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f-electron correlations in nonmagnetic Ce studied by means of spin-resolved resonant photoemission

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We have studied the spin-spin coupling between two f electrons of nonmagnetic Ce by means of spin-resolved resonant photoemission using circularly polarized synchrotron radiation. The two f electrons participating in the 3d_{5/2}→4f resonance process are coupled in a singlet while the coupling is veiled in the 3d_{5/2}→4f process due to an additional Coster-Kronig decay channel. The identical singlet coupling is observed in the 4d→4f resonance process. Based on the Ce measurements, it is argued that spin-resolved resonant photoemission is one approach to study the correlation effects, particularly in the form of spin, in the rare earths.

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In photoemission investigations of nonmagnetic materials, the measurement of electron spin polarization, with respect to a suitably chosen quantization direction, can provide insight into the electronic structure of the systems under study, beyond that which can be gleaned from merely the energy and momentum relationship of the photoelectrons.1,2 For example, the spin-polarized photoemission experiment performed with circularly polarized light, in which spin polarization is aligned with photon propagation direction,3 is an essential method to characterize the symmetries of the states. These measurements permit the performance of the symmetry resolved band mapping of solids4-6 and the determination of all dipole matrix elements and phase-shift differences of continuum wave functions describing the photoelectron emission from free and absorbed atoms.6 The combination of a spin-orbit interaction and circularly polarized light induces an anisotropic distribution of the m_j in the final state wave function via the relativistic dipole selection rules. To a great extent, these measurements are extensions of the Fano effect, predicted7 and observed8 over 30 years ago.

The photoexcitation of core electrons by circularly polarized light in nonmagnetic materials will not only result in the spin polarization of the excited photoelectrons9 but also in spin-polarized core holes aligned with photon spin. These spin-polarized core holes can decay by emission of polarized photons or by spin dependent Auger processes.10-18 For example, consider the CVV (core valence valence) Auger process in a solid. The initial state is two valence electrons and the final state is two electron states in which one of them has filled the polarized hole and another one is ejected as an Auger electron. If the two valence electrons are coupled in a singlet, the spin of the outgoing electron has to be antiparallel to the spin of the primary core hole, due to the selection rules governing the Auger process ∆S=0.19 If the two valence electrons are coupled in a triplet, the spin of the outgoing electron has to be parallel to the spin of the primary core hole. Therefore, the spin analysis of the photoelectrons and the corresponding Auger electrons allows the study of the spin-spin coupling between the two valence electrons participating in the CVV decay process from the nonmagnetic materials.

In the above context, this paper reports on the spin-resolved measurements of the 3d→4f resonant photoemission in nonmagnetic Ce, as a means of studying electron correlations between f electrons. Furthermore, because Ce is the first element of the rare earths and the location of the delocalized/localized transition in the rare earths,20 the results from Ce may serve as a prototype for the other rare earths, in which strong correlation effects originating from highly localized f electrons may play a crucial role for the determination of the physical properties.21-24 Here, it will be shown that spin-resolved resonant photoemission spectroscopy, using circularly polarized light, can provide a direct means of investigating electron correlation in nonmagnetic materials. In this case, it will be shown that electron-electron coupling in the Auger-like decay process can be observed in a powerful manner.

The experiments were performed at the beamline 4-ID-C of the Advanced Photon Source, at Argonne National Laboratory.25 The beamline provides circularly polarized x-rays, with switchable helicities (s-polarized) on demand, with a degree of polarization >96% in the energy range of 500–3000 eV. The total resolution (beamline+analyzer) of 0.8 eV was chosen to allow a reasonable signal in the Mott detector, since the efficiency of the Mott detector (S^2/I_o) is of the order 10^{-5}. The experimental apparatus used in the present study has been previously described.26-28 Uncrystallized cerium samples were made by evaporation in situ on a W(110) substrate at room temperature.29 Our samples are close to the γ-Ce.20 The base pressures were in the low 10^{-11} torr range, rising to ~6×10^{-10} torr during evaporation. The cleanliness of the Ce samples was monitored by an O 1s signal. In order to make sure the samples are free from oxygen, fresh samples were made approximately every 6 h, and cleanliness was controlled before and after the measurements. No oxygen contaminations were indicated in the samples. Photon energies were calibrated using the 3d_{5/2}→4f transition in x-ray absorption spectroscopy (XAS), at an energy of 882.2 eV (Ref. 30.)

