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Spin-resolved Fe $L_3M_{2,3}M_{2,3}$ and $L_3M_{2,3}V$ Auger transitions on and off resonance by excitation with linearly polarized photons

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Spin polarized Fe $L_3M_{2,3}M_{2,3}$ and $L_3M_{2,3}V$ Auger-electron spectra were measured and calculated using on-resonance excitation energies of linearly polarized photons (at the $2p_{3/2}$ threshold) as well as in the off-resonance region. Both the intensity distribution and the spin polarization in the experimentally obtained Auger spectra are explained by means of the theoretical model taking into account spin-orbit splitting of core levels and exchange interaction of the core states with the polarized valence band. Different contributions to the Auger spin polarization of both CCC (Fe $L_3M_{2,3}M_{2,3}$) and CCV (Fe $L_3M_{2,3}V$) Auger spectra are discussed. Auger electron spin polarization is positive by transitions to the singlet final two-hole states and is increased with increasing core state-valence state exchange interaction, whereas for the triplet final states the opposite behavior is found.

I. INTRODUCTION

Recently the magnetic behavior of the metallic surfaces, adlayers and thin films is intensively studied by means of spin-resolved spectroscopic techniques.[1–4] In particular, the pioneering work of Landolt and co-workers[5–8] has shown that spin-resolved Auger electron spectroscopy (SRAES) is a powerful probe of local, element specific magnetic properties of surfaces.

When Auger electron emission is caused by an unpolarized electron beam or by linearly polarized photons the spin polarization (SP) of the Auger electrons could in principle arise either from the mixing of different channels for the Auger decay or from spin-orbit effects and magnetism in the material being studied. The former mechanism is based on the interference of emitted electronic partial waves and leads to so-called dynamical SP (Ref. 9) that can be observed especially in the resonance case.[10] The necessary condition is that at least two decay channels with comparable amplitudes exist like in the photoelectron emission where we have the mixing between the $l_c \pm 1$ channels. If one channel dominates in the Auger decay, the contribution from dynamical effects to the SP of the Auger electrons is small. Then the SP in AES is caused predominantly by the magnetic properties and the spin-resolved measurements can provide us with valuable information on the electronic and magnetic structure of the investigated samples.

In order to utilize SRAES as a powerful probe of local magnetism in ferromagnetic solids and surfaces it is necessary to fully understand all the underlying mechanisms which contribute to net spin polarization of the Auger electrons. A simple picture of the Auger process involving valence electrons (CCV or CVV transitions) suggests that net spin polarization of the Auger electrons results directly from the spin polarization of density of states (DOS). On the other hand the CCC Auger transitions, as for example, $L_3M_{2,3}M_{2,3}$, of magnetic $3d$ elements[8] is expected to carry no net spin polarization. However, large SP of different sign are observed[5] in spite lack of involvement of the valence electrons, suggesting that other interactions during the Auger process are important. Some attempts to give a theoretical explanation for the observed Fe $L_3M_{2,3}M_{2,3}$ Auger spectra were done[11–13] just after the first experimental results had been published. Bennemann[11] first presented a simple (more qualitative) theory based on the idea of the coupling of the core shells involved into Auger process to the ferromagnetic $3d$ band. Subsequent model calculations performed by Kotani and Mizuta[12] have taken into account only spin-spin interactions of the electron states and included neither the spin-orbit interaction in the core shells, which is evidently larger than the exchange interaction with the valence states, nor the direct Coulomb interaction between two final-state holes in the $3p$ level, which play the key role in the formation of the energy structure of the observed Auger spectrum. The possible effect of these interactions is only briefly discussed by the authors in Ref. 13.

The aim of our paper is to study the formation processes of the Fe $L_3M_{2,3}M_{2,3}$ and Fe $L_3M_{2,3}V$ Auger spectra in more detail and to analyze the factors which contribute to observed strong SP of the escaping Auger electrons. On the contrary to the previous works[11,12] both spin-orbit and exchange interactions have been taken into account in the calculations. It has been shown that this approach leads to the quantitative agreement with the experimental data.

In order to resolve some of these issues it is essential to obtain SRAES spectra with higher-energy resolution. This is difficult to achieve with electron excitation because of the high secondary electron background making accurate spin-resolved measurements with high energy resolution not feasible. More importantly, the electron and photon (above core-level threshold) excited Auger spectra show broad peaks because the core-hole created in such process is not well defined (as evident from multiplets in the core-level photoemission spectra) giving raise to superposition of the Auger processes. Both of these difficulties can be overcome with the use of on-resonant excitation leading to sharper AES spectra. However, when using the on-resonant excitation the spin polarization of Auger electrons is strongly modified. Although this has been attributed to the spin asymmetry of the core hole due to promotion of core electron into the spin polarized unoccupied valence state[14] such effects need to be further analyzed.

The paper is organized as follows. Section II is devoted to the description of the experimental setup and measurements. The theoretical model and the details of calculations are presented in Sec. III. In Sec. IV we discuss the results obtained for Fe $L_3M_{2,3}M_{2,3}$ Auger spectrum whereas the Fe $L_3M_{2,3}V$ spectrum is discussed in Sec. V. Conclusions are drawn in Sec. VI.

