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Temperature dependence of x-ray absorption spectra in the ferromagnetic Heusler alloys Mn_2VAl and Co_2FeAl

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We investigate the temperature dependence of the spin-resolved unoccupied density of states (DOS) in ferromagnetic Co_2FeAl and ferrimagnetic Mn_2VAl epitaxial films on MgO(100) using x-ray magnetic circular dichroism. We observe an unexpected strong temperature dependence of the DOS beyond the change expected from the Fermi distribution function. An increase in spectral weight is observed for majority states below the Fermi energy in the case of Mn_2VAl and for minority states above the Fermi energy in the case of Co_2FeAl. Reduced atomic order near the interface suppresses the unexpected temperature dependence of the DOS for Mn_2VAl.

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Half-metallic ferromagnetism (HMF) plays a major role for spintronics[1] providing a special state of itinerant magnetism. While one spin channel is semiconducting with an energy band gap at the Fermi level the opposite spin channel provides metallic conductivity. Heusler alloys X_2YZ (transition metal X, Y, and main group element Z) have attracted much attention in the field of spintronics since *ab initio* theory based on a single-electron model predicted HMF for many of these compounds.[2–4] Spintronic devices consisting of tunneling magnetoresistors (TMR) that exploit the high spin polarization of HMF Heusler alloys have been fabricated[5–10] and confirm the concept of HMF at low temperatures. However, the strong temperature dependence of the effective spin polarization is not fully understood, yet.

The temperature dependence of the electronic states in half-metals might contribute to the strong decrease in the spin polarization and its experimental investigation is highly interesting. In a single-particle model changes in the electronic structure are only expected in the vicinity of the Fermi level due to thermally excited occupation of previously unoccupied states which is in agreement with a recent high-energy photo emission study for a half-metallic Co_2MnSi/MgO interface.[11] Instead, a model beyond the single-particle picture of the local density approximation considering finite-temperature many-body effects[12,13] results in a considerable temperature dependence of the density of states (DOS).

In this paper we report on a spectroscopic measurement of the temperature dependence of the spin-resolved DOS in two different kinds of Heusler alloys, which were predicted to be half-metallic comprising band gaps in the majority spin band (Mn_2VAl) (Ref. 14) and in the minority spin band (Co_2FeAl),[15] respectively. Spin-resolved photo emission,[16,17] appearance potential spectroscopy,[18] and scanning tunneling spectroscopy can directly probe the spin polarization at a half-metal surface. However, these methods have no access to the crucial buried interfaces in spintronic devices. We apply x-ray absorption spectroscopy (XAS) providing an element-specific information of near-surface and bulk properties.[19,20]

The ferrimagnetic Heusler alloy Mn_2VAl has been investigated previously revealing atomic moments of 1.5 μ_B for Mn and −0.9 μ_B for V.[21] X-ray magnetic circular dichroism (XMCD) (Ref. 22) confirmed the existence of a ferrimagnetic state in epitaxial Mn_2VAl films comprising atomic moments that agree with the *ab initio* calculations.[14] The Curie temperature of Mn_2VAl is far above room temperature at about 760 K.[21] The half-metallicity of Mn_2VAl is predicted to be rather robust against a small degree of local disorder. An intermixing between V and Al atoms still results in an almost half-metallic behavior.[14] Recently, an advanced local density approximation plus dynamical mean field (LDA +DMFT) approach[12] predicted the occurrence of additional states in the majority band gap caused by many-body effects. However, at present there is no experimental proof that Mn_2VAl is a HMF.

