PRAMANA
— journal of
physics

Vol. 58, Nos 5 & 6
May & June 2002
pp. 725–730

Stoner vs. spin-mixing behavior in the bulk magnetism of Gd: A spin-resolved photoemission study

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Abstract. The temperature dependence of the rare-earth Δ_2-bulk band has been regarded as an exemplary case which realizes the simple Stoner behavior. We examined the evolution of GdΔ_2 bulk bands with temperature in the range $0.5 \leq T/T_C \leq 1$ with spin-resolved, photoemission spectroscopy. The direct observation of the spin-dependent spectral line shapes reveals a complex temperature dependence and manifests a clear inadequacy of the Stoner model to the description of the magnetism in rare earths.

Keywords. Spin-resolved photoemission; Stoner model; spin-mixing model; rare-earth magnetism.

PACS Nos 75.70.Ak; 75.25.+z; 79.60.Bm

1. Introduction

Understanding the relation between electronic and magnetic properties at finite temperatures is a central question for many branches of solid state physics. Two simple models are usually considered in order to schematically describe the limiting cases, depending on the degree of the electron localization. The Stoner model [1] predicts that the exchange splitting of delocalized states parallels the temperature dependence of the magnetization. In this case, the exchange-split electronic sub-bands gradually merge together and become degenerate at the Curie temperature. The spin-mixing model [2], on the other hand, describes strongly localized systems where thermal fluctuations reduce the magnetization, but leave the local moments and the local electronic structure unchanged.

Rare earths provide apparently a simple case for these studies where the magnetism appears due to indirect exchange coupling between localized $4f$ moments via the highly delocalized ($5d6s$)-valence states [3]. Gd is the most investigated case, being a prototype Heisenberg ferromagnet with a large moment ($S = 7/2$) in the half-filled $4f$ levels. Various investigations [4–6] report that the exchange splitting of the bulk valence bands vanishes at the Curie temperature ($T_{Cb} = 293$ K) as also observed in other rare earths [7,8].

These are interpreted as a first evidence for the applicability of the Stoner model to real ferromagnetic systems. Interestingly, a higher degree of localization for the surface states of Gd(0001) leads to controversial results. A scanning tunneling spectroscopy study reports the persistence of the surface state exchange splitting, $\Delta_{ex}^{surface}$ at and above T_{Cb} [9]. Photoemission and inverse photoemission studies on the surface states reached strongly contrasting conclusions, ranging from pure spin-mixing to Stoner behavior [6,10,11].

While most attention has been devoted to the surface states magnetism, the bulk bands have been until now indisputably believed to follow a Stoner behavior. The spin-resolved measurements in the present study exhibit a complex temperature dependence of the spectral line shapes suggesting clear inadequacy of the Stoner model in describing the bulk band behavior. These results establish that changes in both the spin polarization and the exchange splitting of valence band states play a significant role on the magnetism of rare earth metals.

2. Experimental

Spin and angle-resolved photoemission spectroscopic (PES) measurements were carried out at the TGM5 wiggler-undulator beamline at BESSY, Berlin. The experimental resolution was 160 meV at 34 eV photon energy. The spin polarization was measured by means of a Mott detector operating at 100 keV. 30 monolayer (ML) thick Gd(0001) films were epitaxially grown by e-beam evaporation on a clean W(110) surface at room temperature ($P < 2 \times 10^{-10}$ mbar) and subsequently annealed at 600 K. The cleanliness and crystalline structure of the samples were confirmed by the negligible spectral intensities of impurity-related features in PES and by sharp LEED patterns, respectively.

3. Results and discussion

We show normal emission spectra of Gd measured at various temperatures in figure 1. The intense feature close to E_F is due to $5d_{z^2}$ surface states and has a predominantly up-spin character. With increase in temperature, the spin polarization of these states continuously decreases along with a small shift towards E_F. A simulation of the spin-integrated surface feature by a Lorentzian convoluted with the Fermi function and energy resolution (solid lines in the figure) estimates a small increase (~ 0.1 eV) in the linewidth with temperature and the peak position remains below E_F till room temperature, consistent with previous observations [9,11].

