Helical magnetic ordering in Tb completely suppressed by uniaxial tension: Evidence of electronic topological transition and support for the nesting hypothesis

A. Vl. Andrianov and D. I. Kosarev
Department of Physics, Moscow State University, Moscow 119899, Russia

A. I. Beskrovnyi
Laboratory of Neutron Physics, Joint Institute for Nuclear Research, Dubna, Moscow District 141980, Russia
(Received 8 September 2000)

A single crystal of Tb was examined under uniaxial tension applied along the hexagonal axis by means of magnetic susceptibility vs temperature. The temperature range within which helical ordering occurs was determined from $\chi(T)$ curves. This range diminishes rapidly as uniaxial tension is applied, and at tensions over $p^*=680$ bar the helical ordering is completely suppressed. The experiment supports the hypothesis that the type of magnetic ordering in rare-earth metals is determined by Fermi surface topology, which can in turn be altered by uniaxial deformation.

It was suggested more than 30 years ago that the properties of long-periodic magnetic structures in heavy rare-earth metals are dictated by ''nesting'' phenomenon.[1] According to this approach, referred to hereafter as the ''nesting'' hypothesis, the wave vector of a long-periodic magnetic structure in a metal is exactly equal to the known extreme diameter q of the Fermi surface (FS). The introduction in Ref. 2 presented a brief explanation of the phenomenon.

In the case of heavy rare-earth metals, this diameter was supposed to be a diameter of the so-called ''webbing'' feature in the L point of the Brillouin zone;[1] see Fig. 1. (Note that this diameter intersects the Brillouin-zone face.) There was no direct experimental proof of this hypothesis for a long time. Nevertheless, there was a number of indirect proofs, so that the ''nesting'' hypothesis is now well accepted in rare-earth physics.

The less studied subject is that the FS in heavy rare-earth metals possesses properties that are not typical of ''ordinary'' metals. That is, all these materials are on the verge of change in the FS topology. We are going to demonstrate that a combination of the nesting hypothesis with the hypothesis of an electronic topological transition in FS's might provide a fruitful approach to rare-earth magnetism.

The classical calculations of FS's (Ref. 1) in rare-earth metals revealed that minor variations in initial parameters might cause a change in the FS topology. Particularly, the calculated FS's of Tb, Ho, Dy, Lu, and Y exhibit the above-mentioned webbing feature, while there is no such feature in the FS of Gd. Further investigations of FS in these materials, both experimental[2–6] and theoretical[7,8] never affected the main conclusions.

The two plausible topologies of the Fermi surfaces in these materials are presented on Fig. 1. These sketches summarize those of Refs. 1–8. The sketches of the FS hereafter relate to the paramagnetic state; distortions caused by magnetic structure not presented, for the sake of clarity.

It should be emphasized that all these substances are similar enough to be considered just variations of the same entity. The well-localized $4f$ shell is the only difference, while the outer electronic shells and hence the chemical and crystalline properties are almost the same. Nevertheless, the FS topology turns out to be different in different heavy rare-earth metals.

Let us consider a typical system Y_xGd_{1-x}. As the FS topology is different in Y and Gd, the change in FS topology would definitely occur in this system at some x. In other words, this system is in the vicinity of an electronic topological transition.

Actually, the recent direct observations of FS's in YGd alloys[2] and pure Y,[3] by positron annihilation, experimentally confirmed the change in FS topology under content variations in these systems. Hence both the simplest considerations and the experiment suggest that the electronic topological transition is plausible for heavy rare-earth metals. Now we consider direct consequences.

The governing parameter that determines the shape of FS's in all heavy rare-earth metals and Y in the simplified model is just the c/a ratio of lattice parameters of hcp crystalline structure.[9] All these materials are hcp metals with 3^+ ions. The influence of the internal $4f$ electrons can be neglected as a first approximation; thus the c/a ratio remains the only free parameter.

It immediately follows that all these metals and their alloys are on the verge of an electronic topological transition in c/a parameter. Two plausible topologies both occur in various substances; a FS with the webbing feature corresponds to lower c/a values. Note that the variation in the c/a parameter in these substances is as low as 1%: from 1.570 in Y and Ho to 1.590 in Gd. Hence all these substances and their alloys are remarkably close to this transition.

To summarize our conclusions, if the hcp metal is on the verge of a change in the FS topology, then this change will definitely occur under a proper variation in the c/a ratio, the reason being simply that the c/a ratio is the primary parameter that determines the FS shape in such metal (see, for example, Ref. 9).

