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Spin-Polarized Auger Spectroscopy from Magnetically Ordered Solids

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> Auger electrons from magnetically ordered solids are spin polarized. As an example ferromagnetic $Fe_{83}B_{17}$ glass was studied. The polarization of the Fe Auger electrons exhibits structures on the order of 20% , both parallel and opposite to the magnetization of the sample depending on the particular transition. Auger electrons from B are unpolarized. The technique opens new perspectives in the study of magnetic solids, particularly alloys and compounds.

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High-resolution Auger spectroscopy from solids yields rich information on electron and hole interactions.¹ LMM spectra from $3d$ metals contain the extent of delocalization of 3d holes in metals with unfilled d bands² and reveal the effective Coulomb interaction between two 3d holes at the same site.³ Ferromagnetism, on the other hand, is probably the most important quality of the $3d$ transition metals and their compounds. In the present work we hope to open new perspectives in both the Auger spectroscopy on 3d transition metals as well as in the ferromagnetism of pure metals, compounds, and alloys, by studying the spin polarization of Auger electrons from a ferromagnetic solid. The information gained is manifold depending on the particular Auger transition. LVV (e.g., $L_{3}M_{45}M_{45}$) transitions provid a local probe of the spin-dependent density of valence states where special relevance has to be given to electron-electron correlation effects depending on the strength of the effective Coulomb interaction relative to the bandwidth. IMM transitions with no valence electrons involved, on the other hand, are spin polarized through the coupling of partly filled inner shells with the net spin of the magnetic $3d$ electrons,⁴ and thus provide information on the local magnetization (i.e., average magnetic moment).

We wish to emphasize the element specificity of the Auger transition as well as its $local$ character. We point out that spin-polarized Auger spectroscopy from solids therefore will be a powerful technique to study not only pure ferromagnetic metals but also magnetic alloys and compounds. Also ferrimagnetic systems will be of interest where sublattice specific information can be gained.

As a first example we present in this Letter spin-polarized Auger spectra from a ferromagnetic glass of composition $Fe_{83}B_{17}$. The sample was kindly provided to us by R. C. O'Handley at the Massachusetts Institute of Technology. In Figs. 1-3 we present intensity $I(E_s)$ and spin polarization $P(E_s)$ versus secondary-electron kinetic energy E_s in the vicinity of all the prominent Auger lines of iron and boron.

The spin polarization is defined as $P(E_s)$ $=[j*(E_s) - j*(E_s)]/j*(E_s) + j*(E_s)],$ with $j*(E_s)$ being the intensity of majority-spin electrons, i.e., electrons with magnetic moment parallel to the magnetization of the ferromagnetic sample. Polarization due to spin-orbit coupling in the

scattering process⁵ was averaged out by reversing the magnetization. $P(E_s)$ was measured using a high-efficiency Mott detector⁶ combined with an energy analyzer run at constant energy resolution of 1.² eV and constant angular resolution of $\pm 2.5^{\circ}$ cone angle. The intensity $I(E_s)$ $=j \uparrow (E_s) + j \uparrow (E_s)$ is the total backscattered intensity in the Mott detector. The sample was magnetically saturated parallel to the emitting surface by means of a horseshoe magnet with closed magnetic circuit. The Auger transitions were excited with electrons of various kinetic energies E_p up to 2900 eV at grazing incidence (20° off surface) and the secondary electrons were collected at normal emission. The sample was cleaned by 6-keV Ne^+ sputtering at grazing incidence and mild annealing (up to 60° C). Carbon contamination was found to be below 2 at.%, and the oxygen impurity level was below the Auger detection limit.