The structures at higher binding energies, derived from ($5d6s$)-bulk states, exhibit $\Delta_{ex}^{bulk}(\approx 0.9$ eV) at 120 K to be close to the theoretical estimations (~ 1 eV at $T = 0$ K) [5,12]. Interestingly, the distinct signature of the up- and down-spin states in figure 1a gradually disappears with the increase in temperature. This is the key observation which led, in previous studies, to the conclusions of a Stoner behavior. We note, however, that the linewidth of this feature at 293 K is more than two times larger than that of non-magnetic La [13]. Since the crystal structure and the valence configuration $(5d6s)^3$ are very similar for La and Gd, the above comparison suggests that the larger linewidth of the Gd Δ_2-band might have a magnetic origin.

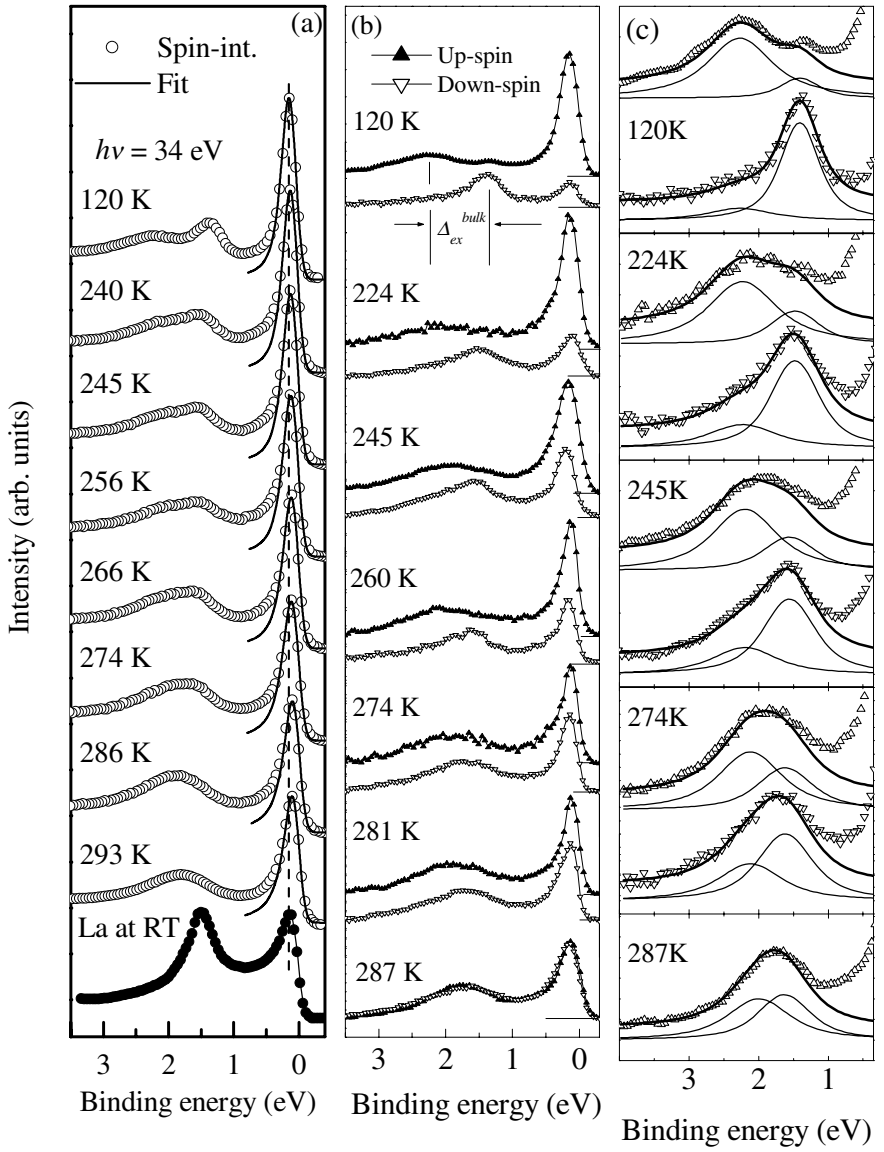

Figure 1. (**a**) Spin-integrated and (**b**) spin-polarized spectra of 30 ML Gd at different temperatures. The solid lines are the fit to the surface peak. (**c**) Spin-resolved spectra of (5*d*6*s*)-bulk emissions at different temperatures. The thick solid line is the simulated total spectra and the thin lines are the components.

