PHYSICAL REVIEW B VOLUME 36, NUMBER 13 1 NOVEMBER 1987

Suppression of superconductivity by antiferromagnetism in $Tm_2Fe_3Si_5$

J. A. Gotaas
Institute for Materials Science and Engineering, National Bureau of Standards, Gaithersburg, Maryland 20899

J. W. Lynn
Department of Physics, University of Maryland, College Park, Maryland 20742 and National Bureau of Standards, Gaithersburg, Maryland 20899

R. N. Shelton* and P. Klavins*
Ames Laboratory and Department of Physics, Iowa State University, Ames, Iowa 50011

H. F. Braun
Institut für Physik, Universität Bayreuth, D-8580 Bayreuth, Federal Republic of Germany

(Received 24 July 1987)

Powder neutron-diffraction experiments at pressures up to 8.2 kbar and temperatures down to 0.3 K have been performed on the antiferromagnetic ternary compound $Tm_2Fe_3Si_5$. For pressures between 2 and 21 kbar, this system becomes superconducting at $T_{c1} > T_N$, with a subsequent reentrance to the normal conducting state at $T_{c2} \approx T_N$. Our measurements demonstrate that the antiferromagnetic structure is unchanged under pressure, with no evidence for a ferromagnetic component at any pressure that would compete with the superconducting phase. This is the first experimental observation of the quenching of superconductivity by a purely antiferromagnetic state.

There has been intense interest in ternary rare-earth (R) compounds since the discovery in the RRh_4B_4 and $RMo_6(S,Se)_8$ systems of both the interesting competition between ferromagnetism and superconductivity and the coexistence of antiferromagnetic ordering with superconductivity.[1] The ternary system $R_2Fe_3Si_5$ displays a variety of unusual phenomena, including several examples of the occurrence of superconductivity in the absence of magnetic ordering (R = Y, Lu, or Sc),[2] which is possible since the iron atoms carry no moment.[3,4] In the case of many of the magnetic rare earths, antiferromagnetic ordering is found without superconductivity (R = Gd, Tb, Dy, Ho, Er, Tm, or Yb).[3,5] The particular case of $Tm_2Fe_3Si_5$ is interesting because it orders antiferromagnetically at ambient pressure with a rather low transition temperature of $T_N = 1.1$ K,[5,6] while the application of pressure drives the system superconducting, as shown[6] in Fig. 1. The most interesting feature of this phase diagram is the line forming the lower boundary of the superconducting phase (SP), where the SP phase is suppressed once the antiferromagnetic ordering takes place. If the ordered magnetic state is truly antiferromagnetic, then this represents a unique experimental observation of the destruction of superconductivity by antiferromagnetism in rare-earth ternary compounds. In order to verify this important result, we have examined $Tm_2Fe_3Si_5$ via neutron diffraction under applied pressures up to 8.2 kbar and temperatures down to 0.3 K.

The neutron experiments were performed at pressures of 2, 4.5, 5.8, and 8.2 kbar, which span the region of interest up to the maximum T_c. In each case, the powder sample was placed in an aluminum pressure cell which was pressurized at room temperature and clamped.[7] The pressure was calibrated by directly measuring the superconducting transition temperature via ac susceptibility. For the diffraction experiments, the pressure cell was enclosed in an aluminum sample chamber, which was filled with an STP of helium gas to ensure good thermal conductivity at low temperatures, and then mounted in a pumped ^3He cryostat capable of reaching 0.3 K. The diffraction measurements were carried out on a variety of spectrometers at the National Bureau of Standards Reac-

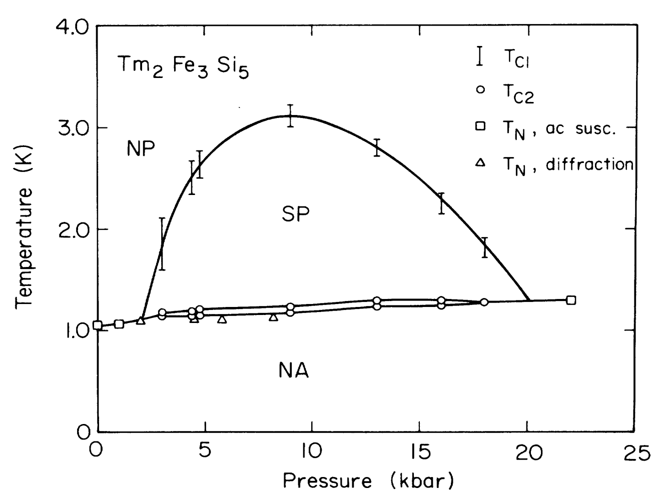

FIG. 1. Temperature-pressure phase diagram of $Tm_2Fe_3Si_5$, showing the normal-paramagnetic (NP), superconducting-paramagnetic (SP), and normal-antiferromagnetic phases (NA). The transition temperatures have been determined by ac susceptibility (T_{c1},(I); T_{c2},○; T_N,□) and neutron diffraction (T_N,△).

tor, utilizing a pyrolytic graphite monochromator and filter and a typical incident neutron wavelength of 2.35 Å. After completion of the neutron measurements, the pressure was checked via ac susceptibility. In each case the pressure in the cell was found to be stable with time and thermal cycling.