In order to obtain the net spin polarization of Auger electrons one has first to consider both the intensity and spin polarization of the background secondary electrons. Generally, in absence of Auger electrons, the secondary electrons from a ferromagnet are spin polarized. This has recently been reported for very-low-energy secondaris been reported for very-low-energy secondar-
ies,⁷ namely for E_s < 25 eV or E_s/E_p < 0.05. With increasing E_s beyond the steep drop at low energies reported in Ref. 7 the polarization goes to a plateau which presumably reflects the band polarization and then monotonically decreases to zero for E_s/E_p+1 . The spectra do depend on the kinetic energy of the primary electrons E_a and presumably on the scattering angle. A detailed discussion of these rather interesting effects is out of place here and will be the subject of a further communication. At present we used a somewhat arbitrary background polarization, as indicated with dashed lines in the figures, to find a coarse estimate of the net spin polarization of the leading Auger peaks. The Auger polarization is $P_{\text{Auger}} = P_0 + \Delta P I_{\text{tot}} / I_{\text{Auger}}$, with P_0 = background polarization, and $\Delta P = \overline{P}_{observed} - P_{0}$. We note that this estimate depends more critically on the estimated relative intensity of the Auger emission than on the polarization of the background. The spin polarizations of the prominent peaks are compiled in Table I.

The various Auger transitions all have different final-state hole configurations and thus carry a variety of information. In the following we will briefly discuss each line group separately starting with the Fe lines in the order of decreasing

TABLE I. Estimated net spin polarization of the Auger electrons in the prominent peaks (vertical lines in Figs. 1-3).

 $\sqrt[3]{\text{Error } \Delta P \leq \pm 3\}}$.

kinetic energy.

(1) Fe $L_3M_{45}M_{45}$, shown in Fig. 1 at 700 eV: Here two additional holes in the 3d valence band are left behind. For $3d$ transition metals the magnitude of the effective Coulomb interaction U_{eff} as compared to the one-electron bandwidth Γ , together with the number of unoccupied $3d$ states,⁸ is very important. Band effects in the Auger spectra will appear³ for $U_{\text{eff}} \leq 2\Gamma_{45}$. This is the case in the ferromagnetic transition metals Fe, spectra will appear³ for $U_{\text{eff}} \leq 2\Gamma_{45}$. This is the case in the ferromagnetic transition metals Fe, Co, and Ni; particularly in Fe, $U_{\text{eff}}/2\Gamma_{45} \leq 0.2^{3.8}$ The two holes generated in this Auger process thus can be regarded as nearly independent, and the Auger spectrum is then given by a self-convolution of the one-particle density of states. A self- convolution of spin- split rectangular densities of states indeed yields a polarization which is in qualitative agreement with the observation: a positive peak of polarization $(+ 100\% \text{ at } T = 0)$ lying a few electronvolts below the intensity maximum and zero spin polarization on the high kinetic energy side of the line. For a more realistic interpretation the local density of states for each symmetry, $s, p, \text{ or } d$, has to be taken into account. Furthermore, the correlation of the generated holes with the ones already present in the ground state has to be considered as was recently done by Tréglia $et~al.^9$ We note that in the case of metallic Fe the modification of the two-particle spectrum is not severe.⁹ We wish to emphasize that the $L_3M_{45}M_{45}$ spectrum reflects modified spin-polarized local band features of the ferromagnet.

(2) Fe $L_3M_{23}M_{45}$, shown in Fig. 1 at 645 eV: In this Auger transition a $3p$ hole and an additional $3d$ hole are generated. The spectra basically reflect the valence-band density of states in the presence of a $3p$ hole. The double structure resolved in the intensity spectrum is ascribed to various $3p^{5}3d^{n}$ multiplet lines. The detailed assignment, however, of the two prominent peaks

FIG. 1. Spin polarization P and intensity I vs kinetic energy of secondary electrons from $Fe_{83}B_{17}$, excited with electrons of $E_p = 2900 \text{ eV}$. Each datum point represents the average of three consecutive runs. The statistical error is $\Delta P = \pm 0.2\%$.

is uncertain in this case with an unfilled d band. The spin polarization spectrum is expected to contribute valuable information for a detailed treatment of these Auger transitions.