II. EXPERIMENTAL PROCEDURE

The spin-resolved Auger electron spectra presented here were obtained with an electron spectrometer[14] consisting of a commercial (100 mm radius) hemispherical energy analyzer (±16° acceptance cone)[15] and a version of the NIST low-energy spin detector.[16] The on-resonance Auger electron spectra were measured utilizing linearly polarized soft x-ray photons from the U13UA wiggler beamline[17] at the National Synchrotron Light Source. The spectra were taken at $\hbar\omega=707$ eV, with electron and photon energy resolution of 1.25 eV and 0.75eV, respectively. The *e*-beam exited Auger spectra (off-resonance) were taken with primary electron beam of 4 keV and electron energy resolution of 4.0 eV. The photons and primary electrons were incident at 45° with respect to the surface normal and the Auger electrons were collected along the surface normal. The Fe sample was a relatively thick Fe film (>30 Å) epitaxially grown on a Ag(001) substrate. The contamination level after Fe film growth was below 3% of a monolayer and no Ag $3d$ photoelectrons were visible. The Fe films were magnetized in the photoemission plane along the [100] direction using a small pair of coils. The magnetization saturation of the Fe films was confirmed by *in situ* magneto-optical Kerr measurements. The spin detector was calibrated by comparing measured spin polarization of secondary, Fe$3p$ and Fe$2p$ photoelectrons with published Fe data.[18–21] All measurements were performed at room temperature.

III. THEORETICAL MODEL

The basic theoretical model and the main approximations used for the description of the Auger process have been given in Refs. 22–24. Here we present briefly the general expressions for the calculation of the spin-resolved Auger electron intensity. We pay more attention to the modifications of the theoretical model made with the aim to apply it to the ferromagnetic solids.

We disccuss the spectra where an initial core hole state c is created by photon absorption. The SP of the hole states is transferred to the Auger electron via the matrix element M of the Auger process

$$M(L\sigma,c|g_1,g_2)=\langle f_{L\sigma},c|V|\{g_1,g_2\}\rangle. \tag{1}$$

The escaping Auger electron having the energy ε_f is described by a sum over spherical waves (or different channels) characterized by quantum numbers $L(=l,m)$ and σ. The final two-hole state denoted as $\{g_1,g_2\}$ can belong either to the core shell or to the valence band. Usually $\{g_1,g_2\}$ is written as a product of two one-particle states and its exchange expression, thus, M contains the expectation value of the Coulomb interaction and the corresponding exchange integral. The essential point of our treatment is that $\{g_1,g_2\}$ can be a correlated two-particle state as explained later on.

The expression for the spin-resolved intensity of the Auger electrons in a direction $\mathbf{e}$ $(\mathbf{e}=\mathbf{r}/r,\mathbf{k}=k\mathbf{e},k=\sqrt{\varepsilon_f})$ can be written in the form

$$I_\sigma(\mathbf{k})=\sum_{g_1g_2}\langle M_\sigma^2(\mathbf{k})\rangle_{g_1g_2}\delta[\varepsilon_f+E(c)-E(g_1,g_2)], \tag{2}$$

where $E(c)$ and $E(g_1,g_2)$ are the total energies of the system with emitter having a core hole in the state c (initial state of the Auger process) and two holes in the state $\{g_1,g_2\}$, respectively. In Eq. (2) the sum over g_1,g_2 still contains the integration over different energies $E(g_1,g_2)$. We avoid to simplify this expression as in Eq. (2) of Ref. 24 to an explicit integral containing the densities of states.

Auger transition probabilities $\langle M_\sigma^2\rangle_{g_1g_2}$ can be expressed as

$$\langle M_\sigma^2(\mathbf{k})\rangle_{g_1g_2}=\sum_{\mu_c} w_{\mu_c}(\vec{\epsilon},\hbar\omega)$$
$$\times\left|\sum_L B_{L\sigma}(\mathbf{k})M(L\sigma,c|g_1,g_2)\right|^2. \tag{3}$$

$w_{\mu_c}(\vec{\epsilon},\hbar\omega)$ is the photoionization probability for the electron state μ_c in the core shell c. It depends on the polarization $\vec{\epsilon}$ and the energy $\hbar\omega$ of incoming photons. $B_{L\sigma}(\mathbf{k})$ is the scattering path operator.[25–28]

The spin polarization of the Auger electrons is obtained from the calculated spin-resolved intensities (2) as

$$P_{\mathrm{AES}}=\frac{I_\uparrow-I_\downarrow}{I_\uparrow+I_\downarrow}. \tag{4}$$

The axis of quantization coincides with the direction of the magnetization $\mathbf{M}$.

In order to calculate Auger transition matrix elements at first we determine one-particle wave functions $|\psi\rangle$ that include both the spin-orbit interaction and the exchange interaction. The latter can be treated by means of a spin field. These contributions to the one-particle Hamiltonian are given by

$$H_{\mathrm{int}}=\lambda \boldsymbol{l}\cdot s+\xi s_z, \tag{5}$$

so that the spin-orbit and exchange splitting of the electron states are equal to $\Delta_{\mathrm{so}}=\frac{3}{2}\lambda$ and $\Delta_{\mathrm{ex}}=\xi$, respectively. It is reasonable to choose the eigenfunctions of the spin-orbit part of the Hamiltonian

$$|jl\mu\rangle=\sum_{m\sigma} C^{j\mu}_{lm,(1/2)\sigma}|lm\sigma\rangle \tag{6}$$

as basis functions. Here $C^{j\mu}_{lm,(1/2)\sigma}$ are the Clebsh-Gordan coefficients. This approach is similar to that applied in Refs. 29–31 to the photoemission process. However, in the Auger process additionally the correlation in the final two-hole state has to be taken into account.

