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**LETTER TO THE EDITOR**

# Stabilization of the helical phase in Tb and in $Dy_xTb_{1-x}$ alloys films grown epitaxially on Y

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**Abstract.** Neutron diffraction experiments and macroscopic magnetization measurements show evidence of the stabilization of the helical magnetic phase over large temperature ranges for Tb and $Dy_xTb_{1-x}$ alloy films grown epitaxially on yttrium. In particular, the temperature of the transition between the helical and the ferromagnetic states is shifted from 220 K for bulk terbium to 160 K for a pure terbium film grown epitaxially on yttrium, despite the low stability of the helical phase in the bulk element. The decrease of the Curie temperature is due to the negative *c*-axis strain induced by the epitaxial growth on yttrium. The epitaxial strains also induce modifications of the Fermi surface, which leads to an increase of the turn angle even at $T_N$. At low temperature, a long-wavelength-modulated phase, whose origin still remains to be explained, has been observed.

Hexagonal-close-packed heavy rare earths are known to exhibit *c*-axis-modulated magnetic phases. These phases are the consequence of quasi-parallel flat 'nesting' portions of the Fermi surface which give rise to a maximum of the conduction electron susceptibility along the *c*-direction for a wave vector $q_m$. This wave vector $q_m$ becomes larger and larger on increasing the atomic number [1]. For gadolinium $q_m$ is 0, which leads to a ferromagnetic structure at the ordering temperature. Then $q_m$ shifts from $0.22\pi/c$ for terbium to $0.49\pi/c$ for dysprosium, and to $0.57\pi/c$ for erbium. Correlatively, the temperature range of stability of the modulated phase increases with $q_m$: for example, the helical phase is stable only between 230 K and 220 K in terbium but extends from 179 K to 89 K in dysprosium.

On decreasing the temperature, these materials undergo a first-order transition giving rise to a ferromagnetic component. This transition is due to a gain in the magnetostrictive energy of the ferromagnetic phase which compensates the difference $\Delta E_{ex}$ between the exchange energies in the ferromagnetic and the modulated phases [2–4]. As a result, the ferromagnetic transition is accompanied by strains of the lattice.

Additionally, it is now well known that the epitaxial growth of dysprosium [5], holmium [6] and erbium [7] layers on yttrium shifts the ferromagnetic transition to a lower temperature or even suppresses it. This stabilization of the helical phase is due to the occurrence of a negative $\alpha$-strain induced by the epitaxial growth of the magnetic rare earth on yttrium, whose lattice parameters are larger. In a more precise analysis, it has been shown that epitaxy is responsible for the variation of the Curie temperature via three effects: clamping of the rare-earth lattice to the substrate one, epitaxial strains, and modification of the Fermi surface [8–10].

  

However, probably due to the weak stability of the helical phase of bulk terbium [11], very few results have been published on epitaxially grown terbium and terbium-based alloys [12]. In this letter, we present the main features of the magnetic behaviour of a 400 Å thick terbium film grown on yttrium, in the framework of the evolution of the properties of 400 Å $Dy_xTb_{1-x}$ epitaxial layers grown on yttrium. Using magnetization measurements and neutron diffraction experiments, we show that the helical phase is systematically stabilized over a large temperature range, in the alloy films as well as in the pure terbium film.

Terbium and dysprosium are neighbouring elements in the periodic table; they are miscible in all proportions and exhibit similar magnetic behaviours. As mentioned above, they present a helical magnetic phase followed by a ferromagnetic order. Furthermore, the magnetic behaviour of bulk terbium is still currently under investigation [13]. In these two rare earths, the anisotropy keeps the magnetic moments in the (0001) basal plane. One difference between these elements is that in the in-plane easy-magnetization direction: it is the *a*-axis for dysprosium and the *b*-axis for terbium. Another difference concerns the ordering temperatures mentioned above: the helical phase appears at higher temperature and is stable over a smaller temperature range in terbium than in dysprosium. In bulk $Dy_xTb_{1-x}$ alloys, the Néel temperature and the Curie temperature vary linearly with the dysprosium composition $x$, between the values given for pure dysprosium and terbium [14].

