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Angle and spin resolved Auger and photoelectron spectroscopy on Rb-layers by means of circularly polarized VUV radiation

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Abstract. Normal incidence circularly polarized VUV radiation with energies around 23 eV creates spin polarized photoelectrons from thick layers of Rb on Pt(111) and thus excites oriented $4p$ hole states. The preferential spin direction of the Auger electrons and its dependence upon the emission angle has been measured and is compared with the corresponding angular dependence of the primary photoelectron spin polarization also measured. Since the CVV Auger decay relates to a s^2 pair of valence electrons, the cross comparison of results for photoelectrons and Auger electrons studies the questions on whether photoemission and Auger decay occur in sequence, assuming an independent two step model, and whether the valence s-electrons couple to a singlet state configuration.

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Photon impact on solids often creates core level holes with subsequent valence electron Auger decay. 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 in most cases with ferromagnetic material [2, 3], where the electrons to be emitted are highly spin oriented in the ground state. Very recently first experimental results of spin dependent Auger emission from non-magnetic Rb-layers [4] as well as from free unoriented Ba-atoms [5] exposed to circularly polarized radiation have been reported. The spin polarized radiation creates spin polarized primary photoelectrons and thus spin oriented hole states whose decay results in spin polarized Auger electrons. It is worth noting that the Auger electrons from the CVV decay of metallic Rb-layers could not be observed for an isolated Rb atom since the alkalis only have one outermost s electron. The second s electron for the decay has to be supplied in the condensed matter by the Rb neighbours via the joint valence band. This gives rise to the question on whether they only couple to a singlet state configuration or also partially to a triplet state configuration.

Figure 1 shows the spin polarization of the Auger electrons averaged across the Auger peak measured [4] at rubidum thick layers on Pt(111) in normal emission versus the energy of the exciting circularly polarized VUV-radiation in normal incidence, after having subtracted the unpolarized inelastic background in the spin-resolved Auger electron spectra. Figure 1 also denotes the thresholds for $p_{\frac{3}{2}}$ and $p_{\frac{1}{2}}$ hole creation by the photons. The $p_{\frac{1}{2}}$ hole states excited above the $p_{\frac{1}{2}}$ threshold decay via a Koster-Kronig transition into $p_{\frac{3}{2}}$ hole states [6, 7]. Since at higher photon energies phototransitions into d states are more probable, the creation of $p_{\frac{3}{2}}$ holes with magnetic sublevels $m_j = +\frac{1}{2}$ and $m_j = +\frac{3}{2}$ is preferred against creation of $p_{\frac{3}{2}}$ holes with $m_j = -\frac{1}{2}$ and $m_j = -\frac{3}{2}$, result-

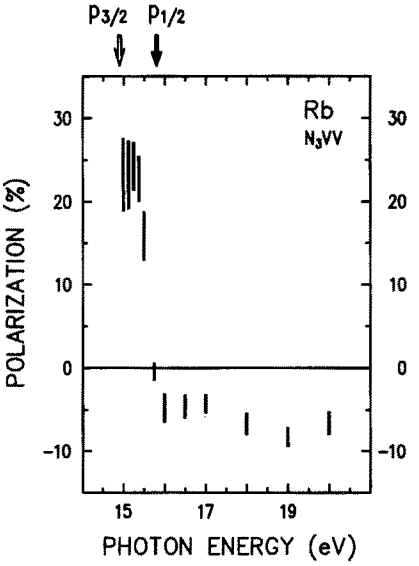

Fig. 1. Spin polarization of the Auger electrons vs energy of the exciting polarized photons [4], with the corresponding thresholds $p_{\frac{3}{2}}$ and $p_{\frac{1}{2}}$ for hole creation

ing in a negative Auger electron spin polarization measured to be nearly constant with photon energy.

It is worth noting, that the primary electrons which are excited by the circularly polarized radiation and which correspond to the Auger electrons shown in Fig. 1 cannot be detected as photoelectrons since their energy is too low. Thus for the data in Fig. 1 there is no direct quantitative evidence for the hole orientation created by the photons, regardless of the subsequent Auger decay. In order to get this information and to have a tool to study the two-electron emission process independently with respect to their spin polarizations, the photoemission process had to be studied at higher photon energies. It is the goal of this contribution to present first studies of the angular dependences of the spin polarizations of photoelectrons as well as of the corresponding Auger electrons from Rb layers and their cross comparison.

