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Measurement of K–L radiative vacancy transfer probabilities in rare earth elements bombarded with 3 MeV-4 MeV protons

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Abstract. K-shell X-ray intensity ratios for rare-earth elements were measured aftr irradiation with proton beams having energies between 3 MeV and 4 MeV. Using the X-ray intensity ratios, the radiative vacancy transfer probabilities from the shell K to the L sub-shells were determined. The experimental data were compared to theoretical predictions and semiempirical fits for X-ray line intensities and radiative vacancy transfer probabilities. The results showed a good agreement between theory and experiment.

Keywords: Radiative vacancy transfer; intensity ratio, rare earths, proton beam.

INTRODUCTION

One of the main difficulties in the measurement of concentration of rare earth elements in several kinds of samples using Particle Induced X-ray Emission (PIXE) is the overlap of the L-lines emitted by these elements with the K X-rays from more abundant lighter elements, such as Mn and Fe. Therefore, the possibility of using proton beams with energies higher than the usual 2 MeV to 3 MeV to induce the K X-rays of the rare earths might be explored, although information about ionization cross sections or other atomic parameters is still scarce. In this regard, accurate knowledge of X-ray relative intensity ratios, fluorescence yields and ionization cross-sections is increasingly important, both in the analytical ambit and in fundamental studies of atomic and nuclear processes. Because of this, the present work is aimed to measure the K–L radiative vacancy transfer probabilities for a selected group of rare earth elements irradiated with protons in the energy range 3 MeV to 4 MeV. A further comparison with published predictions and measurements is also carried out.

RADIATIVE VACANCY TRANSFER PROBABILITIES

When an ion, X-ray photon or electron impinges on a target atom, it is possible for an electron to be ejected out of the atom creating a vacancy in the K shell. The vacancy created in the K shell is filled through a radiative or a non-radiative transition. The total vacancy transfer probability for the K shell to any of the L shells, η_{KLi}, is the sum of the radiative vacancy transfer probability $\eta_{KLi}(R)$ and non-radiative vacancy transfer probability $\eta_{KLi}(A)$

$$\eta_{KL_i} = \eta_{KL_i}(R) + \eta_{KL_i}(A) \qquad (1)$$

The radiative vacancy transfer probability is given by

$$\eta_{KL_i} = \omega_K \left[\frac{I(KL_i)}{I_K(R)} \right]. \qquad (2)$$

Here, $I(KL_i)$ is the K to L_i-X-ray intensity, $I_K(R)$ is the total intensity of K-X-rays, while ω_K is the K shell fluorescence yield. As the radiative transition from L_1 to K is forbidden, it is only necessary to determine the radiative vacancy transfer probabilities η_{KL2} and η_{KL3} for L_2 to K and for L_3 to K, respectively, as follows:

$$\eta_{KL_2} = \omega_K \left[\frac{I(K_{\alpha 2})}{I(K_{\alpha 1})}\right]\left\{\left[1+\frac{I(K_{\alpha 2})}{I(K_{\alpha 1})}\right]\left[1+\frac{I(K_{\beta})}{I(K_{\alpha})}\right]\right\}^{-1}, \quad (3)$$

$$\eta_{KL_3} = \omega_K \left\{\left[1+\frac{I(K_{\alpha 2})}{I(K_{\alpha 1})}\right]\left[1+\frac{I(K_{\beta})}{I(K_{\alpha})}\right]\right\}^{-1}, \quad (4)$$

where $I(K_{\alpha 2})/I(K_{\alpha 1})$ and $I(K_\beta)/I(K_\alpha)$ are the K-X-ray intensity ratios of the corresponding lines. From these equations it is apparent that by measuring the intensity of the K X-ray lines it is possible to determine the radiative vacancy transfer probabilities η_{KLi}. Although numerous measurements of these quantities have been published when photons or electrons are used as primary radiation, finding that the theoretical predictions [1, 2] usually agree well with experiments, information obtained using ions is largely unknown, except for lighter elements. Kasagi et al. [3] measured the radiative vacancy transfer probabilities for elements with atomic numbers between 62 and 82, irradiated with 3.5 MeV protons. Ximeng et al. [4] published a similar study, but using 3 MeV protons on elements with $39 \leq Z \leq 68$. No studies considering the variation of the beam energy on rare earth elements have been published. It must be mentioned that different results should be expected for ions than those for electrons or photons, due to the increased probability of multiple ionization after ion bombardment, thus altering the number of L-shell electrons available for K vacancy filling.