The spin-resolved data in figure 1b show a distinct modification of the line shape with temperature in addition to the decrease of $\Delta_{\mathrm{ex}}^{\mathrm{bulk}}$. The linewidth of each spin-component becomes much broader at intermediate temperatures (see figure 1c at an expanded scale),

which cannot be visualized in the spin-integrated measurements. As the center of gravity of the down-spin emission shifts to higher binding energies, its overall linewidth first increases (120 K $< T <$ 245 K) and then decreases (for $T \geq$ 274 K). A similar trend is also found for the linewidth of the up-spin features. These changes are not expected in the Stoner picture and also cannot be described by electron or hole lifetime broadening and/or phonon broadening ($\sim$ 0.1 eV).

The changes in the spectral asymmetry with temperature indicates that a single-peak structure in each spin channel cannot account for the evolution of the spectral line shapes. Two components are most evident at 120 K. Thus, we simulate the bulk features in terms of two Voigt (Lorentzian + Gaussian) functions after a linear background subtraction by least squared error method. The Gaussian and Lorentzian components account for the resolution and lifetime broadening, respectively. The simulated spectra are shown by thick solid lines in figure 1c. All the spectra could be described consistently only with two Voigt functions (figure 1c). The relative integrated intensity and the spectral width of the two components in the down-spin spectra is exactly reproduced (with reversed intensity ratio) in the up-spin ones, providing confidence to our simulations. It is evident from the figure that, besides a reduction of the exchange splitting, the spin polarization of the individual Δ_2-emission features decreases with increasing temperature. The Lorentzian widths exhibit a linear dependence (figure 2a), $\gamma = \gamma_0 + \alpha E^2$ ($\gamma =$ FWHM, $E =$ binding energy) suggesting a Fermi liquid behavior. The values of γ_0(=0.25 eV) and α(=0.15 eV^{-1}) are very similar to the energy dependence of the quasi-particle lifetime broadening in other systems [14]. The open diamond representing the linewidth of the bulk feature of La [13] agrees well with the simulated data. The consistency of the above analysis with the experimental results suggests that the large linewidth of the Gd bulk feature at $T \cong T_C$ is mainly due to the persistence of exchange splitting.

The qualitative scenario that emerges from these results is shown schematically in figure 2b. In the ground state ($T_1 = 0$ K), the bands with up- and down-spins are separated by exchange splitting with 100% spin polarization. At finite temperatures, the exchange splitting reduces, and, as in the spin-mixing model, also the spin polarization decreases, as shown successively for $T = T_2$ and T_3 ($T_2 < T_3$). Thus, the spectra in each spin channel will show two components with relative intensities depending on the temperature. Finally, the spin polarization vanishes at the Curie temperature ($T_4 = T_C$). In this case, the spectral shape has to be identical in both the spin channels and corresponds to the spin integrated spectrum. The presence of an exchange splitting at this temperature or above cannot be detected in the spectra, unless it is comparable with the intrinsic linewidth of the individual peaks. In fact, the lifetime broadening for the bulk feature is too large ($\geq$ 0.7 eV) to observe a distinct signature of the exchange splitting ($\approx$ 0.5 eV at T_{Cb}).

We now compare the exchange splitting and the spin polarization for surface and bulk states in figure 2c and d, respectively. The values of $\Delta_{ex}^{surface}$ are reproduced from Bode *et al* [9]. It is clear that the magnetization vanishes near T_{Cb} for *both* surface and bulk [15]. A more rapid decrease of the surface spin polarization compared to the bulk is consistent with the observations on other ferromagnetic systems [16], and also with a recent investigation on Gd [15].