The Hamiltonian for the core p level in the basis

$$|3/2,-3/2\rangle=Y_{1-1}\chi_-,$$

$$|3/2,-1/2\rangle=\sqrt{1/3}Y_{1-1}\chi_+ +\sqrt{2/3}Y_{10}\chi_-,$$

$$|3/2,+1/2\rangle=\sqrt{2/3}Y_{10}\chi_+ +\sqrt{1/3}Y_{11}\chi_-,$$

$$|3/2,+3/2\rangle=Y_{11}\chi_+,$$

$$|1/2,-1/2\rangle=-\sqrt{2/3}Y_{1-1}\chi_+ +\sqrt{1/3}Y_{10}\chi_-,$$

$$|1/2,+1/2\rangle=-\sqrt{1/3}Y_{10}\chi_+ +\sqrt{2/3}Y_{11}\chi_-$$

becomes

$$H_{int}=\begin{pmatrix} \frac{1}{2}\lambda-\frac{1}{2}\xi & 0 & 0 & 0 & 0 & 0 \\ 0 & \frac{1}{2}\lambda-\frac{1}{6}\xi & 0 & 0 & -\frac{\sqrt{2}}{3}\xi & 0 \\ 0 & 0 & \frac{1}{2}\lambda+\frac{1}{6}\xi & 0 & 0 & -\frac{\sqrt{2}}{3}\xi \\ 0 & 0 & 0 & \frac{1}{2}\lambda+\frac{1}{2}\xi & 0 & 0 \\ 0 & -\frac{\sqrt{2}}{3}\xi & 0 & 0 & -\lambda+\frac{1}{6}\xi & 0 \\ 0 & 0 & -\frac{\sqrt{2}}{3}\xi & 0 & 0 & -\lambda-\frac{1}{6}\xi \end{pmatrix}. \tag{7}$$

The exchange interaction leads to the mixing of the $j=3/2$ and $j=1/2$ states having the same value of μ which is still a good quantum number:

$$|\psi_1(\pm 1/2)\rangle=\frac{1}{\sqrt{1+\alpha^2}}(|3/2,\pm 1/2\rangle-\alpha|1/2,\pm 1/2\rangle),$$

$$|\psi_2(\pm 1/2)\rangle=\frac{1}{\sqrt{1+\alpha^2}}(|1/2,\pm 1/2\rangle+\alpha|3/2,\pm 1/2\rangle). \tag{8}$$

Equation (7) can be easily diagonalized to obtain the eigenstates ψ, and the mixing parameter α can be expressed in terms of λ and ξ:

$$\alpha=\frac{1}{x}(\sqrt{1+x^2}-1),\quad x=\frac{4\sqrt{2}\xi}{9\lambda\pm 2\xi}, \tag{9}$$

where the sign in denominator corresponds to the sign of the μ value. In the limit $\lambda\gg\xi$ an approximation for α could be used which is not dependent on μ:

$$\alpha=\frac{2\sqrt{2}\xi}{9\lambda}. \tag{10}$$

The mixing parameter α has been estimated from the data for the spin-orbit and exchange splittings published in Ref. 29, where they were obtained by fitting the calculated results for magnetic circular dichroism in photoemission to the experimental spectra. The values used in the present calculations are $\alpha_{2p}=-0.033$ and $\alpha_{3p}=0.243$. For the sake of simplicity we have used values calculated from Eq. (10) in spite of weak validity of this approximation for $3p$ level [Eq. (9) gives the values $\alpha_{3p}=0.205$ for $\mu=-1/2$ and $\alpha_{3p}=0.282$ for $\mu=+1/2$]. Different signs for the α_{2p} and α_{3p} values are caused by the fact that in the initial state of the Auger process the hole-state is considered in the $2p$ shell whereas in the $3p$ shell there is an electron state.

To calculate the correlated two-hole final state $\{g_1,g_2\}$ we have to take into account the Coulomb interaction leading to the energy splitting of the individual LS terms. These two-particle states can be constructed as linear combinations of $|LS;JM_J\rangle$ functions created by means of angular momentum summation and transformation relations

$$|j_1j_2;JM_J\rangle=\sum_{\mu_1\mu_2} C^{JM_J}_{j_1\mu_1,j_2\mu_2}|j_1l_1\mu_1\rangle|j_2l_2\mu_2\rangle, \tag{11}$$

$$|LS;JM_J\rangle=\sum_{j_1j_2}\zeta\begin{Bmatrix} l_1 & \frac{1}{2} & j_1 \\ l_2 & \frac{1}{2} & j_2 \\ L & S & J \end{Bmatrix}|j_1j_2;JM_J\rangle,$$

$$\zeta=[(2L+1)(2S+1)(2j_1+1)(2j_2+1)]^{1/2}. \tag{12}$$

Finally, the spin-orbit interaction and the exchange interaction with the valence electrons (5) have to be included for both electrons and this two-particle Hamiltonian has to be diagonalized. The most important results are the eigenfunctions whereas the energy eigenvalues are mainly determined by the exchange interaction between the two electrons. Details of this procedure will be described elsewhere. These two-particle functions inserted into Auger transition matrix elements (1) for the corresponding configuration of the final two-hole state $\{g_1, g_2\}$ give us the possibility to calculate the spin-resolved intensities (2), and, consequently, the SP (4) for each LS component of the Auger spectrum. In the present work we have not taken into account effects of the magnetic scattering of outgoing Auger electrons, for simplicity, therefore only the direct wave contributions were retained in Eq. $B_{L\sigma}(\mathbf{k})$ in Eq. (3). For the calculations we consider a geometry used in the experiment described in Sec. II.

Auger transition selection rules provide for the $L_3M_{2,3}M_{2,3}$ transition outgoing electron waves of p and f symmetry. For the $L_3M_{2,3}V$ transitions (if we take into account only valence d electrons involved in the Auger process) there are outgoing s, d, and g waves. Our calculations of the partial Auger intensities show that for the considered transitions in Fe only one decay channel dominates, namely the outgoing p wave for the Fe $L_3M_{2,3}M_{2,3}$ transition (between 90 and 99 % of the total intensity for different multiplet components) and the outgoing d-wave for the Fe $L_3M_{2,3}V$ transition (between 79 and 95 % of the total intensity). Thus, dynamical effects on the SP should be negligible and are not taken into account.

