Angular Distribution Measurements of the Xenon $N_{4,5}O_{2,3}O_{2,3}$ Auger electrons

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INTRODUCTION

We measured the angular distribution of the Xe $N_{4,5}O_{2,3}O_{2,3}$ Auger lines, after ionization of free Xe atoms by monochromatized synchrotron radiation of 140-210 eV photon energy. In the framework of the two-step model of Auger decay the alignment of the primary hole states $4d^{-1}\,{}^2D_{3/2}$ and $4d^{-1}\,{}^2D_{5/2}$ could be derived. This quantity yields important information about the behavior of the dipole matrix elements around the Cooper minimum of the $4d$ photoionization cross section. Angular distribution and spin polarization measurements of Auger electrons give valuable insight into the primary photoionization process which is governed by the dipole interaction and the succesive radiationless decay process which is governed by the Coulomb interaction.

The transition from the single-hole to the double-hole state is described by the Coulomb operator, which is a scalar operator. This means, that Auger electrons from an isotropic initial state can have only an isotropic angular distribution and no spin polarization. The photoionization process usually leaves the singly charged photoion in a polarized state, i.e. with an anisotropic charge distribution. Thus, decay of a polarized photoion may lead to an anisotropic angular distribution and also to spin polarization of Auger electrons [1, 2].

Up to now a thorough investigation of the angular distribution of the Auger decay process which follows the 4d photoionization was done only up to 142 eV photon energy [3]. Between 150 and 190 eV four data points with large uncertanities exist [4]. The major difficulty in performing these measurements above 150 eV photon energy is the relatively low 4d photoionization cross section (<1 Mb) which results in very weak Auger lines. An additional problem is posed by the $4p$ photolines ($E_{bind}\approx 146$ eV) which overlap with the $N_{4,5}O_{2,3}O_{2,3}$ Auger group in the 175-188 eV photon energy range.

EXPERIMENT

The experiment was carried out at the AMO undulator beamline 10.0.1 of the Advanced Light Source. Gratings with groove density of 925 lines/mm and 2100 lines/mm were used for operating at photon energies below and above 160 eV, respectively. High photon energy resolution was not required for the present measurements; photon bandwidths in the range of 40-150 meV were obtained using the 925 lines/mm grating in combination with entrance and exit slit widths of 100 and 100 μm. The 2100 lines/mm grating with entrance/exit slit settings of 100 and 800 μm provided photon bandwith between 380 and 600 meV at the photon energies from 160 to 210 eV.

The electron spectra were measured using an endstation designed for gas-phase angle-resolved studies and based on the Scienta SES-200 hemispherical analyzer [5]. The analyzer is rotatable in a plane perpendicular to the propagation direction of the beam of linearly polarized photons (the degree of linear polarization is estimated to be higher than 99%), allowing studies of the angular distribution of electrons. The analyzer was operated at the constant pass energy of 40 eV with the electron energy resolution of 25-30 meV. The spectra were not corrected for the transmission of the analyzer, which was assumed to be independent of the measurement angle. The target gas was introduced into a differentially pumped gas cell, achieving about 10^{-3} mbar pressure in the interaction region.

