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Vacancy rearrangement processes in multiply ionized atoms

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Abstract. We demonstrate that in order to interpret the x-ray satellite structure of Pd $L\alpha_{1,2}(L_3M_{4,5})$ transitions excited by fast O ions, which was measured using a high-resolution von Hamos crystal spectrometer, the vacancy rearrangement processes, taking place prior to the x-ray emission, have to be taken into account. The measured spectra were compared with the predictions of the multi-configuration Dirac-Fock (MCDF) calculations using the fluorescence and Coster-Kronig yields which were modified due to a reduced number of electrons available for relaxation processes and the effect of closing the Coster-Kronig transitions. We demonstrate that the vacancy rearrangement processes can be described in terms of the rearrangement factor, which can be calculated by solving the system of rate equations modelling the flow of vacancies in the multiply ionized atom. By using this factor, the ionization probability at the moment of collision can be extracted from the measured intensity distribution of x-ray satellites. The present results support the independent electron picture of multiple ionization and indicate the importance of use of Dirac-Hartree-Fock wave functions to calculate the ionization probabilities.

1. Introduction

The x-rays emitted in collisions of fast heavy ions with atoms show the x-ray satellite structure reflecting the multi-vacancy configurations at the moment of the x-ray emission resulting from a rearrangement of initial vacancies formed in the collision. Consequently, high-resolution measurements of ion-excited x-ray satellites give thus access to study the structure of multi-vacancy configurations in atoms as well as the dynamics of the multiple ionization process. The structure of x-ray satellites can be interpreted in terms of the multi-configuration Dirac-Fock (MCDF) calculations giving the shapes of x-ray multiplets corresponding to a given multi-vacancy configuration which, to be compared with the measured spectra, have to be further convoluted with the natural Lorentzian width of x-ray transitions and experimental broadening of the spectrometer. In this way the probabilities of observing specific multi-vacancy configurations at the moment of emission of x-ray satellites can be extracted from the data by using the MCDF calculations. However, in order to extract the ionization probabilities for the moment of collision, which carry an information on the dynamics of the multiple ionization

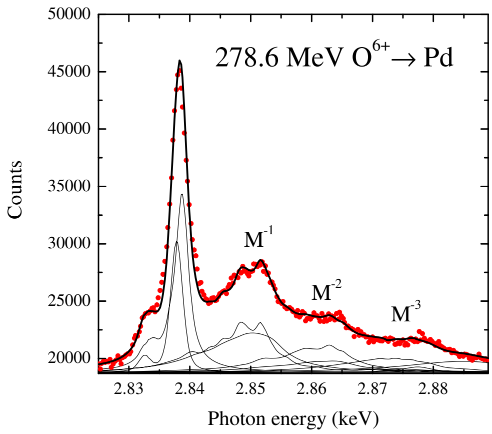

Figure 1. Measured x-ray satellite structure of L$\alpha_{1,2}$ transitions in palladium excited by 278.6 MeV O^{7+} ions. The data are compared with the MCDF calculations.

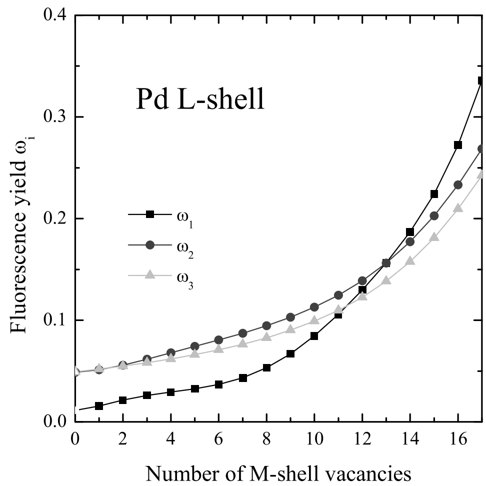

Figure 2. Calculated L-shell fluorescence yields (ω_1, ω_2 and ω_3) for palladium for different number of vacancies in the M-shell.

process, the vacancy rearrangement processes taking place between the moments of ionization and x-ray emission have to be taken into account.

