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Spin-resolved Fe $L_3M_{45}M_{45}$ Auger transition on and off resonance: The effect of exchange correlation

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Spin-resolved Fe $L_3M_{45}M_{45}$ Auger-electron spectra were recorded with on- and off-resonant ($2p_{3/2}\rightarrow 3d^*$) photons. The spectra agree with convolution of spin-resolved Fe density of states, except in the degree of spin polarization which was found to be higher (lower) than the spin polarization of Fe valence electrons when using on- (off-) resonant photon excitation. This is consistent with a reduction of the Auger transition probability of the spin-parallel two-hole final state as compared to spin-antiparallel state, which indicates the importance of exchange correlation in $3d$ itinerant ferromagnets such as Fe.

Studies of resonant processes at the L_3 edges of $3d$ transition metals and their compounds including x-ray absorption and emission, Auger-electron emission, and resonant photoemission, have recently attracted much attention. In particular, there has been much interest in employing resonant behavior in the study of magnetic materials, most notably using circularly polarized photons to yield magnetic circular dichroism effects[1–3] with the potential of providing local, *element specific* magnetic properties.[4–6] This information can also be provided by spin-resonant electron emission techniques involving core levels. In particular, the pioneering work of Landolt and co-workers[7–10] has shown that spin-polarized Auger-electron spectroscopy (SPEAS) is a powerful probe of local, *element specific* magnetic properties of thin films. Although it has been demonstrated that the spin polarization of core-valence-valence (CVV) Auger-electron spectra (AES) is a direct manifestation of the local polarization of the valence electrons, the question of how these two are related remains open. One way of testing whether CVV SPAES is a direct measure of valence spin polarization or is complicated by electron correlation effects is to determine if the AES spin polarization depends on the polarization of the initial core hole. This can be done most conveniently by performing SPAES experiments on and off resonance, which is the topic of this paper.

In the past two decades much understanding has been gained regarding the ways in which electron correlation effects influence CVV AES spectra. The work of Sawatzky, Cini, and others[11–15] has shown that CVV AES in fact provides a good measure of the effective correlation energy of two valence electrons [Hubbard's U^{eff} (Ref. 16), and that the CVV Auger spectra of strongly correlated $3d$ metals ($U^{\text{eff}} > 2W$) like Cu and Ni are dominated by atomic multiplets, while more weakly correlated ones, such as Fe and more bandlike ($U^{\text{eff}} < 2W$) and are best represented by a convolution of the density of states (DOS). The question we address in this paper is to what extent spin-dependent interactions, or *exchange correlations*, are present in bandlike ferromagnetic metals such as Fe. Such correlations, or "polar fluctuations," have been shown by Herring[17] to be important in the development of magnetism in $3d$ metals.

In this paper we report spin-resolved measurements of Fe $L_3M_{45}M_{45}$ Auger-electron transitions excited either with off- or on- the Fe L_3-resonant ($2p_{3/2}\rightarrow 3d^*$) excitation using synchrotron radiation at $h\nu=707$ and 820 eV, respectively. The work of Landolt and co-workers employing electron excitation identified the resonant behavior of the Fe $M_{23}M_{45}M_{45}$ feature.[10] Similarly, Kisker and co-workers[18] have measured the spin polarization of the same transition using on-M_{23}-resonance photon excitation. Although both of these studies have indicated that the on resonance Auger-spin polarization is greater than the valence-band spin polarization, these estimates were made difficult by the uncertainties of high or unknown background. In this work, we employ the L_3 resonance, which is an order of magnitude stronger than the M_{23} resonance (photon or electron excited). This greatly reduces the effect of background on the estimate of spin polarization and also eliminates possible complexities due to interference with the direct photoemission channel. Furthermore, we make direct comparison between on- and off-resonant Auger spectra, as well as with electron excited spectra, from the same sample and instrumentation, thereby eliminating any systematic errors.[19]

The spin-polarized Auger-electron-emission experiments reported here were carried out on a newly constructed electron spectrometer consisting of a commercial (100-mm radius) hemispherical energy analyzer ($\pm 6°$ acceptance cone)[20] and a version of the NIST low-energy spin detector.[21] $L_3M_{45}M_{45}$ Auger electrons were excited using linearly polarized soft-x-ray photons provided by the U13UA wiggler beamline[22] at the National Synchrotron Light Source. The spectra presented here were taken with electron energy resolution of 1.25 eV and photon energy resolution of 0.75 eV. The photons were incident at 45° with respect to the surface normal and the 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 polarizations of secondary,

Fe3p, and Fe2p photoelectrons with published Fe data.[23–26] All measurements were performed at room temperature.