EXPERIMENT

The experimental setup is similar to that used previously for the measurement of L X-ray production cross sections [5, 6]. In this case, thick foils (thickness between 125 μm and 250 μm) of the pure elements (Ce, Nd, Gd, Sm, Ho), were irradiated using protons with energies in the range 3 MeV to 4 MeV, in 0.1 MeV steps. X-rays were registered by a Canberra LEGe detector, covered with a 35 μm thick Al absorbing filter. The detection efficiency was measured using a calibrated ^{241}Am point source (Amersham, England). The proton beam was produced by the 9SDH-2 Pelletron accelerator at Instituto de Física, UNAM. To have a reference of the total incident number of protons, an Au layer was deposited onto the surface of the foils. The thicknesses of these layers, measured separately through Rutherford Backscattering (RBS) of a 0.7 MeV proton beam, were of the order of 50 μg/cm^2. The Au L_α lines were then used as reference for the beam integrated charge after comparison with the X-ray emission of a thin Au MicroMatter standard. Thus, the K X-rays intensity ratios between lines K_i and K_j is given by:

$$\frac{I(K_i)}{I(K_j)} = \frac{\varepsilon(K_j)N_X(K_i)\beta(K_j)}{\varepsilon(K_i)N_X(K_j)\beta(K_i)}, \quad (5)$$

where $\varepsilon(K_i)$ is the detection efficiency for line K_i, $N_X(K_i)$ is the number of X-ray photons registered in the spectrum, and $\beta(K_i)$ is an X-ray transmission factor, including absorption in the Au layer and in the thick rare earth substrate. The X-ray mass attenuation coefficients were obtained with the XCOM computer code [7], while ion stopping powers were evaluated using the SRIM program [8]. Spectra were analyzed with the QXAS code [9], using the included line shape correction, to consider the appropriate peaks in the manner explained by Papp [10].

RESULTS AND DISCUSSION

Fig. 1 presents a spectrum of the K X-rays of Dy, after irradiation with a 3.3 MeV proton beam. It is apparent that the $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1}$ and $K_{\beta 2}$ lines are resolved. Then, the radiative vacancy transfer probabilities η_{KL2} and η_{KL3} for Gd as a function of the energy are plotted in fig. 2. It is possible to see that no trend in these quantities, within experimental uncertainties, is observed when the beam energy varies. This observation was repeated for the other four elements. Therefore, fig. 3 shows the averages η_{KL2} for all these elements, compared to the theoretical predictions from the tables published by West [11] and those of Scofield, based on the relativistic Hartree–Slater theory [12], and the experimental results of Kasagi et al. [3] for Gd, Dy and Ho, and those of Ximeng et al. [4], for Ho. The same is shown for η_{KL3} in fig. 4.

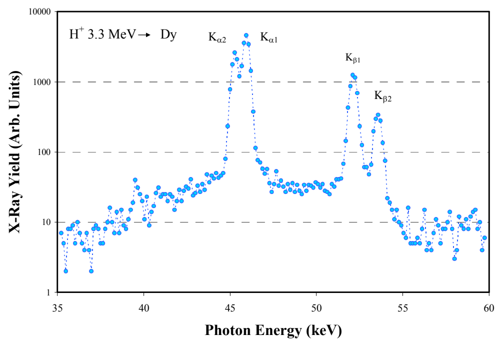

FIGURE 1. Spectrum of the K X-rays of Dy, after irradiation with a 3.3 MeV proton beam.

In all cases, the theoretical predictions of radiative vacancy transfer probabilities using different databases are in excellent agreement.

