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High-resolution X-ray study of the multiple ionization of Pd atoms by fast oxygen ions

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Abstract. The multiple ionization of the L- and M-shells of Pd by fast oxygen ions has been studied by measuring with high-resolution the satellite structures of the $L\alpha_{1,2}$ X-ray transitions. Relativistic multi-configuration Dirac-Fock (MCDF) calculations were used to interpret the complex X-ray spectrum, allowing to derive the number of L- and M-shell spectator vacancies at the *moment of the X-ray emission*. After correcting these numbers for the atomic vacancy rearrangement processes that take place prior to the X-ray emission, the ionization probabilities corresponding to the *collision time* were obtained. The latter were compared to predictions of the semiclassical approximation (SCA) and the geometrical model. The SCA calculations were performed using relativistic hydrogenic and self-consistent Dirac-Hartree-Fock (DHF) electronic wave functions. It was found that the use of the more realistic DHF wave functions in the SCA calculations leads to a much better description of the measured ionization probabilities for both the L- and M-shells.

1 Introduction

In high-resolution measurements of X-ray spectra produced in collisions of heavy ions with atoms, complex X-ray satellites resulting from the radiative decay of the multi-vacancy states characterizing the target atoms at the moment of the X-ray emission are observed. By correcting the intensities of the X-ray satellites for the atomic vacancy rearrangement processes that take place prior to the X-ray emission, the vacancy distribution at the moment of the collision can be deduced and the corresponding probabilities can be determined. Consequently, high-resolution studies of ion-excited X-ray satellites give insight into the dynamics of the multiple ionization process.

In the present paper the high-resolution $L\alpha_{1,2} + L\beta_1$ X-ray spectrum of Pd excited by 278.6 MeV O^{6+} ions is reported and discussed in the context of the multiple L- and M-shell ionization. The measurements were carried out by means of the Fribourg von Hamos crystal X-ray spectrometer [1]. The experimental X-ray spectrum, characterized by rich M-shell satellite and L-shell hypersatellite structures, was interpreted with the help of multi-configuration Dirac-Fock (MCDF) calculations [2]. The distribution of the M-shell vacancies at the collision time was determined from the observed X-ray M-satellite intensities that were corrected beforehand for the vacancy rearrangement processes (radiative, Auger and Coster-Kronig) and the changes of the L-shell decay rates in multiply ionized atoms. The ionization probabilities for nearly zero impact parameters were extracted from the measured relative L-hypersatellite yield and M-satellite intensity distribution, respectively, and compared to the values predicted by the semiclassical approximation (SCA) [3,4] and the geometrical model (GM) [5]. The SCA calculations were performed using relativistic hydrogenic (SCA-DH) [3] and self-consistent Dirac-Hartree-Fock (SCA-DHF) [4] electronic wave functions, whereas nonrelativistic hydrogenic wave functions were employed in the GM calculations. A detailed comparison between the experimental L-shell and M-shell ionization probabilities obtained in the present work and the ones predicted by the SCA and GM models is presented.

2 Experiment

The high-resolution measurements of the Pd $L\alpha_{1,2}$ ($L_3 \rightarrow M_{4,5}$) and $L\beta_1$ ($L_2 \rightarrow M_4$) X-ray transitions were performed at the Paul Scherrer Institute (PSI), in Villigen, Switzerland. As the experiment was already presented in detail elsewhere [6], only the main characteristics of

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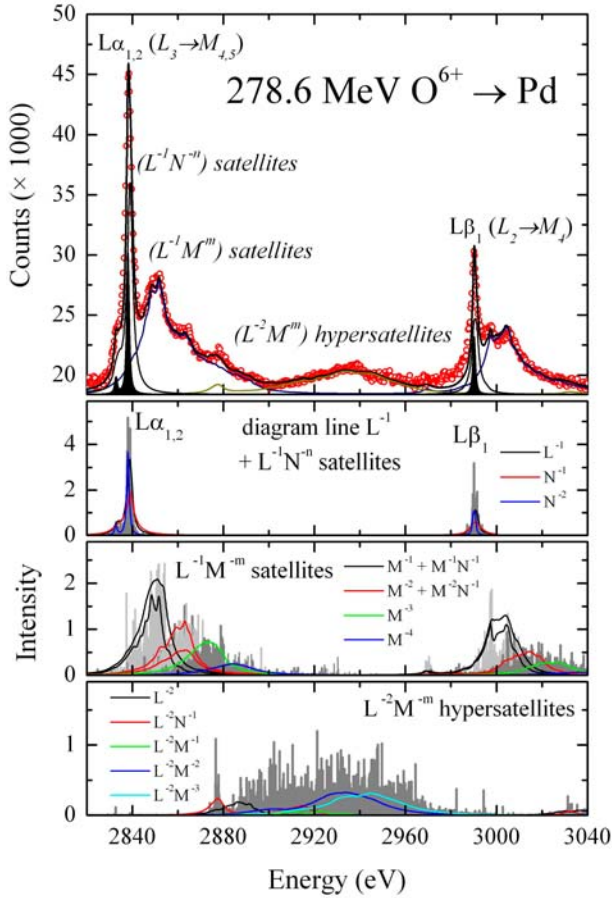