First, we briefly discuss the spin-integrated data. A series of Fe $L_3M_{45}M_{45}$ Auger-electron spectra taken from our Fe/Ag(001) samples using a range of photon energies near the Fe L_3 edge (700–735 eV) closely resembles the resonant Auger behavior reported previously.[27] The on-L_3-resonant ($h\nu$=707 eV) Fe $L_3M_{45}M_{45}$ Auger intensity was found to be 35 times greater than the direct Fe valence band photoemission at $h\nu$=700 eV and 5 times greater than off this resonance ($h\nu$=735 eV). No significant changes in Fe $L_3M_{45}M_{45}$ Auger spectral line shape were observed as a function of photon energy: the only measurable differences are a slightly higher low-kinetic-energy background in the off-resonance Auger spectrum. The question we address in this paper is whether or not the spin polarizations of the two Auger spectra are also similar. If the two 3d holes created in the Auger process are not correlated, then the polarization of the emitted Auger electron should not change as the polarization of the initial core hole is changed, e.g., by performing the Auger measurement on and off resonance.

We start with a discussion of the off-L_3-resonance spin-resolved FeAg(001) $L_3M_{45}M_{45}$ Auger-electron spectra taken using 820 eV photons, as shown in Fig. 1(a). The spectra are very similar to spin resolved Fe $L_3M_{45}M_{45}$ Auger spectra measured earlier by Landolt and co-workers[10] employing electron excitation and our own electron excited measurements. As expected, Fig. 1(a) shows that the majority-spin ($I_\uparrow$) Fe $L_3M_{45}M_{45}$ spectrum has larger spectral weight than the minority-spin spectrum ($I_\downarrow$). The majority-spin spectrum is somewhat broader than the minority-spin spectrum and is centered at ~0.5 eV lower kinetic energy. The maximum measured spin polarization $P=[I_\uparrow-I_\downarrow)/(I_\uparrow+I_\downarrow)]$ of 0.27 occurs at ~2 eV lower kinetic energy than the intensity maximum, in agreement with previous data. Although this maximum polarization has been compared to the spin polarization of the Fe valence band (P_{vb}=0.27), a more appropriate quantity for comparison is the total (energy-integrated) Auger polarization, which may be different from the maximum value since the polarization varies across the Auger spectrum. Energy integration also increases the statistical accuracy of the resultant spin polarization, eliminates the dependence on energy resolution, and is relatively insensitive (~±0.01) to the small low-kinetic-energy background. Therefore, all the Auger-spin polarizations reported here are electron-energy-integrated values. The energy-integrated spin polarization of the off-resonance Fe $L_3M_{45}M_{45}$ Auger spectrum, P_{off}=0.20, is considerably smaller than P_{vb}(=0.27). This difference is significant and cannot be accounted for by a small contribution from the $L_2L_3M_{45}$ super Coster-Kronig (SCK) Auger process.[28]

In Fig. 1(b) we show spin-resolved Fe $L_3L_{45}M_{45}$ Auger-electron spectra of Fe/Ag(001) taken on-L_3-resonance at 707 eV photon energy. By simple inspection, these spectra are very similar to the off-resonance spectra taken at 820 eV photon energy [Fig. 1(a)]. Indeed, when the majority-spin and minority-spin spectra on and off resonance are scaled to the same height, they overlap within experimental uncertainty except for the small shoulder on the high-kinetic-energy side of the on-resonance spectra (E_{kin}~701 eV) which is ascribed to direct Fe valence-band photoemission.[27]

