Multiple Ionization Effects in X-Ray Emission Induced by Heavy Ions

M. Czarnota, M. Pajek, D. Banaś,
Institute of Physics, Świętokrzyska Academy, 25–406 Kielce, Poland

J.-Cl. Doussie, Y.-P. Maillard, O. Mauron, P. A. Raboud, M. Berset,
Physics Department, University of Fribourg, CH-1700 Fribourg, Switzerland

D. Chmielewska, J. Rzadkiewicz, Z. Sujkowski,
Sołtan Institute for Nuclear Studies, 05-400 Otwock-Świerk, Poland

J. Hoszowska,
European Synchrotron Radiation Facility (ESRF), F-38043 Grenoble, France

K. Slabkowska, and M. Polasik
Faculty of Chemistry, Nicholas Copernicus University, 87-100 Toruń, Poland

Received on 29 July, 2005

The x-ray satellite structure of Pd L\(\alpha\)1,2(L3M4,5) transition excited by an impact of O7\(^{+}\) and Ne6\(^{+}\) ions with energies 279 and 178 MeV, respectively, which were measured using a high-resolution von Hamos crystal spectrometer, is discussed in terms of the multi-configuration Dirac-Fock (MCDF) calculations. We demonstrate, by using the arguments of the general central limit theorem (GCLT), that a structure of complex M-shell satellites of Pd L\(\alpha\)1,2(M\(^{m}\)) transitions for a higher number of spectator vacancies \((m > 4)\), which consists of hundreds of thousands of individual x-ray transitions as obtained from the MCDF calculations, can be well described by a single Voigtian profile. The Lorentzian width of such Voigtian line can be well modeled by using the results of the MCDF calculations for simpler configurations with a number of vacancies \(m \leq 4\). This method allows one to describe realistically a complex structure of M-shell satellites, thus extending the applicability of the MCDF calculations, which are limited by an increasing complexity of numerical calculations.

Keywords: Multiple ionization; X-ray satellite structure; MCDF calculations

I. INTRODUCTION

The x-rays emitted from atoms multiply ionized by heavy ions exhibit, apart from the well known x-ray diagram lines, the satellite structure corresponding to different multi-vacancy configurations present at the moment of the x-ray emission. High-resolution measurements of excited x-ray satellites give thus access to study the structure of multi-vacancy configurations in atoms. However, in order to extract from such complicated spectra the x-ray transitions the structure of x-ray multiplets and their Lorentzian widths for a given multi-vacancy configuration as well as the experimental Gaussian broadening have to be known. This results from the fact that for heavy ion impact the x-ray spectra, containing x-ray satellites with up to several spectator vacancies in the inner-shells, become extremely complex and, consequently, cannot be fitted uniquely without performing the MCDF calculations of the structure of individual x-ray multiplets and realistic modelling of their widths. In this paper we discuss a new method of analysis of complex x-ray spectra which, using the general central limit theorem arguments, describes the complex x-ray satellites as the smooth Voigtian profile having known mean value and width, which can be obtained from MCDF calculations. This method, which is based on our earlier works [1, 2], is presently extended to high-resolution spectroscopy. An alternative approach of analysis of x-ray spectra emitted from multiply ionized atoms can be found in a recent work by Horvat et al. [3].

In this paper we discuss the M- and N-shell satellites of Pd L\(\alpha\)1,2(L3M4,5) x-ray transitions excited by fast O7\(^{+}\) and Ne6\(^{+}\) ions [4], which were measured with high-resolution (\(\sim 1\) eV) using crystal diffraction spectrometer [5]. The measured x-ray spectra were compared with predictions of the multi-configuration Dirac-Fock (MCDF) calculations. The details concerning the MCDF calculations adopted here are described in Ref. [6]. In fact, the x-ray satellites of the L\(\alpha\)1,2(L3M4,5) transitions which are dominated by a small number of M-shell satellites, as for instance for O7\(^{+}\) ion impact on palladium \((m \leq 4)\), can be well reproduced by MCDF calculations (see Fig. 1). However, for more complex configurations with a higher number of spectator vacancies \((m > 4)\) the MCDF calculations become too complex numerically to be performed in practice. This is the case of Ne6\(^{+}\) impact on palladium (see Fig. 2), for which up to \(m = 7\) M-shell satellites have to be calculated in order to reproduce the measured x-ray spectrum of Pd L\(\alpha\)1,2(L3M4,5). We demonstrate, by using the general central limit theorem arguments, that the M-shell satellites for a higher number of spectator vacancies \((m > 4)\) can be approximated by a single Voigtian profile for which a mean energy and width can be obtained by extrapolating the MCDF calculations for configurations with a smaller number of spectator vacancies.
II. EXPERIMENT