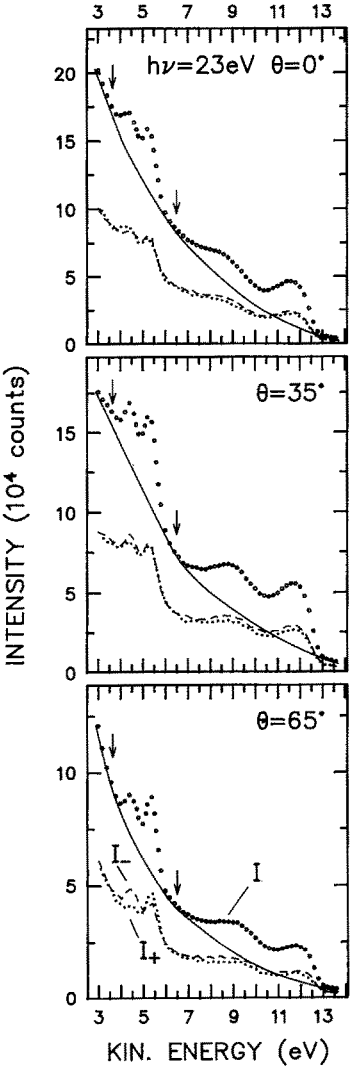

Fig. 2. Spin resolved electron spectra of Rb layers (*open circles:* total intensity; *dotted and dashed lines:* partial intensities with spin parallel and antiparallel to the light helicity, respectively) under normal light incidence (23 eV, circularly polarized) and different electron emission angles. The *solid curve* denotes the background, the *arrows* mark the positions where the background spin polarization is determined

The measurements have been performed at the 6.5 m normal incidence monochromator [8] for circularly polarized off-plane radiation at the electron storage ring BESSY in Berlin using an apparatus described elsewhere [4, 9, 10]. The degree of polarization and the band width of the circularly polarized VUV radiation was 90% and 0.2 nm, respectively; all data were recorded for normal light incidence. Electrons emitted were collected by a rotatable simulated 180° spherical field energy analyzer followed by a UHV Mott polarimeter for electron spin polarization analysis. The angular and energetic resolution was $\pm 5°$ and 0.5 eV, respectively. The Rb layers were evaporated onto a clean Pt(111) crystal cooled by means of liquid helium to about 90 K using dispensers. The Rb layers have been prepared to be so thick that no UPS and Auger signals from the Pt substrate have been observed. LEED from the prepared layers did not reveal structured patterns.

Figure 2 shows the spin resolved electron spectra of Rb/Pt obtained at the energy of 23 eV of the normally incident circularly polarized photons for three different electron emission angles, 0°, 35°, 65°. In the spectra for the total electron intensity (open circles) as well as in the partial spectra for electron spin parallel and antiparallel to the photon spin (dotted and dashed lines, respectively) we can observe the Auger peak at around 12 eV, a corresponding plasmon loss peak at around 9 eV and the direct photoelectron peaks $p_{\frac{3}{2}}$ and $p_{\frac{1}{2}}$ at 5.5 eV and 4.5 eV, respectively. While the Auger electrons and the corresponding plasma loss peak are slightly negatively spin polarized, the spin polarizations of the photoelectrons

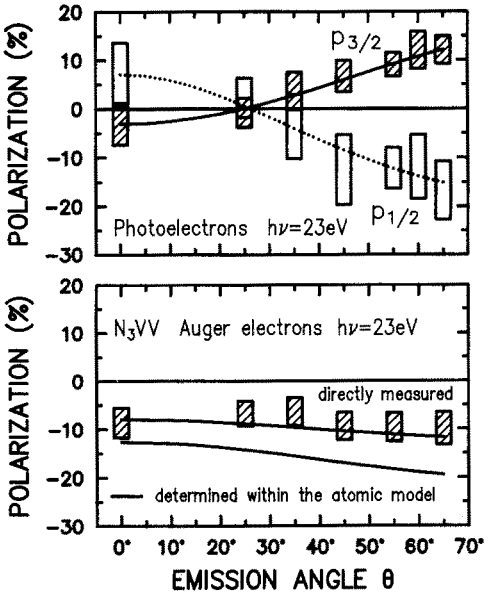

Fig. 3. Measured angular dependences of the spin polarization component in the direction of the helicity of circularly polarized 23 eV-radiation for photoelectrons and Auger electrons: *Upper part:* photoelectrons leaving a $p_{\frac{3}{2}}$ or $p_{\frac{1}{2}}$ hole state with a fit using the atomic model (*full* and *dotted curve*, respectively). *Lower part:* Auger electrons in cross comparison with the prediction of the atomic model using the data of the upper part (two curves as upper and lower limit). The rectangles describe the experimental uncertainties, including uncertainties in the background correction performed