Co_2FeAl is ferromagnetic with a large magnetization of 5 μ_B/f.u. and a high Curie temperature in accordance with the predicted half-metallic properties.[15] However, reports on HMF properties of Co_2FeAl are controversial.[23] Using LDA calculations Ref. 24 reported a spin polarization of just 30% in both disordered B2 and ordered $L2_1$ structures. Using a similar calculation scheme Ref. 2 reported a reduction of magnetization from an integer value (in Bohr magnetons per formula unit), indicating that Co_2FeAl is not a half-metal. Magnetic tunneling junctions with Co_2FeAl electrodes showed large TMR values of up to 330% at room temperature (700% at 10 K), indicating a high effective spin polarization.[25] However, as was shown in Ref. 26 the high effective spin polarization can be explained by coherent tunneling and therefore cannot be regarded as a proof for half-metallicity. Reference 27 reported a TMR ratio of approximately 50% at room temperature in an optimized $Co_2FeAl/Al\text{-}O/CoFe$ magnetic tunneling junction. A more recent experimental study[28] resulted in spin polarization values of 55%. These findings are consistent with theoretical predictions of the absence of HMF for Co_2FeAl.

We deposited epitaxial Al/60 nm $Mn_2VAl/MgO(100)$ (Ref. 29) by rf sputtering and post-deposition annealing at 550 °C and Al/MgO/30 nm $Co_2FeAl/MgO(100)$ (Ref. 25) films using dc sputtering and post-deposition annealing at 480 °C, respectively. The characterization of the films involved x-ray diffraction, reflection of high-energy electrons, scanning tunneling microscopy, and vibrating sample magnetometry. Mn_2VAl films predominantly revealed a $L2_1$ and Co_2FeAl films a B2 atomic order. XAS/XMCD measurements were carried out at the German light source BESSY II (beam line UE56/1-SGM). Results were derived from total electron yield (TEY) data (300 K) providing an information depth of 2.5 nm and from the transmitted (TM) x-ray intensity with infinite information depth. A magnetic field of 1.22 T was applied normal to the film surface and parallel to the incident photon beam.[30] The energy resolution of the x-ray monochromator was set to 0.1 eV (0.25 eV in the case of Mn).

The XAS/XMCD measurements for Mn_2VAl films at the V and Mn $L_{2,3}$-absorption edges show features similar to those observed in other metallic compounds.[31] Surface and bulk sensitive measurements are similar to each other and no traces of oxide-induced multiplet features are visible. We take this as evidence that the Mn-derived states have an itinerant character for Heusler compounds with Mn at the X position.

The partial DOS (PDOS) can be directly calculated from the XAS/XMCD spectra following the procedure described in Refs. 19 and 20. The absorption of circularly polarized x-rays is explained by a two-step process considering the dipole matrix elements. In the first step, the electron is excited from a spin-orbit split $2p_{3/2}$ or $2p_{1/2}$ level ($L_{3,2}$ edge) and absorbs the angular momentum of the photon in part to its spin due to spin-orbit coupling. In a second step the unoccupied valence band states serve as a detector for the spin and orbital polarization. As the orbital magnetic moment is negligible at least in the case of the transmission signal the orbital polarization of the photo electrons is neglected in the following. Effects from anisotropic charge and spin densities may be neglected due to the high crystal symmetry. Since the radial matrix elements show in general only a small energy dependence, the absorption spectra can be interpreted as a direct image of the spin-resolved PDOS above the Fermi energy.

The spin-resolved unoccupied PDOS $D^{\uparrow(\downarrow)}(1-f_F)$ (Fermi function f_F) results from the L_3 edge XAS spectra μ^+ and μ^- according to Ref. 19

$$D^{\uparrow(\downarrow)}(1-f_F) \propto \mu_{iso} - s + (-)\frac{1}{P_{L3}}\frac{\mu^+ - \mu^-}{2}, \qquad (1)$$

where μ_{iso} denotes the isotropic absorption coefficient $(\mu^+ + \mu^-)/2$, s the step function and $P_{L3}=-0.25$ the spin polarization of the excited photo electrons.

The result for the Mn related PDOS in Mn_2VAl is shown in Figs. 1(a) and 1(b) and compared to theoretical data. The minority states clearly reproduce the *ab initio* calculation considering the lifetime broadening of the spectral features. The large maximum at $E-E_F=1.9$ eV corresponds to the peak in the theoretical data at 1.7 eV. The pronounced shoulder closer to E_F is the remainder of the maximum in the theoretical data just below E_F. The pronounced maximum of the majority states observed at 2.2 eV above E_F [see Figs. 1(a) and 1(b)] is at variance with theory. This might be explained by static electron correlation effects that are not properly considered by the evaluation procedure or by the theoretical calculation.