For our purposes the 3d→4f transition of Ce is an excellent choice because the spin-orbit split 3d_{5/2} and 3d_{3/2} levels are well separated by 18.5 eV in binding energy.21 Thus well
defined and oppositely spin-polarized core holes can potentially be generated by photoexcitation using circularly polarized light into the 4f final states. The 3d → 4f resonance arises when an interference occurs between the direct photoemission channel,

\[ 3d^{10}4f^4 + h\nu \rightarrow 3d^{10}4f^6 + \text{electron} \quad (1) \]

and an indirect channel in which there is the 3d → 4f excitation with subsequent CVV Auger-like decay,

\[ 3d^{10}4f^4 + h\nu \rightarrow 3d^64f^2 \rightarrow 3d^{10}4f^0 + \text{electron} \quad (2) \]

which has the identical initial and final state with the direct channel. Since the condition for the interference is coherence between the direct and the indirect channels, the loss of coherence can be due either to delocalization of the 4f electrons in the intermediate state, which produces an energy difference between the final states of the two channels or to a different time for the two channels.\(^{31,32}\) Therefore, the sharp resonance in the 3d → 4f transition is consistent with the strongly localized nature of 4f wave functions in rare earths.\(^{30}\)

A spin-resolved core level spectrum is shown in Fig. 1.

Here, the Ce 3d_{5/2} level at a binding energy of 883.8 eV is undergoing interrogation with photons of 1375 eV. With photons of 1375 eV, 3d direct photoelectrons have a kinetic energy of \(-500\) eV, which is good energy for an optimum operation of the Mott detector, and are well separated from the 3p direct photoemission peaks and MNN Auger peaks, and any peaks associated with 3p core holes. The 3d_{5/2} level will be one of those used in the resonant photoemission study. Measurements of its spin polarization is essential to establishing the initial polarizations of the core holes and testing the validity of our relativistic models. In order to eliminate the instrumental asymmetry, the electron spin polarization \(P_z\) aligned to the photon spin is determined from the raw spectra using the following formula:

\[ P_z = \frac{1}{SP_\sigma \cos 55^\circ} \left( \frac{\sqrt{N_1^2 N_3^2} - \sqrt{N_2^2 N_4^2}}{\sqrt{N_1^2 N_3^2} + \sqrt{N_2^2 N_4^2}} \right). \]

Here, \(N_1^\sigma(3)\) is the intensity with \(\sigma^s\) at the counters 1(3) located at equal polar angles 120° in the reaction plane defined by the incoming and scattered electron beam, \(P_\sigma\) is the light polarization, and \(\cos 55^\circ\) is due to the 55° off-normal incidence of light and the normal emission of electrons. The Sherman function \(S\), which describes the sensitivity of the Mott detector, is 0.12 (Ref. 26). It should be noted that the instrumental asymmetry \(A_Z\) defined in the following way:

\[ A_Z = \frac{\sqrt{N_1^2 N_3^2}}{\sqrt{N_2^2 N_4^2}}, \]

has to be monitored for every run to determine the electron spin polarization correctly.\(^{33} If A_Z\) differs from 1, this means that there is an instrumental asymmetry. The instrumental asymmetry can be eliminated by using Eq. (3) only if \(A_Z\) does not vary in time.\(^{33}\) Therefore, the monitoring of \(A_Z\) provides an important check on the performance of the Mott detector. If \(A_Z\) is not constant over runs, the spin polarization determined by using Eq. (3) cannot be claimed to be accurate. For example, the bottom panel of Fig. 1 shows that we have an instrumental asymmetry because \(A_Z\) differs from 1, and that the instrumental asymmetry can be eliminated by using Eq. (3) because \(A_Z\) is constant over runs. The middle panel of Fig. 1 shows the \(P_z\) measured by using Eq. (3).

