Observation of the stabilization of a second lock-in phase in holmium at 42 K with a wave vector of 0.2 reciprocal lattice units

D. A. Tindall
Department of Physics, Dalhousie University, Halifax, Nova Scotia, Canada B3H 3J5

M. O. Steinitz
Department of Physics, Saint Francis Xavier University, Antigonish, Nova Scotia, Canada B2G 1C0

T. M. Holden
Neutron and Condensed Matter Science, AECL Research, Chalk River, Ontario, Canada K0J 1J0
(Received 20 July 1992)

We report the observation of the stabilization of a second lock-in phase in holmium by a c-axis magnetic field. Neutron scattering has been used to investigate the behaviour of holmium in a magnetic field of 2.95 T applied along the c-axis in the temperature range 30–50 K. The variation of the position of the fundamental magnetic satellites with temperature is described and the lock-in at a wave vector of 0.2 reciprocal-lattice units is observed at 42 K.

The magnetic structure of holmium, a c-axis helical antiferromagnet, was determined nearly 30 years ago by Koehler *et al.*[1] The wave vector of the helix (τ) varies with temperature from about 0.167 reciprocal-lattice units (r.l.u.) at 20 K to about 0.28 r.l.u. at the Néel point. In most of this temperature range the period is incommensurate with the lattice but, beginning with the discovery[2,3] of effects near 96 K where τ passes through 1/4 r.l.u. (making the period of the spiral equal to four lattice constants), many other commensurate effects have been found[4,5] using neutron scattering in this temperature range as well as by magnetic x-ray scattering[6,7] below 30 K. This led to the development of the spin-slip model[6,8,9], which successfully accounted for many of the phenomena seen in neutron-diffraction studies. Magnetization measurements[10] in the range between 4 K and the Néel temperature (132 K) have allowed the preliminary mapping of a complicated magnetic phase diagram for fields along the three crystal axes.

The simplest commensurate values of the wave vector τ, available between the end points of the spiral phase at 0.167 r.l.u. and 0.28 r.l.u., are 1/5 r.l.u. and 1/4 r.l.u. Our recent neutron-diffraction studies[11,12] on holmium in a c-axis magnetic field have revealed that the magnetic field has a stabilizing effect on the locked-in phase with τ=1/4 r.l.u. near 96 K. We have now undertaken a similar investigation of the lock-in at $\tau = 1/5$ r.l.u., in conditions identical to those used in our previous studies, i.e., a c-axis magnetic field of 2.95 T. This wave vector occurs near 42 K in zero field.[1]

The same single crystal of holmium (about 10 mm$\times$10 mm$\times$20 mm), as used in our previous studies at 96 K and by Pechan and Stassis,[13] was mounted on the N5 triple-axis spectrometer at the NRU reactor at the Chalk River Laboratories of AECL Research in such a way that we were able to measure reflections in the ($h0l$) plane. The field of 2.95 T was applied along the c axis using a superconducting magnet which allows the neutrons 350° access in the horizontal plane.[14] The measurements were made at zero energy transfer with an analyzer crystal in front of the detector. The neutron energy used throughout these experiments was 8.225 THz, using the (113) reflection from a silicon monochromator. The beam divergence in the scattering plane, after the monochromator collimator, was 0.45°, with matching collimation on the analyzer side.

As is shown in Fig. 1, the wave vector, as defined by the difference between the position of the (1,0,0) nuclear peak and the (1,0,τ) magnetic satellite, is locked in at a value of 0.2 r.l.u. over a temperature range of about 1.8 K in the vicinity of 42 K. This is almost the identical temperature width as that which we found for the locked-in region around 96 K in the same field (see inset to Fig. 1).

These results indicate that the assignment of branches in the phase diagram for c-axis fields in Fig. 5 of the

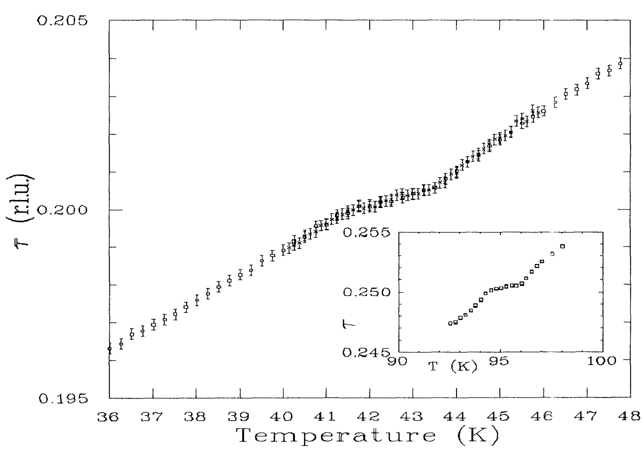

FIG. 1. Variation with temperature of τ [obtained from the (10τ) satellite] in a c-axis magnetic field of 2.95 T. The inset shows the similarity of the behavior under the same conditions, in the same sample, near the 96-K ($\tau = 1/4$ r.l.u.) lock-in.

paper by Willis *et al.*[10] should probably be altered so that the line beginning at 42 K in zero field should be progressively split in increasing fields in a manner similar to that in the vicinity of 96 K. Thus the points shown in that figure at fields above about 3 T, near 37 K, and connected to a branch originating at about 20 K, should be reassigned to the branch discussed here.