In Ref. 10 we emphasized that the shape and the very FS topology in heavy rare-earth metals are extremely sensitive to the content variation as well as to the crystalline lattice deformation, in contrast to ''ordinary'' metals. Moreover, a

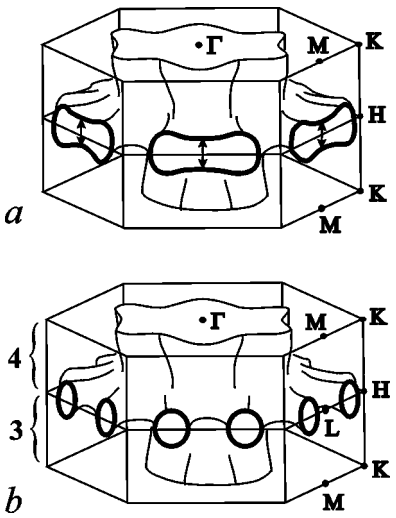

FIG. 1. Two plausible topologies of the Fermi surface in heavy rare-earth metals (double-zone presentation, semiquantative sketch): (a) A ''webbing feature'' is present (indicated by bold line), and long-periodic magnetic ordering is expected. Arrows mark the extreme diameter responsible for ''nesting.'' (b) No ''webbing feature,'' and ferromagnetic ordering is expected. Zones are labeled by numbers. The doubtful ''arm'' at the *M* point is not presented.

slight change in the c/a ratio caused by thermal expansion or by external pressure may be enough to change the FS shape distinctly.

According to the nesting hypothesis the magnetic structure wave vector duplicates the behavior of the extreme FS diameter. The evolution of a FS due to a variation in the c/a ratio with thermal expansion may therefore be responsible for the thermal dependence of the magnetic structure wave vector. A qualitative agreement with this hypothesis was observed in Ref. 10.

Since the c/a ratio is a governing parameter for the FS shape, a change in its value may cause a change in the FS topology of the given material. This change would be a classical example of an electronic topological transition, also known as the Lifshitz transition.[11] One should expect a square-root dependence of an extreme FS diameter on the c/a ratio, which is characteristic of such a transition. According to the nesting hypothesis, the helical wave vector should behave the same way.

Similarly, one can expect a square-root dependence of the helical wave vector on the x parameter in binary alloys like Ho_xGd_{1-x}, Y_xGd_{1-x}, Tb_xY_{1-x}, etc. In other words, the change in the c/a ratio is expected to have exactly the same effect on the magnetic structure as the chemical content variation.

Our general conclusion is as follows: the helical wave vector q shall depend on the c/a ratio as a square root $q \propto [(c/a)_{cr} - (c/a)]^{1/2}$, with no helical ordering at $(c/a) > (c/a)_{cr}$. The critical value $(c/a)_{cr}$ at first approximation appears to be the same for all heavy rare-earth metals and yttrium (see above).

Obviously this is a simplification, and it would be more realistic to expect a unique value of $(c/a)_{cr}$ for any given substance, while the other aspects of the dependence shall be common. Nevertheless the existing experimental data (gathered in Ref. 10) suggest that these critical values for different rare-earth metals are close enough, and are all located in the range $(c/a)_{cr} = 1.582 \pm 0.002$ (with the possible exception of the GdY system[13]).

It immediately follows that one can switch a rare earth metal from a ferromagnetic state to an antiferromagnetic one, and vice versa, by driving the c/a ratio through its critical value by outside action. This is what we are going to demonstrate experimentally.

The simplest way to change the c/a ratio without breaking the symmetry of the lattice is to apply uniaxial pressure along the hexagonal axis c. In our recent work,[14] we demonstrated that uniaxial compression of the ferromagnetic $Ho_{0.4}Gd_{0.6}$ single crystal causes the occurrence of a presumably helical magnetic phase.

In this work we are going to demonstrate the opposite, i.e., that uniaxial elastic tension may completely suppress the helical ordering in a rare-earth metal. The obvious choice for such an experiment is terbium, with its remarkably narrow temperature range within which helical ordering occurs (as low as 10 K, while the magnetic ordering temperature $T_N = 230$ K).

Recently the FS for terbium was recalculated with modern computer power. The presence of the ''webbing feature'' was confirmed. Moreover, direct calculation supported the credibility of the ''nesting'' hypothesis in the case of Tb.[8]

Experiments with Tb films deposited on Y (Ref. 15) revealed a broadening of the temperature range within which helical ordering occurs, and an increase in the helical wave vector. This broadening is presumably caused by an expansion in the basal crystalline plane caused by deposition, i.e., by a decrease in the c/a ratio. The only known experiment with uniaxial compression on Tb (Ref. 16) revealed a broadening of the temperature range within which a helical structure occurs under compression along the c axis.

