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

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The x-ray satellite structure of Pd $\alpha_{1,2}(L_3M_{4,5})$ transition excited by an impact of $O^{7+}$ and $Ne^{6+}$ 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 $\alpha_{1,2}(M_{7m})$ 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 $\alpha_{1,2}(L_3M_{4,5})$ x-ray transitions excited by fast $O^{7+}$ and $Ne^{6+}$ 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 $\alpha_{1,2}(L_3M_{4,5})$ transitions which are dominated by a small number of M-shell satellites, as for instance for $O^{7+}$ 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 $Ne^{6+}$ 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 $\alpha_{1,2}(L_3M_{4,5})$. 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.
The high-resolution measurements of the M- and N-shell satellites of Pd Lα1,2(L3M4,5) x-ray transitions excited by fast O7+ and Ne6+ 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 Lα1,2(L3M4,5) transitions were excited by O7+ and Ne6+ 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 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 Lα1,2(L3M4,5) transitions were measured for several settings of the spectrometer. The energy calibration of the spectrometer has been performed by measuring well resolved Kα1,2 x-ray lines of vanadium excited by photons from x-ray tube with Cr anode.

II. EXPERIMENT

In order to interpret quantitatively the measured x-ray satellite structure of Pd Lα1,2(L3M4,5) transitions excited by Ne6+ 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 Lα1,2 (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. 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 V oigtian profile resulting from a convolution of natural Lorentzian and experimental Gaussian widths.

In order to verify this idea, the calculated MCDF structure of x-ray transitions for Pd Lα1,2 (M−m) configuration, convoluted with natural Lorentzian widths of individual transitions, assumed to scale approximately with a number of spectator vacancies m as \( \Gamma(m) = \Gamma(0) + 2\Gamma_{\text{spec}} \cdot m \), 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 L$\alpha_{1,2}$($M^{-4}$) x-ray transitions obtained by using the MCDF calculations.

FIG. 4: A profile of L$\alpha_{1,2}$($M^{-4}$) 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 L$\alpha_{1,2}$($M^{-m}$) 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 M$^{-m}$ vacancy configurations with vacancies in 3p and 3d states are also shown in the figure.

IV. CONCLUSIONS

A novel approximate description of x-ray spectra for complex multi-vacancy M$^{-m}$ configurations has been proposed, which is based on the general central limit theorem. The calculated profiles of L$\alpha_{1,2}$ (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: $\Gamma_{\text{bin}}(m) \propto \sqrt{m}$.

The fitted effective widths of x-ray transitions for 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 L$\alpha_{1,2}$($L_3M_4,5$) 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.

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