L X-ray anisotropy and L_3-subshell alignment in rare earths induced by ^{12}C ion impact

O.G. de Lucio and J. Miranda
Instituto de Física, Universidad Nacional Autónoma de México,
Apartado Postal 20-364, 01000, México D. F., México

Recibido el 4 de noviembre de 2003; aceptado el 1 de marzo de 2004

The alignment parameter A_{20} of the L_3-subshell of Ce, Nd, Eu, Dy, and Yb has been calculated by measuring the anisotropic emission of the L_l X-ray line induced by ^{12}C ion impact at 9 MeV bombardment energy. A new Coster-Kronig transition probabilities database is used, as well as more recent theories for ionization cross sections. These experimental results are in better agreement with the theory than previously published data.

Keywords: Anisotropy; X-ray emission.

Midiendo la emisión anisotrópica de rayos X de la línea L_l inducidos por impacto de iones de ^{12}C a 9 MeV, se calculó el parámetro de alineación A_{20} para la subcapa L_3 de los elementos Ce, Nd, Eu, Dy e Yb. Se utilizó una nueva base de datos para obtener las probabilidades de transiciones Coster-Kronig así como teorías más recientes para calcular las secciones eficaces de ionización. Se observa que los resultados experimentales obtenidos en este trabajo concuerdan mejor con la teoría en comparación a resultados obtenidos con anterioridad.

Descriptores: Anisotropía; emisión de rayos-X.

PACS: 32.30.Rj; 32.70.-n; 32.80.Hd

1. Introduction

Ionization of the L_3 subshell by proton and helium impact has been studied to a great extent, but much less has been investigated for heavy ion impact. A problem of particular interest is that the ionized target atom can actually be aligned in the direction of the incident ion beam, thus setting a preferential symmetry. The alignment results from the fact that the ionization cross section has different values for different projections of the total angular momentum of the ionized atom along the beam direction. The study of such alignment generally involves measurements of the angular distribution [1-5] or polarization of the induced X-rays [6-10]. This alignment provides information on the process of ionization, the wavefunctions of inner-shell electrons, and results in a sensitive testing ground for theoretical models.

Moreover, there has been an extensive research on L-shell X-ray production cross sections by ion impact [11], but no consideration of the aforementioned asymmetry has been taken, hitherto. To be accurate, quantitative analysis based on X-Ray emission may require this correction.

The alignment is defined as [12]:

$$A_{20} = \frac{\sigma_{L3}\left(\frac{3}{2}\right) - \sigma_{L3}\left(\frac{1}{2}\right)}{\sigma_{L3}\left(\frac{3}{2}\right) + \sigma_{L3}\left(\frac{1}{2}\right)} \tag{1}$$

where $\sigma_{L3}(3/2)$, $\sigma_{L3}(1/2)$ denotes the ionization cross section corresponding to the magnetic substates $|m_j| = 3/2, 1/2$, respectively.

For characteristic dipole radiation, the intensity I as a function of the emission angle θ relative to the direction of the primary beam is given by [12]:

$$I(\theta) = I(90^\circ)(1 - P\ \cos^2\theta) \tag{2}$$

where P, known as the degree of polarization, is defined as:

$$P = \frac{3\alpha A_{20}}{\alpha A_{20} - 2} \tag{3}$$

The constant α depends on the angular momentum of the initial (J) and final state (J_f) and it can be calculated as:

$$\alpha = (-1)^{J+J_f+1}\sqrt{\frac{3}{2}(2J+1)}\begin{Bmatrix} 1 & J & J_f \\ J & 1 & 2 \end{Bmatrix} \tag{4}$$

the factor

$$\begin{Bmatrix} 1 & J & J_f \\ J & 1 & 2 \end{Bmatrix}$$

corresponds to a Racah coefficient.

From previous investigations of the L_3-subshell alignment for medium and heavy elements by proton impact, it seems that the experimental data are in agreement with theoretical PWBA predictions. However, in the case of heavy ions it deviates from those calculations; this disagreement becomes larger in the low-velocity region, even in the case of protons [1,9].

Furthermore, X-ray emission involves non-radiative electronic transitions known as Coster-Kronig. The existing databases are rather old [1,13-15], and it has been shown that they are not accurate. Therefore, it is convenient to use more recent and reliable data for this magnitude.