The experiments with hydrostatic pressure on Tb (Refs. 17 and 18) also revealed a slight broadening of the temperature range within which a helical structure occurs under pressure increase. A calculation that takes into account elastic moduli suggests there will be a slight decrease in the c/a ratio under hydrostatic pressure. Hence all the existing experimental data confirm a stabilization of the helical structure, and an increase in helical wave vector under decrease in c/a ratio. This agrees qualitatively with our expectations.

However, our goal was to suppress the helical ordering in Tb completely by uniaxial tension, i.e., by an increase in the c/a ratio. In other words, we expected terbium to behave ''like gadolinium'' under certain tension applied along the c axis. We also expected a sort of square-root-like dependence of helical structure parameters on tension.

The Tb single crystal was prepared at the Moscow Institute for Metals in 1972. Its declared purity is 99%. X-ray examination revealed that the sample is a single-phase crystal with a hcp crystalline structure, a mosaic spread less than 1°, and lattice parameters $c = 5.696$ Å, $a = 3.605$ Å, and $c/a = 1.580$ (at room temperature).

The neutron scattering examination performed in the Laboratory for Neutron Physics of JINR (Dubna, Russia) on DN-2 time-of-flight spectrometer on an IBR-2 pulse reactor confirmed the helical ordering in the sample. The typical neutron pattern and temperature dependence of a helical

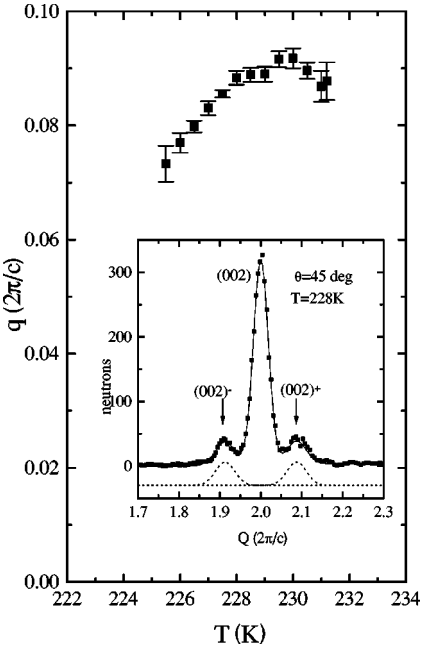

FIG. 2. Helical wave vector in the sample measured by neutron diffraction (ambient pressure). Inset: typical neutron pattern ($\theta = 45°, T = 228$ K).

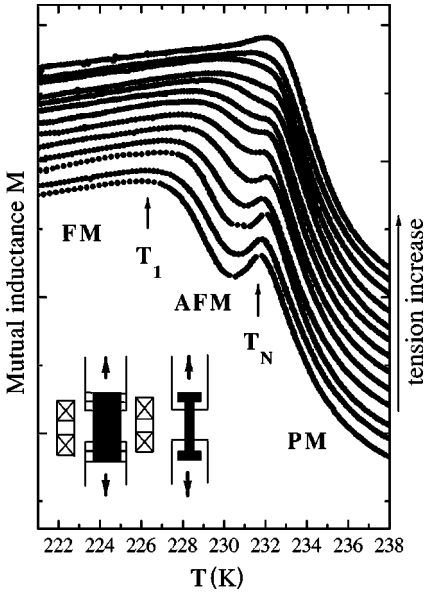

FIG. 3. Mutual inductance $M(T)$ dependencies at different tensions. Onsets are omitted, and the vertical shift is proportional to the tension applied. Arrows indicate characteristic temperatures. Tensions, from bottom to top, are 0, 60, 120, 190, 250, 310, 380, 440, 470, 530, 590, 660, and 720 bar. Inset: sketch of the experiment (front and side views).

wave vector are presented in Fig. 2 (for Bragg angle $\theta = 45°$). The helical wave vector happened to be smaller than that for pure Tb,[12] presumably due to the sample impurity. The temperature range where helical ordering occurs, 5.5 K wide, also turned out to be significantly more narrow than that in pure terbium. All these features were typical of the early Tb samples (see the review in Ref. 19). In our experiment this was an advantage, as we had better chances to suppress the helical structure completely before the sample rupture.

An I-shaped sample *en face* was cut out of this single crystal by spark erosion. Two holders of beryllium-copper were also cut by spark erosion so as to fit the sample shape. The *c* crystalline axis was parallel to load direction within $\pm 2°$.

The operational fragment of the sample was 3 mm long, with a cross section of 0.57×3.87 mm^2; the total length of the sample was 5.5 mm. The sketch of the experiment is presented in the inset of Fig. 3. The cell design was suitable for a neutron scattering experiment.