Fig. 1. (Color online) High-resolution $L\alpha_{1,2}+L\beta_1$ X-ray spectrum of Pd excited by 278.6 MeV oxygen ions (top panel). For comparison, the M- and N-shell satellite and L-shell hypersatellite spectra obtained from extensive MCDF calculations are also shown (three lower panels).

the experimental setup will be described below. The 278.6 MeV O^{6+} ion beam provided by the variable energy PSI Philips cyclotron was focused by means of dipole and quadrupole magnets to a 4 mm-high × 1 mm-wide spot on the 1.8 mg/cm^2 thick metallic palladium foil. The beam current was of the order of 100 nA. The L X-ray emission of the Pd target was measured by means of the high-resolution von Hamos crystal spectrometer [1] with an instrumental energy resolution of 0.6 eV. The spectrometer was equipped with a quartz $(1\bar{1}0)$ crystal bent cylindrically to a radius $R = 25.4$ cm. The diffracted X-rays were detected with a front illuminated deep depleted charge-coupled device (CCD) camera having a pixel resolution of 27 μm and a depletion depth of 50 μm. With the chosen crystal, the 27.6 mm long CCD-detector permitted us to cover an energy domain of about 70 eV, so that four CCD lengths were needed to collect the Pd L X-ray spectrum depicted in Figure 1. The spectrometer was calibrated in energy with the $K\alpha_{1,2}$ X-ray lines of vanadium, using the bremsstrahlung of a Cr-anode X-ray tube for the fluorescence production. The uncertainty in the energy calibration was estimated to be about 0.3 eV.

3 Results and discussion

The Pd $L\alpha_{1,2}$ + $L\beta_1$ X-ray spectrum was analysed by means of a least-squares fit program using the theoretical line shapes obtained from the MCDF calculations (see Fig. 1). The relative intensities of the MCDF components belonging to each group of the diagram, satellite and hypersatellite lines were fitted to the total yield of the experimental spectrum. The intensities provided by the fit were then corrected for the vacancy rearrangement processes and the modified fluorescence yields characterizing multiply ionized atoms (see Ref. [7]). From these corrected intensities which reflect the relative numbers of spectator holes at the collision time, the initial distributions of the L- and M-shell vacancies were determined. As the multiple ionization resulting from impact with heavy ions is expected to be described within the independent electron picture, binomial distributions were assumed for the numbers of initial vacancies $P_X(b,n)$. The latter which depend only on the ionization probabilities per electron $p_X(b)$ are given by:

$$P_X(b,n) = \binom{N}{n} p_X(b)^n [1-p_X(b)]^{N-n}, \qquad (1)$$

where X stands for the L- or M-shell, b represents the impact parameter, n the number of vacancies in the shell X and N the original number of electrons in this shell.

In order to determine the L-shell ionization probability, the $L\alpha_{1,2}$ hypersatellite-to-diagram+satellite intensity ratio was considered. Because L hypersatellites are characterized by two L-shell vacancies in the initial state, whereas their parent diagram and satellite lines have a single L-hole in the initial state, this ratio indeed depends on the L-shell ionization probability. Furthermore, as the L-shell ionization probability extends over a wide impact parameter range (see Fig. 2), the intensity ratio $R = I(L^h\alpha_{1,2})/[I(L\alpha_{1,2}) + I(L\alpha_{1,2}M^{-m})]$ can be written as follows:

$$R = \frac{[\binom{4}{1}^2 + \binom{4}{2}] \int_0^\infty b p_L^2(b)[1-p_L(b)]^6 db}{\binom{4}{1} \int_0^\infty b p_L(b)[1-p_L(b)]^7 db}. \qquad (2)$$