As seen in Fig. 2, the integrated intensity of the satellite peak has an anomalous behavior, increasing in intensity at the beginning and end of the locked-in region, in a manner entirely similar to the behavior of the intensity in the vicinity of the 96-K lock in (see lower part of Fig. 2). The reduction of intensity at the center of the locked-in region might be explained by increased extinction due to the greater perfection of a commensurate structure over an incommensurate structure, but the increases at either end of the locked-in region cannot be explained on the basis of such arguments. We have also observed these effects at various neutron energies [3.52 (Ref. 11) and 8.212 (Ref. 12) THz], thus laying the extinction argument to rest, but equally we have not yet found evidence of additional peaks[12] that would account for the intensity lost from the τ peaks. The intensity of the (100) peak is rather constant through this region, as is the case in the vicinity of 96 K. This indicates that any effect of a *c*-axis moment, occurring due to a tilting of the moments in the locked-in phase, is below the limits of our detection.

Cowley *et al.*[15] have explored the temperature region just below that studied here and have interpreted their results in terms of a devil's staircase of spin-slip structures. However, that explanation does not appear to be applicable to our results.

Our recent work[16] has shown the intimate connection between the spontaneous magnetostriction (and thus the strain dependence of the exchange interaction) and the wave vector at the 96-K lock in and similar connections are to be expected here. Strain measurements at 42 K in a *c*-axis field are planned, as well as a study to determine if the behavior of the intensity of the 2-τ peaks relative to that of the τ peaks is as anomalous at the 42-K lock-in as that which we observed around 96 K.[17]

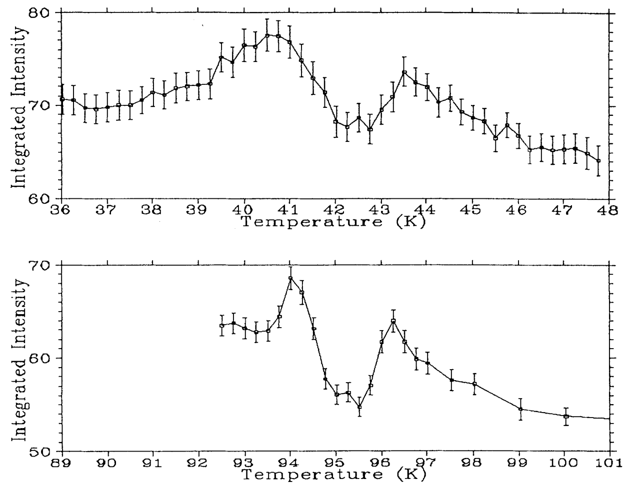

FIG. 2. Upper figure shows the temperature variation of the integrated intensity (arbitrary units) of the (10τ) satellite in the vicinity of the 42-K lock-in. The lower figure shows the corresponding variation, again under the same conditions, near the 96-K (τ = 1/4 r.l.u.) lock-in.

A puzzling question is why the attenuation of ultrasonic shear waves reported by Lee, Treder, Levy[18] shows no anomaly at all in the neighborhood of 42 K, while showing a large anomaly at 96 K. This is particularly strange in the light of the similarities between the two locked-in phases reported here.

We are grateful to M. J. Pechan and C. Stassis for the loan of the sample and to M. L. Plumer for helpful discussions. This work was supported in part by grants from the Natural Sciences and Engineering Research Council of Canada and the Canadian Institute for Neutron Scattering. We gratefully acknowledge the provision of facilities and skilled assistance by AECL Research, Chalk River.

[1]W.C. Koehler, J.W. Cable, M.K. Wilkinson, and E.O. Wollan, Phys. Rev. **151**, 414 (1966).
[2]M.C. Lee, R.A. Treder, and M. Levy, J. Phys. Chem. Solids **36**, 1281 (1975).
[3]A.M. Simpson, M.H. Jericho, and M.C. Jain, Can. J. Phys. **54**, 1172 (1976).
[4]J. Baruchel, A. Drillat, D. Fort, D.W. Jones, S.B. Palmer, and M. Schlenker, J. Magn. Magn. Mater. **31-34**, 183 (1983).
[5]A. Drillat, J. Baruchel, S. Bates, and S.B. Palmer, J. Magn. Magn. Mater. **44**, 232 (1984).
[6]J. Bohr, D. Gibbs, D.E. Moncton, and K.L. D'Amico, Physica **140A**, 349 (1986).
[7]D. Gibbs, D.E. Moncton, K.L. D'Amico, J. Bohr, and B.H. Grier, Phys. Rev. Lett. **55**, 234 (1985).
[8]R.A. Cowley and S. Bates, J. Phys. C **21**, 4113 (1988).
[9]S. Bates, C. Patterson, G.J. McIntyre, S.B. Palmer, A. Mayer, R.A. Cowley, and R. Melville, J. Phys. C **21**, 4125 (1988).
[10]F. Willis, N. Ali, M.O. Steinitz, M. Kahrizi, and D.A. Tindall, J. Appl. Phys. **67**, 5277 (1990).
[11]D.R. Noakes, D.A. Tindall, M.O. Steinitz, and N. Ali, J. Appl. Phys. **67**, 5274 (1990).
[12]D.A. Tindall, M.O. Steinitz, M. Kahrizi, D.R. Noakes, and N. Ali, J. Appl. Phys. **69**, 5691 (1991).
[13]M.J. Pechan and C. Stassis, J. Appl. Phys. **55**, 1900 (1984).
[14]D.C. Tennant, N. Kerley, and N. Killoran, Rev. Sci. Instrum. **60**, 136 (1989).
[15]R.A. Cowley, D.A. Jehan, D.F. McMorrow, and G.J. McIntyre, Phys. Rev. Lett. **66**, 1521 (1991).
[16]M.O. Steinitz, D.A. Tindall, and M. Kahrizi, J. Magn. Magn. Mater. **104-107**, 1531 (1992).
[17]D.A. Tindall, M.O. Steinitz, and D.R. Noakes, Physica B **180/181**, 79 (1992).
[18]M.C. Lee, R.A. Treder, and M. Levy, J. Phys. Chem. Solids **36**, 1281 (1975).