The load produced by steel string of rigidity 13.8 N/mm was transmitted to the cell by two long coaxial plungers made of stainless steel. The elasticity limit for polycrystalline Tb is some 2 Kbar; thus we never exceed half of this value for fear of destroying the sample.

The magnetic state of the sample was examined by magnetic susceptibility, that was measured by the ac mutual inductance technique at a frequency of 6030 Hz. Two mutual inductance coils were placed parallel to each other around the sample. The ac magnetic field was parallel to the sample surface and perpendicular to the crystal hexagonal axis *c* (see the inset of Fig. 3). The shape factor had no visible influence on the curve shape; hence we can accept the mutual inductance to be exactly proportional to the sample magnetic susceptibility.

The temperature dependences of mutual inductance $M(T)$ were taken on cooling, with a rate of less than 0.5 K/min in N_2 atmosphere. The sample temperature was monitored by a copper-constantan thermocouple, with ± 0.2 K accuracy.

A set of $M(T)$ curves at different tensions is presented in Fig. 3. At ambient pressure and at tensions $p < 700$ bar, the well-known characteristic splitting occurs on the curves. This splitting marks the Neel point at T_N and the point, often referred to as Curie point, where the sample converts to a ferromagnetic phase at T_1 (marked by arrows). These values at ambient pressure (231.5 and 226 K, respectively) agree well with those obtained by neutron scattering.

This splitting is obviously narrowed under tension increase, and it never occurs at tensions larger than 700 bar. It is hard to doubt that this phenomenon shows the complete suppression of helical ordering in the sample under uniaxial tension.

The tension dependences of characteristic temperatures are presented in Fig. 4. The magnetic ordering temperature T_N depends on tension almost linearly, and its slope $dT_N/dp = 0.9 \times 10^{-3}$ K/bar is of the same sign and order as the 1.6×10^{-3} K/bar value measured in the uniaxial compression experiment.[16]

The tension dependence of T_1 is, in contrast, essentially unlinear. Moreover, it can be fitted fairly well by the square-root dependence $(p^* - p)^{1/2}$ with parameter $p^* = 680$ bar (the curve in Fig. 4). Such behavior is typical of the electronic topological transition expected.

Hence it was demonstrated that helical ordering in rare-earth metal can be suppressed completely by a uniaxial tension as low as 680 bar. We speculate that this critical tension p^* corresponds to a change in FS topology caused by uniaxial deformation. The proposed evolution of the FS shape with applied tension is sketched in Fig. 4.

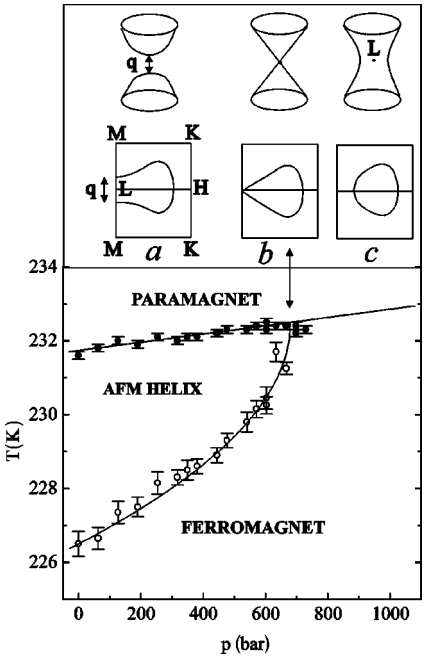

FIG. 4. Tension dependencies of characteristic temperatures T_N (black dots) and T_1 (open dots). Lines represent a linear fit for T_N, and a square root fit for T_1. Arrow marks the critical tension value $p^*=680$ bar. Insets: sketches of the proposed FS shape near the L point of the Brillouin zone. Top row: axonometry. Bottom row: FS intersection with MKHL face of the Brillouin zone. (a) $p<p^*$. (b) $p=p^*$. (c) $p>p^*$. q is the helical wave vector.

The numerical estimations of changes in the c/a value under uniaxial tension, of the order of 10^{-3} in our case, are as follows. The c/a ratio in our sample, measured by x-ray diffraction, was 1.580 ± 0.001. A straightforward calculation of the c/a variation under uniaxial tension 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 values for Tb—$c_{11}=0.68$, $c_{12}=0.25$, $c_{13}=0.21$, $c_{33}=0.71\times10^{12}$ din/cm^2 (see Ref. 20)—and critical pressure $p=p^*=680$ bar, we obtain $\Delta(c/a)=0.002$, and hence the critical value $(c/a)_{cr}=1.582\pm0.001$.