In the case of the resonant excitation there are two possible decay processes of the excited state. First, the electron that has been excited into the state just above the Fermi level goes back to the core hole (a so called participator transition which contributes to the intensity of the direct photoemission peak), and second, spectator Auger decay where the core hole is filled by an electron from the states below the Fermi level. Because of the extended character of the Fe band state above the Fermi level and a high DOS in the occupied part of the valence band, it could be expected that the probability of the participator decay is small as compared to the spectator decay. The latter can be treated theoretically as a pure Auger decay that differs from the off-resonance case only in an additional spin polarization of the core hole caused by the primary photoexcitation of the core electron into the polarized state. For the on-resonance excitation the Fe $3p$-photoelectron peak (maybe enhanced by participator transitions) overlaps partly the $L_3M_{2,3}V$ Auger spectrum from the high-kinetic-energy side. However, the experimental measurements as well as the theoretical estimations show that its intensity is negligibly small in comparison to the $L_3M_{2,3}V$ (spectator) Auger intensity and thus its effect on the SP was not taken into account. It should be noted, that this is the case for the L_3 resonance but not for the $M_{2,3}$ resonance which is about a factor 100 weaker and true interference effects need to be included.

If valence electrons are involved in the Auger process, the shape of the spectrum is determined (together with the other effects discussed above) also by the DOS in the valence band. However, the interaction of two final-state holes, U, plays a significant role in the formation of the spectral shape: the bandlike spectrum observed for small values of U transforms to an atomiclike for large U (for CVV transitions the shape of the Auger spectrum can be expressed in terms of the valence DOS and the parameter U by means of the Cini-Sawatzky model[32,33]). For the $L_3M_{2,3}V$ (i.e., CCV) spectrum, however, the effect of the final-state interaction on the valence DOS is properly included by considering the atom with the $3p$-hole as a point defect. The calculated DOS curve (distorted by the core-hole potential) can be used to construct the spectrum, as this is discussed below in Sec. V.

The local partial DOS and the occupation numbers for the Fe valence subbands were calculated by means of the self-consistent linear muffin-tin orbital (LMTO) method[34,35] in the atomic sphere approximation and including combined corrections. For the calculation of the local electronic structure of atoms with a core hole the LMTO Green function method has been used.[36,37]

IV. FE $L_3M_{2,3}M_{2,3}$ SPECTRUM

For the $L_3M_{2,3}M_{2,3}$ transition there are two final-state holes in the $3p$ shell giving rise to three LS components: two singlet states ^1S, ^1D, and a triplet state ^3P. Their energy can be described by the following relations:

$$E(^1S)=F_0+10F_2,$$

$$E(^1D)=F_0+F_2,$$

$$E(^3P)=F_0-5F_2, \tag{13}$$

where the values of F_0 and F_2 are proportional to the Coulomb integrals for the $3p$-hole pair.[38,39] As it can be seen from the experimental results (Fig. 1) the shape of the Auger spectra is determined by ^1D and ^3P peaks. The contribution from the ^1S final state cannot be clearly resolved due to its small intensity. The ^1D component has a positive SP whereas that of the ^3P component is negative.

The calculated results presented in Table I show the role of the exchange interaction in the formation of the SP of the LS components in the spectrum. For the off-resonance excitation (the excitation energy of 1360 eV was used in the calculations, i.e., the energy of the photoelectron state was about 660 eV above Fermi threshold) the calculations give practically no SP for all components if the exchange interaction of the core states with the polarized valence band is not taken into account ($\xi=0$, case A). Small values of the SP given in Table I are caused by the fact that photoelectrons with majority and minority spins have slightly different radial parts of the wave function and, consequently, different dipole matrix elements due to the different potentials for spin-up and spin-down electron states. As a result the polarization of the $2p_{3/2}$ hole is equal to $P_{\mathrm{hole}}=-0.07\%$. The Coster-Kronig L_2L_3V decay of the $2p_{1/2}$ hole can also contribute to the initial polarization of the $2p_{3/2}$ hole, but our estimations show that these contributions are small and P_{hole} does not exceed 1% in all off-resonance cases.

If the exchange interaction is switched on for the $2p$ shell (case B) the SP of individual LS components is changed only in few percent. It follows from these results that effect of the exchange corrections in the $2p$ shell on the spin polarization of the Auger electrons is small, although it is not negligible.

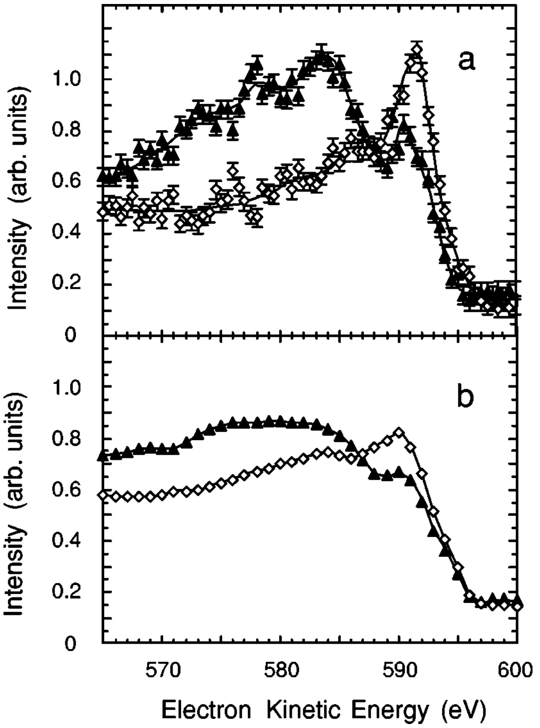

FIG. 1. Spin-resolved experimental Fe $L_3M_{2,3}M_{2,3}$ Auger spectra obtained for on-resonance (a) and off-resonance (b) excitation: majority-spin contributions are denoted by solid triangles, minority-spin contributions by open diamonds.