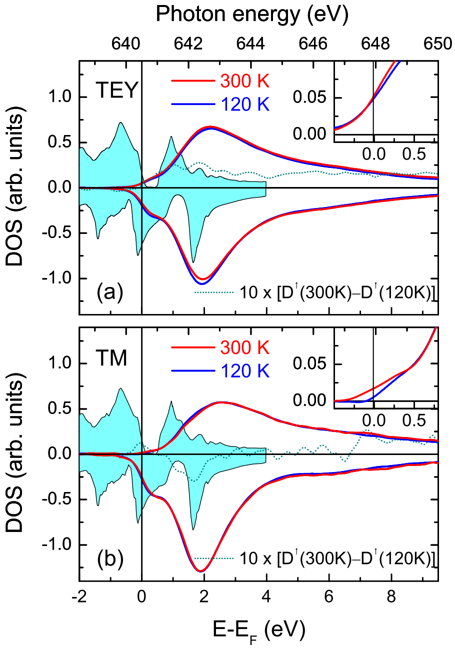

FIG. 1. (Color online) (a) Spin-resolved unoccupied PDOS calculated from the XAS/XMCD TEY data measured at the Mn L_3 edge of the Mn_2VAl film. The majority and minority PDOS is shown on a positive and a negative scale, respectively. The difference of the low- and high-temperature majority PDOSs is tenfold enhanced, emphasizing the temperature induced spectral changes near the majority band gap. The theoretical data from Ref. 12 are shown as shaded areas. The inset shows the magnified energy region around E_F for majority states. (b) Spin-resolved PDOS derived from the transmission data.

The majority state data are different for surface and bulk sensitive measurements. While the TEY data [Fig. 1(a)] reveals a pronounced nonzero PDOS at E_F, the transmission data [Fig. 1(b)] is almost zero at E_F for low temperatures and shows a small positive value at room temperature. The difference $D^{\uparrow}(300\ \text{K})-D^{\uparrow}(120\ \text{K})$ has clearly a maximum below E_F. This temperature dependence cannot be explained by a thermal broadening of the Fermi function, which is much smaller at 300 K and would show up antisymmetrically below and above E_F. Similar temperature dependence is clearly absent for the case of the TEY related data. This fact coincides with the strong decrease in the Mn and V interface magnetic moments by a factor of 2. The origin of the reduced interface magnetization is unclear. One might speculate that it is caused by a reduced atomic order at the interface since films deposited at room temperature without annealing do show a strongly reduced atomic and magnetic order in the bulk of the films. Therefore we suggest that the interface region is metallic instead of half-metallic.

Figure 2(a) shows results similarly evaluated from the TEY data of Co_2FeAl films. The small x-ray intensity caused by the higher energy resolution chosen for this experiment produces increased noise of the transmission data Fig. 2(b). From measurements with lower resolution we infer that in contrast to the Mn_2VAl films the difference between surface

