Ferromagnetic order at Tb surfaces above the bulk Curie temperature

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The magnetic order at surfaces of the 4*f* rare-earth metal terbium is investigated using electron capture spectroscopy (ECS), which probes the electron spin polarization (ESP) of the *topmost* surface layer. In ECS, the capture of spin-polarized electrons during grazing-angle ion-surface interaction is used to determine the ESP due to long- and short-ranged surface magnetic order. It is found that long-ranged ferromagnetic order exists far above the bulk Curie temperature, measured to be $T_{Cb} = 220$ K. At 140 K, the long-ranged ESP amounts to 24%. With increasing temperature, the ESP first decreases montonically up to the bulk Néel temperature $T_{Nb} = 228$ K, then exhibits a pronounced maximum at $T = 238$ K, and ultimately vanishes at the surface Curie temperature $T_{Cs} = 248$ K. These striking results on enhanced magnetic order at Tb surfaces suggest the presence of very large surface anisotropies.

INTRODUCTION

Recent studies have shown that the magnetic properties of surfaces may differ strongly from those of the bulk.[1-3] For sufficient enhancement of the interactions between the surface spins, an ordered surface phase is expected to coexist with a disordered bulk phase.[4] This remarkable phenomenon was first observed in Gd.[5,6]

The magnetic order at the *topmost* surface layer of a sample is conveniently studied with electron capture spectroscopy (ECS).[7] For Gd, ECS was used to investigate the temperature dependence of the surface magnetic order. Over the whole investigated temperature region, which ranged from well below the bulk Curie temperature to well above the surface Curie temperature, the surface magnetization was found to decrease monotonically with increasing temperature.[5,8]

In this work, we have studied the surface magnetization of another rare-earth, Tb, which, for reasons described below, is expected to behave differently from Gd.

In the bulk, Tb is ferromagnetic at low temperatures, up to the bulk Curie temperature $T_{Cb} = 220$ K, at which it undergoes an antiferromagnetic transition with helical spin structure. The antiferromagnetic (helical) order disappears at the bulk Néel temperature T_{Nb}, above which bulk Tb is paramagnetic.[9]

Contrary to Gd, which has zero total orbital angular momentum, Tb has eight electrons in the 4*f* shell, so that its total orbital angular momentum is equal to 3. This very high anisotropy is expected to strongly affect the temperature dependence and variation of the magnetic ordering at the topmost surface layer of Tb samples.

These expectations are confirmed by the present study: We first find that ferromagnetic order exists at the topmost surface layer of Tb *far above* the bulk Curie temperature $T_{Cb} = 220$ K. With increasing temperature, starting well below T_{Cb}, the electron spin polarization first decreases monotonically up to the bulk Néel temperature $T_{Nb} = 228$ K; it then exhibits a very sharp maximum at $T = 238$ K, which is almost independent of the applied magnetizing field, then decreases steeply and ultimately vanishes at and above the surface Curie temperature $T_{Cs} = 248$ K. This T dependence of the electron spin polarization of the topmost layer of Tb contrasts sharply with that found for Gd in which the electron spin polarization is a monotonic function of temperature from T well below T_{Cb} up to T well above the surface Curie temperature T_{Cs}.

EXPERIMENT

The basic process in electron capture spectroscopy (ECS) is the capture of spin-polarized electrons during small-angle surface reflection of deuterons D^+. For an angle of incidence of 0.3°, the distance of closest approach of the deuterons to the surface is about 0.8–1.5 Å. Therefore, the deuterons probe the spin-polarized local electron densities of state of the topmost surface layer of the sample.[7]

Typically, a well-collimated beam of 150-keV D^+ ions is reflected at grazing angle on the target, which is magnetized parallel to the surface plane and perpendicular to the incoming beam. Next, specularly reflected particles enter a transverse electric field, which separates the remaining D^+ from the neutral D^0 and ionic D^- created in one- and two-electron capture processes, respectively. Finally, the D^0 atoms then hit a tritium(^{3}H) target inducing the nuclear reaction $^3H(d,n)^4He$ ($D^0 + {}^3H = n + {}^4He$). The emitted ^{4}He particles are detected using two perpendicular surface barrier solid-state detectors.

