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Spin Resolved Auger-Spectroscopy on Rb-Layers after Core-Hole Creation by Circularly Polarized VUV-Light

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Abstract

Circularly polarized light from BESSY with energies from 15 eV to 20 eV in normal incidence is used to excite oriented $4p_{3/2}$-hole states in thick layers of Rb on Pt(111). The *CVV*-Auger decay of these hole states is studied in a normal-emission set-up. The preferential spin direction of the Auger-electrons has been measured to be parallel to the spin of the exciting photons near the excitation threshold and changes to be antiparallel at somewhat higher photon energies. This behaviour is consistent with an excitation varying with the photon energy and an atom-like model for the decay of the primary hole state. Thereby the two valence electrons participating in the decay are assumed to be *s*-like and to be coupled to a singlet state.

As the Auger-decay of the excited hole-state does not depend on the excitation, spin dependent Auger-spectroscopy opens a new method of studying excitations and unoccupied electronic states.

1. Introduction

In this paper we report on first experimental results of spin dependent Auger-emission from non-magnetic solids. In Auger-spectroscopy usually the energetic position and the shape of Auger-lines in spin independent intensity spectra are studied [1]. Spin dependent effects have been investigated experimentally only with ferromagnetic materials [2, 3].

The theory of Auger-emission from solids involves spin coupling and spin interactions [4–6], but in view of the existing experiments, up to now only the evaluation of the position and the shapes of Auger lines in spin averaged intensity spectra have been of interest.

The theory of Auger-processes in atoms predicts a transfer of the orientation of an inner shell hole to the outgoing Auger-electron showing up in a non-vanishing electron spin-polarization [7–9]. We have studied this transfer of orientation in solids at thick layers of Rb. Thereby we have used the possibility to create oriented core holes in the outermost *p*-shell by circularly polarized synchrotron radiation [7, 8, 10, 11].

2. Experiment

The measurements were performed at the 6.5 m normal incidence monochromator [12] for circularly polarized off-plane radiation at BESSY in Berlin using an apparatus described previously [13, 14]. The degree of circular polarization was $(90 \pm 3)\%$, the band width of the radiation 0.5 nm. For the used photon energies $h\nu \leqslant 20\,\mathrm{eV}$ this band width results in an energy width $\Delta E_{h\nu} < 0.16\,\mathrm{eV}$. The calibration uncertainty of the monochromator is about 0.1 eV.

All data were recorded for normal incidence of the radiation. Electrons emitted into an acceptance cone with half angle 5° around the surface normal were collected by a simulated 180° spherical-field energy-analyser [15] followed by a UHV Mott-polarimeter for electron spin-polarization. The energy analyser was operated with fixed energy resolution $\Delta E = 0.5\,\mathrm{eV}$. Apparatus asymmetries of the Mott-polarimeter were avoided by changing the helicity of the radiation.

The Rb-layers were evaporated onto a clean Pt(111)-crystal using dispensers [16]. During the evaporation and during the measurements the Pt-substrate was cooled using liquid helium to about 90 °K. The appropriate thickness of the layers was controlled by photoemission and standard Auger-spectroscopy such that all Pt-signals disappeared. LEED from the prepared layers do not reveal structured patterns. The pressure during the measurements was below 1×10^{-10} mbar. To avoid interference caused by reactions of the layers with the residual gas, fresh layers were prepared after heat cleaning the substrate approximately every two hours.

3. Results and discussion

In Fig. 1 spin averaged intensity spectra measured in the normal-incidence/normal-emission set-up at a thick Rb-layer are given. The spectra clearly point out the $N_3\,VV$-Auger-peak at about a kinetic energy $E_{\mathrm{kin}} = 11.7\,\mathrm{eV}$ and the attached plasmon loss-peak. Although the threshold for creation of a $4p_{3/2}$- and a $4p_{1/2}$-hole is $h\nu = 14.9\,\mathrm{eV}$ and $h\nu = 15.8\,\mathrm{eV}$, respectively [17–19], there is no detectable intensity of a $N_2\,VV$-Auger-peak in going from $h\nu = 15.5\,\mathrm{eV}$ to photon energies above the $4p_{1/2}$-threshold. This is due to the most probable decay of the $4p_{1/2}$-hole via a Koster–Kronig-transition resulting in a $4p_{3/2}$-hole and a conduction-band electron lifted above the Fermi-level [18, 20].