In the top of Fig. 1, the spin-separated partial intensities \(I_+\) and \(I_-\) are connected with the measured spin polarization \(P_z\) and the measured total (spin-independent) intensity \(I\) by \(I_+ = I/2(1 \pm P_z)\). Correspondingly, \(I_+\) and \(I_-\) are the partial intensities polarized totally parallel and antiparallel to the photon spin, respectively. From the total intensity \(I\), a strong main peak A centered at a binding energy of 883.8 eV and a weak peak B centered at a binding energy of \(-878\) eV are visible. The physical origin of the two peaks can be understood in terms of the configuration interaction as follows. Because 4f states are localized in the 3d core region, they are extremely sensitive to the attractive Coulomb potential of the 3d hole.
Consequently, the creation of a $3d$ core hole by photoexcitation causes an additional empty $4f$ state to be pulled down below the Fermi energy. Thus, in a very simple picture, two final states are produced; when the empty $4f$ state is not yet occupied and the screening is produced by the $(5d6s)^3$ valence band, the result is a final state with a configuration of $3d^24f^1$ (peak $A$). If the empty $4f$ state is filled by an extra $f$ electron from the conduction band and it screens the core hole, this is another final state with a configuration of $3d^24f^2$ (peak $B$). The second final state with the configuration of $3d^24f^2$ has a much weaker intensity and smaller binding energy.\textsuperscript{21,34–37} From the spin separated partial intensities $I_y$ and $I_z$, it can be seen that $I_z$ is dominant over peak $A$, with an approximately +20% spin polarization. It is interesting to note that peak $B$ is also slightly positively polarized.

For convenience, the configuration interaction model has been used to explain the double $3d_{5/2}$ structures of Ce in this paper. But, it should be noted that there is another description for strongly correlated electron systems using a modified Anderson impurity model\textsuperscript{38–40} which describes the features of the valence band spectra and of the core-level spectra as well.

We use the three-step model of photoemission in the interpretation of the spin-resolved spectra. It should be noted that even though the three-step model explains the experimental data for the bulk photoemission surprisingly well, there is a conceptual deficiency. The disadvantages of the three-step model are overcome by the one-step model of photoemission, in which the photoemission process is considered as a one-step quantum mechanical event.\textsuperscript{41–43} However, in our case, the dominant effect is the spin-dependent optical pumping process induced by the circularly polarized x-rays, which can be dealt with most easily in the first step of the three-step model. The three-step model consists of (i) the primary excitation process in the bulk by absorption of the incident photon, (ii) the transport of the excited electron to the surface which includes the possibility for inelastic scattering by the electron-electron and the electron-phonon, and (iii) the escape of the electron through the surface into the vacuum.\textsuperscript{44} In principle, every one of these three steps may result in a nonzero contribution to the net spin polarization. For centrosymmetric nonmagnetic materials with no preferential population of one spin state in the ground state, however, it is demonstrated that the first step, the primary excitation process in the bulk by absorption of the incident photon, is mainly responsible for the non-zero spin polarization if the exciting light is circularly polarized.\textsuperscript{2} The second step, transport to the surface, produces no net spin polarization in nonmagnetic materials.\textsuperscript{2} In the third step, off-normally emitted photoelectrons may require a nonzero contribution to the net spin polarization due to the spin-dependent diffraction of the photoelectrons at the surface.\textsuperscript{45–47} However, this contribution to the net spin polarization vanishes for the symmetry reasons for normal emission to the surface.\textsuperscript{45} Therefore, nonmagnetic materials and normal emission of electrons ensure that the second and the third step of the three-step model of photoemission are spin-independent, and the spin polarization is dominated by the primary excitation known as optical spin orientation.\textsuperscript{1,2}
that of 3d/H20849izations of the final states. Using Eq. be calculated using the following expression:

\[ P \approx \frac{\sum_{i=J}^{J} a_i \delta_i}{\sum_{i=J}^{J} a_i} \]  