In the present paper we discuss the x-ray satellites of Pd L$\alpha_{1,2}$($L_3M_{4,5}$) transitions excited by 278.6 MeV O ions, which were measured using a high-resolution von Hamos crystal spectrometer. The observed M-shell x-ray satellites, having up to $m = 4$ M-shell vacancies (see Fig. 1), are interpreted in terms of the MCDF calculations. Furthermore, a simplified vacancy rearrangement correction, which is obtained by solving the system of rate equations for vacancy relaxation processes, is discussed here to extract from the data the ionization probabilities at the moment of collision. The ionization probabilities obtained in this way are compared with the theoretical predictions indicating the importance of description of the electronic wave function for the M-shell electrons using relativistic Dirac-Hartree-Fock approach.

2. Experiment

The high-resolution measurements of the Pd L$\alpha_{1,2}$($L_3M_{4,5}$) x-ray transitions excited by fast O^{7+} ions of energy 278.6 MeV have been performed at the Philips cyclotron in the Paul Scherrer Institute (PSI) in Villigen, Switzerland, using a von Hamos high-resolution diffraction spectrometer. Since this experiment has been discussed in more detailed way elsewhere [1], here only a short description is given. The x-ray diagram and satellite transitions were measured by means of a high-resolution, reflecting von Hamos spectrometer [2] with an energy resolution of about 1 eV for studied Pd L-x-rays (3 keV), resulting from a natural Lorentzian width of transition and experimental Gaussian resolution of about 0.7 eV. The spectrometer was equipped with a quartz $(1\bar{1}1)$ crystal curved with a radius of $R = 25.4$ cm. The energy calibration of the spectrometer has been performed by measuring K$\alpha_{1,2}$ x-ray lines of vanadium excited by photons from an x-ray tube with Cr anode and the absolute x-ray energies were accurate within 0.3 eV.

3. Results and discussion

The measured x-ray spectra of Pd L$\alpha_{1,2}$(L$_3$M$_{4,5}$) transitions were compared with the predictions of the MCDF calculations by assuming that the measured data can be reproduced by a linear combination of calculated MCDF multiplets for different multi-vacancy configurations. In this way the ionization probabilities for the moment of X-ray emission can be obtained by fitting the weights for the vacancy configurations with a given number of vacancies. In order to derive the ionization probabilities for the moment of collision the measured intensities of individual M-shell satellites were corrected for the modification of L$_3$-subshell fluorescence yield for a given number of M-shell vacancies as well as the vacancy rearrangement process. The modified fluorescence yield ω_3 for L$_3$-subshell (see Fig. 2) and the Coster-Kronig yields for the M-shell, needed for the rearrangement correction, were calculated for palladium following the procedure described in Ref. [4], which includes both the statistical scaling of the decay rates for a reduced number of available vacancies as well as the important effect of closing the Coster-Kronig transitions in multiply ionized atoms [5].

In order to describe the vacancy rearrangement process modifying an initial distribution of vacancies in the M-shell we have developed a simplified approach describing an evolution of the number of vacancies after a moment of collision based on the system of rate equations relating the number of vacancies $n_i(t)$ at the moment t in a given subshell i with the total decay rates Γ_i of the states involved in the rearrangement process, namely

$$\frac{dn_i(t)}{dt} = \sum_{m=1}^{i-1} \Gamma_m n_m(t) - \Gamma_i n_i(t) \tag{1}$$

The total decay rates Γ_i above, which include all possible radiative and radiationless transitions, were calculated for given initial multi-vacancy configurations following Refs. [4, 5]. Using this approach the average number of vacancies $\langle n_i \rangle_j$ with respect of a radiative decay of a vacancy in the j-state, here L$_3$-subshell, can be obtained by solving recurrently the rate equations with the initial condition $n_i(0)$ denoting the initial number of vacancies in the i-th subshell in the moment of collision ($t = 0$). Finally, the average vacancy rearrangement correction factor for the M-shell $f_M = \sum_i \langle n_i \rangle_j / \sum_i n_i(0)$, being a ratio of the numbers of actual and initial vacancies in all subshells, was calculated.

