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MAGNETIC ORDERING OF Tb OVERLAYERS ON Ni(111)

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Abstract

The $5p_{3/2}$ to $5p_{1/2}$ shallow core level branching ratios in different photoemission geometries provide a measure of the magnetic ordering in rare earth overlayers as a result of final state effects in photoemission. This $5p$ level anisotropy can be used to probe magnetic ordering across the Tb Curie temperature as well as magnetic ordering induced by a ferromagnetic substrate in a paramagnetic overlayer. Results are shown for paramagnetic Tb overlayers on Ni(111). The Ginzberg–Landau theory can be used to accurately model terbium thin films for $T > T_c$.

Introduction

We have recently introduced a technique for probing local magnetic ordering using conventional angle resolved photoemission spectroscopy [1–4], without spin detection of the photoelectrons. By measuring the 5p core levels in different photoemission geometries, the magnetic ordering of Gd [1,3] and Tb [4] overlayers have been characterized. While most investigations of rare earth metal interactions with the transition metals have been directed toward Gd overlayers, Tb has a low Curie temperature (213 K) which permits investigation of ferromagnetic and paramagnetic Tb without the complications associated interdiffusion and alloy formation [5].

Experimental

The experiments were performed in a U.H.V. system equipped with a hemispherical analyzer for angle resolved photoemission [1–3]. The crystal was prepared by repeated 2 KeV Ar⁺ ion bombardment and careful annealing, as previously described [3]. The deposited thickness of our films was determined using an oscillating crystal thin film monitor, but the absolute thickness of our films was calibrated based upon changes in the core level binding energy shifts as seen for gadolinium [6]. This latter procedure is based upon surface to bulk core level shift arguments. Data has been reduced [4] following procedures developed for gadolinium overlayers [3].

Changes in the Tb 5p branching ratios across the Curie temperature

The initial state eigenspinors of the Tb $5p_{3/2}$ level, $m_j = \pm 3/2$, are of well defined electron spin character, while the $5p_{1/2}$, and $5p_{3/2}$, $m_j = \pm 1/2$, eigenspinors have mixed electron spin character [7]. As a result of these differences, there are differences in the final state valence 5d interactions with these shallow core levels. These changes in the final state interactions are easily observed in angle resolved photoemission by changing the incident light polarization [1,3,4]. The strongest manifestation of this 5d–5p coupling is a change in the 5p branching ratio with changes in the incident light polarization. Changing the incident light polarization selectively excites different symmetry states in the initial state eigenspinors. This can occur even without alignment of the spatial coordinate system with the magnetic coordinate system [4].

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This probe of the magnetic ordering can be quantified on a relative scale by the ratio of the 5p branching ratios for p and s polarized light (β_p/β_s), which we denote as the p level anisotropy [4]. The p level anisotropy has been measured and plotted for a variety of Tb film thicknesses at temperatures well above (300 K) and well below (170 K) the Tb Curie temperature (220 K). These results are summarized in Fig. 1, for Tb overlayers on Ni(111).
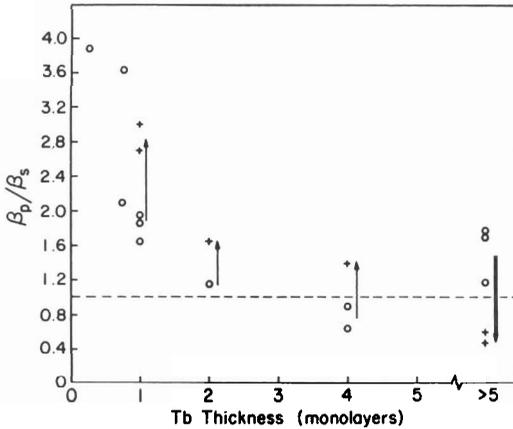

Figure 1. The p level anisotropy $\frac{\beta_p}{\beta_s}$ as a function of Tb overlayer thickness on Ni(111). The (+) indicates data taken at 170 K while the (o) indicate 300 K. The arrows indicate the changes that occur as a result of magnetic ordering. The changes that occur with a $d_{xz,yz}$ occupation at E_f are indicated by the thin arrows while the changes that occur with a $d_{3z^2-r^2}$ occupation at E_f are indicated by a thick arrow.