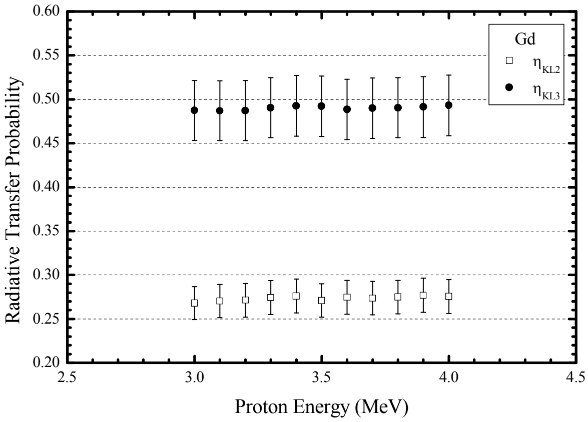

FIGURE 2. Measured radiative vacancy transfer probabilities η_{KL2} and η_{KL3} for Gd as a function of the proton incident energy.

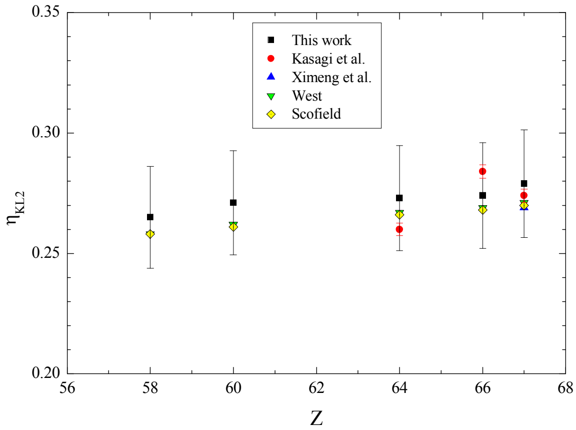

FIGURE 3. Comparison of radiative vacancy transfer probabilities η_{KL2}. Experimental data is taken from the works by Kasagi et al. [3], and Ximeng et al. [4], while theoretical predictions were obtained using the tables published by West [7] and Scofield [8].

In Fig. 3 it is apparent that the experimental data for radiative vacancy transfer probabilities η_{KL2} from this work agree well, within experimental uncertainty, with all the other results. However, in all the cases the theoretical predictions lie below the present experimental points.

A similar situation is observed for the η_{KL3} vacancy transfer probabilities, but in this case the experimental results by Kasagi et al. are even lower than those predicted by the theories.

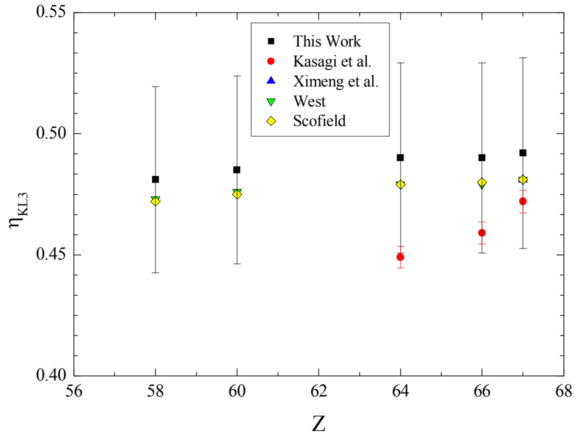

FIGURE 4. Same as Fig. 2, but for the radiative vacancy transfer probabilities η_{KL3}.

Although the works by Kasagi et al. [3] and Ximeng et al. [4] do not discuss any particular reason of possible disagreements between the theory and their experiments, one explanation may be the occurrence of multiple ionizations in the higher electronic shells, which is a weaker effect when electrons or photons are used as primary radiation.

CONCLUSIONS

This work stresses the fact that very little information exists for the emission of K X-rays of rare earth elements under ion beam irradiation. Intensity ratios and other quantities related to them, such as the radiative vacancy transfer, provide useful information to understand atomic inner-shell processes. No evidence for a dependence of the radiative vacancy transfer probabilities on the proton incident energy was noticed. It was found that experimental averages for the radiative vacancy transfer probabilities η_{KL2} and η_{KL3} have higher values than those predicted by the theoretical models, although this is not a significant conclusion due to the large uncertainties. A possible explanation is that those models do not consider the multiple vacancy creation in higher electronic shells during the ion impact. Improvements in the experimental setup must be performed, to reduce the uncertainty. Further investigations will include measurement of K X-ray production cross sections in the rare earth elements, and the use of heavier ions as projectiles.

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