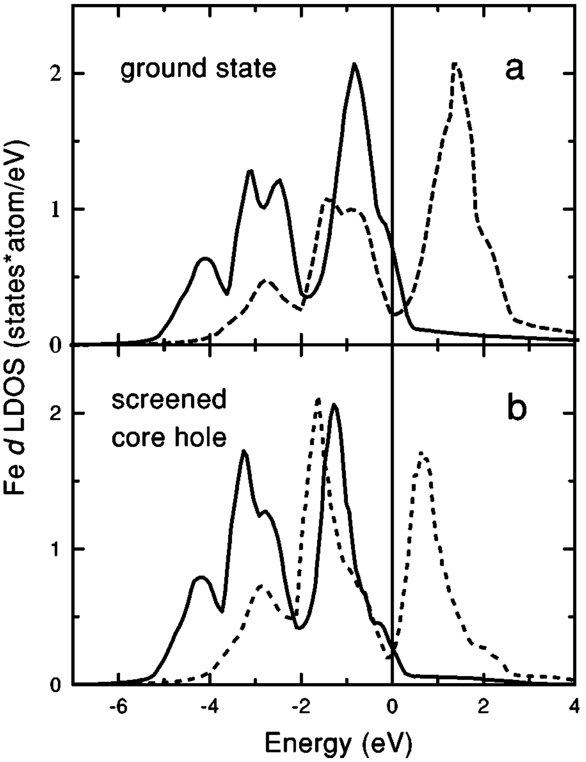

FIG. 2. Local density of the valence d-states for an atom in Fe metal: ground state (a) and an atom with $3p$ hole (b) for majority (solid line) and minority (dashed line) spin.

In case C the calculations include the exchange interaction of the core states with the valence electrons for both $2p$ and $3p$ shells. It could be seen from the results given in Table I that just this modification of the wave functions of the $3p$ states determine the values of P. These calculated values agree well with the experimental ones obtained at the maxima of ^1D and ^3P components. The ^1S component has a large positive SP over 22%, but its intensity is only 11% of the total $L_3M_{2,3}M_{2,3}$ intensity whereas its position is almost 10 eV to the low-energy side from the main peaks. Due to the weak intensity and a higher background in this energy region the SP of the ^1S component can not be estimated experimentally with sufficient accuracy. The individual J contributions to the triplet ^3P component are not resolved in the experiment, and we have compared with the experiment the total values evaluated from the superposition of all J contributions. It should be noted, however, that $^3P_0$, $^3P_1$, and $^3P_2$ terms have quite different behavior if the exchange interaction is switched on. The most sensitive is the $^3P_1$ final state with a drastic increase of the absolute SP value to $|-40\%|$. On the other hand the $^3P_2$ term which shows only an increase of the SP to $|-3\%|$ contributes the main part to the intensity. The $^3P_0$ component has small intensity and cannot significantly influence total values for ^3P triplet. Due to the different behavior of the $^3P_1$ and $^3P_2$ terms the calculated Auger-electron SP may be sensitive to the small changes in the values of exchange corrections. This sensitivity could be useful for deducing interaction parameters from experimental data, and these possibilities will be studied in our forthcoming work.

TABLE I. Itensities and spin polarizations (in %) of two-hole final-state components in the off- and on-resonance Fe $L_3M_{2,3}M_{2,3}$ spectra. The experimental data are the values at the maxima of the ^1D and ^3P components in Fig. 1: A: without exchange corrections, B: $\alpha_{2p}=-0.033$, $\alpha_{3p}=0$, C: $\alpha_{2p}=-0.033$, $\alpha_{3p}=0.243$.

Final state	$^1S_0$	$^1D_2$	$^3P_0$	$^3P_1$	$^3P_2$	Total $^3P_{0,1,2}$
Intensity	11.0	40.7	2.7	12.1	33.5	48.3
P(off)						
A	$+0.15$	$+0.005$	$+0.02$	-0.20	-0.12	-0.13
B	$+2.43$	$+2.67$	-4.44	-0.93	-1.11	-1.25
C	$+22.13$	$+11.67$	-19.74	-38.95	-2.91	-13.00
Exp.		$+8$				-15
P(on)						
A	$+18.00$	$+16.21$	$+17.87$	-14.92	-2.56	-4.51
B	$+20.28$	$+18.82$	$+13.16$	-15.50	-3.53	-5.59
C	$+40.50$	$+27.20$	-3.98	-49.26	-4.47	-16.30
Exp.		$+22$				-21

In the case of L_3-resonance excitation the photoelectron goes to the state just above the Fermi level. As it can be seen from Fig. 2(a), in this small energy region the ratio of the majority and minority spin states changes considerably. These changes can cause a strong dependence of the $2p_{3/2}$ core-hole SP on the excitation energy. In the ground state at the Fermi level the SP of the valence states is still positive, and only 0.3 eV above the Fermi energy the minority-spin states begin to dominate. Taking into account the finite energy resolution in the experiment, we have assumed in our calculations the final energy of the photoelectron about of 0.6 eV above the Fermi level. In this case the polarization of