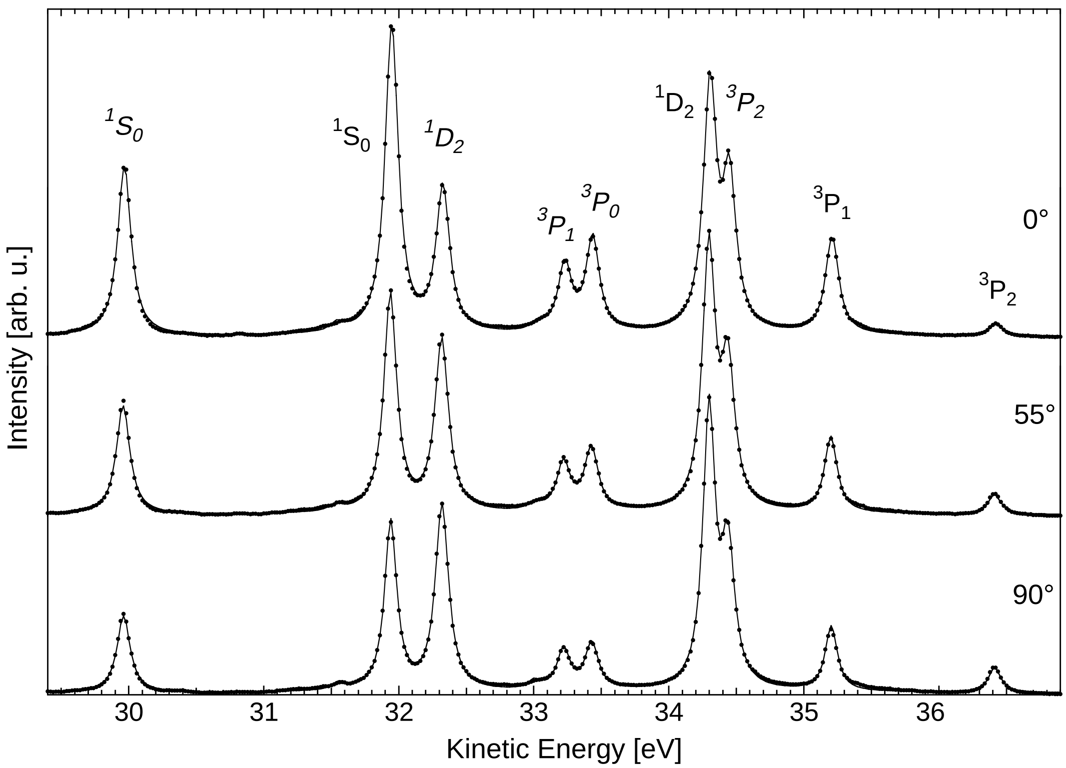

Figure 1. Xe $N_{4,5}O_{2,3}O_{2,3}$ Auger group after excitation by 160.4 eV photons taken at three different angles, after subtracting a linear background. The resolution of the electron spectrometer was approx. 30 meV. The solid lines represent the sum of fitted Voigt-profiles. All three spectra are normalized to the same scale. Notations in italic refer to N_5 lines.

THEORETICAL BACKGROUND

In the two-step model of Auger decay, i.e. the photoionization and Auger decay processes are assumed to proceed subsequently and independently of each other, the angular anisotropy parameter can be factorized into a parameter describing the anisotropy of the primary hole state and into an 'intrinsic' parameter α_2 describing the Auger decay itself [1]:

$$\beta = \alpha_2 A_{20} . \tag{1}$$

A_{20} is the alignment parameter of the primary hole state and is proportional to its electric quadrupole moment. A_{20} is given here with respect to the direction of the electric field vector **E**. Whereas the Auger anisotropy parameter α_2 depends on the Coulomb matrix elements, A_{20} is connected to the dipole matrix elements.

The description of the photoionization process in the framework of the dipole approximation limits the possible values of the orbital angular momenta of the outgoing photoelectrons through the selection rules. Thus, in the case of the 4*d* ionization, the electrons can leave the atom as εp or εf continuum waves. In the nonrelativistic approximation, i.e. when the spin-orbit interaction in the continuum is neglected, there are only the two outgoing waves εp and εf and the photoionization cross section is determined by the single-particle radial matrix elements $R_{4d,\varepsilon p}$ and $R_{4d,\varepsilon f}$ [6]. Using the density-matrix and statistical-tensor formalism, Berezhko et al [7] showed, that the alignment tensor depends only on the ratio of the dipole amplitudes

$$A_{20}^{5/2} = -\frac{1}{5}\sqrt{\frac{2}{7}}\frac{2+7\lambda^2}{1+\lambda^2},\; A_{20}^{3/2} = -\frac{1}{10}\frac{2+7\lambda^2}{1+\lambda^2} \text{ with } \lambda = \sqrt{\frac{2}{3}}\frac{R_{4\mathrm{d},\varepsilon p}}{R_{4\mathrm{d},\varepsilon f}} . \tag{2}$$

In the nonrelativistic approximation the ratio of the alignments for the different final states is a fixed number

$$A_{20}^{5/2}/A_{20}^{3/2} = \sqrt{8/7}\,. \qquad (3)$$

The extrema of the alignment can be easily derived, when either the εp or the εf wave completely vanishes. For $R_{4d,\varepsilon p}=0$, $A_{20}^{5/2}=-0.214$ and for $R_{4d,\varepsilon f}=0$, $A_{20}^{5/2}=-0.748$ [6].
Theoretical A_{20} values in dependency of the photon energy were published by several authors. Hartree-Fock calculations were performed by Kennedy and Manson [6] and by Berezhko et al [7]. Cherepkov [8] carried out calculations using the random-phase approximation with exchange (RPAE). All these authors treated the photoionization process in the nonrelativistic approximation.