The crystallographic structure of $Tm_2Fe_3Si_5$ is primitive tetragonal with space group $P4/mnc$,[8] and remains unchanged under application of pressures up to 8 kbar, as we have determined by high-resolution neutron-diffraction patterns taken at room temperature and 78 K. Figure 2(a) shows the diffraction pattern at 8.2 kbar and 1.6 K, which is below T_c but above T_N. The broad lump of scattering peaking at $\sim 11°$ is temperature independent and originates from the pressure cell, while the sharp peaks are nuclear Bragg reflections. Figure 2(b) shows the diffraction pattern in the ordered magnetic state at 0.4 K, which consists of both nuclear and magnetic Bragg peaks. Note in particular the additional strong reflections at low angles which result from the antiferromagnetic order. A subtraction of these two data sets isolates the magnetic Bragg peaks;[9] these are shown in Fig. 2(c). Note in particular that the broad scattering at low angles has subtracted out, leaving a general negative background due to the shifting of the high-temperature diffuse paramagnetic scattering into the antiferromagnetic Bragg peaks. The magnetic pattern at 8.2 kbar is the same as that found at ambient pressure. We remark that a dramatic change in the magnetic structure was not expected since T_N shows little change with pressure.

The model for the spin structure proposed by Moodenbaugh, Cox and Vining[8] is a complicated four-sublattice structure with the moments lying in the ⟨110⟩-type directions. Both sets of neutron data are in reasonable agreement with the model, although there are some systematic discrepancies between the model and the data. For the present considerations, it is essential to determine if there is any ferromagnetic component in the actual structure (at any pressure) which might be responsible for the destruction of the superconductivity. As a result of the multisublattice magnetic structure there is magnetic intensity at virtually all the nuclear peak positions, which complicates the detection of a possible ferromagnetic component. Two peaks, which have no antiferromagnetic contribution