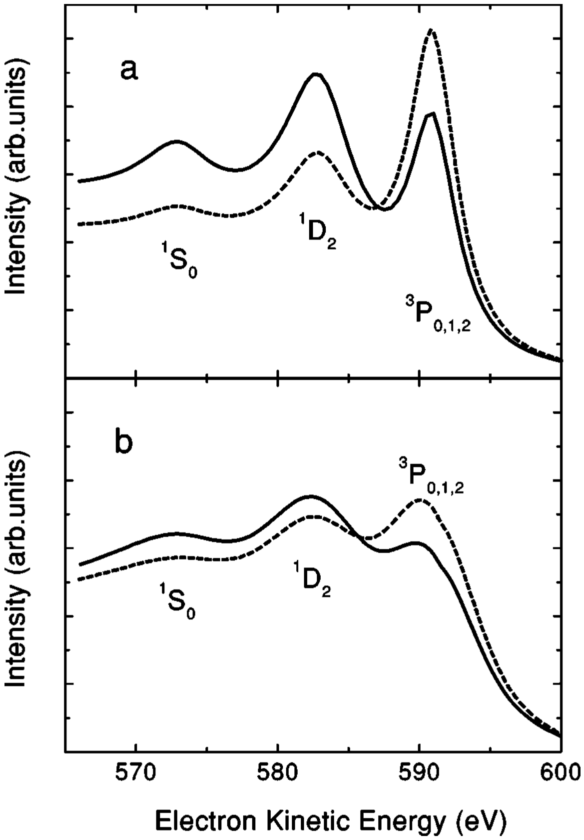

FIG. 3. Majority-spin (solid line) and minority-spin (dashed line) contributions to the Fe $L_3M_{2,3}M_{2,3}$ Auger spectra calculated for on-resonance (a) and off-resonance (b) excitation. An integral background was calculated in the manner to reproduce the experimental signal-to-background ratio.

the core hole is equal to -14.7%. It should be noted, that further small increasing of the excitation energy makes it available for the photoelectron to reach the energy region of maximal density of unoccupied minority-spin states (at 1.4 eV above Fermi level) providing the core hole SP about -50%.

Considering the calculated results for the on-resonance $L_3M_{2,3}M_{2,3}$ spectra (Table I) we note that a significant SP of the Auger electrons is obtained even without exchange corrections, due to the initial spin polarization of the core hole. It is interesting to note, that switching on the exchange interaction causes practically the same changes in the SP values of the Auger electrons as in the off-resonance spectra (only ''starting values'' are others): the differences of values given in Table I for the cases A, B, and C for every multiplet component are very close to those calculated for the off-resonance excitation.

Calculated spin-resolved Fe $L_3M_{2,3}M_{2,3}$ Auger spectra are given in Fig. 3. For the line shapes, Lorentzian curves with 3.0 eV FWHM (lifetime and finite resolution effects) for each multiplet component was assumed. Comparison with the experimentally obtained spectra (Fig. 1) shows that calculated and measured results agree well also in the line shapes of the spectra.

Fe $L_3M_{2,3}M_{2,3}$ Auger spectra have been studied theoretically by Mizuta and Kotani[13] using similar ideas to develop the theoretical models as in the present work. However Mizuta and Kotani describe spin-orbit, exchange and final-state interactions with adjustable parameters while these interactions were obtained from direct calculations in our case. The most remarkable disagreement of the Mizuta-Kotani results with the present theoretical results (as well as with the experimental spectra) is the position of zero SP point in the spectrum: in Ref. 13 it is directly under the triplet peak whereas its correct position can be found between ^1D and ^3P peaks (see the crossing point of majority-spin and minority-spin curves in Figs. 1 and 3). To consider the resonance case the authors of Ref. 13 need to introduce an additional adjustable parameter, whereas in the present work we perform calculations for both off-resonance and on-resonance cases within the same model. Finally, the formalism from[13] could not be immediately applied to the $L_3M_{2,3}V$ transitions.

V. FE $L_3M_{2,3}V$ SPECTRUM

In the case of the $L_3M_{2,3}V$ transitions the valence electrons are directly involved into Auger process. Taking into account the Auger transition probabilities as well as the occupations of the sp and d subbands (or the partial DOS), it could be seen that their contributions to Auger electron intensity differ in three orders of magnitude. Thus, the effect of the sp subbands on the intensity and, consequently, on the spin polarization of the Auger electrons is negligible, and in the following consideration it is not taken into account. In this case it is possible to talk about $L_3M_{2,3}M_{4,5}$ spectra.

In the initial state of the Auger process the Fe atom has a $2p$ hole which is screened by the valence electrons. The screening processes lead to the increasing of the local valence charge and to the redistribution of the valence DOS. From the calculations of the local electronic structure of the Fe atom with a core hole embedded into Fe metal we have obtained for the occupation number of the valence d-state 7.65 whereas in the ground state of the Fe atom the calculations give 6.58. It should be noted that processes of the core-hole screening leads to small decreasing of the s contribution, the p contribution is practically unchanged. Thus, it could be concluded that the core-hole screening in Fe has d character. Owing to the core-hole screening the local SP of the valence states is also changed to 22.6% (4.69 electrons with majority spin and 2.96 electrons with minority spin) from in the ground state polarization of 35.0% (4.44 majority spin and 2.14 minority spin). These occupation numbers were used for the calculation of the spin-resolved Auger electron intensities. Here we have assumed the same occupation numbers for all d orbitals. The effect of different occupation for d states of t_{2g} and e_g symmetry was discussed in Ref. 23.

The interaction of two final-state holes gives rise to the energy splitting of the individual multiplet components. Because these two states belong to different subshells, the picture of the two-particle p-d configurations is more rich than in the $L_3M_{2,3}M_{2,3}$ case considered above. Similar to Eq. (13) the energies of the multiplet components can be defined in terms of Coulomb and exchange integrals as[38,39]

TABLE II. Intensities and spin polarizations (in %) of two-hole final-state components in the off- and on-resonance Fe $L_3M_{2,3}V$ spectra. The experimental data are the values at the maxima of the singlet and triplet components in Fig. 4: *A*: without exchange corrections, integral $3d$ contributions ($n_\uparrow=4.69, n_\downarrow=2.96$), B: $\alpha_{2p}=-0.033$, $\alpha_{3p}=0.243$, integral $3d$ contributions, C: $\alpha_{2p}=-0.033$, $\alpha_{3p}=0.243$, contributions from different regions of the valence band: C_1: low-energy half of the VB ($n_\uparrow=2.69, n_\downarrow=0.95$), C_2: high-energy half of the VB ($n_\uparrow=2.00, n_\downarrow=2.01$).