Here, \( a_i \) are the transition probabilities and \( \delta_i \) the spin polarizations of the final states. Using Eq. (5) and Fig. 2, the transitions \( d_{5/2} \rightarrow f_{7/2} + f_{5/2} \) and \( d_{3/2} \rightarrow f_{5/2} \) result in the spin polarizations of +60% and −50%, respectively. It should be noted that the \( d_{5/2} \rightarrow f_{5/2} \) transition gives a negative spin polarization, but it has 20 times weaker transition probability than that of the \( d_{5/2} \rightarrow f_{7/2} \). Therefore, \( d_{5/2} \rightarrow f (f_{7/2} + f_{5/2}) \) transition gives a net positive spin polarization of photoelectrons.

We notice that there is a poor agreement between the measured spin polarization of +20% and the calculated one of +60% for \( d_{5/2} \rightarrow f (f_{7/2} + f_{5/2}) \) transition. For this discrepancy, we should note the following two points. Firstly, the measured partial intensities \( I_s \) and \( I_0 \) include the unpolarized background. A proper subtraction of the background from the partial intensities \( I_s \) and \( I_0 \) results in a spin polarization of approximately +44% in the peak A which is much closer to +60%. Secondly, the atomic model used in Fig. 2 oversimplifies the real system. For example, the spin polarization of \( 3d_{4/2} \rightarrow 4f \) transition depends on the photon energy through the radial parts of the matrix elements,7,9 but the dependency is not included in the atomic model. In any case, even though the atomic model given in Fig. 2 describes the system under study qualitatively only, it is extremely useful because it presents a simple picture how photoelectrons are spin polarized without any complicated calculations.

Another aspect of the spin-resolved direct photoemission of core levels is that it provides an important illustration of the generation of a spin polarized core hole, which is the
the main message of this paper. As shown in Fig. 4, the 3d_{5/2} \rightarrow 4f_0 transition with \sigma^+ gives rise to an anisotropic excitation in the m_j of 3d_{5/2}, generating a net +47% spin-polarized core hole in the 3d_{5/2} state. Here, Eq. (5) has been used by substituting s_i with the spin polarizations of m_j of a 3d_{5/2} state. The meaning of spin-polarized core hole is that the sum of the m_j of 3d_{5/2} core holes is not equal to zero. 10 In the same way, the 3d_{3/2} \rightarrow 4f_1 transitions create a −30% spin polarized core hole in the 3d_{3/2} state. These spin-polarized core holes will decay by the spin-dependent CVV Auger processes, contributing to the resonant photoemission through the indirect channel.

Now, let us consider the resonant photoemission itself. Figures 3(b) and 3(c) present spin-resolved resonant photoemission measured at the 3d_{5/2} \rightarrow 4f_0 transition at h\nu = 882.2 eV and the 3d_{3/2} \rightarrow 4f_1 transition at h\nu = 899.4 eV. At the 3d_{5/2} \rightarrow 4f_0 resonance, the partial intensity I_0 is dominant over the two peaks labeled as f^0 and f^1 21,48 with a monotonic −20% spin polarization. This observation and the impact of the negative spin polarization at the 3d_{5/2} \rightarrow 4f_0 resonance is the main message of this paper. As shown in Fig. 4(a), we propose a simplified physical explanation for the measured spin polarization. The positively spin-polarized core holes in the 3d_{5/2} state decay obey the selection rule for the CVV Auger-like process; the two valence f electrons coupled in a singlet participate in the decay process, result in a −20% spin polarization. In principal, the spin polarization from the direct photoemission and the indirect CVV part can both contribute to the spin polarization measured at resonance. As shown below, however, the contribution from the direct photoemission can be suppressed because uncrystallized Ce samples were used instead of a single crystal in the measurements. In photoemission from nonmagentic valence bands, a significant nonzero spin polarization can be expected only at points and along lines of high symmetry in the Brillouin zone because there are spatially degenerated bands and spin-orbit interaction splits the bands. These bands modified by the spin-orbit interaction and circularly polarized light are the necessary conditions for nonzero spin polarization. However, at the general points of the Brillouin zone, where there are no spatially degenerated bands to be modified by spin-orbit interaction, photoelectrons are unpolarized giving equal amounts of “up” and “down” spins. 1,49 An example is the spin-resolved valence band photoemission from Ge; electrons emitted from a Ge(001) single crystal using circularly polarized light are highly spin polarized while electrons emitted from amorphous Ge film at the same photon energy are unpolarized. 1 Since we used uncrystallized Ce samples, which have no well defined points and lines of symmetry, we expect that there is no contribution in the measured spin polarization from the direct photoemission. Therefore, we conclude that the −20% spin polarization measured in the resonant spectra results from correlations in the spins between the polarized core holes and the outgoing Auger electrons, mediated by two valence f electrons coupled in a singlet.

