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Rapid Note

Observation of magnetic splitting in XPS MnL-spectra of Co_2MnSn and Pd_2MnSn Heusler alloys

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Abstract. The XPS MnL-spectra of Co_2MnSn, a nearly half-metallic ferromagnet (HMF) and Pd_2MnSn were investigated. The most drastical feature of the spectra observed is the well-defined magnetic splitting of the Mn $2p_{3/2}$, $2p_{1/2}$ lines. This gives direct evidence of the existence of well-defined local magnetic moments in Heusler alloys in comparison with other itinerant-electron ferromagnets. The calculations of Mn2p XPS spectra of these materials were carried out using a fully relativistic generalization of the one-step model of photoemission and show excellent agreement with experiment.

PACS. 71.20.Be Transition metals and alloys – 75.20.Hr Local moments in compounds and alloys; Kondo effect, valence fluctuations, heavy fermions

1 Introduction

The problem of local magnetic moments, that is, localized behaviour in some aspects of itinerant d-electrons, is one of the most important in the physics of magnetic metals (the contemporary state of the problem is described, *e.g.*, in Refs. [1–3]). The spectroscopy of electron core levels gives us important informations of this behaviour which is complementary to that from static magnetic measurements. The reason is that in spectroscopic experiments we deal with rather short-lived states and, hence, investigate the behaviour of the magnetic subsystem on a short time scale. Here we report about first direct observations of magnetic exchange splitting of the L-spectra of itinerant-electron magnets using non-polarized radiation.

2 Experimental details

The specimens were prepared from a melt in an atmosphere of purified argon, annealed at 720 °C 24 h in argon and quenched in water. According to X-ray diffraction patterns (Cu K_α) samples were single $L2_1$ phase. The X-ray photoelectron spectra (XPS) were measured using a PHI 5600ci spectrometer with an energy resolution of 0.35 eV with monocromatized Al K_α radiation. The Heusler alloys were crushed *in situ*. The surface cleanlyness was tested by monitoring the C1s and O1s peaks. During the measurements no significant increase in the contamination was observed.

3 Results and discussions

The splitting of core states is the consequence of the existence of spin polarization in 3d states. But, because of the finite natural linewidth Γ, we probe the magnetic moment on a characteristic time scale of order of $\tau = \Gamma^{-1}$. Local spin S_0 on site 0 may be expanded in the Fourier integral over the Brillouin zone: $S_0(t) = qS(q,t)$. But it is well-known (see *e.g.* [3] and references therein) that for most of the itinerant-electron magnets Stoner damping is dominated in most parts of the Brillouin zone which leads to the decay of $S(q,t)$. That is why it is so difficult to observe spin splitting for L-spectra where characteristic values of order of 1-2 eV are typical. Note that the splitting in M-spectra with essentially larger values of typical Γ is well observable [4]. Among all of the itinerant-electron ferromagnets, so-called half-metallic ferromagnets (HMF) may

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be separated, with zero or almost zero density of states at the Fermi level for one spin direction [3]. For these magnets Stoner damping is absent in the whole Brillouin zone and the magnetic moments are as well defined as for magnetic dielectrics [3]. Note that Mn is a special element in the phenomenon of HMF since almost all known HMF (except CrO_2) are Mn-based compounds. In particular, according to band-structure calculations [5,6] the intermetallics Co_2MnX (X = Si,Ge) are HMF, while Co_2MnX (X = Sn, Al) are nearly half-metallic. The theoretical approach to the problem of core-level photoemission is done within a fully relativistic generalization of the one-step model of photoemission [7,8]. The photoemission intensity can be expressed by

$$I(\varepsilon_f, \mathbf{k}_\parallel) = -\frac{1}{\pi}\Im \int \mathrm{d}r \int \mathrm{d}r' \Psi_f^{*t}(r)\Delta(r) \times G_1(E, r, r')\ \Delta^\dagger(r')\Psi_f^t(r'). \quad (1)$$

The final state in this expression appears as a so-called time reversed SPLEED-state and is given by

$$\Psi_f^t(\mathbf{r}) = \langle \mathbf{r}|G_2^+|\varepsilon_f, \mathbf{k}_\parallel\rangle \quad (2)$$

with G_2^+ being the Green function for the final state. The initial state is described by the core-level Green matrix

$$G_1(E, \mathbf{r}, \mathbf{r}') = \sum_{\mathcal{K}_m} |\mathcal{K}_m\rangle\ \frac{\Sigma_c}{(E - \varepsilon_{\mathcal{K}_m})^2 + \Sigma_c^2}\ \langle \mathcal{K}_m| \quad (3)$$

where $|\mathcal{K}_m\rangle$ is the relativistic core-level wavefunction for $\mathcal{K}_m = (\kappa_m, \mu_m)$ with the energy eigenvalue $\varepsilon_{\mathcal{K}_m}$. Possible damping processes in the initial state are considered *via* the imaginary part Σ_c of the complex self-energy Σ. The perturbation operator Δ mediates the coupling to the electromagnetic field. In its fully relativistic ferromagnetic implementation it has the form

$$\Delta \sim \mathbf{A}_0 \bigtriangledown V(r) + \frac{i\omega}{c}\hat{\alpha}\mathbf{A}_0 V(r) + \mathbf{A}_0 \beta \bigtriangledown \hat{\sigma}\mathbf{B}(r) - \frac{\omega}{c}\beta\hat{\sigma}(\mathbf{A}_0 \times \mathbf{B}(r)). \quad (4)$$