The binomial factors in the numerator of equation (2) reflect the fact that for the $L\alpha_{1,2}$ hypersatellite transition, at least one of the two initial L-shell vacancies should lie in the L_3-subshell which contains originally four electrons. If there is a single vacancy in the L_3-subshell, the second one should be present in one of the $L_{1,2}$-subshells which contain in total also four electrons. Thus, the squared binomial factor corresponds to the case of a single vacancy in the L_3-subshell, while the second one to the case of two vacancies. Using equation (2) the ratios R can be calculated from the theoretical $p_L(b)$ values as a function of $p_L(0)$. Results of such calculations performed within the SCA for both the DH and DHF wave functions are presented in Figure 3. As expected from equation (2) for $p_L(b) \ll 1$ this formula predicts a nearly linear scaling of R with $p_L(0)$, i.e., $R \propto p_L(0)$.

From the measured $L\alpha_{1,2}$ spectrum interpreted with the aid of MCDF calculations (see Fig. 1), and after

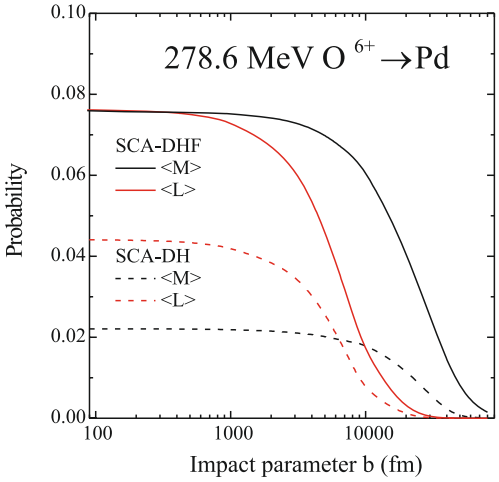

Fig. 2. (Color online) Variation of the L- and M-shell ionization probabilities per electron as a function of the impact parameter for Pd atoms bombarded by 278.6 MeV oxygen ions. The curves were obtained from SCA-DH and SCA-DHF calculations.

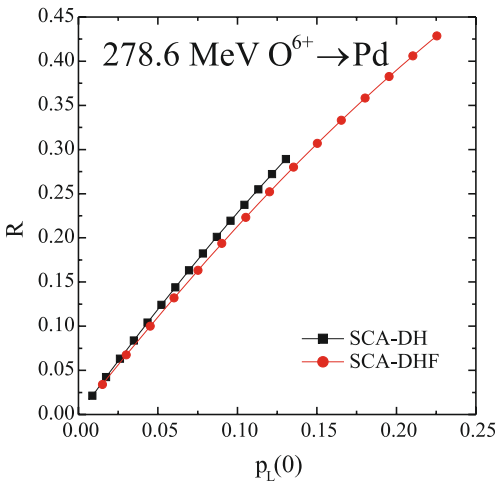

Fig. 3. (Color online) Calculated intensity ratios R plotted as a function of $p_L(0)$ for the SCA-DH and SCA-DHF approaches.

correcting the satellite intensities for the rearrangement processes and fluorescence yield changes, the intensity ratio $R_{exp} = 0.214 \pm 0.032$ was obtained. This result can be compared to values of 0.104 and 0.164 from the SCA-DH and SCA-DHF calculations, respectively (see Tab. 1). The SCA-DHF value lies closer to the experimental ratio compared to the SCA-DH prediction. This indicates the importance of using self-consistent wave functions for the description of the Pd L-shell electrons. The experimental ionization probability $p_L(0)$ was then determined from the experimental ratio R_{exp} and the calculated ratios plotted in Figure 3 by solving for $p_L(0)$ the equation $R(p_L(0)) = R_{exp}$. Using the SCA-DHF intensity ratio (which is, as mentioned before, more reliable than the SCA-DH one) and the experimental ratio $R_{exp} = 0.214 \pm 0.032$, a probability $p_L(0) = 0.093 \pm 0.012$ was found. As shown in Table 1, this result is in quite

Table 1. Experimental and theoretical intensity ratios $R = I(L^h\alpha_{1,2})/[I(L\alpha_{1,2}) + I(L\alpha_{1,2}M^{-m})]$ and zero impact parameter ionization probabilities $p_L(0)$ and $p_M(0)$ for Pd bombarded by 278.6 MeV oxygen ions. Theoretical values were obtained using SCA-DH, SCA-DHF and GM calculations.