RESULTS

Fig. 2 shows $A_{20}^{5/2}$ together with other experimental data and theoretical curves. The agreement between all experimental data is very good up to 142 eV photon energy. At higher energies a considerable dicrepancy between our results and those of Southworth et al. [4] can be seen. The earlier data has large uncertainties and doesn't show a pronounced minimum.
The overall shape of all theoretical curve is similar, showing a clear minimum at higher energies. Whereas the RPAE calculation of Cherepkov gives the position of the minimum at 6 eV lower energy than experiment, all other calculations predict the minimum at either 30 eV lower energy Hartree-Slater model of Ref. [7] or at 30 eV higher energy Hartree-Fock model of Ref. [7] and Ref. [6].
To get a better idea about the course of the A_{20} curve, we modified the RPAE curve by stretcthing it to higher energies to match the minimum of the experimental data. In this way we obtain a good agreement with a slight deviation around 145 eV. This deviation might be due to the fact that the 4p threshold is approx. at 146 eV photon energy and this was not taken into account during the calculations.
As described above, A_{20} can reach two extrema for vanishing p- or f-waves. (cf. Eq. (3)). Due to the potential barrier for f-electrons near threshold ($E_{bind}(4d_{5/2})=67.54$ eV), the outgoing p-waves dominate the photoionization. At increasing energies, the f-electrons overcome the barrier and carry almost all the cross section in the maximum of the shape resonance (~100 eV). In the Cooper minimum $R_{4d,\varepsilon f}$ changes sign, i.e. only the p-wave is present at ~176 eV. Whereas the first two extrema were confirmed by the previous experiments, we could prove the existence of the A_{20} minimum in the Cooper minimum of the cross section.

SUMMARY

Using the framework of the two-step model of Auger decay we determined the alignment A_{20} of the $4d^{-1}\ ^2D_{3/2}$ and $^2D_{5/2}$ photoions at different excitation energies including the very important region of the 4*d* photoionization Cooper minimum around 180 eV. By obtaining an A_{20} value of –0.73(4) at 176 eV photon energy, we could experimentally prove for the first time, the vanishing of the εf component of the outgoing electron wave. This is in good agreement with theoretical predictions, taking into account a small deviation in the position of the predicted minimum. An important implication of the present result is that with $R_{4d,\varepsilon f}=0$ at 176 eV all dynamical parameters of photoionization have fixed values at this energy.

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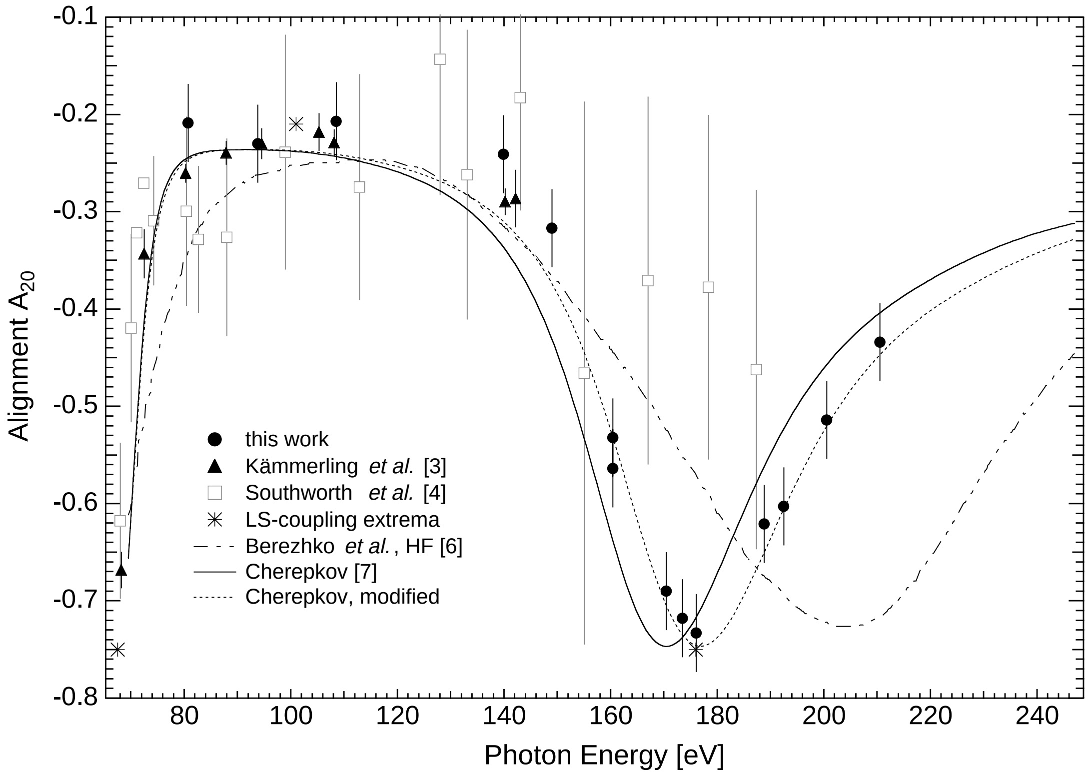

Figure 2. Alignment A_{20} of the Xe $4d^{-1}\,{}^2D_{5/2}$ hole state. The stars show the extreme values of A_{20} in LS-coupling for vanishing f-wave (at threshold and in the Cooper minimum) and vanishing p-wave in the cross section maximum (cf. Eq. (3) and text). The dashed curve was obtained by stretching the calculation of Cherepkov to match the A_{20} minimum.

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