For thin films, the p level anisotropy is observed to increase with a decrease in temperature from above T_c to below T_c, while for thick films, the p level anisotropy is observed to decrease. This is a result of the changing symmetry of the 5d bands near the Fermi energy [4]. For thin films the 5d bands are of $5d_{xz,yz}$ character near E_f while for the thicker films they are of $d_{3z^2-r^2}$ character [4]. The p level anisotropy changes are revisible for several cycles of temperature change above and below T_c, as can be seen in Fig. 2.

It is also clear from Fig. 1, that the p level anisotropy deviates away from unity, the expected value, even for temperatures above T_c. This is particularly true as the film thickness decreases. While there are substantial p level anisotropies above the Tb Curie temperature, this can be easily reconciled with magnetic ordering imposed by the substrate. At 300 K, the system is above the Tb Curie temperature, but well below the Ni Curie temperature (627 K). Thus for very thin films, the Tb overlayer is in the applied magnetic field of the substrate. This will induce magnetic ordering as in a paramagnet:

$$M = \mu_B^2 HN(Ef). \qquad (1)$$

This ordering will damp away from the interface and can be understood in terms of Ginzburg–Landau theory.

Ginzburg–Landau theory applied to paramagnetic Tb on Ni(111)

Recently there have appeared theoretical studies in which the spatially dependent magnetization, $M(\vec{r})$, has been computed in films, alloys, and ferromagnetic multilayers, applying the Ginzburg–Landau formulation [8–17]. J. Mathon [8] has argued that Ginzburg–Landau theory can be applied to paramagnetic overlayers on ferromagnetic substrates if the overlayer is less than 10 monolayers thick.
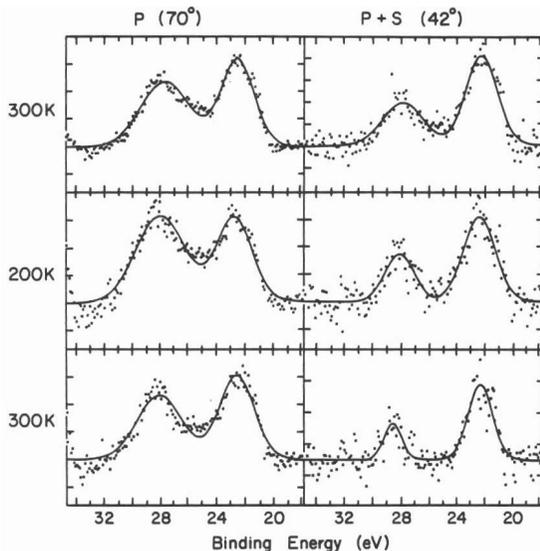
Figure 2. Photoemission spectra of the Tb 5p levels for two light polarizations. The spectra were taken at various temperatures from the same 1 monolayer thick Tb film on Ni(111). The photon energy was 60 eV.

The Ginzburg–Landau equation in the Gaussian approximation with no external field may be written as [13]

$$-C\,\frac{d^2M(y)}{dy^2} + AM(y) + BM^3(y) = H^m, \qquad (2)$$

where y is the distance of the considered point in the film to the interface. B and C are positive constants with $0 \leq y \leq z$ (z is the film thickness). The values A, B, and C are actually temperature dependent [12], but we shall apply the above equation at only one temperature. The correlation length $\kappa^{-1} = (C/A)^{\frac{1}{2}}$. The term H^m on the right hand side is the demagnetization field. Integrating this equation [9] gives,

$$\frac{1}{\kappa^2} \left(\frac{d\mathrm{M}(y)}{dy}\right)^2 = \mathrm{M}(y)^2 + b\mathrm{M}(y)^4 + a, \tag{3}$$

where a is a constant of integration and $b = \frac{B}{2A}$. To obtain this result we have set H^{m} to zero. This is easily shown to be justified when the film is in the paramagnetic state.

