INFLUENCE OF COSTER-KRONIG TRANSITIONS ON THE POLARIZATION OF L-SHELL X-RAYS INDUCED BY PROTON IMPACT

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The alignment parameters for L_3 -subshells of Cd and Sb atoms are obtained by measuring the degree of polarization of the L_1 -lines excited by proton impact in the range from 0.1 to 0.5 MeV. To compare the experimental alignment parameter with theory, either experimental or calculation results must be corrected for Coster-Kronig (CK) transitions. The uncertainty in CK transitions yields makes this comparison difficult. In this work the semiempirical values of the corrections have been derived from measured L-line intensities. The semiempirical correction factors exceed the theoretical ones. For constant reduced velocity, the semiempirical correction factor depends on the atomic number of the target. The semiempirical correction factor obtained in the same experiment improves the agreement between the theoretical alignment parameter and the experimental data.

1. INTRODUCTION

The alignment of atomic inner shells induced by ion impact has been the object of extensive theoretical and experimental investigations, because it provides a more sensitive test of theoretical models and atomic wave functions than the total excitation and ionization cross sections. Alignment leads to emission anisotropy and X-ray polarization, which can influence measurements of the X-ray production cross sections in ion-atom collisions. We have studied proton-induced cadmium and antimony L_3 -subshell alignment by measuring the $L_1 X$ -ray line polarization. To better test theory, an effort should be made to eliminate effects that hinder comparison of theory and experiment. To compare the experimental alignment parameter with theory, either experimental or numerical results must be corrected for Coster-Kronig (CK) transitions. Present knowledge of the CK rates f_{12} , f_{13} and f_{23} , which describe the probability of the vacancy transfer from L_1 - to L_2 - and L_3 -subshells and from L_2 - to L_3 -subshells, respectively, is unsatisfactory, given the paucity and low accuracy of the experimental and numerical data. This is particularly true in the region of the periodic table around atomic number Z = 50 where the onset of the $L_1 - L_2 - M_{4,5}$ and cut off of the $L_1 - L_3 - M_{4,5}$ CK transitions occur [1]. The onset and cut off of CK transitions at certain atomic numbers cause sharp discontinuities in the initial-state lifetimes as functions of atomic number. This results in a dramatic change in the associated yields [1-3]. Rosato's results [4] show no sharp discontinuities in the range 47 $\leq Z \leq$ 53. The exact location of these cut offs is also somewhat uncertain.

The uncertainty in the Coster-Kronig yields makes an accurate comparison of experiment and theory difficult. To avoid these uncertainties, we derive the correction factor from measurements of the L X-ray line intensities [5]. In this work we also study the effect of an abrupt change of the CK transition probability around Z = 50 on the measured alignment of L_3 -subshell vacancies.

2. EXPERIMENT

The experimental work was carried out at the 0.5 MeV Cockcroft-Walton generator at the Institute of Nuclear Physics of Moscow State University. A Soller type (flat crystal) X-ray spectrometer-polarimeter is used to measure the intensity and polarization of the L X-ray lines. The experimental setup is described in detail elsewhere [6]. Protons of 0.1 MeV to 0.5 MeV energy have been used. The collimated proton beam is stopped by a thick target. The target is positioned at 45° with respect to the incoming beam. The incident beam intensity is monitored by a current integrator. The X-rays emitted at 90° with respect to the beam axis are analyzed by the spectrometer, which is equipped with a graphite crystal (2d = 6.71 Å). The energy resolution of the spectrometer is $E/\Delta E = 600$. A polarization experiment with the crystal spectrometer can be carried out due to the linear polarization dependence of the crystal diffraction. The polarization P is defined by

$$P = \frac{1}{Q} \frac{(J_{\parallel} - J_{\perp})}{(J_{\parallel} + J_{\perp})},\tag{1}$$

where Q is the polarization sensitivity of the crystal. The alignment parameter A_{20} can then be deduced from the polarization P:

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

where α is a constant determined by the angular momentum of the initial and final states [7]. In our experiment we used a thick target, so the experimental alignment parameter is corrected for the energy loss of protons and for the absorption of X-rays in the target [8]. Errors in the alignment parameter are mainly due to errors in the determination of the L_1 and L_β yields, which are caused by statistical fluctuations, background substraction, and fitting procedures. The EWA code [9] was used to evaluate the spectra. As a rule, the statistical error in the measurements of the degree of polarization does not exceed 1%.

