Magnetic effects in the band structure of ferromagnetic and antiferromagnetic lanthanide–metal films

C. Schüßler-Langeheine[a],*, E. Weschke[a], H. Ott[a], A.Yu. Grigoriev[a,b], A. Möller[a],
R. Meier[a], Chandan Mazumdar[a], G. Kaindl[a]

[a]*Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin-Dahlem, Germany*
[b]*Institute of Physics, St. Petersburg State University, 198904 St. Petersburg, Russia*

Abstract

Systematic magnetic effects in the band structure of the heavy lanthanide metals Gd, Tb, Dy, and Ho were studied by angle-resolved photoemission from epitaxial films. The temperature-dependent exchange splitting exhibits a different behavior for delocalized and localized band states. For the delocalized Δ_2 bulk band states, a Stoner-like temperature dependence of the splitting is observed in ferromagnetic and helical antiferromagnetic phases. A more localized oxygen-induced surface state shows a temperature-independent contribution to the splitting. All observed magnetic splittings scale with the 4f spin moment. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

The magnetic properties of lanthanide elements are strongly determined by their local 4f moments, which persist in the solid state. Long-range magnetic order in these systems is caused by an indirect-exchange interaction between 4f moments via the polarization of valence electrons (RKKY interaction). The oscillatory character of this interaction together with strong anisotropies leads to a variety of periodic magnetic structures, including ferromagnetic, helical antiferromagnetic, and conical fer-

rimagnetic phases [1–4], well understood in terms of the shape of the Fermi surface [2]. The influence of magnetic order on the valence-electronic structure, on the other hand, is a current topic of research [5–14]. Theoretical calculations by Nolting et al. [8] for Gd metal point out the importance of two different contributions to the magnetic splitting, with their relative weights determined by the degree of localization of the respective band state. The first contribution, determining the behavior of fully localized valence states, reflects the local exchange with the essentially temperature-independent 4f moment and is thus expected to result in a temperature-independent energy splitting. The second contribution applies to strongly delocalized states averaging the 4f moment over a larger crystal volume. They are expected to show a mean-field behavior with a

*Corresponding author. Tel.: +49-30-838-52988; fax: +49-30-838-56560.

E-mail address: christian.schuessler@physik.fu-berlin.de (C. Schüßler-Langeheine).

splitting that reflects the net magnetization. The latter kind of behavior has been experimentally observed by photoemission (PE) for the Δ_2 band of ferromagnetic Gd metal [5], while the observation of resonances in the optical reflectivity of lanthanide metals has been interpreted in terms of a temperature-independent splitting [15]. Such a behavior was also expected for the highly localized d-like surface state found on all close-packed lanthanide metal surfaces [16], which is known to be exchange-split for ferromagnetic Gd [6] with only the majority component occupied. While this state cannot be probed by PE only, a similar behavior has been observed for the fully occupied localized surface state that can be formed by oxygen adsorption on Gd(0001) [17].

Taking into account that in a local-moment system at finite temperatures the spin of a band state is not a good quantum number, a second theoretical study by Sandratskii and Kübler [7] simulated these systems by a non-collinear helical spin structure. In this model, higher temperatures correspond to a larger angle between 4f moments of neighboring planes. The calculation predicts a magnetic splitting of delocalized states for finite angles, i.e. even though the overall magnetization of the sample is zero. Interestingly, such a helical structure can be investigated experimentally for the antiferromagnetic phases of Tb, Dy, and, in particular, Ho. In this contribution, we report on a systematic investigation of the exchange splitting in ferromagnetic and antiferromagnetic lanthanide metals as observed for the above-mentioned Δ_2 band and for the oxygen-induced surface state.

2. Experimental

An established method to prepare atomically clean lanthanide metal surfaces is growth of epitaxial films on a refractory metal surface in ultra-high vacuum (UHV) [6,9–12,14,16]. For the experiments presented here, single-crystalline films of thicknesses between 100 and 300 Å were prepared in situ on W(110) as described in Ref. [10]. The high crystalline quality of the films was established by LEED and X-ray diffraction [13]; cleanness of the films was checked by surface sensitive PE. Measurements were performed at the TGM 1,3,5, and 6 beamlines of the

Berliner Elektronenspeicherring für Synchrotronstrahlung (BESSY) using a VSW-ARIES spectrometer as well as with a SCIENTA SES200 analyzer and monochromatized light from a He light source. The total-system energy resolution for the measurements at BESSY was between 100 and 150 meV and for experiments with the SCIENTA analyzer 60 meV (FWHM); the acceptance angle was $2°$ in both setups. The existence of a bulk-like helical magnetic structure in the epitaxial Ho films was checked by resonant magnetic X-ray scattering in a separate UHV setup at the ESRF beamline ID10A (Troïka) [18,19]. For the preparation of the surface-oxide samples, oxygen was dosed at room temperature. Subsequent annealing led to well-ordered surface-oxide phases with sharp 1×1 LEED patterns [17,20].