Theory	R	$p_L(0)$	$p_M(0)$
SCA-DH	0.104	0.044	0.023
SCA-DHF	0.164	0.075	0.076
GM	–	0.098	0.055
Experiment	0.214 ± 0.032	0.093± 0.012	0.062 ± 0.007

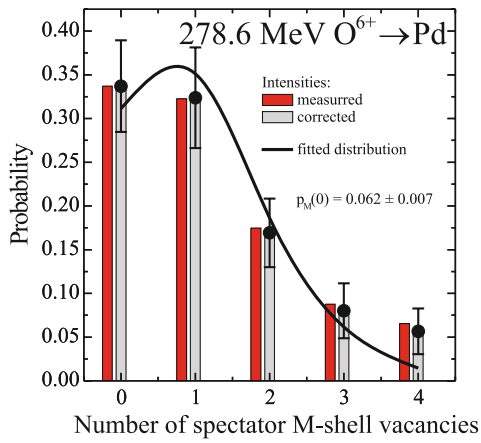

Fig. 4. (Color online) Measured intensities of the M-shell satellites as a function of the number of spectator holes in the M-shell. For comparison, the intensities before and after correction for the rearrangement processes and the changes in the fluorescence yields are shown. As the two corrections have opposite effects on the satellite yields, the differences are small and they are only observed for the 3rd and 4th order M-satellites. The solid line represents the best fit to the corrected data using a binomial distribution.

satisfactory agreement with the SCA-DHF prediction of 0.075, whereas the SCA-DH approach yields a significantly lower probability of 0.044. On the other hand, it is somewhat surprising that the best agreement is observed for the geometrical model, a simple approach based on the binary encounter approximation (BEA), which provides a value of 0.098 that is very close to the result obtained in the present experiment.

In contrast to the case of the L-shell discussed above, the M-shell ionization probability $p_M(0)$ could be extracted directly from the intensity distribution of the $L\alpha_{1,2}$ M-shell satellites depicted in Figure 4. The relative yields of the M-satellites were fitted with a binomial distribution (see Eq. (1)), using the M-shell ionization probability per electron $p_M(0)$ for the zero impact parameter as single free fitting parameter. The fact that the fitted probability was assumed to correspond to an impact parameter equal to zero can be explained as follows: the production of $L\alpha_{1,2}$ M-shell satellites requires the simultaneous ionization of one L_3-subshell and one or several M-shell electrons. However, as shown in Figure 2, the M-shell ionization probability is nearly constant, i.e.

$p_M(b) \cong p_M(0)$, over most of the impact parameter range for which the L-shell ionization probability is not negligibly small. From the binomial fit of the M-shell satellite intensities $p_M(0) = 0.062 \pm 0.007$ was obtained. This value is in good agreement with the SCA-DHF (0.075) and GM (0.056) predictions, whereas the SCA-DH model, which predicts a value of 0.023, underestimates the M-shell ionization probability to a larger extent than the L-shell probability. Note that similar observations were done in former multiple ionization studies of mid-heavy elements bombarded by light and heavy ions [8–10].

4 Summary and concluding remarks

In this paper the L- and M-shell ionization probabilities corresponding to nearly-central collisions of 276.8 MeV bare oxygen ions with neutral palladium atoms were reported. The probabilities were determined using MCDF calculations to interpret the complex Pd $L\alpha_{1,2}$ X-ray spectrum measured with a high-resolution von Hamos crystal spectrometer. The $p_L(0)$ probability was deduced from the relative intensity of the X-ray $L^h\alpha_{1,2}$ hypersatellite transitions by comparing the measured and SCA-DHF theoretical intensity ratios $R = I(L^h\alpha_{1,2})/[I(L\alpha_{1,2}) + I(L\alpha_{1,2}M^{-m})]$, while the $p_M(0)$ probability was determined by fitting the measured yields of the $L\alpha_{1,2}M^{-m}$ M-shell satellites with a binomial distribution for $m \leq 4$. The experimental probabilities were found to be in reasonable agreement with the predictions of SCA-DHF calculations. In contrast, it was observed that the SCA-DH model predicts systematically too low probabilities, in particular for the M-shell. This result evidences the importance of using self-consistent DHF electronic wave functions in SCA calculations dealing with the L-shell and especially with the M-shell. Finally, it was also found that the geometrical model, taking into account its simplicity, predicts surprisingly well the zero impact parameter ionization probabilities for both the L- and M-shells.

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