$\mathbf{A}_0$ is the spatially constant amplitude of the electromagnetic vector potential, $V(r)$ and $\mathbf{B}(r)$ are the crystal potential and the magnetic field respectively.

A comparison of experimental and theoretical MnL-spectra for Co_2MnSn and Pd_2MnSn is shown in Figure 1. For both materials an excellent overall agreement has been found for the line widths as well as for the relative intensities. A detailed analysis of the calculated spectra reveals that the line widths are closely connected with the amount of exchange splitting in the Mn $2p_{3/2}$ sublevels. As it can be seen from Figure 1 the exchange splitting in the Mn $2p_{3/2}$ peaks of Pd_2MnSn is larger than the Mn $2p_{3/2}$ splitting in Co_2MnSn. This can be simply explained by the difference in the local magnetic moments for Mn in Co_2MnSn and for Mn in Pd_2MnSn. In detail the local magnetic moment $\mu_{\mathrm{Mn}}^{\mathrm{Pd_2MnSn}} = 4.3\mu_\mathrm{B}$ is significantly larger than the local moment $\mu_{\mathrm{Mn}}^{\mathrm{Co_2MnSn}} = 3.6\mu_\mathrm{B}$ and is directly correlated with the ratio of the line widths of the Mn $2p_{3/2}$ peaks in both materials. The different line shapes of the Mn $2p_{3/2}$ peaks in Co_2MnSn and Pd_2MnSn can be understood by analysing the relative intensities in the $2p_{3/2}$ sublevels. The calculation results in a completely different distribution of spectral weights in the four Mn $2p_{3/2}$ sublevels of both crystals. The energetic positions and relative intensities are marked by solid lines in Figure 1. The intensity distributions in both Mn $2p_{3/2}$ peaks can be explained mainly due to direct transitions from the Mn $2p_{3/2}$ states into final d-bands and therefore can be classified as matrix element effects induced by the dipole selection rules. The broader structure in the Pd_2MnSn spectrum has two reasons. At first Pd_2MnSn is not a HMF and, what is more important, its Curie temperature is below room temperature where the measurements were carried out. So the observation of exchange splitting in Pd_2MnSn shows that the local magnetic moments are well defined in Heusler alloys at a time scale of order of Γ^{-1} even in the paramagnetic phase. A detailed discussion and results for other Heusler alloys will be presented in a forthcoming publication.

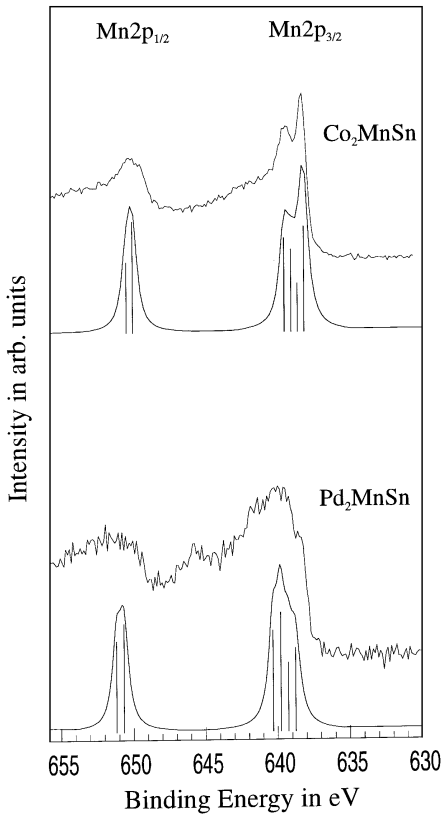

Fig. 1. Measured and calculated Mn 2p spectra of Co_2MnSn and Pd_2MnSn.

4 Conclusion

To summarize, we observed the exchange splitting in Heusler alloys Mn $2p_{3/2}$-spectra and were able to describe it by theory. The splitting is directly correlated to the local magnetic moment at the Mn sites. The changes in the relative intensities are dominated by the dipole transition matrix elements. The agreement between theory and experiment is rather good which demonstrates the existence of well defined local magnetic moments in Heusler alloys under consideration.

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