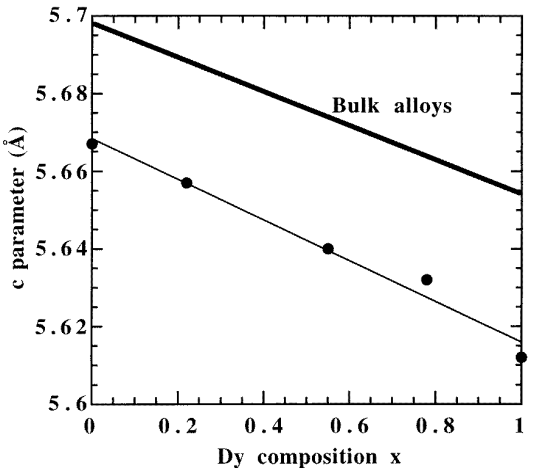


**Figure 1.** *c*-axis parameters deduced from x-ray diffraction measurements versus the dysprosium composition $x$ for various Y/400 Å $Dy_xTb_{1-x}$/Y epitaxial trilayers. The continuous bold line represents the *c*-axis parameters for bulk alloys.

$Dy_xTb_{1-x}$ films are grown on yttrium by molecular beam epitaxy on sapphire substrates using the method developed by Kwo *et al* [15]. The general structure of the samples is: $(11\bar{2}0)$ sapphire/500 Å (110) Nb/500 Å (0001) Y/400 Å (0001) $Dy_xTb_{1-x}$/500 Å (0001) Y. Films with the following concentrations have been prepared: $x = 0.78$, $x = 0.55$, $x = 0.22$ and $x = 0$. The crystallographic quality of the samples was checked *in situ* by RHEED and *ex situ* by high-angle x-ray diffraction. The coherence length along the *c*-axis was found to be equal to the film thickness and the in-plane mosaicity is around 0.25°. The *c*-parameters deduced from the position of the (0002) Bragg peak for the epitaxial $Dy_xTb_{1-x}$ alloys are presented versus the dysprosium composition in figure 1. As expected from a solid solution of the elements, the *c*-parameter varies linearly with

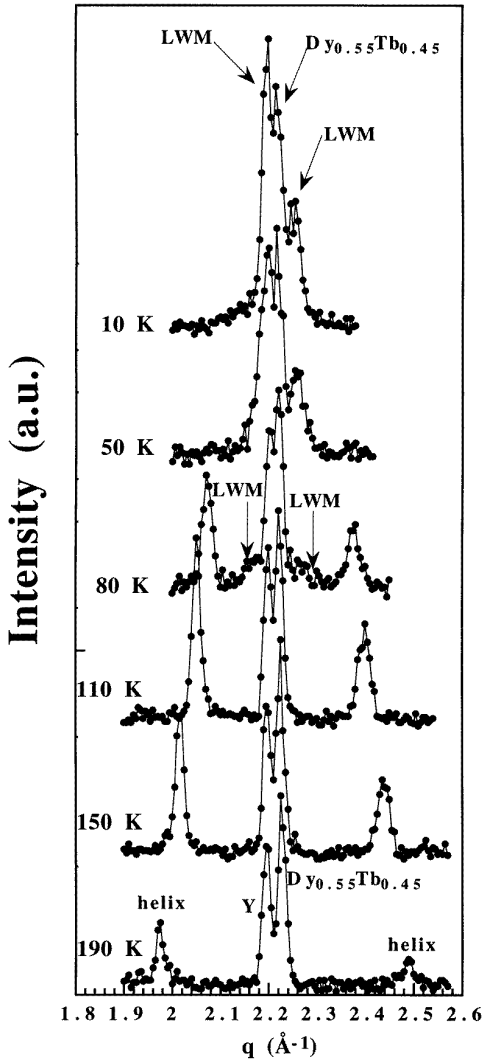


**Figure 2.** Neutron scattering spectra measured at different temperatures along the $\boldsymbol{c}^*$-direction around the (0002) reflection for a Y/400 Å $Dy_{0.55}Tb_{0.45}$/Y trilayer.

the composition of the thin-film alloy. However, the values are systematically about 0.5% lower than the bulk values. This indicates the occurrence of a negative $c$-axis strain induced by epitaxial growth on yttrium. This $c$-parameter reduction is consistent with the fact that yttrium presents a larger in-plane parameter than terbium and dysprosium: the epitaxy increases the in-plane parameter of the $Dy_xTb_{1-x}$ alloy and consequently reduces its $c$-parameter.

Magnetization measurements were carried out with a standard SQUID magnetometer. Neutron diffraction experiments were performed on the G4.3 triple-axis spectrometer at LLB (Saclay, France). The incident neutron wavelength was 4.245 Å and the collimations in front of and beyond the monochromator and the analyser were 60′, 30′, 30′ and 30′, yielding an elastic resolution width of 0.008 $Å^{-1}$.