A spin-dependent measurement across the Auger-peak is shown in Fig. 2. It is performed using circularly polarized radiation with an energy $h\nu = 15.3\,\mathrm{eV}$ well below the $4p_{1/2}$-threshold to suppress the influence of the $4p_{1/2}$-level. As is usual in spin-dependent photoemission from solids, the partial intensities I_+ and I_- for emitted electrons with spin parallel and antiparallel to the spin of the exciting photons, respectively, are displayed together with the total intensity $I = I_+ + I_-$. I_+ and I_- are connected with the spin polarization P of the electrons by $P = (I_+ - I_-)/(I_+ + I_-)$.

A strong preference of the partial intensity I_+ is obvious in Fig. 2, which means emission of Auger-electrons with spin parallel to the spin of the exciting photons is preferred. If an unpolarized secondary electron background is considered to underlie the Auger peak as indicated in Fig. 2, the electron

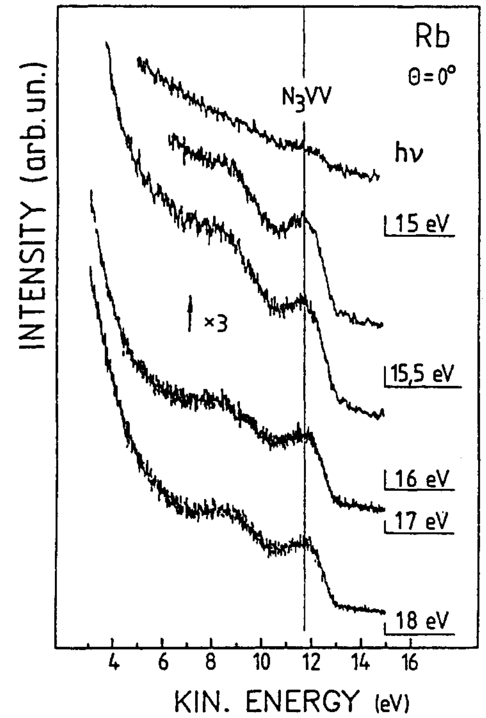

Fig. 1. Intensity spectra $I(E_{kin})$ of electrons emitted from a thick Rb-layer on Pt(111) irradiated with photons of varied energy $h\nu$ in a normal-incidence/normal-emission set-up. The N_3VV-Auger peak is marked by the vertical line

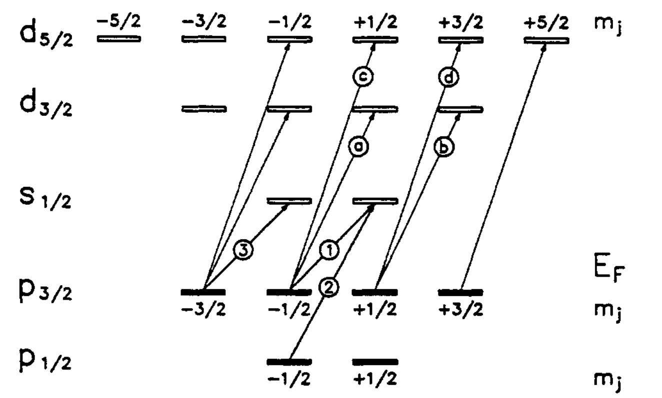

Fig. 3. Scheme of the excitation $4p^6 + h\nu \rightarrow 4p^5 5s^1$, $4p^5 5d^1$ for σ^+-polarized radiation. Numbers in the circles at the transition arrows to the s-states give the relative transition probabilities normalized at the $|p_{3/2}, m_j = -1/2\rangle \rightarrow |s, m_j = +1/2\rangle$ transition assuming identical radial parts of the $p_{1/2}$- and $p_{3/2}$-wave functions [23]. Lower case letters in the circles at transition arrows to d-states mark the following transition probabilities normalized to transition (c): (a) 8/27, (b) 2/9, (c) 1, (d) 2. Thereby identical radial parts of the $d_{3/2}$- and $d_{5/2}$-wave functions are assumed. Energy differences are not to scale

spin polarization averaged across the Auger-peak will be $P(h\nu = 15.3\,\text{eV}) = (24 \pm 3)\%$. To understand this result we use a three-step model of the CVV-Auger process with the first step being the excitation of a primary hole, the second step being the decay of the primary hole by creating two holes in the occupied region of the conduction band and an outgoing Auger-electron above the vacuum level [1], and the third step being the transport to the surface and the transmission into the vacuum. The third step does not change the spin state of the emitted electrons as we are using a normal emission set-up. The two first steps we treat within an atom-like model, since in both steps core states favouring local interactions are involved. A scheme of the excitation is displayed in Fig. 3, a scheme of the decay in Fig. 4. In these schemes all p- and d-states are assumed to be spin-orbit coupled.