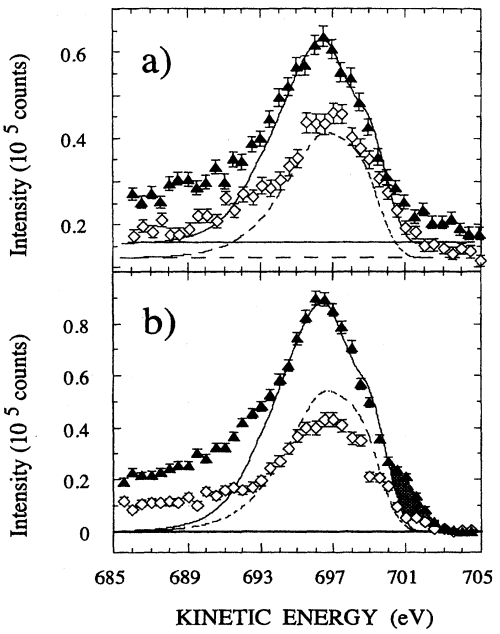

FIG. 1. (a) Majority-spin (▲) and minority-spin (◇) off-resonance Fe $L_3M_{45}M_{45}$ Auger-electron spectra ($h\nu$=820 eV) from an Fe/Ag(001) film, and the convolution of majority-spin (—) and minority spin (---) Fe DOS[32] with total Fe DOS (all DOS are broadened with a 1.5 eV Gaussian). The convolutions are scaled so that the majority-spin convoluted DOS matches the majority-spin Auger spectrum at ~696.5 eV (b) as in (a) except using on-resonant excitation ($h\nu$=707 eV). The shaded region represents the direct Fe valence-band photoemission.

We note that the spin polarization of this shoulder (~0.60) is substantially greater than that of the angle-integrated Fe valence-band photoemission spectra (~0.20).[29] This suggests that the on-L_3-resonance valence-band spectrum exhibits resonant behavior, which is not evident from spin-integrated data. Future work will examine this behavior with higher energy resolution.

The energy integration of on-resonance Fe $L_3M_{45}M_{45}$ spectra yields Auger-spin polarization (P_{on}=0.37), which is considerably higher than both the off-resonance value (P_{off}=0.20) and the Fe valence-band spin polarization (P_{vb}=0.27). In the remainder of this paper, we concentrate on the interpretation and ramifications of these photon-energy-dependent Auger-spin polarization differences.

The Fe $L_3M_{45}M_{45}$ Auger transition is thought to be band-like ($U_{eff}<2W$), so the majority-spin $[I_\uparrow(E)]$ and minority-spin $[I_\downarrow(E)]$ spectra should be well represented by a convolution of the appropriate spin-resolved DOS:[7,18,30]

$$I_\uparrow(E)=M_{\uparrow\uparrow}\int D_\uparrow(E-\varepsilon)D_\uparrow(\varepsilon)d\varepsilon + M_{\uparrow\downarrow}\int D_\uparrow(E-\varepsilon)D_\downarrow(\varepsilon)d\varepsilon,$$

$$I_\downarrow(E)=M_{\downarrow\downarrow}\int D_\downarrow(E-\varepsilon)D_\downarrow(\varepsilon)d\varepsilon + M_{\downarrow\uparrow}\int D_\downarrow(E-\varepsilon)D_\uparrow(\varepsilon)d\varepsilon, \quad (1)$$

where $D_\uparrow$ ($D_\downarrow$) is the majority-spin (minority-spin) DOS and $M_{\uparrow\uparrow}$ ($=M_{\downarrow\downarrow}$) and $M_{\uparrow\downarrow}$ ($=M_{\downarrow\uparrow}$) are the Auger matrix elements for spins of the two-hole Auger final state being parallel and antiparallel, respectively. $D_\uparrow(E-\varepsilon)$ and $D_\downarrow(E-\varepsilon)$ refer to the emitted Auger electron (whose spin and energy are measured) while $D_\uparrow(\varepsilon)$ and $D_\downarrow(\varepsilon)$ refer to the electron that drops into the initial core hole. Summation over the majority-spin $[D_\uparrow(\varepsilon)]$ and minority-spin $[D_\downarrow(\varepsilon)]$ core-hole DOS is necessary when the core hole is unpolarized, which is a good approximation for the case of *regular*, electron excited or off-resonant Auger spectra.