Final state	$^1F_3$	$^1P_1$	$^3D_{1,2,3}$	$^3P_{0,1,2}$	$^1D_2$	$^3F_{2,3,4}$
Intensity	42.7	13.0	26.9	12.3	1.9	3.2
P(off)						
A	+2.35	+5.78	+18.30	+21.08	+51.95	+62.19
B	+10.42	+22.61	+4.94	+15.76	+68.31	+26.44
aver.	+13.3		+8.3			
C_1	+13.25	+28.92	+26.31	+38.20	+93.48	+45.20
C_2	+8.04	+16.92	−12.92	−5.52	+25.63	−20.01
	+14.0		+9.0			
Exp.	+15		+6			
P(on)						
A	+17.59	+19.61	+14.00	+15.56	+60.94	+59.44
B	+25.49	+35.88	+1.04	+10.37	+74.98	+28.05
aver.	+28.0		+4.0			
C_1	+28.04	+41.49	+22.85	+34.33	+94.95	+50.09
C_2	+23.32	+30.92	−16.67	−11.26	+37.83	−19.82
	+29.0		+8.0			
Exp.	+33		+11			

$$E(^1P)=F_0+7F_2+G_1+63G_3,$$

$$E(^3P)=F_0+7F_2-G_1-63G_3,$$

$$E(^1D)=F_0-7F_2-3G_1+21G_3,$$

$$E(^3D)=F_0-7F_2+3G_1-21G_3,$$

$$E(^1F)=F_0+2F_2+6G_1+3G_3,$$

$$E(^3F)=F_0+2F_2-6G_1-3G_3. \tag{14}$$

For the sake of simplicity we have considered here the interaction of the $3p$ core hole with *one* $3d$ hole in spite the final-state valence band configuration could give rise to the much more complicated multiplet structure. Thus, in our simplified picture we discuss neither the individual *J* contributions to the triplet components nor an individual occupation of different *d* states. This is possible, because the most important effect is the change of the two-hole state wave function.

The multiplet components are given in Table II in order of increasing Auger electron kinetic energy. The strong singlet ^1F and ^1P contributions (about 55% of the total $L_3M_{2,3}V$ intensity; the energy separation of these lines is about 1 eV) determine the peak of the maximal intensity in the experimental spectra (see Fig. 4). The triplet ^3D and ^3P contributions are shifted about 5 eV to the higher energies. They provide almost 30% of the total intensity, and their energy separation is equal to 0.6 eV. These contributions are responsible for the high-energy shoulder clearly seen in the experiment. Two last components, ^1D and ^3F, lie about 4 eV to the higher kinetic energies from the main triplet contributions. They have only 5% of the total intensity and are not resolved in the experimental spectra.

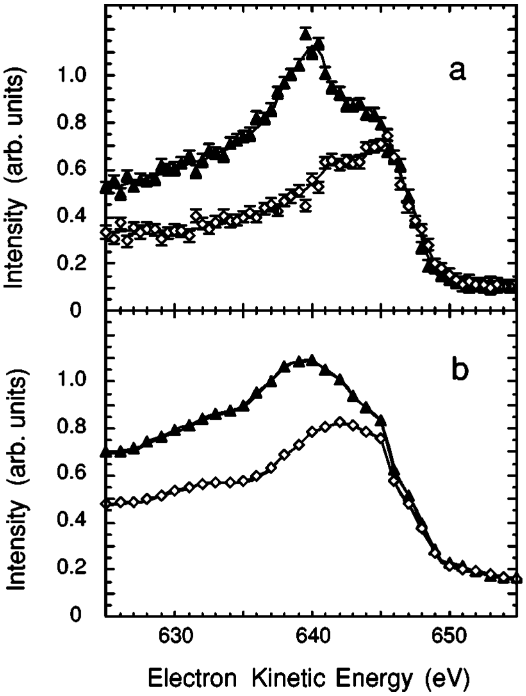

FIG. 4. Spin-resolved experimental Fe $L_3M_{2,3}V$ Auger spectra obtained for on-resonance (a) and off-resonance (b) excitation. Notations as in Fig. 1.

Let us consider the effect of the exchange corrections on the Auger electron SP. The positive values given in the Table II for the off-resonance excitation in the case A (without exchange corrections) are determined by the polarization of the valence $3d$ states. It can be seen that these values show no agreement with the experimental results. The singlet final states provide smaller SP of Auger electrons, and the triplet final states show much higher P values, than those measured in the experiment. Note that the weak ^1D and ^3F components have very high SP values.

Taking into account exchange corrections (case B) leads to the strong increasing of the SP calculated for the singlet final states, whereas the SP values for the triplet final states are decreased. In order to compare the values calculated for individual multiplet components with the experimental results one has to average the contributions from close singlet (^1F and ^1P) or triplet (^3D and ^3P) lines as shown below case B. Despite the simplifications mentioned above it is easy to see that including exchange interaction brings the SP to much better agreement with the experiment.