The high-resolution measurements of the M- and N-shell satellites of Pd L$\alpha_{1,2}$ (L$_3$M$_4\alpha$) x-ray transitions excited by fast O$^{7+}$ and Ne$^{6+}$ ions have been performed [8] at the Philips cyclotron in the Paul Scherrer Institute (PSI) in Villigen, Switzerland, using a von Hamos high-resolution diffraction spectrometer [5]. The x-ray spectra of L$\alpha_{1,2}$ (L$_3$M$_4\alpha$) transitions were excited by O$^{7+}$ and Ne$^{6+}$ ion beams of energies 279 and 178 MeV, respectively, bombarding thin metallic palladium foils. The x-rays were measured by means of a high-resolution von Hamos spectrometer [5] with a precision of about 1 eV for studied Pd L-x-rays (\(~\sim\) 3 keV), including an experimental Gaussian resolution of about 0.7 eV. The von Hamos spectrometer was equipped with a quartz (111) crystal curved with a radius of 25.4 cm. The x-rays were measured with the CCD detector covering in one setting the x-ray energy range of about 50 eV. Consequently, the x-ray spectra of Pd L$\alpha_{1,2}$ (L$_3$M$_4\alpha$) transitions were measured for several settings of the spectrometer. The energy calibration of the spectrometer has been performed by measuring well resolved K$\alpha_{1,2}$ x-ray lines of vanadium excited by photons from x-ray tube with Cr anode.

III. RESULTS AND DISCUSSION

In order to interpret quantitatively the measured x-ray satellite structure of Pd L$\alpha_{1,2}$ (L$_3$M$_4\alpha$) transitions excited by Ne$^{6+}$ ions of energy 178 MeV, MCDF calculations involving up to seven M-shell spectator vacancies are needed. However, the MCDF calculations for multi-vacancy Pd L$\alpha_{1,2}$ (M$^{-m}$) configurations become, in practice, numerically intractable for \(m > 4\). For instance, the MCDF calculations for palladium for M$^{-4}$ configuration contain 244953 transitions (see Fig. 3) and for mixed M$^{-1}$N$^{-2}$ configuration 268210 transitions. Such numerical limitation of the applicability of the MCDF calculations asks for developing of alternative approximate methods to treat the complex satellite structures of x-rays excited in ion-atom collisions.

Following the idea presented in our earlier works (see Refs. [1] and [2]) on multiple ionization effects in ion-induced x-ray spectra we suggest that the x-ray profile for complex x-ray transitions can be well approximated by an effective single profile resulting from a convolution of natural Lorentzian and experimental Gaussian widths applied to the calculated MCDF x-ray multiplets consisting of large number of transitions. This observation is based on the firm ground of the general central limit theorem (see Ref. [7]) suggesting a Voigtian type profile as the limiting distribution in our case.

In order to verify this idea, the calculated MCDF structure of x-ray transitions for Pd L$\alpha_{1,2}$ (M$^{-m}$) configuration, convoluted with natural Lorentzian widths of individual transitions, assumed to scale approximately with a number of spectator vacancies \(m\) as \(\Gamma(m) = \Gamma(0) + 2\Gamma_{spec} \cdot m\), and experimental Gaussian widths of about 0.7 eV, are shown in Fig. 4. The effective widths of the resulting Voigtian profiles for complex x-ray multiplets are expected to follow approximately a simple scaling rule, namely,

\[
\Gamma(m) = \Gamma_0 + \alpha m + \beta \sqrt{m}
\]

where \(\alpha\) and \(\beta\) are constants which can be fitted for a calcu-
FIG. 3: The structure of Pd $\text{L}_{\alpha_1,2}(\text{M}^{-4})$ x-ray transitions obtained by using the MCDF calculations.