The present results, thus, provide a direct evidence that the valence band in bulk Gd cannot be described within a purely Stoner or spin-mixing model. Theoretical calculations based on the combination of many-body interactions and *ab initio* band structure by Nolting *et al* [17] predict a temperature behavior strongly dependent on the degree of itineracy

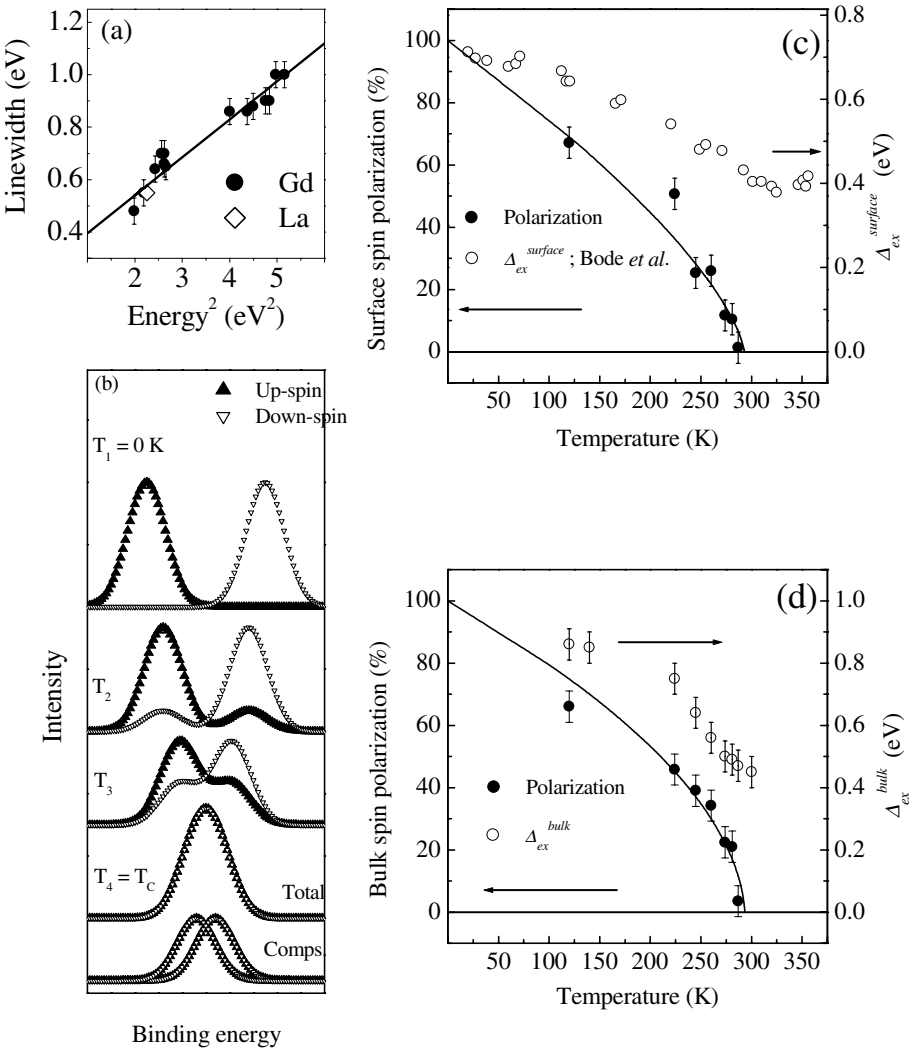

Figure 2. (**a**) Lorentzian widths as a function of $(BE)^2$. (**b**) Schematic model describing the evolution of magnetic properties with temperature, T where $T_1(=0\,K) < T_2 < T_3 < T_4(=T_C)$. Temperature dependence of the spin polarization and exchange splitting for (**c**) the surface and (**d**) bulk electronic structure. The solid lines show a guide to the eye.

of the corresponding electronic states and on the specific k point in the Brillouin zone. It is shown that weakly correlated s-like states exhibit a temperature dependence closer to the Stoner picture, while relatively more localized d-like states approach a spin-mixing behavior. The general behavior is, however, intermediate between these two extreme cases, as reported in the present study. The bearing of these findings is likely to be significant for the other rare-earth metals as well, due to the very similar character of their band states. It

is interesting to note here that other magnetic rare earths also exhibit a large linewidth of the Δ_2-band near T_C, with only a small difference above and below the transition temperatures [7,8,13].

4. Conclusion

In summary, we have investigated the temperature dependence of the electronic structure in Gd by means of spin-resolved photoemission spectroscopy. The spin-resolved spectra reveal a complex temperature dependence, that could not be detected in spin-integrated measurements. In contrast to previous interpretations, the present results firmly establish that the Stoner picture cannot describe the Gd bulk bands.

Acknowledgements

KM thanks Alexander von Humboldt Stiftung and DLR, the Federal Republic of Germany, for financial assistance.

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