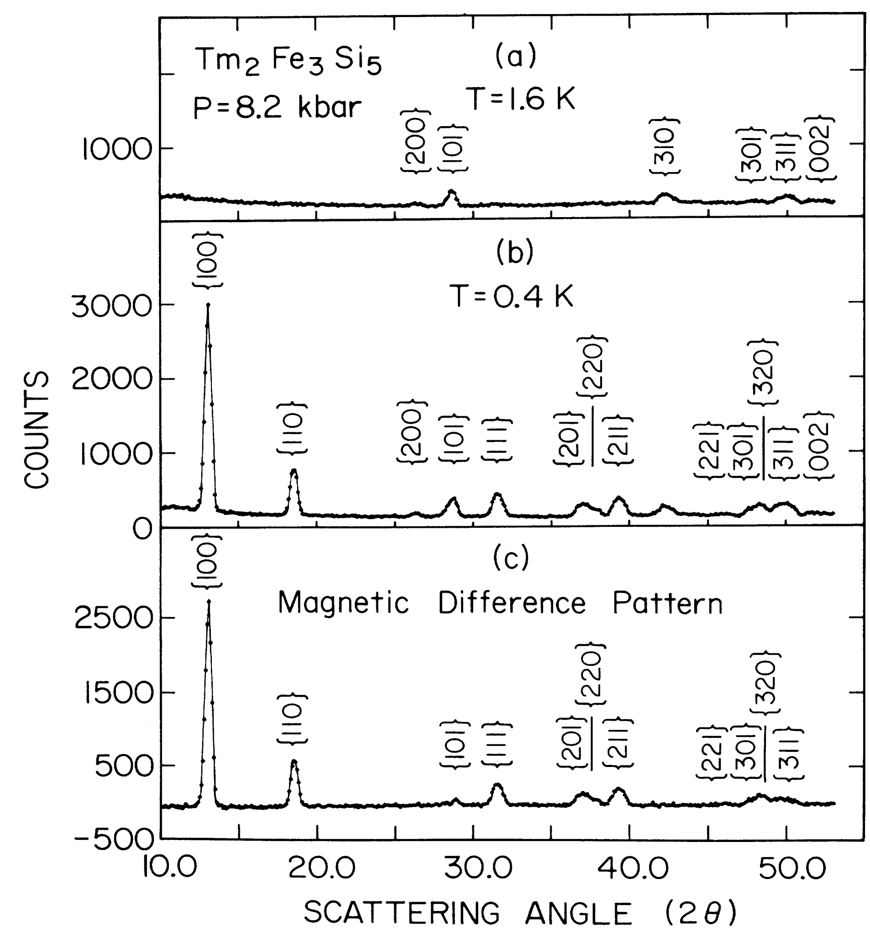

FIG. 2. Observed diffraction data at 8.2 kbar. Only peaks with observable intensity are labeled. (a) Nuclear-diffraction pattern observed at 1.6 K (SP phase). (b) Diffraction pattern observed at 0.4 K, showing additional peaks due to antiferromagnetic ordering (NA phase). (c) Difference pattern obtained by subtracting the data at 1.6 K from the data at 0.4 K. Only the peaks which are magnetic in origin survive the subtraction.

({200} and {002}), were examined carefully and found to have no ferromagnetic contribution, but the experimental precision was only $1.5\mu_B$. To improve the precision of this important result, an additional experiment was carried out with polarized neutrons. If a polarized neutron beam is transmitted through a sample which orders ferromagnetically, the local magnetization will tend to rotate the polarization of the beam; the polarized beam transmission measurement, thus, serves as a sensitive indicator of any ferromagnetic ordering in a system.[10] There was no observed change in the beam polarization as a function of temperature in $Tm_2Fe_3Si_5$ at 8.2 kbar and we estimate from these data that any ferromagnetic component must be $\lesssim 0.1\mu_B$. We have also used small-angle neutron scattering as well as conventional diffraction techniques to search for ferromagnetic fluctuations or possible oscillatory magnetic states in the vicinity of the transition, but no evidence was found for such additional effects which might accompany the destruction of the superconducting phase.

To further characterize the magnetic state, the peak intensity of the {100} antiferromagnetic peak was measured as a function of temperature. The normalized intensities for 2 and 8.2 kbar are displayed in Fig. 3. The magnetic intensity is proportional to the square of the sublattice magnetization, and hence, is directly related to the magnetic order parameter. The behavior of this peak intensity is very similar at all pressures measured, as can be seen in Fig. 3. For the purposes of estimating the antiferromagnetic transition temperature, we may employ the mean-field approximation, wherein the magnetic intensity will be linear with temperature in the vicinity of the transition. Fitting a straight line to the peak intensity between 1.0 and 1.1 K yields the estimates of the transition temperatures shown in Fig. 1. The values range from 1.086 K at 2 kbar to 1.122 K at 8.2 kbar, a change which is three times smaller than the change in T_{c2} observed via ac susceptibility. In addition, there is clear evidence of hysteresis in the reentrant transition temperature in the susceptibility measurements, growing as large as 50 mK between 4.4 and 16 kbar,[6] while there is no measurable hysteresis observed in the {100} intensity. The only noticeable difference in the present measurements at the two pressures is the additional scattering between 1.1 and 1.3 K at higher pressures. If we associate the reentrant transition with this extra scattering, rather than the linear extrapolation as estimated above, then the magnitude of the change in T_N is approximately the same as the change in T_{c2}. However, there is still no evidence of hysteresis in the neutron scattering data. We remark that this additional scattering could be due to a distribution of T_N's in the sample, or it could originate from unresolved satellites arising from a very long wavelength modulated state, or it could be critical scattering. In any case, the magnetic transition shows no first-order character. We can contrast this behavior with the case of the reentrant ferromagnetic superconductors $ErRh_4B_4$ and $HoMo_6S_8$, which display substantial hysteresis in the ferromagnetic, sinumagnetic, and superconducting order parameters.[1,11,12]

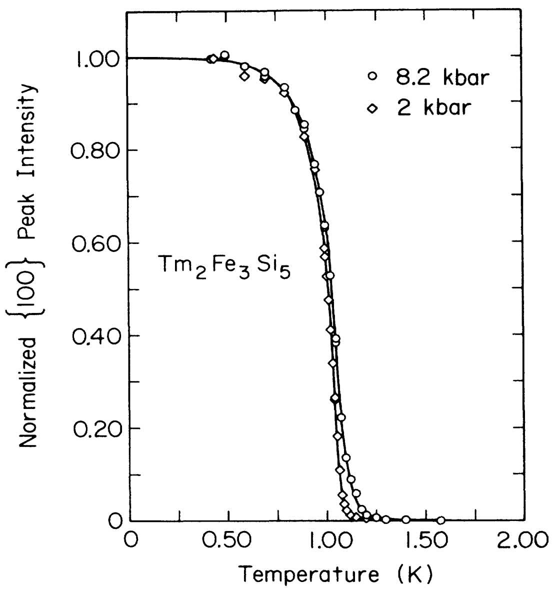

FIG. 3. Normalized {100} antiferromagnetic peak intensity, for $P=2$ kbar (◇) and $P=8.2$ kbar (○). Solid lines are guides for the eye.