To determine the L_{α} X-ray production cross sections, we measured the L_{α} X-ray line yields as a function of incident particle energy. The X-ray production cross section can be determined from these measurements by the method and formulas described by Merzbacher and Lewis [10]. The proton stopping powers were taken from Andersen and Ziegler [11]. The absorption coefficient was calculated by averaging the data of Storm and Israel [12]. The X-ray production cross sections were determined to 15% accuracy.

3. RESULTS AND DISCUSSION

The alignment parameter can be inferred from measurements of the proton-induced L_1 -line polarization. To compare experimental and theoretical alignment parameters, we should take

into account possible changes in the L subshell populations due to Coster-Kronig transitions (indirect formation of a vacancy in the L_3 -subshell). In proton-atom collisions, vacancies are also produced in the L_1 - and L_2 -subshells, which then decay via Coster-Kronig transitions, so the number of L_3 vacancies increases. The alignment of L_1 and L_2 vacancies is zero, so that this two-step process leads to a decrease in L_3 vacancy alignment. The alignment parameter correction factor F is

$$A_{20} = F A_{20}^{exp}, (3)$$

$$F = 1 + f_{23}\frac{\sigma_2}{\sigma_3} + (f_{13} + f_{12}f_{23})\frac{\sigma_1}{\sigma_3},$$
(4)

where σ_i is the L_i -subshell ionization cross section, and f_{ij} are the Coster-Kronig yields.

The uncertainty in the Coster-Kronig yields makes it difficult to compare accurately experiment and theory. In order to eliminate this uncertainty, we determine the correction factor F from our measured L_{α} X-ray production cross sections. This cross section is defined as

$$\sigma_{\alpha} = \omega_3 \frac{\Gamma_{\alpha 1} + \Gamma_{\alpha 2}}{\Gamma_{R_3}} \left(1 + f_{23} \frac{\sigma_2}{\sigma_3} + (f_{13} + f_{12} f_{23}) \frac{\sigma_1}{\sigma_3} \right) \sigma_3.$$
(5)

From this equation, the correction factor is

$$F_{exp} = \frac{\Gamma_{R_3}}{\Gamma_{\alpha 1} + \Gamma_{\alpha 2}} \frac{\sigma_{\alpha}}{\omega_3 \sigma_3}.$$
 (6)

As experimental data on ω_3 for elements around Z = 50 are incomplete, we have taken the fluorescence yield ω_3 from Ref. [3] and the radiative transition rates $\Gamma_{\alpha 1}$, $\Gamma_{\alpha 2}$, Γ_{R_1} from Ref. [13]. Here we have not used the experimental values of σ_3 , because these data are found with Eq. (5) based on measurements of L_{α} -lines intensities, using the CK yields, and the basic parameters $(\omega_i, f_{ij} \text{ and } \Gamma)$ can strongly affect the final results. The ionization cross section σ_3 was calculated in terms of the so-called ECPSSR theory of Brandt and Lapicky [14, 15], which is a derivative of the plane wave Born approximation with corrections for ion energy loss effects (E), Coulomb repulsion (C), polarization and binding-energy changes via perturbed stationary states (PSS), and relativistic effects (R). Obviously, that the uncertainty in the ω_3 , which can change in the course of the collision, increases the uncertainty in the semiempirical correction factor. References [16] and [17] examine the influence of the line shape and the satellite contribution on the interpretation of data in the measurements of X-ray spectra, and show that ignoring line shape effects can lead to systematic errors. In present work it has been assumed that satellite structure (in particular, the CK satellites) can be ignored in analyzing the spectra. It is clear that disregard of line shape effects and the fact that satellites contribute to polarization measurements can lead to systematic errors in measurements of the alignment parameter and in the semiempirical correction factor.

Figure 1 presents the semiempirical correction factors F_{exp} for Cd and Sb atoms obtained using Eq. (6) with the σ_{α} values measured in this experiment. The same figure shows the correction factors calculated with Eq. (4), using σ_i calculated within the ECPSSR theory and the CK yields taken from Ref. [3]. It can be seen that the semiempirical correction F_{exp} exceeds its theoretical value. It is also seen that the correction factors depend on collision velocity $(v/v_0)^2$ (v is the proton velocity and v_0 is the Bohr velocity of L_3 -electrons), and reach their minimum when the reduced velocity is about $(v/v_0)^2 = 0.1$. This minimum results



Fig. 1. Correction factor for CK transitions vs. reduced velocity. Δ — Cd, o — Sb. Dashed lines are drawn through the experimental data to guide the eye. The solid lines are the result of the calculation

Fig. 2. L_3 -subshell alignment parameter vs reduced velocity. Experimental data: ∇ — Cd, • — Sb without correction; Δ — Cd, • — Sb with correction for CK transitions. The curve presents the result of the calculation in the Born approximation

from a minimum in the ratio σ_1/σ_3 of cross sections for ionization of L_1 - and L_3 -subshells of atoms [18]. The minimum in the σ_1/σ_3 ratio is due to the node of the 2*s*-electron wave function. The discrepancy between F_{exp} and the theoretical value is partly due to the incorrect calculated ionization cross sections. Nevertheless, we believe that the semiempirical correction factor is more accurate, as it contains more accurate basic parameters.

