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 after 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, \( \eta_{KL_i} \), is the sum of the radiative vacancy transfer probability \( \eta_{KL_i} (R) \) and non-radiative vacancy transfer probability \( \eta_{KL_i} (A) \)

\[
\eta_{KL_i} = \eta_{KL_i} (R) + \eta_{KL_i} (A) \tag{1}
\]

The radiative vacancy transfer probability is given by

\[
\eta_{KL_i} = \omega_k \frac{I(KL_i)}{I_K(R)} \tag{2}
\]

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 \( \omega_k \) is the K shell...
fluorescence yield. As the radiative transition from L₁ to K is forbidden, it is only necessary to determine the radiative vacancy transfer probabilities \( \eta_{KL2} \) and \( \eta_{KL3} \) for L₂ to K and for L₃ to K, respectively, as follows:

\[
\eta_{KL2} = \frac{\omega_k}{(1 + I(K_{\alpha2})/I(K_{\alpha1}))^{1/2}} \left[ \frac{I(K_{\alpha2})}{I(K_{\alpha1})} + \frac{I(K_{\beta})}{I(K_{\alpha2})} \right]^{-1}, \quad (3)
\]

\[
\eta_{KL3} = \frac{\omega_k}{(1 + I(K_{\alpha3})/I(K_{\alpha1}))^{1/2}} \left[ \frac{I(K_{\alpha3})}{I(K_{\alpha1})} + \frac{I(K_{\beta})}{I(K_{\alpha3})} \right]^{-1}, \quad (4)
\]

where \( I(K_{\alpha i})/I(K_{\alpha1}) \) and \( I(K_{\beta})/I(K_{\alpha i}) \) 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 \( \eta_{KL2} \) and \( \eta_{KL3} \). 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 four elements. Therefore, fig. 3 shows the averages \( \eta_{KL2} \) 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 \( \eta_{KL3} \) 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 \( \eta_{KL3} \) in fig. 4.

**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 Kα1, Kα2, Kβ1 and Kβ2 lines are resolved. Then, the radiative vacancy transfer probabilities \( \eta_{KL2} \) and \( \eta_{KL3} \) 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 \( \eta_{KL2} \) 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 \( \eta_{KL3} \) in fig. 4.

![FIGURE 1. Spectrum of the K X-rays of Dy, after irradiation with a 3.3 MeV proton beam.](image-url)
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 $\eta_{KL2}$ and $\eta_{KL3}$ for Gd as a function of the proton incident energy.

**FIGURE 3.** Comparison of radiative vacancy transfer probabilities $\eta_{KL2}$. 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 $\eta_{KL2}$ 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 $\eta_{KL3}$ vacancy transfer probabilities, but in this case the experimental results by Kasagi et al. are even lower than those predicted by the theories.

**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 $\eta_{KL2}$ and $\eta_{KL3}$ 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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