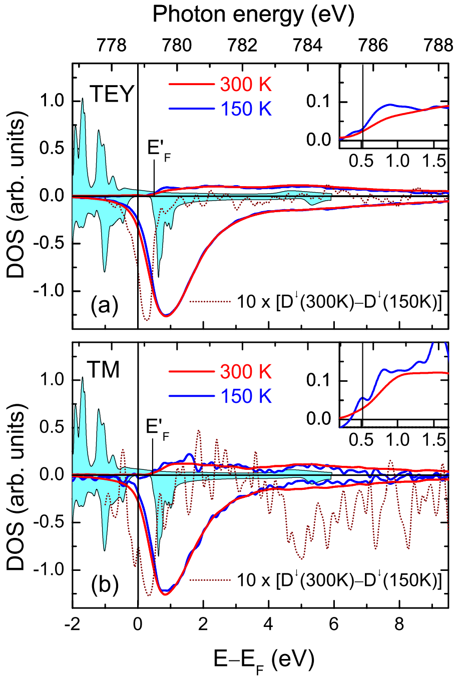

FIG. 2. (Color online) (a) Spin-resolved unoccupied PDOS calculated from the XAS/XMCD TEY data measured at the Co L_3 edge of the Co_2FeAl film. The majority and minority PDOS is shown on a positive and negative scale, respectively. The difference of the low- and high-temperature minority PDOSs is tenfold enhanced, emphasizing the temperature induced spectral changes near the minority band gap. The theoretical data from Ref. 15 are shown as shaded areas. The inset shows the magnified energy region around E_F for the majority states indicating further spectral changes for the conducting spin channel. (b) Spin-resolved PDOS derived from the transmission data.

and bulk sensitive information is almost negligible for this sample and the electronic properties of the bulk and interface are identical. We observe a large maximum of the minority PDOS at $E-E_F=0.9$ eV followed by a weak shoulder at $E-E_F=1.2$ eV and a second peak at $E-E_F=5$ eV. The position of the minority maximum is in good agreement with theory for this sample in contrast to the Co_2FeAl films investigated earlier[19] where the maximum was shifted closer to E_F probably due to reduced local atomic order. The onset of the majority states appears 0.5 eV above E_F (E_F'), which is a consequence of the fact that an itinerant $3d$-t_2 band dominates the unoccupied majority states near E_F in Co_2FeAl.[4] The core hole in the final state attracts the electron in the localized $3d$ states thus lowering the photon energy needed for the transition. For an itinerant state the energy decrease is smaller since the electrons from neighboring atoms screen the core hole to some extent.[32] The different core hole screening thus produces an energy shift $\Delta E_c=0.5$ eV between itinerant and localized states.[19] This is in contrast to the case of Mn_2VAl where the corresponding itinerant state is shifted far above E_F.

In contrast to Mn_2VAl we do not observe a significant difference for surface (TEY) and bulk (TM) related spectra for Co_2FeAl. We attribute this fact to the optimized preparation procedure for Co_2FeAl films resulting in similar electronic properties of interface and bulk of these films. A comparison of the low-temperature and room-temperature measurements [Fig. 2(a)] reveals a difference of the minority PDOS with an increased value near E_F at room temperature. A similar behavior might be inferred in the case of transmission data as well [Fig. 2(b)], despite the reduced signal to noise ratio. In both cases the maximum of the difference occurs slightly above E_F. As XAS fully measures the unoccupied part of the PDOS, the observed small effect which is enhanced by the core hole correlation[33] corresponds to a tiny change in the spin polarization at E_F in good agreement with the recently observed small temperature dependence of the TMR effect for Co_2FeAl based tunnel junctions.[25] In addition, the majority states reveal a redistribution of the state occupancy at 0.3 eV above E_F, which is emphasized by the insets in Fig. 2 (0.3 eV above E_F').

Our experimental observation of the increase in spectral weight just below the Fermi level for the case of the TM data of Mn_2VAl with a majority band gap is in qualitative agreement with the model of nonquasiparticle (NQP) states, calculated for real systems using (LDA+DMFT).[12] The observed decrease in the majority PDOS above E_F agrees with the predicted half-metallic properties of Mn_2VAl. As up to now no experimental results of nonhalf-metallic properties of Mn_2VAl were reported, we may assume that our Mn_2VAl films are half-metallic in the bulk, thus fulfilling the precondition for the existence of nonquasiparticle states. At the interface, the magnetization is clearly decreased and moreover shows a stronger temperature dependence. We attribute this difference to a less ordered structure at the interface probably due to a nonstoichiometric composition in this region originating from the large diffusion coefficient of Mn. Please note, that the films were optimized for large magnetization and structural properties measured by bulk-sensitive methods. The interface disorder obviously leads to an increased majority PDOS at E_F combined with a strong reduction in spin polarization. In this case no extra peak appears at E_F with increasing temperature. The remaining temperature dependence of the spectra can be explained by a temperature induced spin mixing (spin fluctuations) according to a Heisenberg model. The absence of temperature-induced DOS changes beyond the single particle model is expected for a normal metal. Thus, for the case of Mn_2VAl all available experimental observations are consistent with the model of nonquasiparticle states.