3. Results

Low-temperature photoemission data recorded from the clean surfaces of ferromagnetic Gd, Tb, Dy, and antiferromagnetic Ho are presented in Fig. 1, showing the Δ_2 band around 2-eV binding energy and the surface state, S. In all spectra, a splitting of the Δ_2 band can be observed, systematically decreasing from Gd to Ho. Except for Tb, the spectra have been taken in normal-emission geometry at 35-eV photon energy, corresponding to transitions from Δ_2 states near the center of the Brillouin zone (Γ) [5]. For Tb, the Δ_2-band emission overlaps with the ^8S 4f final-state emission at Γ. Therefore, an angle of $8°$ off normal along $\overline{\Gamma K}$ and 40-eV photon energy was chosen in order to separate these contributions by making use of the dispersion of the band. From the analogy with Gd, no noticeable change of the splitting in this range is expected.

For a quantitative comparison, least-squares fit analyses were applied (solid lines through the data points in Fig. 1). All spectra could be described in a consistent way using the same model of two Lorentzians for the Δ_2-band components, a smooth background to account for inelastic processes and emissions from other parts of the Brillouin zone (broken lines), and additional lines for S and the Tb-4f emission. As known for Gd [5], the splitting de-
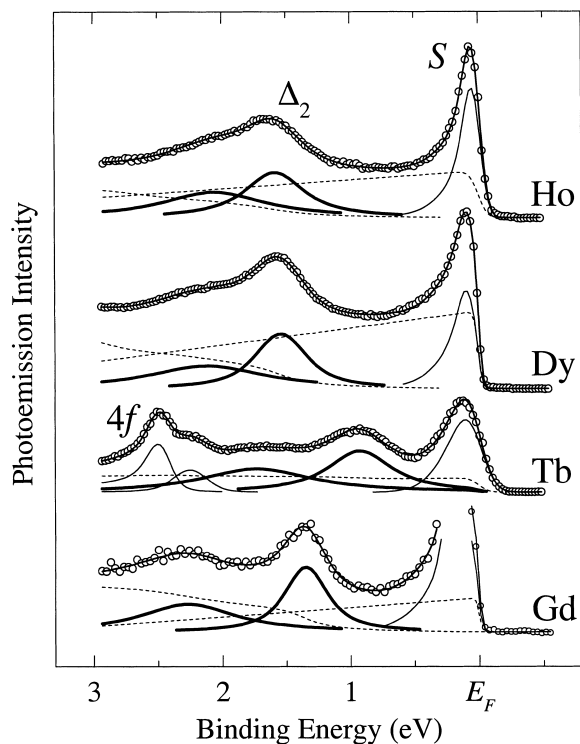
Fig. 1. Valence-band photoemission spectra of Gd (63 K), Tb (26 K), Dy (65 K), and Ho (43 K) showing the Δ_2 band, the surface state, S, and for the case of Tb the ^{8}S component of the 4f multiplet. Solid lines through the data points and subspectra represent the results of a least-squares fit analysis.
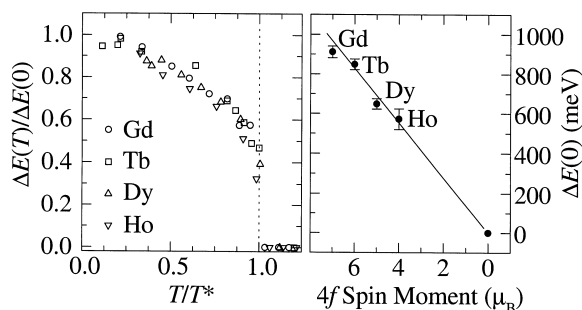

Fig. 2. Left: temperature dependences of the Δ_2 magnetic splittings for Gd, Tb, Dy, and Ho. Right: zero-Kelvin splitting plotted versus 4f spin moment.

splitting is not diminished by the non-collinear structure. Therefore, the Δ_2 splitting seems to reflect the ferromagnetic order within the (0001) planes of the lanthanide metals rather than the total magnetization.

Low-temperature PES data from the oxygen-induced surface state on Gd, Tb, Dy, Ho and, for comparison, nonmagnetic Lu are presented in Fig. 3. Except for Lu, these states show a temperature-dependent splitting, however, with a considerable temperature-independent residual contribution, persisting above T^*. For Lu, the symmetric emission shape does not change with temperature and exhibits only some slight thermal broadening. The magnetic splitting extracted from variable-temperature measurements for all metals (spectra not shown here) are presented in the left panel of Fig. 4 normalized in the same way as for the Δ_2 band in Fig. 2. The behavior is again very similar for all magnetic systems and even the ratio between the temperature-dependent and the temperature-independent residual contributions is the same in all cases. This is also illustrated in the right panel of Fig. 4, which shows the extrapolated zero-Kelvin splittings and the residual splittings in the paramagnetic phase. Both quantities scale linearly with the 4f spin moment.

