7] D. E. Moncton, D. B. McWhan, P. H. Schmidt, G. Shirane, W. Thomlinson, M. B. Maple, H. B. Mackay, L. D. Woolf, Z. Fisk, and D. C. Johnston, to be published.

[8] P. W. Anderson and H. Suhl, Phys. Rev. 116, 898 (1959).

[9] E. I. Blount and C. M. Varma, Phys. Rev. Lett. 42, 1079 (1979); C. M. Varma, in *Superconductivity in d- and f-Band Metals*, edited by H. Suhl and M. B. Maple (Academic, New York, 1980), p. 391.

[10] R. A. Ferrell, J. K. Bhattacharjee, and A. Bagchi, Phys. Rev. Lett. 43, 154 (1979).

[11] M. Tachiki, H. Matsumoto, and H. Umezawa, Phys. Rev. B 20, 1915 (1979).

[12] M. V. Jaric and M. Belic, Phys. Rev. Lett. 42, 1015 (1979).

[13] S. Maekawa, M. Tachiki, and S. Takahashi, J. Magn. Magn. Mater. 13, 324 (1979).

[14] C. G. Kuper, M. Revzen, and A. Ron, Phys. Rev. Lett. 44, 1545 (1980).

[15] M. Tachiki, H. Matsumoto, T. Koyama, and H. Umezawa, Solid State Commun. 34, 19 (1980).

Dispersive Transport and Recombination Lifetime in Phosphorus-Doped Hydrogenated Amorphous Silicon

J. M. Hvam[a] and M. H. Brodsky
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

(Received 14 May 1980)

The recombination lifetime in phosphorus-doped hydrogenated amorphous silicon is determined from the dispersive photocurrent decay following a short-pulse excitation. The electron drift mobility and dispersion parameter α are obtained as well. It is found that α is temperature dependent as expected for extended-state transport controlled by multiple trapping. A lower limit to the extended-state mobility is determined: $\mu_c \gtrsim 1\ \mathrm{cm^2/V\cdot s}$.

PACS numbers: 72.40.+w, 72.80.Ng

Transient photocurrent (TP) with a short pulse excitation is a simple but powerful technique for separating transport parameters that are lumped together in ordinary dc measurements. By phosphorus doping of hydrogenated amorphous Si (a-Si:H), we have observed TP decays extending over several orders of magnitude in time, allowing for a detailed analysis of the form of the decay. We introduce a new and direct measure of the carrier lifetimes and our results confirm that the increase in photoconductivity with phosphorus doping, previously observed,[1,2] is mainly due to an increase in recombination lifetime. For n-type a-Si:H, the dispersive nature[3] of the transport of the photoexcited carriers (electrons) is clearly revealed, and we have been able to investigate its doping and temperature dependence in detail. We observe for the first time that the dispersion is temperature dependent. We take this as evidence for trap-controlled[3-5] transport in extended states and obtain a lower limit to the extended-state mobility.

The TP decay was studied in a-Si:H films produced by glow discharge of SiH_4 with PH_3 added for n-type doping. The contacts were predeposited sputtered Mo stripes, 1 mm apart. The deposition of the a-Si:H films took place in an inductively excited 0.07-Torr plasma onto 250 °C fused-quartz substrates. The TP was excited by a N_2-laser-pumped dye laser (Rh6G) with peak power, pulse width, and repetition rate of 1 kW, 3 ns, and 20 Hz, respectively. The laser (photon ener-