The aim of this work is to investigate the alignment of the induced L X-Ray by ^{12}C ion bombardment on Ce, Nd, Eu, Dy, and Yb targets in the low velocity region. Of particular interest is the L_l line, which is expected to show a high degree of anisotropy, because for this transition the α coefficient has a maximum value.

2. Experimental

Experiments were carried out with a ^{12}C ion beam from the 9SDH-2 NEC Pelletron Accelerator at the Instituto de Física, UNAM. The beam energy was 9 MeV, and it was collimated at the entrance of the scattering chamber, to a diameter of about 1.5 mm. The scattering chamber allowed measurement of the L X-ray spectrum from an angle of 26° up to an angle of 66° in 4° steps. The X-ray detector was an XR-100CR AMPTEK, with 158 eV FWHM resolution at 5.9 keV. The detector was mounted on a goniometer inside the chamber, so the angles could be measured accurately. The photons entering the detector were also colimated to a size of about 0.5 mm diameter. This type of detector was chosen because of its size, which is small enough to fit inside the analysis chamber, so both the detector and the sample are in vacuum during the experiments, thus reducing the X-ray attenuation. A diagram of the experimental setup is sketched in Fig. 1.

Targets were in the form of thin films ($\approx 100\ \mu$g/cm^2) deposited onto pyrolitic carbon discs by evaporating rare earths fluorides in vacuum. Thicknesses of the films were measured with Rutherford backscattering of helium ions [16].

A typical spectrum is shown in Fig. 2; the number of counts in the peak of the L_l line at the maximum was generally of the order 10^3. The emission of the L_γ lines and the unresolved $L_{\beta 1,3,4}$ line is expected to be isotropic, since they result from initial $J = 1/2$ vacancies for which the α coefficient (Eq. 4) vanishes and then equation 2 exhibits no angular dependence. Therefore, their intensities were used to normalize the intensities of the anisotropically emitted lines. In order to check the validity of this statement L_γ and $L_{\beta 1,3,4}$ lines were analized following the same procedure used for the other lines. Results for the $L_{\beta 1,3,4}$ Nd line are shown in Fig. 3. Both lines were found to be isotropic within the uncertainty range.

From Eq. 2, a linear dependence of $I(\theta)$ on $\cos^2\ \theta$ is anticipated. The intensity angular distributions for the L_l X-Ray Line of Nd and Yb are depicted in Fig. 4 and Fig. 5, respectively. Therefore, the slope of the linear fit represents the degree of polarization, from which it is possible to calculate the alignment parameter, as in Eq. 3.

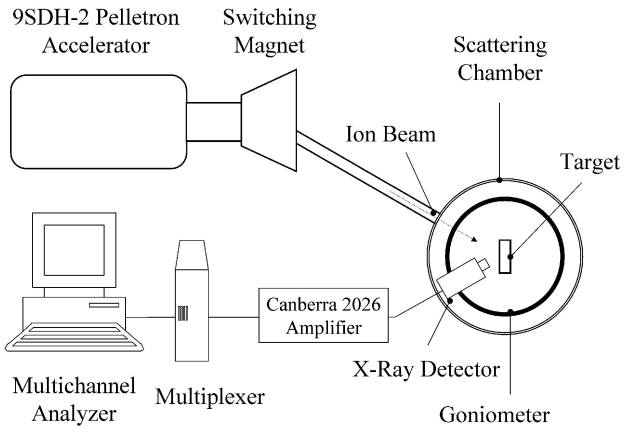

FIGURE 1. Experimental setup.

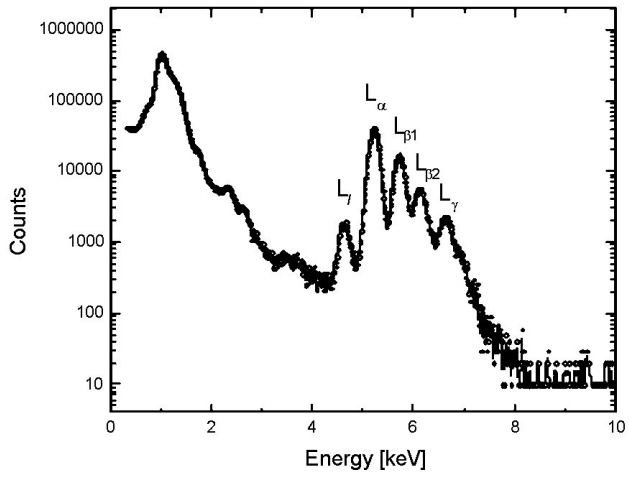

FIGURE 2. X-Ray spectrum from a Nd target for ^{12}C projectile.