The appearance of the two peaks f_0 and f_1 is characteristic of the highly localized nature of 4f electrons assumed to be similarly localized to core electrons. A hole created in the 4f state by photoemission acts as an additional attractive potential for the other electrons in the system, generating complicated final states as f_0 and f_1 (Ref. 21). The monotonic spin polarization measured over the two peaks f_0 and f_1 indicates that the f_0 and f_1 states are resonated with the identical spin structure. It should be noted that the two peaks f^0 and f^1 are also assigned as the lower Hubbard band and the quasiparticle peak, respectively. 40,50-52

In the same vein, a positive spin polarization is expected at the 3d_{3/2} \rightarrow 4f_1 resonance because of a negatively spin-polarized core hole in the 3d_{3/2} state. As shown in Fig. 3(c), however, no spin polarization is observed at the 3d_{3/2} \rightarrow 4f_0 resonance, within the statistics. In addition, a direct comparison of the total intensities of the 3d_{3/2} \rightarrow 4f_0 and the 3d_{3/2} \rightarrow 4f_1 transitions at the position of f_0, indicates that there is an approximately 6.5 times reduction in the intensity at the 3d_{3/2} \rightarrow 4f_0 resonance relative to that at the 3d_{3/2} \rightarrow 4f_1 resonance. It appears that the CVV indirect channel of the 3d_{3/2} core hole has been quenched by a competitive decay channel, a Coster-Kronig (CK) transition, where the spin-polarized core hole in the 3d_{3/2} state is filled by electrons from the 3d_{3/2} state, resulting in spin-polarized holes in 3d_{3/2} and the production of off-resonance electrons with lower kinetic energy. Since the CK transition commonly occurs with 1–3 orders of magnitude greater probability than the usual Auger decay and degrades the spin polarization of the polarized holes in the 3d_{3/2} (Refs. 10 and 53), the consequence is the substantial decrease of the intensity and the spin polarization at the 3d_{3/2} \rightarrow 4f_1 resonance, as shown in Fig. 4(b).

The comparison between the −20% spin polarization measured at the 3d_{3/2} \rightarrow 4f_0 resonance and the −47% spin polarization of the 3d_{3/2} core holes calculated using the atomic model shows clearly a considerable discrepancy, although we understand that the atomic model explains the measured spin polarization qualitatively only. Even though the discrepancy does not prevent us from understanding the spin-spin coupling basically, some possible reasons for the discrepancy are addressed in the following.

(i) Triplet coupled two electrons may participate in the resonant process. Although the singlet coupling is favored, triplet coupling may have some influence.

(ii) A recent publication on Ce compounds 54 indicated that there are incoherent Auger transitions between different multiplet states of a 3d^{8}4f^{2} configuration in the resonant process. Assuming the incoherent Auger electrons are unpolarized, they are overlapped with the main resonant signal as an unpolarized background, thereby lowering the spin polarization. 55

(iii) Because the matrix element of the Auger process is described by the integration of the initial state (two 4f states

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lomb and exchange interaction between 4d and 4f\(^{20,32}\) are observed in the x-ray absorption spectroscopy as shown in Fig. 5(a). Figures 5(b) and 5(c) illustrate spin-resolved resonant photoemission spectra measured at h\(\nu\)=127 eV along the Z direction aligned with the photon propagation direction and along the X-direction perpendicular to that, respectively.