This critical value is in good agreement with those estimated in our recent work,[14] with uniaxial compression on $Ho_{0.4}Gd_{0.6}$ alloy: 1.580 ± 0.001. Hence c/a critical values for these two different substances are almost the same.

Our considerations were based on (i) the well-accepted ''nesting'' hypothesis and (ii) the hypothesis of the electronic topological transition in rare-earth metals that is evident from the classical calculations of FS's as well as from the recent experiments. The combination of these two plausible hypotheses immediately provided a number of transparent consequences, particularly the crucial role of the c/a ratio in the magnetic structure formation, and the existence of a c/a critical value that separates different magnetic phases.

This crucial role was confirmed by two experiments opposite in setting: ferromagnetic $Ho_{0.4}Gd_{0.6}$ was converted to the presumably helical state by uniaxial compression,[14] while helically ordered Tb was turned to a ferromagnetic state by uniaxial tension. In both substances the critical values of the c/a ratio happened to be almost the same: 1.580 ± 0.001 and 1.582 ± 0.001, respectively. These experiments support our considerations as well as our basic hypotheses.

Obviously the considerations presented are rather straightforward. An elaborated approach should consider the self–consistent problem for electronic, elastic, and magnetic phenomena in the vicinity of the electronic topological transition in presence of nesting. We hope this work will stimulate a theoretical treatment of the problem.

We are grateful to the late V. S. Zasimov for the x-ray examination of the sample. We are grateful to O. D. Chistiakov for the crystal provided. The work was partially supported by the Russian Foundation for Basic Research (Grant No. N98-02-17401).

[1] S.C. Keeton and T.L. Loucks, Phys. Rev. **168**, 672 (1968).
[2] H.M. Fretwell *et al.*, Phys. Rev. Lett. **82**, 3867 (1999).
[3] S.B. Dugdale *et al.*, Phys. Rev. Lett. **79**, 941 (1997).
[4] A.R. Mackintosh, Phys. Lett. A **28**, 217 (1968).
[5] R.C. Young, J. Phys. (Paris), Colloq. **40**, C5-71 (1979).
[6] L.I. Vinokurova *et al.*, Pis'ma Zh. Éksp. Teor. Fiz. **34**, 590 (1981) [JETP Lett. **34**, 566 (1981)].
[7] R. Ahuja, S. Auluck, B. Johansson, and M.S.S. Brooks, Phys. Rev. B **50**, 5147 (1994).
[8] L. Nordstrom and A. Mavromaras, Europhys. Lett. **49**, 775 (2000).
[9] A.P. Cracknell and K.C. Wong, *The Fermi Surface* (Clarendon press, Oxford, 1973).
[10] A. Andrianov, Pis'ma Zh. Éksp. Teor. Fiz. **55**, 639 (1992) [JETP Lett. **55**, 666 (1992)]; J. Magn. Magn. Mater. **140-144**, 749 (1995).
[11] I.M. Lifshits, Zh. Éksp. Teor. Fiz. **38**, 1569 (1960) [Sov. Phys. JETP **11**, 1130 (1960)]; for a recent review, see Ya.M. Blanter, M.I. Kaganov, A.V. Pantsulaia, and A.A. Varlamov, Phys. Rep. **245**, 159 (1994).
[12] H.R. Child, W.C. Koehler, E.O. Wollan, and J.W. Cable, Phys. Rev. **138**, A1655 (1965).
[13] S.B. Palmer, G.J. McIntyre, A.V. Andrianov, and R.J. Melville, J. Magn. Magn. Mater. **177-181**, 1023 (1998).
[14] A.Vl. Andrianov and O.D. Chistiakov, Phys. Rev. B **55**, 14 107 (1997).
[15] C. Dufour *et al.*, J. Phys.: Condens. Matter **9**, L131 (1997).
[16] H. Bartolin *et al.*, J. Appl. Phys. **42**, 1679 (1971).
[17] H. Umebayashi, G. Shirane, B.C. Frazer, and W.B. Daniels, Phys. Rev. **165**, 688 (1968).
[18] S. Kawano, N. Achiwa, A. Onodera, and I. Nakai, Physica B **180-181**, 46 (1992).
[19] D.C. Jiles *et al.*, J. Phys. F: Met. Phys. **14**, 3061 (1984).
[20] *Elastic, Piezoelectric, Pyroelectric, Piezooptic, Electrooptic Constants, and Nonlinear Dielectric Susceptibilities of Crystals*, edited by K.-H. Hellwege and A.M. Hellwege, Landolt-Börnstein, New Series, Group III, Vol. 11 (Springer-Verlag, Berlin, 1979), p. 41.