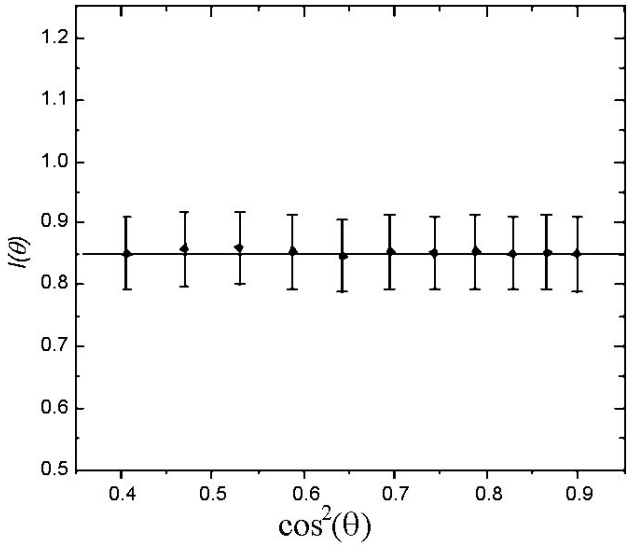

FIGURE 3. Normalized intensity of the Nd $L_{\beta 1,3,4}$ line for 9MeV ^{12}C impact.

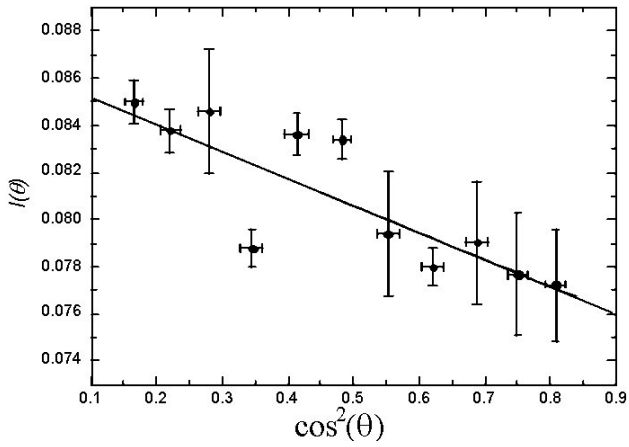

FIGURE 4. Measured angular distribution of the Nd L_l line, normalized to the sum of the L_γ and $L_{\beta 1,3,4}$ isotropic lines for 9 MeV ^{12}C ion bombardment.

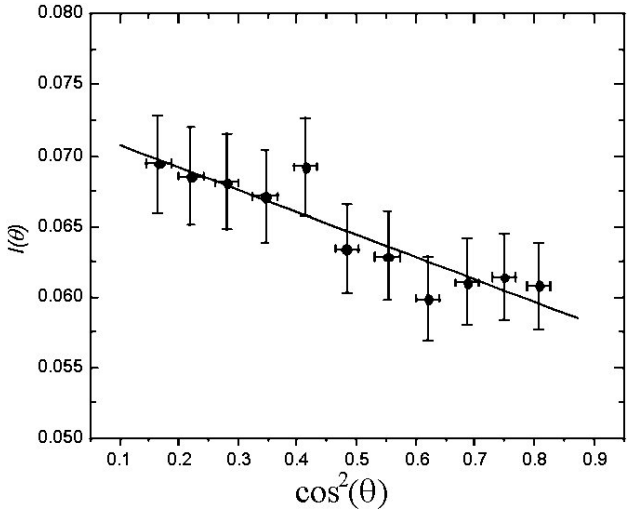

FIGURE 5. Measured angular distribution of the Yb L_l line, normalized to the sum of the L_γ and $L_{\beta 1,3,4}$ isotropic lines for 9 MeV ^{12}C ion bombardment.