The excitation process is determined by the relativistic dipole-selection rules for σ^+ (σ^-)-light: $\Delta l = \pm 1$, $\Delta m_j = +1$ ($\Delta m_j = -1$). Therefore, starting at the $4p_{3/2}$-level only excitations into s- and d-like continuum states (unoccupied conduction-band states) are possible. With bulk Rb, the d-like density of states is small compared to the s-like density of states just above the Fermi-level [21]. Therefore transitions into s-like states will dominate, if σ^+-polarized radiation with energy just above the $4p_{3/2}$-threshold is used, although transition probabilities from $p_{3/2}$-states to d-like states typically are greater by a factor of 2 to 6 as the one for transitions into s-like states [10, 22]. As a result the hole states $|4p_{3/2}, m_j = -3/2\rangle$, $|4p_{3/2}, m_j = -1/2\rangle$ will be excited preferentially. Their relative population will be 3 and 1 [23]. If at somewhat higher photon energies also $4p_{1/2}$-hole states are excited, their relative population would be 2, provided the radial parts of the $p_{3/2}$- and $p_{1/2}$-wave functions are identical.

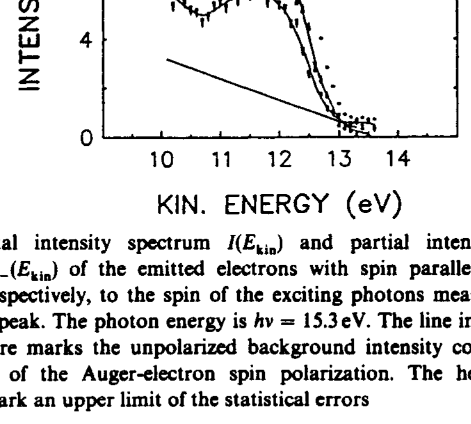

Fig. 2. Total intensity spectrum $I(E_{kin})$ and partial intensity spectra $I_+(E_{kin})$, $I_-(E_{kin})$ of the emitted electrons with spin parallel, spin antiparallel, respectively, to the spin of the exciting photons measured across the Auger peak. The photon energy is $h\nu = 15.3\,\text{eV}$. The line in the bottom of the figure marks the unpolarized background intensity considered for evaluation of the Auger-electron spin polarization. The height of the symbols mark an upper limit of the statistical errors

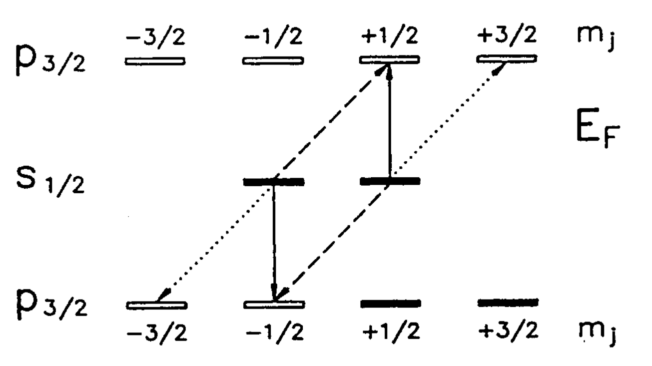

Fig. 4. Decay scheme of $p_{3/2}$-holes. Hole states existing before the decay process are given by open bars, electron states occupied before the decay by filled bars. The different line types at the transition arrows mark the coupling

The core hole in the "simple" metal Rb will be screened by conduction band electrons resulting in a strongly enhanced density of occupied s-like states at the lower edge of the conduction band [24]. Thereby a $(4p^5\,5s^2)$ electron configuration similar to that in an Sr-ion exists locally. This configuration is used in the decay schema shown in Fig. 4. One of the two s-like electrons will fill up the $4p_{3/2}$-hole, the other will leave the system, which is the "Auger-electron". As with the Auger-decay, the involved electrons are coupled by Coulomb-interaction, the parity and the angular momentum J as well as its projection M_J onto the preferential direction must be conserved [1, 7–9]. Therefore with the final two-hole state consisting of the two s-like states, coupled to a singlet state, a hole state $|p_{3/2}, m_j = -3/2\rangle$ will result in an occupied outgoing electron state $|p_{3/2}, m_j = +3/2\rangle$, a hole state $|p_{3/2}, m_j = -1/2\rangle$ in an occupied outgoing electron state $|p_{3/2}, m_j = +1/2\rangle$. The angular parts of these outgoing states have the form

$$|p_{3/2}, m_j = +3/2\rangle\colon\ Y_{11}|\uparrow\rangle \sim \sin\Theta|\uparrow\rangle$$