Now we can try to include into the calculations the finite width of the valence band. Due to this finite width the energy position of each final-state multiplet component depends on the energy of the final $3d$ hole in the valence band. In order to describe the shape of the $L_3M_{2,3}V$ spectrum we have to scan the valence band performing the calculations for different energies of the $3d$ hole. Clearly, for different energies the ratio of the majority and minority spin states is different, and instead of the integral $3d$ occupation numbers (used in case

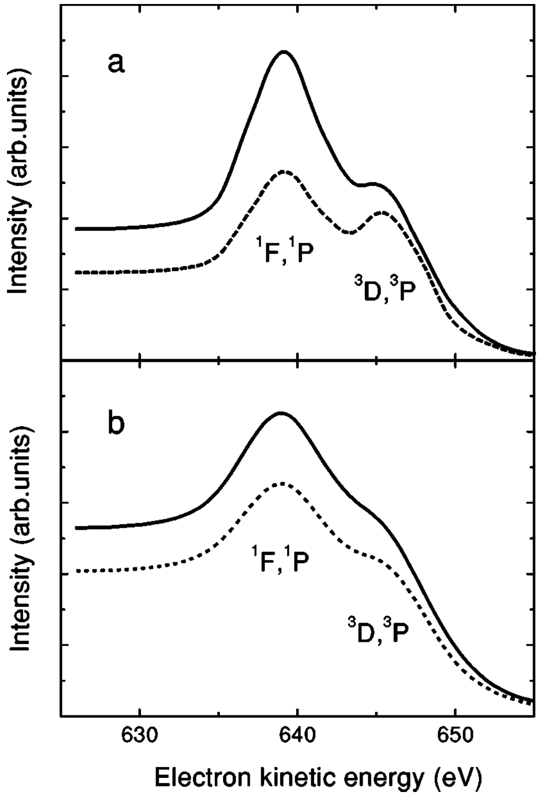

FIG. 5. Majority-spin (solid line) and minority-spin (dashed line) contributions to the Fe $L_3M_{2,3}V$ Auger spectra calculated for on-resonance (a) and off-resonance (b) excitation.

B) the corresponding values taken from the calculated DOS should be used. Consequently, the spin-dependent contributions to the Auger spectrum are quite different, and they are distributed in the energy region of the valence-band width. We have studied this valence-band effect on the Auger electron SP using a simplified approach. The valence DOS screening the core hole [see Fig. 2(b)] could be well separated in two parts: the low-energy part (below of -2.3 eV) where the states with majority spins dominate, and the high-energy part where the contributions from both spin states are almost equal. These two parts were approximated by two Lorentzians splitted in energy by 2 eV. Performing this procedure for each multiplet component we have obtained SP values denoted in Table II as C_1 and C_2. Note how strong is the effect of the polarization in the valence band on the Auger electron SP. In general, the SP of the Auger electrons increases with increasing of the valence-band SP and decreases with its decreasing. The changes in the SP values are more significant for the triplet final states. Decreasing of the positive valence-band SP leads for these states to negative Auger electron SP.

For comparison with the experiment we have calculated the shape of the Fe $L_3M_{2,3}V$ spectrum as a superposition of the Lorentzians (in the case C twelve contributions have been taken instead of six ones in the cases A,B). For the spectrum presented in Fig. 5 the Auger electron SP was calculated at the main peak and at the high-energy shoulder. The results ($+14$ and $+9$ %, respectively) are in excellent agreement with the experimental data.

For the on-resonance excitation the behavior of the SP for different multiplet components in dependence on the exchange interaction or on the valence-band polarization is very similar to that for the off-resonance spectra (as it was discussed above for the case of Fe $L_3M_{2,3}M_{2,3}$ Auger transitions), only the ''starting values'' are different. It should be noted, that these ''starting values'' increase due to the resonant excitation for singlet components whereas for triplet components they decrease (compare off- and on-resonance values of SP given in Table II for case A).

Using the same procedure as for the off-resonance spectrum we have estimated the SP values equal to $+29$ and $+8$ %, for singlet and triplet contributions, respectively. This results agree well with the experimental data.

VI. CONCLUSIONS

We have studied the spin-resolved off- and on-resonance Fe $L_3M_{2,3}M_{2,3}$ and $L_3M_{2,3}V$ Auger spectra both experimentally and theoretically. In the theoretical model the spin-dependent Auger transition matrix elements for excitation with linearly polarized photons and the partial electron-state configuration in the valence band were taken into account. It was shown that it is very important to include a correlated two-hole state into the evaluation of the Auger transition matrix elements, where the exchange interaction between the two electrons as well as the spin-orbit interaction and the exchange interaction with the polarized valence band are considered on the same footing. The calculated Auger electron SP values are in good quantitative agreement with the experiment.

It follows from the calculations that the spin field acting on the core state due to the spin-resolved valence band (exchange interaction) is the most important fact for the formation of the observed values of the Auger electron SP. A satisfactory agreement with the experiment cannot be achieved if only on-resonance excitation or transitions from the polarized valence band are taken into account as possible sources for the Auger electron SP. The increasing exchange interaction causes increasing absolute SP for the singlet and triplet final states whereas the sign is opposite.

It should be noted, that in spite of approximations made in the theoretical model (for example, simplified picture of the multiplet structure for the final state of the $L_3M_{2,3}V$ transition) the calculated shape of the Auger spectra agree well with the experimental one. Some deviations between the calculations and the experiment, for example, relative energy shifts of the majority-spin and minority-spin peaks will be considered in our forthcoming work.

The theoretical model used in the present work does not include some effects which could modify the energy distribution of the Auger electrons and their SP. They are (i) magnetic scattering of outgoing electrons, due to different scattering phase shifts for the electrons with majority and minority spins, (ii) surface effects, because the local electronic structure as well as magnetic moment of the Fe atom in the surface layer is different from those in the bulk, (iii) different symmetry of the valence *d* states, that can play an important role in the orientation dependence of the Auger electron SP. All these effects were discussed in Ref. 23 for Fe L_3VV spectra where it was shown that they could change the SP values in few percent. Furthermore, in an improved theoretical model we have to include the multiplet splitting in more detail.

As mentioned the exchange to spin-orbit splitting ratio for Fe $3p$ level is an important parameter in the theoretical model. However there is little, if any, consensus on the values of these two quantities (see Ref. 29) The sensitivity of the Auger electron SP for the specific multiplet components to the magnitude of the exchange interaction and the selective character of the exchange interaction effects on the individual components in connection with the accurate experimental measurements could be used for estimation of the parameters of electron-electron and spin-spin interactions in ferromagnetic solids.

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