3. Results and Discussion

The polarization values obtained from the L_l line are summarized in Table I. To convert the degree of polarization into the alignment parameter it is necessary to calculate the α coefficient. As this line is the result of a vacancy transition from $J = 3/2$ to $J_f = 1/2$, the correspondent value is $\alpha = 1/2$. The measured alignment values $A_{20,\exp}$ are also presented in Table I. These values are influenced by Coster-Kronig transitions, because a vacancy in the L_3 subshell may be created by a process different from direct ionization, like a Coster-Kronig transition from vacancies induced in L_1 or L_2 subshells by ion impact. As the initial state for such transition corresponds to $J = 1/2$, these transitions have zero alignment. It is possible to describe the direct ionization vacancy formation by the ionization cross section σ_{Li} for each subshell, and Coster-Kronig transitions by the f_{ij} transition rates. Then, the total cross section σ_{L3}^{tot} for L3 vacancy creation can be expressed by:

$$\sigma_{L3}^{tot} = \sigma_{L3} + f_{23}\sigma_{L2} + (f_{12}f_{23} + f_{13})\sigma_{L1} \qquad (5)$$

The alignment A_{20} of an induced L_3 vacancy is given by

$$A_{20} = A_{20,\exp}\frac{\sigma_{L3}^{tot}}{\sigma_{L3}} \qquad (6)$$

Atomic parameters f_{ij} were taken from the Elam *et al.* [17] database. The cross sections σ_{Li} were calculated using the ISICS program [18], considering the United Atom approximation in the ECPSSR picture [19,20].

Results obtained may be compared with some previous experimental work, as well as theoretical calculations. Figure 6 shows a compilation of existing data on the alignment parameter A_{20} induced by carbon impact as a function of $V^2 = (v/v_{L3})^2$; where v is the projectile velocity and v_{L3} the orbiting electron velocity, calculated from $v_{L3}^2 = (2I)/(m_e)$; with I the L_3 binding energy, and m_e the electron mass. For comparison, calculations by Jitschin *et al.* [1] are included in Fig. 6.

The curves presented in Fig. 6 corresponds to ECPSSR and PWBA predictions; as can be seen, both curves are in good agreement, except for the low velocity region, which corresponds to $V^2 < 0.3$ values. Previous experiments developed by Pálinkás *et al.* [21], Jitschin *et al.* [1], and Stachura *et al.* [22] corresponding to C ion bombardment on several targets, are also shown for comparison. It can be seen that, in general, previous results only agree with theoretical predictions in the high velocity region, that is, $0.07 \leq V^2 \leq 0.2$. Opposite to this, experimental results from the present work seem to agree fairly well with theoretical predictions. Main differences between previous works and the present one are, first, the atomic parameters database. In the past, the Coster-Kronig coefficients f_{ij} were taken from the Krause database [13], which has been corrected over the years and now a new compilation exists [17], which is the one used in this work. Second, there is also a difference in the calculations of the σ_{Li} cross sections. In the past these calculations did not take into account effects which appear as a result of bombarding with heavy ions (as is the case of C ion bombardment), and now it is possible to calculate corrections as the Molecular Orbital formation in the modified ECPSSR theory [23] or the United Atom correction [24], which is used in this work.

TABLE I. Polarization $P(L_l)$ and alignment parameter A_{20} of the L_l X-Ray line for ^{12}C ion impact at 9 MeV bombardment energy.

Target	$P(L_l)$ (%)	$A_{20,exp}$ (%)	κ^{-1}	A_{20} (%)
Ce (Z=58)	13 ± 2.5	-18 ± 3.6	1.07	-19 ± 3.8
Nd (Z=60)	16 ± 3.1	-23 ± 4.6	1.07	-25 ± 4.9
Eu (Z=63)	28 ± 2.9	-42 ± 4.7	1.07	-45 ± 5.0
Dy (Z=66)	14 ± 2.7	-20 ± 4.0	1.08	-22 ± 4.3
Yb (Z=70)	24 ± 3.3	-34 ± 5.2	1.05	-36 ± 5.5

