

Evidence of pressure-induced antiferromagnetism in ferromagnetic $\text{Ho}_{0.4}\text{Gd}_{0.6}$

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The rare earth alloy $\text{Ho}_{0.4}\text{Gd}_{0.6}$ was examined under uniaxial pressure applied along the c axis by means of magnetic susceptibility vs temperature. At ambient pressure the compound happened to be ferromagnetic at all temperatures where magnetic ordering occurs. Nevertheless, at pressures larger than $p^*=700$ bars there is clear evidence of an antiferromagnetic phase (likely to be helical one) that occurs just below the magnetic ordering temperature. This result is consistent with the hypothesis that the c/a ratio is the crucial parameter for the occurrence of helical structure in heavy rare-earth metals. [S0163-1829(97)03418-8]

I. INTRODUCTION

Long-periodic magnetic structures in heavy rare-earth metals have been under investigation for more than 30 years. Some new aspects of the problem have opened up as the volume of information collected has increased.¹

In particular, currently there are reasons to think that the role of hexagonal crystalline structure in the occurrence of long-periodic magnetic structures might be more important than had been suspected so far.

Recent investigations of ‘‘lock-in’’ in holmium are illustrative in this respect. These studies revealed that anomalies in the helical wave vector temperature dependence (associated with ‘‘lock-in’’) are accompanied by anomalies in the thermal expansion.^{2,3} It is evidence that the magnetic structure and crystalline lattice are closely tied at least in Ho.

Available data on the crystalline lattice parameters for heavy rare-earth pure metals and their alloys with each other and yttrium were summarized in Refs. 4 and 5.

The main observations^{4,5} are as follows. (i) The period of helical magnetic structure is dictated by the c/a ratio of crystalline lattice parameters. (ii) There is a critical value of $c/a=(c/a)_{cr}$ that separates different magnetic states. Namely, the helical antiferromagnetic structure occurs only at $c/a<(c/a)_{cr}$, while at $c/a\geq(c/a)_{cr}$ all the compounds happen to be ferromagnetic. (iii) The value of $(c/a)_{cr}=1.582\pm 0.001$, likely to be the same for all the heavy rare-earth metals and their alloys with each other and Y.

(It has been proposed in Refs. 4 and 5 that this c/a critical value is associated with the electronic topological transition in the c/a parameter, alias a Lifshitz transition or $2\frac{1}{2}$ type transition.)

The direct conclusion is that one can turn rare-earth metal from the ferromagnetic state to antiferromagnetic one (or vice versa) by changing the c/a ratio by an outside action.

The goal of the work was to check this conclusion experimentally.

II. EXPERIMENT

The simplest way to change the c/a ratio without breaking the lattice symmetry is to apply uniaxial pressure along

the hexagonal c axis. In this case the occurrence of an antiferromagnetic phase in the initially ferromagnetic sample with increasing pressure was expected.

In the other words, an initially ferromagnetic ‘‘like gadolinium’’ sample was expected to behave ‘‘like terbium’’ under certain uniaxial pressure along the c axis.

For this purpose we choose a $\text{Ho}_x\text{Gd}_{1-x}$ system. The contents where the antiferromagnetic phase was observed in this system are $X\geq X_{cr}=0.45\pm 0.01$ at. part.⁶

In consequence of this we chose our sample content as $\text{Ho}_{0.4}\text{Gd}_{0.6}$ to be sure it would be ferromagnetic at ambient pressure.

The $\text{Ho}_{0.4}\text{Gd}_{0.6}$ polycrystal was prepared in Moscow Institute for Metals from the components of 99.99% purity. The single crystal was prepared by means of high-temperature recrystallization. The sample of a shape of rectangular plate of dimensions $0.16\times 0.87\times 1.40$ mm³ was cut by spark erosion with the c axis normal to the plate with $\pm 1^\circ$ accuracy.

X-ray examination revealed that sample is a single-phase crystal with hcp crystalline structure of mosaicity less than 1° .

The uniaxial pressure along the c axis was applied to the sample by two beryllium-copper plugs (see inset of Fig. 1). The load produced by steel string of rigidity 13.8 N/mm was transmitted to plugs by two long plungers made of stainless steel.

The elasticity limit for polycrystalline Ho and Gd is some 2 kbar; thus we never exceeded this pressure value.

The magnetic state of the sample was examined by magnetic susceptibility that was measured by an ac mutual inductance technique of 19 960 Hz frequency.

Two mutual inductance coils were placed parallel to each other around the sample, the ac magnetic field being parallel to the sample’s longest dimension, i.e., perpendicular to the crystal hexagonal c axis. (The shape factor for the sample was $n=0.039$.)

The temperature dependences of mutual inductance M were taken on cooling with a rate less than 1.5 K/min in N_2 atmosphere. The sample temperature was monitored by copper-Constantan thermocouple with ± 0.2 K accuracy.