If this equation is integrated a second time, we obtain the following equation for the dimensionless magnetization $m(y) = \left(\frac{b}{a}\right)^{1/4}\mathrm{M}(y)$:

$$\kappa y = \int_{m(y)}^{m(0)} \frac{dm}{\sqrt{\eta m^4 + m^2 + \eta}} \tag{4}$$

Here, the unitless parameter η is defined by $\eta = (ba)^{\frac{1}{2}}$ and measures the strength of the nonlinear term $B\mathrm{M}^3(y)$ in Eq.(2). The solution in Eq.(4) applies when $m(0) \geq m(y)$, as is the case for paramagnetic overlayers on ferromagnets.

This procedure has been applied [8,9,15] to the case of Pd (a nearly ferromagnetic metal) on Ni, which is similar to our system of Tb on Ni. The solution in both cases assumes $a > 0$, which can be justified from the boundary conditions [8].

The integral in Eq.(4) can be easily cast in the form of incomplete elliptic functions of the first kind, $F(k,\phi)$,

$$ky = \sqrt{\frac{1}{\eta\beta_2}}[F(k,\phi_0) - F(k,\phi_y)], \tag{5}$$

where $k = \sqrt{\frac{\beta_1}{\beta_2}}$, $\phi_i = \sin^{-1}(\frac{m(i)}{\sqrt{\beta_1}})$, $\beta_1 = \frac{-\eta^1 - \sqrt{\eta^{-2} - 4}}{2}$, and $\beta_2 = \frac{-\eta^{-1} + \sqrt{\eta^{-2} - 4}}{2}$.

Using an expansion of the elliptic function valid for small η (appropriate for a paramagnetic film) and keeping only the leading term, the result

$$M(y) = Se^{-\kappa y} - Te^{\kappa y} \tag{6}$$

is obtained. The two constants of integration, S and T, have replaced the original pair of constants, $m(0)$ and a, that appeared in Eq.(4). Also, S and T obey $S > T > 0$.

Since for very thick films the free surface magnetization approaches zero, the constant $T \longrightarrow 0$. This implies that T is small. In what follows, we fit Eq.(6) to the data for paramagnetic Tb, restricting the fit to the case for which $T \ll S$, allowing the second term in Eq.(6) to be omitted. Then

$$M(y) \cong Se^{-\kappa y}. \tag{7}$$

Because the p level anisotropy is unity with no magnetic ordering,

$$\frac{\beta_\mathrm{p}(y)}{\beta_\mathrm{s}(y)} - 1 = p\mathrm{M}(y), \tag{8}$$

where p is an appropriate scaling constant. We have fit equations 7 and 8 to our data as shown in Fig. 3, and the agreement with this exponential profile is good.

If the decay rate is a simple exponential $M(y) \sim e^{-\kappa y}$ over $0 \leq y \leq z$, then $\lambda^{-1} = \kappa$ follows from Eq.(9). Returning to the data for Tb, the fact that its paramagnetic phase follows an exponential decay so well indicates that $\kappa\lambda \sim 1$. Hence, there exists at most, very weak suppression or very weak enhancement at the Tb free surface. The Tb free surface acts like a paramagnet and does not show either strong enhancement or "live" behavior. A similar result (i.e., $\kappa\lambda \sim 1$) was found by Gradmann and Bergholz [20] for Pd overlayers on Ni, at room temperature.

Conclusions

Our results show that Tb overlayers magnetically interact with the Ni substrate and the Ni will induce some magnetic ordering in very thin (< 4 monolayer) Tb films. This is clearly the case for Tb films at temperatures above the Tb Curie temperature where the overlayer is expected to be paramagnetic. The magnetic coupling with the paramagnetic Tb films is damped with increasing distance away from the interface in a manner consistant with Ginzburg–Landau theory. We find the correlation length for Tb to be $\kappa^{-1} = 0.71 \pm 0.19$ monolayers.

Acknowledgements

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