$$|p_{3/2}, m_j = +1/2\rangle\colon\ (\sqrt{2/3}\,Y_{10}|\uparrow\rangle + \sqrt{1/3}\,Y_{11}|\downarrow\rangle)$$
$$\sim (\sqrt{2/3}\cos\Theta|\uparrow\rangle - \sqrt{1/6}\sin\Theta e^{i\phi}|\downarrow\rangle)$$

with Y_{ik} being the spherical harmonics, $|\uparrow\rangle$, $|\downarrow\rangle$ being the two orthogonal spin states, Θ being the emission angle measured from the preferential direction given by the spin of the exciting photon, and φ being the azimuth. In our experimental set-up with normal incidence of the radiation and normal emission, it is $\Theta = 0$. Therefore only the state $|p_{3/2}, m_j = +1/2\rangle$ with its spin up part $\sim\cos\Theta|\uparrow\rangle$ will contribute to the intensity. Assuming pure s-like states to be the final electron states of the excitation step we should measure the Auger-electrons to be totally polarized parallel to the photon spin. The result $P = (24 \pm 3)\%$ measured with photons of energy $h\nu = 15.3$ eV and displayed in Fig. 2 deviates from this prediction with regard to the degree of polarization but not with regard to the sign. This may be due to the small density of d-like states present in the near Fermi-level region [21] resulting in some population of $|p_{3/2}, m_j = +1/2\rangle$-hole states (see Fig. 3). In Ref. [3] with the spin-dependent Auger decay of $3p$-holes states in Fe two-hole states coupled to singlet and triplet states are used to interpret the measurements in a DOS approach neglecting spin orbit coupling with all interacting states. Two hole states coupled to a triplet state also could explain the degree of spin polarization in our measurement by considering $(4p^5\,5s5p)$- and $(4p^5\,5p^2)$-configuration in the atom-like model. But those configurations should have a low weight due to the screening of the hole [24].

To get an enlarged information about the measured effect, we have varied the photon energy used for the excitation step. In Fig. 5 a series of spin dependent partial intensities is displayed. Figure 6 shows the electron spin polarization averaged across the Auger-peaks *vs.* the photon energy. As with the electron spin-polarization given for the measurement shown in Fig. 2, the polarization values are evaluated from the partial intensities I_+ and I_- after subtraction of an unpolarized background and they are averaged across the Auger peaks.

From Figs 5 and 6 it is obvious that the sign of the Auger-electron spin-polarization changes, i.e. the preferential spin direction is inverted if the energy of the exciting photons is raised from values just above the $p_{3/2}$-threshold to energies higher by about 1 eV. In the three-step model of the Auger-process we have assumed in our model the second and third step, the decay of the hole and the transport of the Auger-electron into the vacuum, to be independent of the photon energy used in the first step. Therefore the first step, the excitation by circularly polarized photons, has to be responsible for the measured effects. In our model of the decay displayed in Fig. 4, the $|p_{3/2}, m_j = +1/2\rangle$-hole

