Evidence of pressure–induced antiferromagnetism in ferromagnetic $Ho_{0.4}Gd_{0.6}$

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

75.25.+z, 75.30.Kz, 75.50.Ee

I. INTRODUCTION

Long–periodic magnetic structures in heavy rare–earth metals are under investigation for more than thirty years. Some new aspects of the problem are opened up as the volume of information collected increases [1].

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

Recent investigations of "lock–in" in holmium are illustrative in this respect. These studies revealed that anomalies in helical wavevector temperature dependence (associated with "lock–in") are accompanied by anomalies in thermal expansion [2], [3]. It is evidence that 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 [4], [5].

The main observations [4], [5] are as follows. *i)* The period of helical magnetic structure is dictated by 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 [4], [5] that this c/a critical value is associated with the electronic topological transition in c/a parameter, alias 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 c/a ratio by the outside action.

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

II. EXPERIMENTAL

The simplest way to change c/a ratio without breaking the lattice symmetry is to apply the uniaxial pressure along hexagonal axis c. In this case the occurrence of antiferromagnetic phase in the initially ferromagnetic sample with pressure increasing was expected.

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

For this purpose we choose $Ho_X Gd_{1-X}$ system. The contents where 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 choose our sample content as $Ho_{0.4}Gd_{0.6}$ to be sure it would be ferromagnetic at ambient pressure.

The $Ho_{0.4}Gd_{0.6}$ 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 recrystallisation. The sample of a shape of rectangular plate of dimensions $0.16 \times 0.87 \times 1.40\ mm$ was cut by spark erosion with c axis normal to the plate with $\pm 1\ deg$ accuracy.

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

The uniaxial pressure along c axis was applied to the sample by two beryllium–copper plugs (see inset 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 exceed this pressure value.

The magnetic state of the sample was examined by magnetic susceptibility that was measured by *ac* mutual inductance technique on 19960 Hz frequency.

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

The temperature dependences of mutual inductance M were taken on cooling with 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.

III. RESULTS

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

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

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

The temperature behavior of $Ho_{0.4}Gd_{0.6}$ at pressures $p > p^*$ closely resembles that of Tb. As temperature decreases, the sample apparently undergoes transition from antiferromagnetic to the collinear ferromagnetic state that is typical of the rare–earth metals in helical phase.

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

IV. DISCUSSION

According to [4] the governing parameter in magnetic ordering in heavy rare–earth metals is c/a ratio of crystalline lattice parameters (see above). The numerical estimations of changes in 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 independent [7]). For $Ho_X Gd_{1-X}$ compounds c/a ratio depends on x almost linearly [7].

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

The straightforward calculation of c/a variation under uniaxial pressure p along 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 rare–earth metal values $c_{11} = 0.8$, $c_{12} = 0.3$, $c_{13} = 0.25$, $c_{33} = 0.7 \times 10^{12} din/cm^2$ ([7]) and critical pressure $p^* = 700$ bar we obtain $\Delta(c/a) = -0.002$.

Thus estimated variation in c/a under critical pressure p^* is of the same sign and value that the difference in c/a values between critical $Ho_{0.45}Gd_{0.55}$ and examined $Ho_{0.4}Gd_{0.6}$. It means that formation of antiferromagnetism under pressure as well as under content variation occurs at almost the same $(c/a)_{cr}$ 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 ferromagnetic sample was turned to apparently antiferromagnetic state by uniaxial pressure as low as 700 bar, i.e. under variation of lattice parameters of order of 0.1 percent.

This result is consistent with the hypothesis [4] that 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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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.