Neutron scattering patterns have been collected for each sample along the $\boldsymbol{c}^*$-direction around the (0002) reflection, as a function of temperature. A typical set of neutron

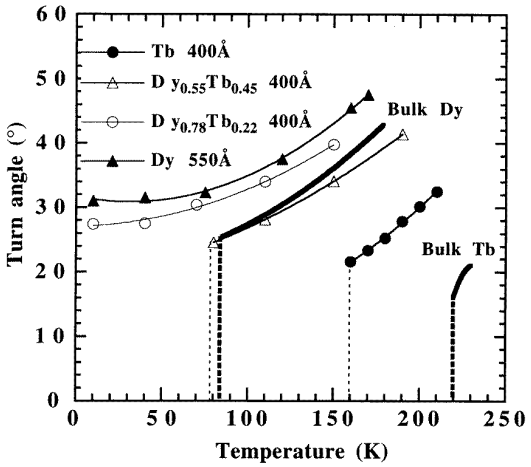


**Figure 3.** The thermal dependence of the turn angle between the magnetic moments in various Y/400 Å $Dy_xTb_{1-x}$/Y trilayers (filled triangles: $x = 1$; open circles: $x = 0.78$; open triangles: $x = 0.55$; filled circles: $x = 0$). The continuous bold lines represent the values for bulk terbium and dysprosium [16].

scattering spectra for the Y/400 Å $Dy_{0.55}Tb_{0.45}$/Y sample is shown in figure 2. As mentioned below, the ordering temperature $T_N$ for this trilayer is 203 K. At 190 K, the (0002) nuclear peaks due to yttrium and to the $Dy_{0.55}Tb_{0.45}$ alloy (at $q = 2.195$ Å$^{-1}$ and $q = 2.228$ Å$^{-1}$) are surrounded by two magnetic satellites located symmetrically about the (0002) nuclear peak of the alloy, at $q = 1.975$ Å$^{-1}$ and $q = 2.488$ Å$^{-1}$. These peaks are unambiguously due to the development of the magnetic helix in the $Dy_{0.55}Tb_{0.45}$ alloy. When the temperature decreases, the intensity of these magnetic peaks first increases, and their positions shift towards the alloy Bragg peak, which is consistent with an increase of the magnetic modulation wavelength. At lower temperature (80 K), the intensity of the magnetic peaks decreases and the satellites have vanished at 50 K. It is obvious that the helix disappears at a temperature definitively smaller than the Curie temperature of the same compositional bulk alloy, which is 150 K. This result is indicative of the stabilization of the helical state by epitaxy, over a temperature range of several tens of degrees.

However, it is to be noted that the magnetic intensity of the satellites is not transferred to the nuclear (0002) peak, as occurs in a classical helical–ferromagnetic transition, but to two new satellites located very close to the (0002) peak. Taking the full width at half-maximum of these satellites into account, we conclude that they are characteristic of a long-wavelength modulation, LWM (170 Å at 50 K). When the temperature decreases, their positions shift towards the alloy Bragg peak; this feature is characteristic of an increase of the wavelength of this modulation which becomes 200 Å at 10 K.

Similar sets of neutron scattering spectra have been collected for the different $Dy_xTb_{1-x}$ films. As expected from the concentration effect, when $x$ decreases, the helical phase is less stable. It is noteworthy that, for all of the epitaxially grown films, the magnetic satellites persist down to a lower temperature than for the corresponding bulk alloys. In particular, they persist down to 160 K for the 400 Å thick pure terbium film, which is a temperature considerably smaller than the one for which the helical state vanishes in bulk terbium (220 K). This shows that, in terbium layers epitaxially grown on yttrium, the

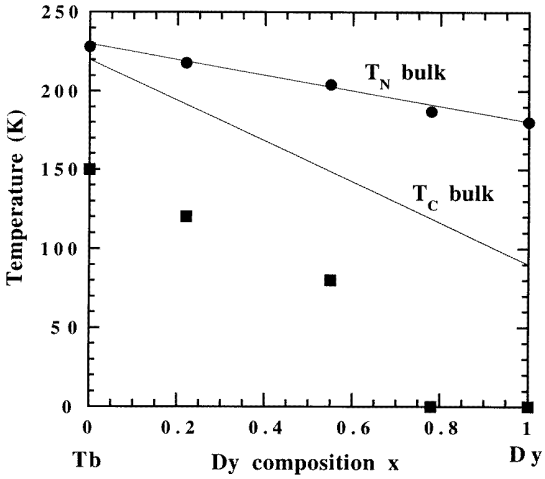


**Figure 4.** The variation of the Néel temperature (filled circles) and of the Curie temperature (filled squares) versus the dysprosium composition $x$ for various Y/400 Å $Dy_xTb_{1-x}$/Y epitaxial trilayers. The solid lines indicate the variations of these temperatures for bulk materials.

helical phase is stabilized by epitaxy over a very large temperature range, despite the fact that this phase presents a weak stability in bulk terbium. Consequently, as regards the shift of the transition temperature between the helical and the ferromagnetic states, terbium layers grown on yttrium present the same behaviour as dysprosium, holmium, or erbium layers grown on yttrium [5–7].