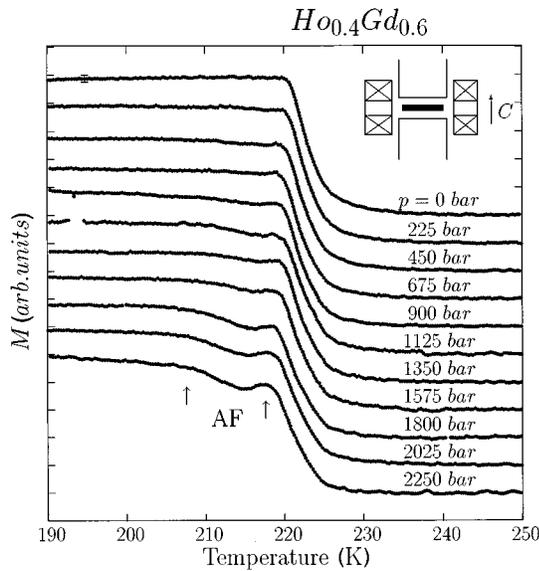


FIG. 1. Mutual inductance M vs T dependences at different pressures. Pressures are indicated on the curves. Curves are shifted vertically for clarity, onsets omitted. Arrows indicate antiferromagnetic area. Inset: sketch of the experiment.

III. RESULTS

A set of M vs T curves at different pressures is presented in Fig. 1. (Dependences are perfectly reversal in pressure.)

At the ambient pressure and at pressures $p < 700$ bars there are no observable anomalies but the Curie point at $T_C = 220$ K. Thus the sample is likely to be “purely” ferromagnetic at low pressures, as was expected.

But at pressures larger than $p^* = 700$ bars this anomaly is obviously split. It would appear reasonable that this splitting is associated with the occurrence of an antiferromagnetic phase—most likely to be a helical one—just below the magnetic ordering temperature. At the largest pressure used, $p = 2250$ bars this antiferromagnetic area is some 10 K wide (indicated by arrows).

The temperature behavior of $\text{Ho}_{0.4}\text{Gd}_{0.6}$ at pressures $p > p^*$ closely resembles that of Tb. As the temperature decreases, the sample apparently undergoes a transition from an antiferromagnetic to a collinear ferromagnetic state that is typical of the rare-earth metals in a helical phase.

Hence it has been demonstrated that the magnetic state of the rare-earth alloy can be altered by uniaxial pressure as low as 700 bars. This suggests that the role of crystalline lattice parameters in magnetic ordering in rare earth metal can be really crucial.

IV. DISCUSSION

According to Ref. 4 the governing parameter in magnetic ordering in heavy rare-earth metals is the c/a ratio of crystalline lattice parameters (see above). The numerical estimations of changes in the c/a value, of the order of 10^{-3} in our case, are as follows.

The c/a ratios for components are 1.569 and 1.590 for Ho and Gd, respectively (in the paramagnetic state, where this ratio is almost temperature independent⁷). For $\text{Ho}_x\text{Gd}_{1-x}$ compounds the c/a ratio depends on x almost linearly.⁷

The calculated c/a value for $\text{Ho}_{0.4}\text{Gd}_{0.6}$ is 1.582, while, for the critical content $\text{Ho}_{0.45}\text{Gd}_{0.55}$, $c/a = 1.581$. The latter value is in good agreement with expected $(c/a)_{\text{cr}} = 1.582 \pm 0.001$ (see above).

The straightforward calculation of the c/a variation under uniaxial pressure p along the c axis reveals

$$\Delta(c/a) = -(c/a) \frac{c_{11} + c_{12} + c_{13}}{(c_{11} + c_{12})c_{33} - 2c_{13}^2} p,$$

where c_{ij} are elastic moduli. Substituting typical for the rare-earth metal values $c_{11} = 0.8$, $c_{12} = 0.3$, $c_{13} = 0.25$, $c_{33} = 0.7 \times 10^{12}$ dyn/cm² (Ref. 7) and critical pressure $p^* = 700$ bars we obtain $\Delta(c/a) = -0.002$.

Thus estimated variation in c/a under critical pressure p^* is of the same sign and value as the difference in c/a values between critical $\text{Ho}_{0.45}\text{Gd}_{0.55}$ and examined $\text{Ho}_{0.4}\text{Gd}_{0.6}$. It means that formation of antiferromagnetism under pressure as well as under content variation occurs at almost the same $(c/a)_{\text{cr}}$ value, estimated as 1.581 ± 0.001 .

V. CONCLUSIONS

The experiment revealed that magnetic ordering in the heavy rare-earth metal alloy is extremely sensitive to the crystalline lattice parameters. Initially the ferromagnetic sample was turned to an apparently antiferromagnetic state by uniaxial pressure as low as 700 bars, i.e., under variation of the lattice parameters of order of 0.1%.

This result is consistent with the hypothesis⁴ that the c/a ratio of crystalline lattice parameters is the crucial parameter for the occurrence of helical magnetic structure in heavy rare-earth metals.

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¹J. Jensen and A.R. Mackintosh, *Rare Earth Magnetism* (Clarendon, Oxford 1991).

²M.O. Steinitz, D.A. Tindall, and M. Kahrizi, *J. Magn. Mater.* **104-107**, 1531 (1992).

³A.S. Bulatov, V.F. Dolzenko, and A.V. Kornietz, in Proceedings of the 21st International Conference on Low Temperature

Physics [Czech. J. Phys. **46**, Suppl. S4, 2119 (1996)].

⁴A. Andrianov, *JETP Lett.* **55**, 666 (1992).

⁵A. Andrianov, *J. Magn. Mater.* **140-144**, 749 (1995).

⁶A.V.I. Andrianov and O.D. Chistiakov (unpublished).

⁷K.N.R. Taylor and M.I. Darby, *Physics of Rare Earth Solids* (Chapman and Hall, London, 1972).