Generally, the theories which have been developed for antiferromagnetic superconductors predict that the magnetic and superconducting states will coexist, with some modifications of the superconducting properties (particularly H_{c2}) in the magnetically ordered states.[13-17] Machida, Nokura, and Matsubara[14] do explicitly consider a choice of parameters in which the superconducting state is destroyed by the antiferromagnetic transition, but then the superconducting state would be expected to reemerge at lower temperatures. We have examined $Tm_2Fe_3Si_5$ at 8.2 kbar via ac susceptibility measurements down to 40 mK, and find no evidence for such an additional transition.

In conclusion, these neutron-diffraction measurements show that there is no ferromagnetic component to the magnetism in the ordered state, and no change in the nature of the antiferromagnetic ordering under increased pressure. Thus, the superconductivity is quenched by purely antiferromagnetic order. This is the first direct experimental observation of this phenomenon, and should place strict constraints on the various theories which have been applied to antiferromagnetic superconductors.

We would like to thank W. B. Daniels (University of Delaware) for his generous assistance. Research at Maryland is supported by the U.S. National Science Foundation under Grants No. DMR 83-19936 and No. 86-20269. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-ENG-82. Research at Ames is supported by the Director for Energy Research, Office of Basic Sciences, under Grant No. WPAS-KC-02-02-02.

*Present address: Department of Physics, University of California-Davis, Davis, CA 95616.

[1]For a comprehensive review of earlier work on these materials, see *Topics in Current Physics*, edited by Ø. Fischer and M. B. Maple (Springer-Verlag, New York, 1983), Vols. 32 and 34.

[2]Hans F. Braun, Phys. Lett. **75A**, 386 (1980).

[3]H. F. Braun, C. U. Segre, F. Acker, M. Rosenberg, S. Dey, and P. Deppe, J. Magn. Magn. Mater. **25**, 117 (1981).

[4]J. D. Cashion, G. K. Shenoy, D. Niarchos, P. J. Viccaro, A. T. Aldred, and Charles M. Falco, J. Appl. Phys. **52**, 2180 (1981).

[5]C. B. Vining and R. N. Shelton, Phys. Rev. B **28**, 2732 (1983).

[6]C. B. Vining and R. N. Shelton, Solid State Commun. **54**, 53 (1985).

[7]T. F. Smith, J. Low Temp. Phys. **6**, 171 (1972). Our cell was made of Al, rather than the standard Be-Cu, for reasons of convenience in the neutron measurements.

[8]A. R. Moodenbaugh, D. E. Cox, and C. B. Vining, Phys. Rev. B **32**, 3103 (1985).

[9]See, e.g., R. Nathans and S. J. Pickart, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic, New York, 1963), Vol. III, Chap. 5.

[10]J. W. Lynn, Q. Li, and R. W. Erwin, Bull. Am. Phys. Soc. **32**, 922 (1987).

[11]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, Phys. Rev. Lett. **45**, 2060 (1980); S. K. Sinha, G. W. Crabtree, D. G. Hinks, and H. Mook, *ibid.* **48**, 950 (1982).

[12]J. W. Lynn, G. Shirane, W. Thomlinson, and R. N. Shelton, Phys. Rev. Lett. **46**, 368 (1981); J. W. Lynn, A. Raggazoni, R. Pynn, and J. Joffrin, J. Phys. (Paris) Lett. **42**, L45 (1981).

[13]A general review of theory is given by P. Fulde and J. Keller, in Ref. 1, Vol. 34, Chap. 9.

[14]Kazushige Machida, Kazuo Nokura, and Takeo Matsubara, Phys. Rev. B **22**, 2307 (1980).

[15]T. V. Ramakrishnan and C. M. Varma, Phys. Rev. B. **24**, 137 (1981).

[16]M. K. Nass, K. Levin, and G. S. Grest, Phys. Rev. Lett. **46**, 614 (1981); and in *Ternary Superconductors*, edited by G. K. Shenoy, B. D. Dunlap, and F. Fradin (North-Holland, New York, 1981), p. 277.

[17]Yoshikazu Suzumura and A. D. S. Nagi, Phys. Rev. B **24**, 5103 (1981).