In the case that the initial core hole is created with nonzero spin polarization ($P_{\text{core}}\neq 0$), the appropriate modification of (1) involves multiplying $D_\uparrow(\varepsilon)$ and $D_\downarrow(\varepsilon)$ by the factors $(1+P_{\text{core}})/2$ and $(1-P_{\text{core}})/2$, respectively. Owing to the strong polarization of the unoccupied Fe DOS, the on-resonant Fe $2d\rightarrow 3d^*$ excitation is expected to create mostly minority-spin core holes. In the limit of $P_{\text{core}}=-1$, it follows from (1) that the spin resolved on-resonant Auger spectra are

$$I_\uparrow(E)=M_{\downarrow\uparrow}\int D_\uparrow(E-\varepsilon)D_\downarrow(\varepsilon)d\varepsilon,$$
$$I_\downarrow(E)=M_{\downarrow\downarrow}\int D_\downarrow(E-\varepsilon)D_\downarrow(\varepsilon)d\varepsilon. \tag{2}$$

Comparison of (1) and (2) indicates that the on-resonant and off-resonant Auger spectra should exhibit slightly different line shape, since the convolution is performed with minority-spin DOS ($D_\downarrow$) or both majority- and minority-spin DOS ($D_\uparrow$ and, $D_\downarrow$), respectively. However, these differences are very small and not detected in our measurement.[31] The results of (1) using calculated spin-resolved Fe DOS (Refs. 32 and 33) are shown in Fig. 1 and the obtained line shapes are in good agreement with both on- and off-resonance measured spectra.

The energy-integrated Auger-spin polarization (P_{AES}) off-resonance and on-resonance [from (1) and (2)], for the case of spin-independent Auger matrix elements ($M_{\downarrow\downarrow}=M_{\downarrow\uparrow}$) is

$$P_{\text{AES}}=\frac{\int I_\uparrow(E)dE-\int I_\downarrow(E)dE}{\int I_\uparrow(E)dE+\int I_\downarrow(E)dE}=\frac{\int D_\uparrow(E)dE-\int D_\downarrow(E)dE}{\int D_\uparrow(E)dE+\int D_\downarrow(E)dE}, \tag{3}$$

which is equal to the spin polarization of the valence band (P_{vb}). In this special case, the Auger spin polarization is given by P_{vb} and does not dependent on the polarization of the initial core hole. Our observation, $P^{\text{on}}_{\text{AES}}\neq P_{\text{vb}}\neq P^{\text{off}}_{\text{AES}}$, suggests that this simple model is not valid.

The polarization of the Auger transition (P_{AES}) can be shown to depend on the polarization of the initial core hole (P_{core}) if the two Auger matrix elements are *not* equal ($M_{\uparrow\uparrow}\neq M_{\uparrow\downarrow}$). For example, in the limit $M_{\uparrow\uparrow}=0\neq M_{\uparrow\downarrow}$ it follows from (1) that the off-resonant ($P_{\text{core}}=0$) polarization is $P^{\text{off}}_{\text{AES}}=0$ whereas, from (2), the on-resonant ($P_{\text{core}}=-1$) polarization $P^{\text{on}}_{\text{AES}}=1$. To illustrate this dependence we show P_{AES} vs the $M_{\uparrow\uparrow}/M_{\uparrow\downarrow}$ ratio for selected values of P_{core} between 0 and -1 in Fig. 2. The actual value of P_{core} in the resonance process is given by the spin polarization of the unoccupied DOS, which in the case of Fe yields $P_{\text{core}}\sim -0.75$.[32,33] On the other hand, P_{core} in the off-resonant case

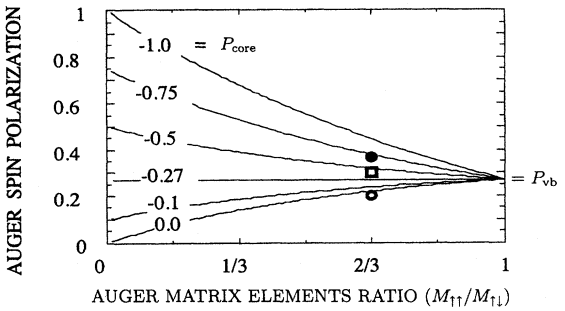

FIG. 2. Spin polarization of appropriately convoluted Fe DOS[32] (see text) as a function of Auger matrix elements for different selected values of the initial core-hole spin polarization: -1.0 -0.75, -0.5, -0.27, -0.1, and 0. The measured energy-integrated spin polarization on-resonance $L_3M_{45}M_{45}$ ($h\nu$=707 eV; ●), off-resonance $L_3M_{45}M_{45}$ ($h\nu$=820 eV; ○), and off-resonance $M_{23}M_{45}M_{45}$ ($h\nu$=260 eV; □) Fe Auger spectra.

can be determined by measuring the polarization of the photoemitted core-level electron. Our measurements of the Fe $2p$ core level yield $P_{\text{core}}\sim -0.08$, similar to recently published data.[26] Thus, the off-resonant Fe L_3 core holes are created with a relatively small degree of polarization.