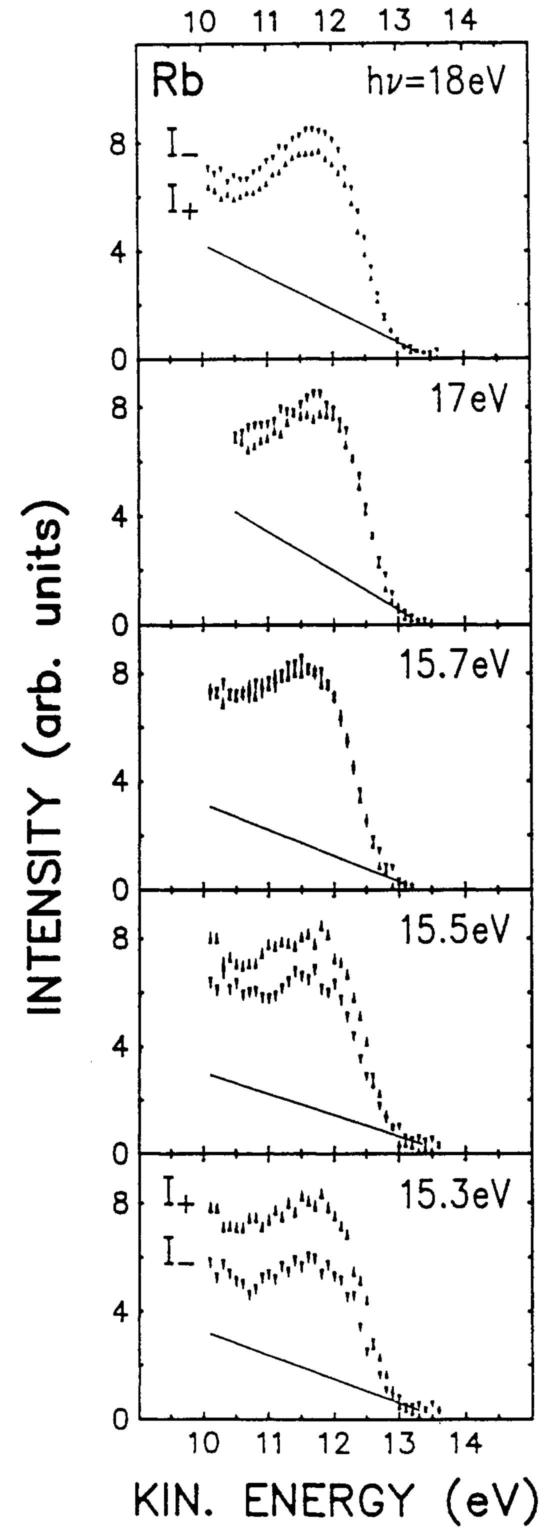

Fig. 5. Partial intensity spectra $I_+(E_{kin})$ and $I_-(E_{kin})$ measured at Rb with varied photon energy $h\nu$. The straight lines represent the unpolarized background used for evaluation of the spin polarization P. (For errors and additional details see Fig. 2)

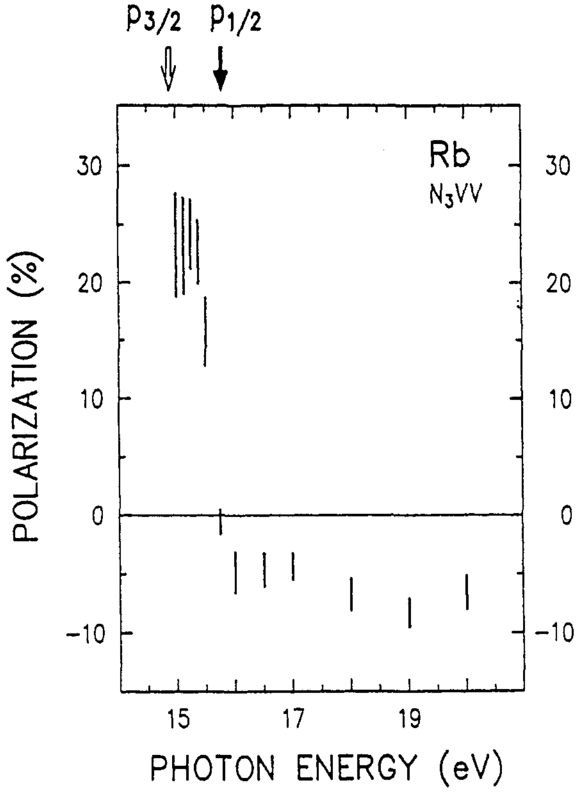

Fig. 6. Spin polarization P of the Auger-electrons *vs.* energy of the exciting polarized photons averaged across the Auger-peak measured at Rb. Details of the evaluation are given in the text. The error bars represent statistical counting errors as well as errors in the background intensity and calibration

state must be preferred against the $|p_{3/2}, m_j = -1/2\rangle$-hole state to end up with a negative Auger-electron spin-polarization. This preference can be caused by primary excitation of $[p_{1/2}, m_j = -1/2\rangle$-hole states which decay via the Koster–Kronig-transition into $p_{3/2}$-hole states [18, 20] or by a dominance of excitations into d-like final states against excitations into s-like final states.

As the zero crossing of the measured spin-polarization coincides with reaching the $4p_{1/2}$-threshold as well as with a significant increase of the d-like density of states [21], the measurements do not allow us to rule out one of these two excitations resulting in $|p_{3/2}, m_j = +1/2\rangle$-hole states. To make a decision data on angle integrated matrix elements for the excitation process not accessible at present would be necessary.

4. Summary

$N_3\,VV$-Auger electrons emitted from Rb show up a significant spin polarization if the excitation is performed using circularly polarized radiation. The degree and the sign of the measured Auger-electron spin-polarization depend strongly on details of the primary excitation. The decay of the excited hole can be described in an atom-like model assuming spin-orbit coupled states and conservation of parity and angular momentum J, M_J.

As the Auger-process can be described in a multiple-step model, i.e. excitation and decay of the primary hole are separable, our experiment opens up a new way to characterize excitation processes and unoccupied electronic states in solids or atoms.

Acknowledgements

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