It is clear that because the incoming circularly polarized light creates spin-polarized core holes aligned with the photon propagation direction, there is no physical driving force for nonzero spin polarization along the X direction. As shown in Fig. 5(c), \(P_X\) is equal to zero within the statistic error bars. The physical origin of the measured spin polarization \(P_Z\) at the 4d\(\rightarrow\)4f resonance is also the spin-polarized core hole at the 4d\(_{5/2}\) created by circularly polarized light. The spin-orbit coupling of 4d\(_{5/2}\) and 4d\(_{3/2}\) is small. However, the core holes generated at the 4d\(_{3/2}\) edge decay via the CK transition into the 4d\(_{5/2}\) core holes. Therefore, the 4d\(_{3/2}\) core holes do not influence the spin polarization measured at the 4d\(\rightarrow\)4f resonance.\(^{10}\) As shown in Fig. 5(b), spin polarization \(P_Z\) measured over the 4d\(\rightarrow\)4f resonance is approximately \(-43\%\), which agrees almost quantitatively with the \(-47\%\) predicted by the atomic model. Thus an “overlap” scenario may explain why the spin polarization measured at the 3d\(\rightarrow\)4f resonance is low. A detailed analysis of spin-resolved resonance spectra at the 4d\(\rightarrow\)4f resonance will be covered in a future work.

More evidently, high resolution photoemission experiments demonstrated that while the 3d\(\rightarrow\)4f resonance probes the bulk 4f states, the 4d\(\rightarrow\)4f resonance probes mainly the surface 4f states which are much more localized and atomic-like than the bulk 4f states.\(^{57,58}\) Due to the strongly localized (atomic) character of the surface 4f states, the 4d\(\rightarrow\)4f resonance may produce a higher spin polarization than that of the 3d\(\rightarrow\)4f resonance.

In conclusion, spin-dependent resonant photoemission using circularly polarized light has been applied to explore the spin-spin coupling between two 4f electrons of strongly correlated nonmagnetic Ce. Positively and negatively spin polarized core holes are created at spin-orbit split 3d\(_{3/2}\) and 3d\(_{5/2}\) states, respectively, by direct photoemission using circularly polarized light. At the 3d\(\rightarrow\)4f resonance, the measured monotonic negative spin polarization reveals a dominant singlet coupling between two 4f electrons participating in the resonant process. Because the Auger process is driven by a Coulomb interaction, the singlet coupling is favored because it allows the two electrons to be in greater proximity than the triplet coupling. At the 3d\(\rightarrow\)4f resonance the spin polarization is quenched via an additional CK process. The negative spin polarization measured at the 4d\(\rightarrow\)4f resonance also supports the singlet coupling. Based on the Ce results, we would like to suggest that spin-dependent resonant photoemission may provide an excellent way to probe the f-electron correlations in rare earth. Finally, from the experimental point of view, theoretical calculations for spin-resolved resonant photoemission are highly desirable to understand the electron correlations quantitatively.

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FIG. 5. (Color) (a) 4d\(\rightarrow\)4f x-ray absorption spectroscopy (XAS) of Ce. The arrow shows the photon energy used for the spin-resolved resonant photoemission measurement given in (b) and (c). (b) Spin-resolved resonant photoemission spectra measured at 4d\(\rightarrow\)4f resonance along the Z-direction (parallel to the photon propagation direction) at h\(\nu\)=127 eV. I is the spin integrated total intensity, and \(I_a\) (red) and \(I_c\) (blue) are the two spin separated partial intensities. The middle and the bottom panels of (b) represent the electron spin polarization \(P_X\) determined by Eq. (3) and instrumental asymmetries \(A_Z\) determined by Eq. (4) for the two independent runs, respectively. (c) Spin-resolved resonant photoemission spectra measured at 4d\(\rightarrow\)4f resonance along the X-direction (perpendicular to the photon propagation direction) at h\(\nu\)=127 eV. The middle and the bottom panels of (c) represent electron spin polarization \(P_X\) determined by Eq. (3) and instrumental asymmetries \(A_Z\) determined by Eq. (4) for the two independent runs, respectively.
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