leaving $p_{\frac{3}{2}}$ and $p_{\frac{1}{2}}$ hole states show opposite signs, as predicted by theory [11] and as measured for many atoms, molecules and adsorbates [12]. In order to be able to compare the photoelectron polarization and thus the hole orientation with the spin polarization of the subsequent Auger-decay electrons it is necessary to quantitatively subtract the inelastic background from the spin resolved electron spectra given in Fig. 2. This inelastic background, indicated by the solid curve in Fig. 2, has been measured as being slightly polarized in the region of the photoemission peaks (typically -1% at the positions marked by arrows in Fig. 2) and is assumed to be unpolarized at the Auger peak.

Figure 3 shows in the upper and in the lower part the angular dependence of the spin polarization component $A(\Theta)$ (in the direction of the light helicity) of the photoelectrons and the corresponding Auger electrons, respectively, after having made the background correction mentioned. While the Auger electrons do not show a pronounced angular distribution of the spin polarization, the photoelectron polarizations switch their sign at around 30°, as usual [10, 12]. It is worth noting that the photoelectrons corresponding to the $p_{\frac{3}{2}}$ and $p_{\frac{1}{2}}$ hole states always have opposite signs. It is also worth noting that as in the case of xenon adsorbates [10] the angular dependence of the spin polarization of photoelectrons can be fitted by the angular distribution valid for free atoms and molecules [13]:

$$A(\Theta) = \gamma \frac{A - \alpha P_2(\cos\Theta)}{1 - (\beta/2) \cdot P_2(\cos\Theta)}$$

with $A = A(\Theta = 54°)$ being the angle integrated spin polarization, γ the light helicity, $P_2(\cos\Theta)$ the second Legendre polynomial and α and β the dynamical asymmetry parameters. For $h\nu = 23$ eV we obtain

$$A_{\frac{3}{2}} = 0.09 \pm 0.024 \quad A_{\frac{1}{2}} = -0.12 \pm 0.04$$

$$\alpha_{\frac{3}{2}} = 0.12 \pm 0.02 \quad \alpha_{\frac{1}{2}} = -0.17 \pm 0.03$$

$$\beta_{\frac{3}{2}} = 0.13 \pm 0.3 \quad \beta_{\frac{1}{2}} = 0.6 \pm 0.4 .$$

It is worth noting that the relationships of the non-relativistic approximation [11] $A_{\frac{3}{2}} = -\frac{1}{2} A_{\frac{1}{2}}$ and $\alpha_{\frac{3}{2}} = -\frac{1}{2}\alpha_{\frac{1}{2}}$ are not fulfilled.

From the dynamical parameters for photoemission given above, the Auger electron spin polarization can be calculated within an atomic model using the non-relativistic LS-approximation (neglecting any influence of the spin-orbit interaction in the continuum states) [14]. Thereby the two step model [15] is applied, treating the emission processes of photoelectrons and Auger electrons as independent and subsequent processes. They are connected only by the aligned and oriented hole states resulting from the spin-dependent photoexcitation [16]. These calculated values for the Auger spin polarization as a function of the emission angle is given in the lower part of Fig. 3 including the range of uncertainty determined by the error bars of the corresponding photoelectron polarization values. They show agreement with the direct experimental values of the Auger electron polarization measured, also given in Fig. 3 lower part, within the experimental uncertainty.

Thus the spin oriented Auger electron emission subsequently following a photoemission process by means of circularly polarized light, also yielding polarized electrons, can be quantitatively explained by use of an atomic model not only with respect to the angle integrated results but also to the complete angular dependence of the electron spin polarization. The circularly polarized photons create oriented holes characterized by the photoelectron spin polarizations observed.

The CVV-decay of the hole states results in spin polarized Auger electrons as a subsequent process; the experimental results are compatible with a coupling of the valence electrons as a singlet state configuration. In agreement with the corresponding photoionization and Auger process of free Ba atoms [14] (also with two outermost s electrons in singlet state configuration) the Auger emission from the Rb adlayer atoms can be quantitatively described within the non-relativistic LS approximation while the photoemission deviates from this model due to non vanishing differences in the dipole matrix elements $p_{\frac{3}{2}} \rightarrow \varepsilon d_{\frac{5}{2}}, \varepsilon d_{\frac{3}{2}}$.

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