In Fig. 2 we compare the experimental alignment parameter A_{20}^{exp} for Cd and Sb atoms with the alignment parameters corrected as described above, and A_{20} calculated in the Born approximation by the method and equations of Ref. [19]. It can be seen that when the semiempirical correction factors F_{exp} are used, the agreement with the experimental data improves.

Based on measurements of L_{α} X-ray production cross sections, we determined the semiempirical correction factors for elements ranging from Z = 45 to Z = 51. The semiempirical correction factors obtained for reduced velocity $(v/v_0)^2 = 0.02$, together with the calculated correction factors, are presented in Fig. 3*a* as functions of the atomic number of the target. The decrease in the theoretical values of *F* around Z = 50 is evident, and correlates with the decrease in f_{13} CK transition yield (see Fig. 3*b*). The f_{13} reduction is due to the fact that the L_1 - L_3 - $M_{4,5}$ CK transition becomes energetically forbidden. From this figure it is evident that there is no agreement of theoretical correction factors with experimental ones. The experimental results do not indicate any visible change in the correction factor around Z = 50.

Summarizing, we conclude that accurately corrected values of the alignment parameter have to be introduced in order to reach good agreement between theoretical and experimental alignments. As the Coster-Kronig yield can change in the course of the collision (post-col-





lision interactions, multiple ionization, chemical effects), the semiempirical correction factor obtained in the same experiment that measured the alignment parameter is a more realistic one. But even so, the uncertainty due to errors in the L_3 -subshell fluorescence yield and ionization cross sections remains. The Z dependence of the correction factor is smooth, and the experimental results do not show sharp discontinuities anywhere within the range $47 \le Z \le 51$.

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References

- 1. W. Bambynek, B. Crasemann, R. W. Fink, H. V. Freud, C. D. Swift, R. E. Price, and P. V. Rao, Rev. Mod. Phys. 44, 716 (1972).
- 2. E. J. McGuire, Phys. Rev. A 3, 587 (1971).
- 3. M. O. Krause, J. Phys. Chem. Ref. Data 8, 307 (1979).
- 4. E. Rosato, Nucl. Instr. Meth. B 15, 591 (1986).
- 5. V. P. Petukhov, E.A. Romanovsky, and H. Kerkow, Nucl. Instr. Meth. B 109/110, 19 (1996).
- 6. V. P. Petukhov, Prib. Tekh. Eksp. 1, 195 (1990).

- 7. E. G. Berezhko and N. M. Kabachnik, J. Phys. B 10, 2467 (1977).
- 8. G. Richter, Doctoral Thesis, Universität Munster (1982).
- 9. J. Vegh, Thesis, ATOMKI, Debrecen (1990).
- 10. E. Merzbacher and H. W. Lewis, Handbuch der Physik, Berlin (1958), vol. 34, p. 166.
- 11. H. H. Andersen and J. F. Ziegler, *The Stopping and Ranges of Ions in Matter*, Pergamon Press, N. Y. (1977), vol. 3.
- 12. E. Storm and H. I. Israel, Nucl. Data Tables A 7, 565 (1970).
- 13. J. H. Scofield, At. Data Nucl. Data Tables 14, 121 (1974).
- 14. W. Brandt and G. Lapicki, Phys. Rev. A 20, 465 (1979).
- 15. W. Brandt and G. Lapicki, Phys. Rev. A 23, 1717 (1981).
- 16. T. Papp, J. L. Campbell, and S. Raman, Phys. Rev. A 49, 729 (1994).
- 17. J. L. Campbell and T. Papp, X-Ray Spectrometry 24, 307 (1995).
- 18. V. P. Petukhov, V. S. Nikolaev, E. A. Romanovsky, and V. A. Sergeev, Zh. Eksp. Teor. Fiz. 71, 968 (1976).
- 19. V. V. Sizov and N. M. Kabachnik, J. Phys. B 13, 1601 (1980).