We now compare our measured Fe $L_3M_{45}M_{45}$ spin polarization on resonance ($P^{\text{on}}_{\text{AES}}=0.37$) and off-resonance ($P^{\text{off}}_{\text{AES}}=0.20$) with the results of the band model described above (see Fig. 2). Indeed, this simple model seems capable of predicting the general trend observed by the experiment: P_{AES} is enhanced on resonance and reduced off resonance compared to P_{vb}, provided that the spin-parallel Auger matrix element ($M_{\uparrow\uparrow}$) is smaller than the spin-antiparallel one ($M_{\uparrow\downarrow}$). The best agreement between the experimental values and the model is found for $M_{\uparrow\uparrow}/M_{\uparrow\downarrow}\sim\frac{2}{3}$.

Since the Fe $L_3M_{45}M_{45}$ and $M_{23}M_{45}M_{45}$ Auger process leads to similar final states, it is of interest to see if spin-resolved Fe $M_{23}M_{45}M_{45}$ data are consistent with the model presented above. Our off-resonant ($h\nu$=260 eV) measurements show that the spin-resolved Fe $M_{23}M_{45}M_{45}$ Auger line shape is very similar to that of Fe $L_3M_{45}M_{45}$, but the energy-integrated Fe $M_{23}M_{45}M_{45}$ polarization is larger ($P^{\text{off}}_{\text{AES}}=0.30$) than that of the off-resonant Fe $L_3M_{45}M_{45}$ transition.[34] This increase is nicely accounted for by the large $3p$ core-hole spin polarization: we measure it to be $P_{\text{core}}=-0.23$, which matches the trend predicted by the band model (see Fig. 2).[35]

The analysis of on- and off-resonant Auger spectra presented here have shown that the physical basis for the differences between the spin polarization of Fe CVV Auger transitions and that of the Fe valence electrons ($P^{\text{on}}_{\text{AES}}>P_{\text{vb}}>P^{\text{off}}_{\text{AES}}$) is the smaller transition probability of creating the spin-parallel two-hole Auger final state ($M_{\uparrow\uparrow}$) than the spin-antiparallel one ($M_{\uparrow\downarrow}$). We suggest this to be a general trend, a consequence of exchange correlation. The Pauli exclusion principle forces two electrons of alike spins to stay further apart than ones with antiparallel spins, which is sometimes thought of as a Fermi hole (sphere): an electron surrounded by a sphere from which other electrons of like spin are excluded. Since the Auger transition is a local process driven

by Coulomb interaction between two electrons, it is expected that the existence of a Fermi hole will reduce the probability of forming the spin-parallel two-hole Auger final state. This description provides an intuitive interpretation of our data in terms of spin-dependent spatial correlations, or exchange correlation. While such effects are visible even in spin-integrated Ni CVV Auger spectra (atomic multiplets),[12,14] the spin-resolved measurements reported in this paper show that they are also present in Fe where no atomiclike features are seen. Therefore, this work demonstrates how the addition of spin analysis to CVV Auger measurements represents a natural extension of previous spin-integrated studies[11–15] which will enable more qualitative analysis of correlation effects, especially in magnetic solids.

In conclusion, we have shown that although the spin-resolved Fe CVV Auger-electron spectra (SPAES) are well represented by convolution of the spin-resolved valence-band density of states, the exchange correlation between the two final state holes needs to be taken into account. That is, SPAES carries important information on both the spin polarization of the valence band and the relative strength of exchange correlation. More quantitative theoretical work is needed in order to use the SPAES technique for measurement of *element specific*, local valence-band spin polarization. On a qualitative level, the present work demonstrates the importance of exchange correlation in itinerant $3d$ metals like Fe.

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[35] Although the magnitude of the intensity-integrated Fe $3p$ spin polarization does depend on the background subtraction procedure used, these differences (± 0.05) do not alter the trend observed in our measurements. We also note that our experimental geometry (p-polarized light and magnetization) does not introduce magnetic linear dichroism effects, as have been observed in some Fe $3p$ photoemission measurements.