For the case of Co_2FeAl we observe an unexpected change of the PDOS with increasing temperature both for interface and bulk related data. For the minority states an extra peak appears slightly above E_F as predicted by the nonquasiparticle model.[13] However, for Co_2FeAl conflicting results in respect to HMF properties have been reported. This is a crucial point since half-metallicity is supposed to be a necessary condition for the emergence of NQP states. The PDOS observed in this work is not conclusive for a determination of half-metallicity in this sample due to the lifetime broadening of XAS (Ref. 19) in combination with the close vicinity of E_F and the upper band edge. Although Ref. 15 predicted Co_2FeAl to be half-metallic, other *ab initio* calculations resulted in a normal metal behavior with E_F crossing the conduction band edge.[24] Reference 24 reported a spin polarization of just 30% which was confirmed by a measurement of the spin polarization using magnetic tunneling junctions with an Al-O barrier (Refs. 27 and 28) considered as a good approximation of Julliere's model. The high TMR ratio reported in Ref. 25 can be explained by coherent tunneling[26] and is therefore consistent with a normal metal behavior of Co_2FeAl. The reported spin-polarization measurements have determined spin polarization at the topmost Heusler layer near an interface. It is known that the surface spin polarization strongly depends on the atomic order and interface termination. Our larger information depth of 2.5 nm (TEY) could lead to different results. This argument still opens the possibility that our films might be half-metallic and that NQP states is a valid model to explain our results even though it is not compulsory.

Further experimental observations in conflict with the NQP model exist. The temperature-induced spectroscopic changes observed for majority states of Mn_2VAl at 1.5 eV above E_F and Co_2FeAl at 0.3 eV above E_F are not reproduced by the nonquasiparticle model.[13] Moreover, the predicted temperature dependence of the DOS originating from NQP states is very small compared to our observation.[13] A recent high energy photo emission study of the HMF Heusler alloy Co_2MnSi (Ref. 11) found no temperature dependence of the valence band spectrum, which is inconsistent with the NQP model. On the other hand, the photoemission study might not be in full conflict with our results as the interaction with the core hole during the x-ray absorption process essentially enhances the spectral weight of the NQP-induced changes in the DOS according to LDA+DMFT calculations,[33] making XAS more sensitive to NQP states compared to photoemission.

Thus, at present the model of NQP states explains our experimental observations for Mn_2VAl while in the case of Co_2FeAl the precondition of HMF is probably not fulfilled and spectral features in addition to the NQP model occur. Therefore, our experimental work cannot justify the NQP model as the exclusive explanation for the observed temperature-induced spectral changes. More detailed LDA+DMFT calculations for Co_2FeAl including B2 order instead of $L2_1$ order would clarify if compounds close to HMF showed temperature dependencies beyond the single particle model. Further experimental work providing a better empirical basis for the occurrence of an unexpected temperature-dependent DOS is needed. As an explanation one might alternatively consider phonon-induced interactions[34] or a temperature-dependent exchange splitting.[35] Also structural changes below the detection limit of x-ray diffraction should be taken into account.

In summary, we report temperature induced changes in the spin-resolved DOS of two Heusler compounds that cannot be explained by thermal excitations within a single-particle model. We observed a temperature-induced spectral change in the PDOS for Mn_2VAl for which theory predicted a majority band gap at E_F. A spectral change was also observed for Co_2FeAl for which theory predicted a minority band gap or at least a pronounced minimum of the minority DOS at E_F. The spectral changes include a peaklike increase in the partial DOS just below E_F in the case of Mn_2VAl and just above E_F for Co_2FeAl. These features are consistent with the main prediction of the NQP model. Admittedly, for the case of Co_2FeAl more details of our experimental results are partly inconsistent with the predictions of the NQP model and, moreover, previously reported results suggest that Co_2FeAl is not a true half-metal which is considered as a necessary condition for the NQP model. Thus, our experimental results do not unambiguously justify the observation of NQP states. Further progress in the understanding of many-body effects is needed to explain the reported results.

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