4. Discussion

All observed magnetic effects occur on a temperature scale given by the respective highest ordering temperature. They are not restricted to ferromagnetic

creases in all cases upon heating (spectra not shown here), with a temperature scale determined by the respective highest ordering temperature, T^*, i.e. the Curie temperature (T_C) in case of Gd and the Néel temperature (T_N) for the other metals. In the left panel of Fig. 2, the resulting splittings $(\Delta E(T))$ are plotted as a function of the reduced temperature T/T^*, normalized to the value extrapolated to zero Kelvin. The behavior is very similar for all metals, which establishes that (i) the relevant temperature scale is given by the highest ordering temperature and that (ii) the splitting is independent of the type of magnetic order. The extrapolated zero-Kelvin splitting, $\Delta E(0)$, is displayed in the right panel of Fig. 2 as a function of 4f spin moment. The obvious linear relationship corresponds to a constant exchange integral along the lanthanide series. The value for Ho fits well into this systematics; the
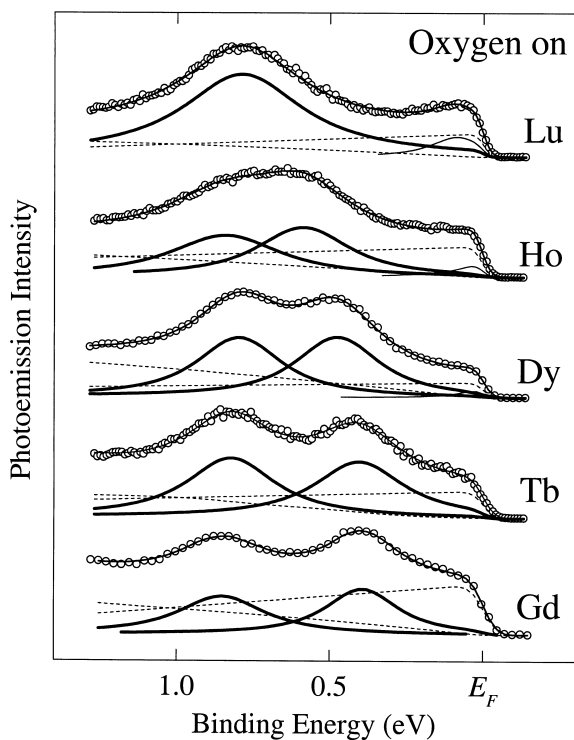
Fig. 3. Photoemission spectra of the oxygen-covered (0001) surfaces of Gd (52 K), Tb (49 K), Dy (70 K), Ho (64 K), and Lu (69 K) showing the oxygen-induced surface state. Together with the data, the results of a least-squares fit analysis are shown (solid lines through the data points and subspectra).

order, but also occur for helical antiferromagnetic Ho without noticeable reduction of the splitting. Applying the model of Ref. [8] to the Δ_2 band, such a behavior with an essentially vanishing splitting,

which, however, is not sensitive to the orientation of the neighboring planes, could be expected for a wave function, which is delocalized within the close-packed planes but localized with respect to the c-axis. Such a view is supported by the observed flat dispersion of this band along c^* [5] and by an estimate of the respective hopping parameters [18]. To some extent, it is also in agreement with the theoretical predictions in Ref. [7], even though the calculations do not explicitly take this anisotropic behavior into account.

In contrast to the Δ_2 band, the oxygen-induced surface state is also localized with respect to the in-plane directions as concluded from the flat dispersion parallel to the surface. The resulting strong influence of the exchange with the local 4f moment leads to a temperature-independent residual contribution to the splitting. The oxygen-induced surface state is therefore well suited for PE studies of localized valence states, and especially spin-resolved measurements would allow an experimental test of the theoretically predicted depolarization [8].

The linear dependence of all magnetic splittings on the 4f spin moment reflects an essentially constant effective exchange coupling along the series of the lanthanide metals, not affected by the lanthanide contraction. Such a linear scaling has been assumed before [21,22], but is shown here explicitly by experiment. The smaller splittings observed for the oxygen-induced states are the result of a weaker coupling in the oxide layer, probably due to strong mixing with O2p states [17].

5. Conclusions

For clean and oxygen-covered Gd, Tb, Dy, and Ho metal films, temperature-dependent magnetic splittings in the band structure were observed as theoretically predicted for delocalized and localized band states, respectively. They occur in ferromagnetic and helical antiferromagnetic phases with no noticeable anomaly in the latter case within the limits of experimental accuracy. All the observed splittings scale linearly with the 4f-spin moment, indicating a constant exchange coupling along the series of the heavy lanthanide metals.
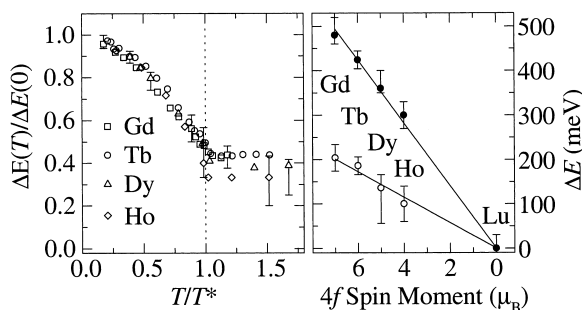

Fig. 4. Left: temperature dependences of the magnetic splittings of the oxygen-induced surface state for Gd, Tb, Dy, and Ho. Right: zero-Kelvin splitting (solid circles) and residual splitting above T^* (open circles) plotted versus 4f spin moment.

Acknowledgements

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