One-electron capture processes at ferromagnetic surfaces yield D^0 atoms with polarized electron shells. A nuclear polarization of the D^0 atoms is then induced by hyperfine interaction and is measured by the asymmetry in the angular distribution of the ^{4}He particles emitted in the nuclear reaction described above. Nuclear polarization is a direct measure of the electron spin polarization of the captured electrons.

A target magnetizing field is applied to align otherwise randomly oriented Weiss domains, thereby producing a

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macroscopic magnetization along which the sign and magnitude of the ESP can be measured. With P the ESP along the target magnetizing field, we have $P=(n^+-n^-)/(n^+ + n^-)$, where n^+ and n^- are the fractional numbers of electrons with spin moment antiparallel (majority-spin electrons) and parallel (minority-spin electrons), respectively, to the target magnetizing field.

Many experimental difficulties must be overcome in order to maintain clean surfaces during multiple heating and cooling cycles. At all temperatures, before and after each measurement, the residual surface contaminations C, S, and O were shown, using a cylindrical mirror analyzer for Auger electron spectroscopy, to be less than 0.01 monolayer. The ESP measurements are performed at working pressures of about 5×10^{-11} mbar. Further details on our experimental procedures can be found in Ref. 7.

The bulk Curie temperature was determined by two independent methods, ferromagnetic induction and the magneto-optical Kerr effect. Details are given in Ref. 10.

RESULTS AND DISCUSSION

The main results are shown in Fig. 1, which illustrates the temperature dependence of the electron spin polarization of the topmost surface layer of a 1-mm-thick Tb sample. The experimental temperatures ranged from 140 to 300 K, respectively, well below and above the bulk Curie and Néel temperatures. The applied magnetic field was varied between $H=30$ and $H=600$ Oe. The ESP data shown in Fig. 1 correspond to $H=60$ Oe.

As the temperature increases from low values, the ESP is found to initially decrease monotonically. The same behavior was found in our earlier studies of Gd. Moreover, as in Gd, the ESP of the surface does not vanish at T_{Cb}. But, whereas for Gd the ESP always decreases monotonically up to the surface Curie temperature at which it vanishes, the ESP of the topmost layer of Tb exhibits a very pronounced nonzero minimum at, or at least very close to, T_{Nb}, beyond which it steeply increases within the next 10° to ultimately suddenly drop to zero at a surface Curie temperature $T_{Cs}=248$ K, remaining zero at higher temperatures.

It is tempting to account for such remarkable behavior of the ESP of the topmost layer by invoking the change of the effective coupling of the latter to the bulk, and assuming that the latter is antiferromagnetically, or at least helically, coupled to the second and deeper surface layers. This assumption is consistent with the observation that the ferromagnetic order, which sets in as temperature is lowered below T_{Cs}, decreases in the neighborhood of T_{Nb}, where antiferromagnetic or helical order appears in the bulk. A similar argument applies to the subsequent increase of the ESP below T_{Nb}, as helical order gradually gives way to ferromagnetic order. Clearly, the validity of this argument relies upon sufficiently strong interlayer couplings. Similar arguments were advanced in Refs. 6 and 11–13.

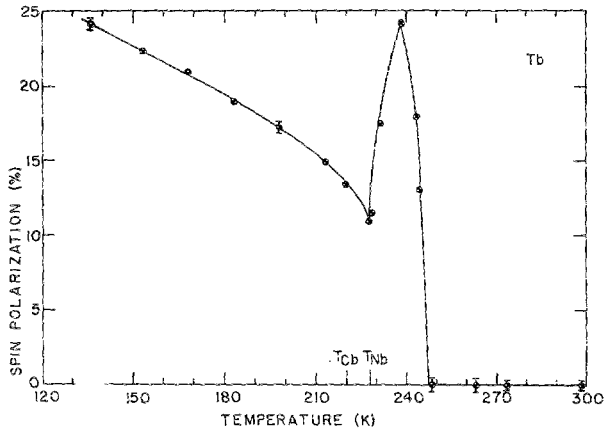

FIG. 1. Electron spin polarization of the topmost surface layer of 1 mm-thick Tb samples as function of temperature. T_{Cb} denotes the bulk Curie temperature as determined using ferromagnetic induction and the magneto-optical Kerr effect. T_{Nb} denotes the bulk Néel temperature of Tb.

It may finally be mentioned that recent work in our group on single crystalline Tb indicates that the behavior observed here is not due to the polycrystalline nature of the present Tb samples.

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