For the whole set of samples, the LWM phase that we observed for the $Dy_{0.55}Tb_{0.45}$ layer is present at low temperature. In contrast, this phase does not appear in pure dysprosium films grown epitaxially on yttrium and prepared in similar conditions [8, 16]. To investigate these features, a thick (7000 Å) terbium film has also been studied by neutron diffraction. This film is directly deposited on niobium and presents the same $c$-parameter as bulk terbium. The neutron spectra exhibit peaks due to the helical phase between 230 K and 219 K, and the peaks characteristic of a long-wavelength modulation appear at around 222 K. As in the above $Dy_{0.55}Tb_{0.45}$ alloy film, their positions rapidly shift towards the Bragg peak; but, unlike in the case of the alloy film, they merge with the Bragg peak at about 200 K, which leads to a ferromagnetic order. Thus, the LWM phase does not depend on epitaxial strains. At this stage, it is difficult to reach a conclusion regarding the origin of this observed long-wavelength modulation, but it seems that it needs to be investigated to achieve a better understanding of the magnetic behaviour of terbium.

The thermal variation of the turn angle has been determined from the position of the magnetic satellites correlated with the helical phase (figure 3). The values of the turn angles in bulk terbium and dysprosium are given for comparison [17]; the values for a 550 Å thick Dy film taken from a previous study are added [16]. As in bulk materials, the turn angle decreases with temperature, which is interpreted in terms of superzone gaps in the Fermi surface, due to the magnetic ordering. What is noticeable is that for all of the films grown on yttrium, the turn angle is larger than in the corresponding bulk materials. This feature is true even at $T_N$, where the magnetic ordering is small and the magnetoelastic effects negligible: for pure terbium, $q_m$ changes from $0.22\pi/c$ for the bulk element to $0.35\pi/c$ for the epitaxially grown film. These higher values of the turn angle at $T_N$ are consistent with

the increased stability of the helical phase and mean that the Fermi surface is changed by epitaxial strains. For the pure Dy film and the $Dy_{0.78}Tb_{0.22}$ film, the turn angle never drops to zero, which means that the helical phase is stable down to low temperature.

Finally, magnetization measurements were performed in order to determine precisely the stability range of the helical phase. The Néel temperatures $T_N$ of the films were obtained from the position of the cusp observed in field-cooled magnetization measurements. As shown in figure 4, they are very close to the Néel temperatures of the bulk alloys and vary almost linearly with the composition $x$.

First magnetization curves as a function of the magnetic field were also collected over a temperature range in which the helix is stable under zero field. They exhibit two domains: (i) a domain of low susceptibility at low field; (ii) a sudden increase of the magnetization which begins at a critical field $H_c(T)$ and is attributed to the field-induced ferromagnetic transition (we do not find any difference between the ferromagnetic order and the so-called LWM phase, which is probably easily destroyed by application of a field). The Curie temperatures presented in figure 4 were determined as the temperatures at which $H_c$ drops to zero. For the Dy and $Dy_{0.78}Tb_{0.22}$ films, the critical fields remain different from zero down to 10 K. This proves again that, for these films, the helical phase is stabilized down to low temperature. As already shown from neutron scattering, the Curie temperature is systematically lower in epitaxial alloys than in bulk materials.

In conclusion, we show evidence of the stabilization of the helical phase of $Dy_xTb_{1-x}$ alloys due to their epitaxial growth on yttrium. It is particularly noticeable that the Curie temperature is shifted from 220 K to 160 K for a pure terbium film grown epitaxially on yttrium, despite the low stability of the helical phase in the bulk element. This observation means that modification of the helical–ferromagnetic transition in epitaxially grown terbium is part of the general behaviour of epitaxially grown heavy rare earths. A long-wavelength-modulated phase appears at low temperature in the alloys and in the 400 Å thick film of pure terbium grown epitaxially on yttrium, as well as in a 7000 Å thick terbium layer. The precise configuration and the origin of this unexpected magnetic phase are still under investigation.

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