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Spectroscopic investigations of *L*-shell ionization in heavy elements by electron impact

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A B S T R A C T

The absolute L subshell specific electron impact ionization cross sections near the ionization threshold ($16 < E < 45$ keV) of lead and thorium are obtained from the measured L X-ray production cross sections. Monte Carlo simulation is done to account for the effect of the backscattered electrons, and the final experimental results are compared with calculations performed using distorted wave Born approximation and the modified relativistic binary encounter Bethe model. The sensitivity of the results on the atomic parameters is explored. Observed agreements and discrepancies between the experimental results and theoretical estimates, and their dependence on the specific atomic parameters are reported.

1. Introduction

Importance of electron impact excitation and ionization data in various materials analysis techniques such as electron probe microanalysis (EPMA), and Auger electron spectroscopy (AES), etc. need not be overemphasized. Precise and accurate knowledge of the corresponding cross sections, either in the form of look-up tables or function dependence on impact energy of electron probe used, is needed for such analysis. The inner shell ionization probabilities, extracted from the above-mentioned data, are also pivotal to many other material analysis techniques, apart from their importance in understanding the physical process of ionization in multi-electron bound systems [1].

Inner shells of atoms can be excited by knocking off the bound electrons to the continuum or unfilled quasi-bound orbitals. Vacancies thus created are filled by the electrons from the outer shells, resulting in the emission of photons or electron. In addition, migration of vacancies through Coster–Kronig (CK) transitions among different subshells (*L*-shell and above) as well as to other inner shells leads to photon emission with different energies and yields. From the observation and quantitative estimation of the related photon yield with high precision, the inner shell ionization cross sections can be obtained, in principle, utilizing the known or pre-determined parameters, such as the fluorescence yield, CK transition probabilities and the sublevel-specific radiative decay probabilities from experiments or theoretical estimates. These important parameters are collectively known as the atomic relaxation parameters.

The above mentioned relaxation parameters are obtained from experiments or from theoretical estimates [2] and are available from various data bases. However, some of these parameters are quoted with large uncertainties due to various processes involved. For example, the fluorescence yield for a specific subshell depends on the primary vacancy distributions, which in turn depends on the mode of vacancy creation in the subshell. It is also expected that migration of vacancies through CK transition

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would alter the primary vacancy distributions and hence the fluorescence yield.

Photon emission by electron impact is also possible as a multistep process through Auger transition, followed by creation of vacancy in the inner subshells by virtual photons [3]. The above process involving virtual photons can only be accounted for by invoking quantum electrodynamics and the associated electromagnetic interaction between the bound electrons, which involve both Coulomb interaction and the magnetic interaction due to the moving electrons. In case of lighter elements, the motion of inner shell electrons are in the non-relativistic regime ($v/c\rightarrow 0$), and therefore, the quantum effects due to magnetic interaction become negligible. Thus, inclusion of the Coulomb interaction alone in estimating the electron impact ionization cross sections results in reasonable agreement with the experimental results for the lighter atoms. For heavier atoms like the ones considered in this experiment, the magnetic interaction can no longer be ignored, and related estimates of the electron impact ionization cross sections should take magnetic interaction into account as well. Theoretical estimates based on above have been done in recent times for Gold (Au) [4].

Cross sections for ionization of inner shells by electron impact can be estimated using the scattering theory for inelastic collisions and applying relativistic kinematics. Static field approximation, direct and local exchange interaction are applied in calculating the transition amplitudes for inelastic scattering of electrons by the bound atomic electrons [5]. For interaction energies from the ionization threshold (U) to approximately 20U, the partial wave expansions in the scattering calculations are taken as distorted waves, modified from the plane waves by the residual interaction. However, in the distorted wave Born approximation (DWBA) formalism, the transverse interaction is neglected in calculating the transition matrix elements and therefore, the DWBA estimates can be considered as semirelativistic [5].

A fully relativistic DWBA formalism was developed by Keller et al. [6] which was successful in the study of relativistic $(e,2e)$ processes leading to inner shell ionization at relativistic electron energies (up to ~ 500 keV) and strong fields of high Z elements (see review by [7]). While this formalism gives more insight into the details of the $(e,2e)$ experiments, including electron coincidence, angular and energy analysis by extracting the triple differential cross sections of the process, we have adopted the semi-relativistic DWBA formalism [5] in the interpretation of our results because of the following reasons: (a) low energy domain (near ionization threshold to ~ 3–$4U$) chosen for our experiment, and (b) measurement and interpretation of absolute subshell resolved ionization cross-sections carried out in this work.

Experiments on electron impact ionization which were done earlier were focused primarily on K-shell ionization cross section, while L and M shell ionization data were seldom reported [8]. One of the major problems faced in the interpretation of experimental results based on established theories is that the extracted subshell specific ionization cross sections do not agree with the theoretical estimates for all the subshells. Recently, many authors have reported L X-ray production cross sections for a few elements, and validation of various theoretical models are done using the data. Comparison between theory and experimental data on L-subshell production cross sections in Gadolinium (Gd, $Z=64$) and Tungsten (W, $Z=74$) were done by Wu et al. [9]. Their experimental results on L_α and L_β lines agree reasonably well with DWBA theory including exchange interaction for W but deviates by 15–20% in case of Gd. Similar comparative studies were done by Varea et al. [10] on Hf, Ta, Re, Os, Au, Pb, and Bi, where experimental results for L_α and L_β lines are explained well by DWBA theory for Ta, Os, Au, Pb and Bi but are lower than the theoretical estimates by $\sim 35\%$ for Hf and Re.

In the present work, the $L_\alpha, L_\beta, L_\gamma$ production cross sections in lead and thorium are measured, and the results are converted to the subshell specific ionization cross sections. Because of the finite thickness of the target materials, single collision condition within the target has to be ensured. In arriving at the ionization cross sections from the production cross sections, corrections due to multiple collisions per beam traverse were done using a Monte Carlo simulation procedure. Parameter dependence in extracting the ionization cross sections are also explored to check the sensitivity to parameter variations. The cross sections obtained from experiment are compared with (a) the theoretical results based on the DWBA formalism [5], as obtained from the PENELOPE [11] code, and (b) the modified relativistic binary encounter Bethe (MRBEB) [12,13] model-based estimates. To the best of our knowledge, the subshell specific ionization cross section for all the L-subshells of thorium are reported here for the first time at the energy values near the corresponding ionization threshold.

2. Experimental details

The experimental set-up consists of an in-vacuum energy dispersive spectrometer with a focusable electron gun(up to 50 keV), electrically cooled silicon PIN diode based X-ray detector, thin film target holder, and Faraday cup. The X-ray detector was placed in the meridian plane at 55° with respect to the beam axis. The pressure maintained inside the vacuum chamber was 5×10^{-7} mbar. Details of the experimental arrangement are described elsewhere in detail [14].

The X-ray detector is specifically placed at an angle 55° so as to avoid the anisotropy effect which may arise due to alignment of vacancy along the incident beam axis [15]. X-rays emitted from the aligned vacancy states will have the anisotropic distribution given as $I=\frac{I_\circ}{4\pi}(1+\alpha\kappa A_{20}P_2(\cos\theta))$, where $I_\circ$ is the total intensity of X-ray line, α is a constant depending on total angular momentum of initial and final vacancy state, κ is the Coster–Kronig correction factor, $P_2(\cos\theta)$ is the second order Legendre polynomial and A_{20} is the alignment parameter for given subshell [16]. Vanishing of $P_2(\cos\theta)$ at 55° makes the intensity distribution independent of alignment parameter.

The targets used in the experiment were made by using two different techniques. Self-supporting lead targets