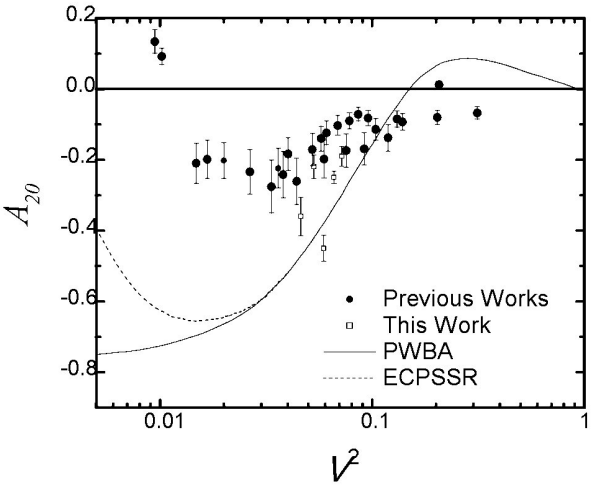

FIGURE 6. L_3-subshell alignment A_{20} for different collision systems.

4. Conclusion

The results obtained in this work are in good agreement with theoretical predictions. The use of a corrected atomic parameters database and, of a more appropriate theory for ionization cross sections provide a better explanation of the L_3-subshell alignment. This demonstrates that more elaborated theories may not be necessary to describe adequately this phenomenon. Finally, more experimental results are needed to decide which theory is the best in the low energy range.

Acknowledgments

The technical assistance of Mr. K. López and Mr. F.J. Jaimes for the accelerator operation and Mr. M. F. Lugo for figure preparation is acknowledged. This work was supported in part by CONACYT (contract 40122-F).

1. W. Jitschin, A. Kaschuba, H. Kleinpoppen, and H.O. Lutz, *Z. Phys. A* **304** (1982) 69.
2. W. Jitschin *et al.*, *J. Phys. B* **16** (1983) 1417.
3. Z. Stachura *et al.*, *J. Phys B* **17** (1984) 835.
4. C.P. Bhalla, *Phys. Rev. Lett.* **64** (1990) 1103.
5. T. Papp, *Nucl. Instr. and Meth. B* **154** (1999) 300.
6. J. Hrdý, A. Henins, and J.A. Bearden, *Phys. Rev A* **2** (1970) 1708.
7. S.C. McFarlane, *J. Phys. B* **5** (1972) 1906.
8. W. Jitschin, H. Kleinpoppen, R. Hippler, and H.O. Lutz, *J. Phys. B* **12** (1979) 4077.
9. J. Pálinkás, L. Sarkadi, and B. Schlenk, *J. Phys. B* **13** (1980) 3829.
10. J. Pálinkás, B. Schlenk, and A. Valek, *J. Phys. B* (1981) 1157.
11. J. Miranda and M. Lugo-Licona, *AIP Conf. Proc.* **576** (2001) 100.
12. E.G. Berezhko and N.M. Kabachnik, *J. Phys. B* **10** (1977) 2467.
13. M.O. Krause, *J. Phys. Chem. Ref. Data.* **8** (1979) 307.
14. W. Jitschin, R. Hippler, R. Shanker, and H.O. Lutz, *Proc. X-82 Int. Conf. on X-Ray and Atomic Inner-Shell Physics*, (Eugene, USA, 1982).
15. E.J. McGuire, *Phys. Rev A* **3** (1971) 587.
16. M. Lugo-Licona and J. Miranda, *AIP Conf. Proc.* **674** (2003) 389.
17. W.T. Elam, B.D. Ravel, and J.R. Sieber, *Rad. Phys. Chem.* **63** (2002) 121.
18. Z. Liu and S. Cipolla, *Comput. Phys. Commun.* **97** (1996) 315.
19. W. Brandt and G. Lapicki, *Phys. Rev. A* **23** (1981) 1717.
20. J.U. Andersen, E. Laegsgaard, M. Lund, and C.D. Mark, *Nucl. Instr. and Meth.* **132** (1976) 507.
21. J. Pálinkás, L. Sarkadi, B. Schlenk, I. Török, and G. Kálmán, *J. Phys. B* **15** (1982) L451.
22. Z. Stachura, F. Bosch, D. Moar, P.H. Mokler, and W.A. Schönfeldt, *GSI Annual Report 1981*. (Darmstadt, West Germany, 1981).
23. O. Benka, M. Geretschläger, and H. Paul, *J. Physique C***9** (1987) 251.
24. J.U. Andersen, E. Laegsgaard, and M. Lund, *Nucl. Instr. and Meth.* **192** (1982) 79.