The calculated vacancy rearrangement correction factor (see Fig. 3) was used to correct the measured intensities of x-ray satellites for the vacancy rearrangement in the M-shell. Additionally, the x-ray intensities were also corrected for a change of the L$_3$-subshell fluorescence yield due to a presence of spectator M-shell vacancies. The corrected intensities of M-shell satellites were fitted with the binomial distribution assuming the independent electron model of the multiple ionization precess. In fact, as it is shown in Fig. 4, the data are reasonable well fitted using the ionization probability in the M-shell $p_M = 0.0636$ at the moment of ionization. This ionization probability can be compared with the predictions of the semiclassical SCA calculations [6] using the relativistic hydrogenic Dirac (HD) wave functions, $p_M^{SCA-HD} = 0.022$, which are much smaller than measured probability. The importance of the self-consistent relativistic Dirac-Hartree-Fock (DHF) wave functions to calculate the ionization probabilities for the M-shell using the semiclassical approximation has been recognized in ion-atom collisions earlier [7]. Following this work, one finds that the ratios of the SCA-DHF and SCA-HD ionization probabilities are close to a factor of three for heavy ions over a wide energy range covering the present ion beam energy. With this assumption the ionization probability according to the SCA-DHF calculations was estimated to be $p_M^{SCA-DHF} \approx 0.066$, being close to the measured probability. This observation demonstrates the importance of the relativistic self-consistent electronic wave functions to be used in the SCA calculations of the ionization probabilities.

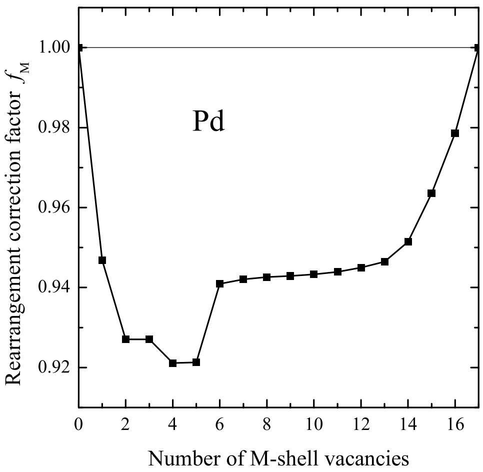

Figure 3. Calculated correction factor for vacancy rearrangement in the M-shell of palladium for different number of vacancies in the M-shell.

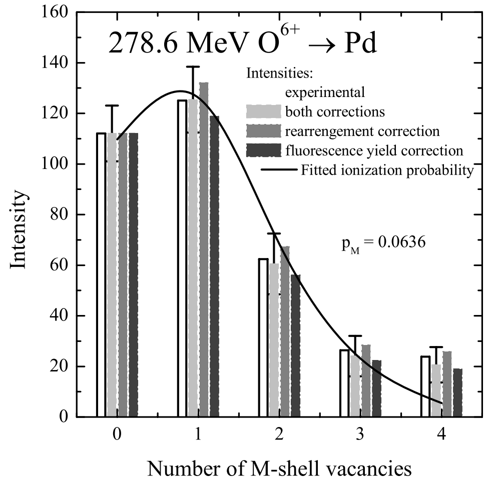

Figure 4. Measured and corrected (see the text) intensity distribution of M-shell satellites of Pd $L\alpha_{1,2}(L_3M_{4,5})$ transitions compared with the fitted binomial distribution.

4. Conclusions

We have demonstrated that the vacancy rearrangement process, which is important to interpret the M-shell satellites of palladium $L\alpha_{1,2}(L_3M_{4,5})$ transitions excited by 278.6 MeV O ions, can be accounted for by using the the vacancy rearrangement correction factor obtained by solving the system of rate equations. With the correction for the vacancy rearrangement, the ionization probabilities for the moment of collision can be extracted from the measured intensity distribution of M-shell satellites. The measured ionization probability indicates the importance of the relativistic Dirac-Hartree-Fock electronic wave function to be used in calculations of the ionization probability.

Acknowledgments

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