(3) Fe $L_3M_{23}M_{23}$, shown in Fig. 1 at 593 eV: The final state of this transition consists of two holes in the $3*b*$ shell. The $3*d*$ valence band is not directly involved in this Auger transition. Therefore the observed spin polarization reflects the coupling of holes in a core subshell to an unfilled valence-electron shell. Such coupling has been observed with x-ray photoemission from various solids containing $3d$ -series atoms. In the simplest case of 3s emission, e.g., from MnO, two lines are identified which are attributed to the two final-state multiplets 'S and 'S originating from a $3d^5$ ⁽⁶S) $3s^1$ configuration.⁴ Also $L_3M_{23}M_{45}$ Auger transitions, e.g., from Cu exhibit $3 \bar{\rho^5} 3 d^3$ multiplets.¹⁻³ However, the characteristic structure of the $L_3M_{23}M_{45}$ transition remains essentially unchanged throughout the transition-metal series. The same holds for the $L_xM_{23}M_{23}$ group. The two-peak structure in the Auger intensity from, e.g., Cu is given by the $3p⁴$ multiplet, the two major components being the singlet ^{1}D and triplet ${}^{3}P$ lines. Only minor changes are encountered when going from Cu to Ni, Co, Fe, etc. This indicates that the number of $3d$ holes is not very important and that the coupling of the itinerant $3d$ electrons to the $3p⁴$ shell is not clearly revealed in an ordinary high-resolution Auger spectrum. The spin polarization reported in this Let-

FIG. 2. Spin polarization P and intensity I vs kinetic energy of secondary electrons from $Fe_{83}B_{17}$, excited with electrons of $E_b = 2000 \text{ eV}$. Each datum point represents the average of two consecutive runs. The statistical error is $\Delta P = \pm 0.2\%$.

ter now demonstrates that such a coupling exists and can be readily observed: *negative* spin polarization for emission in ${}^{3}P$ and positive spin polarization in the ${}^{1}D$ lines. The coupling is to the unpaired spin or magnetic moment at the Fe site. Since s_z along the magnetization of the sample is measured and averaged over many atoms, the signal reflects long-range order. The reported spin polarization thus contains information on the magnetization or, because of its local nature, on the sublattice magnetization in the case of a composite system.

(4) Fe $M_{23}M_{45}M_{45}$, shown in Fig. 2 at 45 eV: The final state in this super-Coster-Kronig tran-The final state in this super-Coster-Kronig transition is identical to that of the $L_3M_{45}M_{45}$ transition dependence on the transition of the superior μ tion described above. The two spectra are expected to be shifted in kinetic energy but otherwise to be similar, if we assume that the initial core hole is unpolarized. The observed spin polarization, however, is markedly different: The polarization is considerably higher in the MMM transition and it exhibits an additional peak on the high kinetic energy side. We do not, at present, understand the origin of the extra polarization maximum which has no visible counterpart in the intensity spectrum. We suggest that the alterations reflect surface-specific features of the sample. This is sensible since the escape depth of $45-eV$ electrons is only ~ 1 monolayer as compared to \sim 3 monolayers for 700-eV elec-

FIG. 3. Spin polarization P and intensity I vs kinetic energy of secondary electrons from $Fe_{83}B_{17}$, excited with electrons of E_b =2900 eV. Each datum point represents the average of three consecutive runs. The statistical error is $\Delta P = \pm 0.2\%$.

trons.¹⁰ In fact we observed that the spin polarie zation in the M/M transition was very sensitive to surface treatment, much more so than in the Auger lines at higher energies. By comparison of the spin polarization of the MMM and LMM transitions it is, in principle, possible to gain detailed information on changes of the local valence-electronic structure in going from the bulk to the surface of a Sd ferromagnet.

(5) B KLL shown in Fig. 3 at 175 eV: This Auger emission leaves behind two holes in the $2s2p$ valence band at the boron site. The polarization spectrum shows a minimum which exactly

lines up with the Auger intensity maximum, and a comparison of the relative intensity and spin polarization shows that these Auger electrons are unpolarized (cf. Table I). This concurs with the fact that the magnetization is zero at the boron site and clearly illustrates how spin-polarized Auger spectroscopy can be a powerful technique in revealing local magnetic features in magnetic alloys and compounds.

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