FIG. 4: A profile of $\text{L}_{\alpha_1,2}(\text{M}^{-4})$ x-ray transitions, as obtained by convolution of calculated MCDF transitions with natural Lorentzian widths, fitted by a single Voigtian profile. Note the smooth shape of the calculated (MCDF) profile and the reasonable good fit by a Voigtian.

FIG. 5: Fitted effective Lorentzian widths of Pd $\text{L}_{\alpha_1,2}(\text{M}^{-m})$ MCDF x-ray transitions for spectator vacancies in s-, p-, and d-states, which are well fitted by the approximate formula $\Gamma(m) = \Gamma_m(0) + \alpha m + \beta \sqrt{m}$. Assumed natural widths for $\text{M}^{-m}$ vacancy configurations with vacancies in 3p and 3d states are also shown in the figure.

FIG. 3: The structure of Pd $\text{L}_{\alpha_1,2}(\text{M}^{-4})$ x-ray transitions obtained by using the MCDF calculations.

FIG. 4: A profile of $\text{L}_{\alpha_1,2}(\text{M}^{-4})$ x-ray transitions, as obtained by convolution of calculated MCDF transitions with natural Lorentzian widths, fitted by a single Voigtian profile. Note the smooth shape of the calculated (MCDF) profile and the reasonable good fit by a Voigtian.

IV. CONCLUSIONS

A novel approximate description of x-ray spectra for complex multi-vacancy $\text{M}^{-m}$ configurations has been proposed, which is based on the general central limit theorem. The calculated profiles of $\text{L}_{\alpha_1,2}(\text{M}^{-m})$ x-ray transitions in palladium are well described by the proposed model. Complex multi-vacancy configurations, involving up to about ten spectator vacancies, $m \leq 4$ in our case. This formula, which uses once more the arguments of the GCLT theorem, has been derived by summing up the natural width $\Gamma(m)$ of the Lorentzian distribution of x-ray transition energy and the width of the binomial distribution of a number of vacancies randomly distributed in the M-shell. In fact, the variance of the binomial distribution $\sigma^2(m) = N_M p_M (1 - p_M)$, where $N_M$ is a number of electrons in the M-shell, scales for $p_M = m/M \ll 1$ as $\sigma^2(m) \approx m$ yielding the following approximate estimate for a width: $\Gamma_{\text{bin}}(m) \approx \sqrt{m}$.

The fitted effective widths of x-ray transitions for $\text{M}^{-m}$ configurations, shown in Fig. 5, fully justify the model assumed. Consequently, such a parameterization of Voigtian widths for complex multi-vacancy configurations, combined with a known linear parameterization of their mean energies adopted in the average MCDF binomial model (Ref. [6]) of x-ray satellite structure, allows one to describe in a realistic way the complex x-ray spectra excited by heavy ions, which include much more spectator vacancies than can be treated numerically in an exact way using the MCDF calculations.

The present findings open a possibility to describe a complex satellite structure of Pd $\text{L}_{\alpha_1,2}(\text{L}3\text{M}4\text{S}5)$ transitions such as in the discussed x-ray spectra excited by Ne$^{6+}$ ions of energy 178 MeV exhibiting M- and N-shell satellites. However, a final interpretation of such x-ray spectra needs further MCDF calculations for the observed L-shell hypersatellite structure overlapping with M-shell satellites. Such MCDF calculations of L-shell hypersatellites are in progress.
vacancies, can be treated within this approach, which significantly extends the applicability of the MCDF calculations for describing multiple ionization effects in x-ray spectra excited by heavy ions.

Acknowledgments

This work was supported by the Polish State Committee for Scientific Research under the Grant No. 1P0301326 and the Swiss National Science Foundation.