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L-subshell ionization of some rare earth elements by $^{7}_{3}$Li ion impact

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Abstract. L x-ray line production cross sections for Eu, Tb and Er elements have been measured as a result of $^{7}_{3}$Li ion bombardment at energies from 4.2 to 19.6 MeV. The corresponding L_1-, L_2- and L_3-subshell ionization cross sections have been calculated and have been compared to the predictions of the ECPSSR and SCA theories. Possible reasons for observed disagreements between experiment and theory are discussed.

1. Introduction

During the past decade experimental and theoretical interest in atomic inner-shell ionization by charged particle bombardment inspired many experimental works. Over 10 000 experimental values concerning K- and L-shell ionization by hydrogen and helium ion impact now exist for an extensive comparison with the predictions of the current theories.

The experimental data of K-shell ionization have been summarized in some works (Rutledge and Watson 1973, Gardner and Gray 1978, Paul and Muhr 1986) and compared extensively with the ECPSSR predictions (i.e. the plane-wave Born approximation with corrections for binding polarization and relativistic effects for target-atom electrons, and for Coulomb deflection and energy-loss effects for incident particles; Brandt and Lapicki 1979, 1981) by Lapicki (1989) and by Paul and Muhr (1986) where, finally, the 'reference' K-shell ionization cross sections have been calculated and with the semiclassical approximation (SCA) by Trautmann and Kauer (1989) and by Pajek *et al* (1990). The comprehensive review on K-shell cross sections by Paul and Muhr (1986) also include a recapitulation of all existing experimental data.

In the case of L-subshell ionization the situation is not so clear as that for K-shell. This is evident from some existing compilation works (Hardt and Watson 1976, Heitz *et al* 1982, Sokhi and Crumpton 1984, Braziewicz and Braziewicz 1988) and from original works where L-subshell ionization has been reported (but in most of them, only in graphical forms). A general review of this field is given by Gray (1980). In the case of L-shell ionization, in spite of the small base of experimental data, there are some experimental problems. In our earlier paper (Braziewicz and Braziewicz 1988) we discussed them more accurately by dividing them into three groups, i.e. (i) concerning the complex structure of the L x-ray spectra, (ii) concerning the different sets of the atomic parameters during evaluation of experimentally measured L x-ray intensities to L_i-subshell ionization cross sections and (iii) concerning the manner of converting the partial x-ray production cross sections to the L_i-subshell ionization cross sections.

Consequently, a detailed comparison of existing L_i-subshell ionization data with the theoretical descriptions is difficult to perform. After all, one can find some comparisons for elements in different regions of the periodic table for proton and helium bombardment (Sokhi and Crumpton 1984, 1985, Braziewicz and Braziewicz 1988, Braziewicz *et al* 1991, Vigilante *et al* 1991, and references quoted there). Generally, good agreement is observed between the experimental data and the predictions of the ECPSSR (perturbed stationary state including corrections for energy loss, Coulomb deflection and relativistic motion of the orbital electrons) theory (Brandt and Lapicki 1979, 1981) and for values of the reduced velocity parameter $\xi^R_{L_i} \geq 0.6$. For lower values of $\xi^R_{L_i}$ serious discrepancies appear, which have been attributed either to an inadequate method of calculating first-order corrections (Vigilante *et al* 1990, Perillo 1990, Cohen 1984), or the presence of second-order effects (Sarkadi and Mukoyama 1984, Jitschin *et al* 1983a) in the ionization process. Some improvements have been obtained by using a modified ECPSSR model leading to a united-atom approximation (UA-ECPSSR) (Vigilante *et al* 1990) or by introducing into the theoretical descriptions the effect of subshell coupling (Sarkadi and Mukoyama 1988).

For a deep understanding of the ionization mechanism, systematic research into different ion–atom systems, especially in the low $\xi^R_{L_i}$ region, are advisable. The required data for some rare earth elements bombarded by hydrogen and helium ions have been obtained recently by our group (Braziewicz *et al* 1984, 1986, 1991). In this paper we report on L_i-subshell ionization of Eu, Tb and Er induced by ${}^7_3$Li projectiles with energy from 4.2 to 19.6 MeV.

2. Experimental procedure

A thin target of Eu(28.5 μg cm^{-2}), Tb(9.4 μg cm^{-2}) and Er(12.5 μg cm^{-2}) (prepared by vacuum evaporation of fluoride compounds onto 0.2 mm thick silicon backing) was bombarded by ${}^7_3$Li projectiles with energy from 4.2 to 19.6 MeV from the Tandem accelerator of Erlangen-Nürnberg University. A maximum energy loss of projectiles inside the target thickness was less than 21 keV (for 4.2 MeV lithium ions in Eu target). To obtain L x-ray production cross sections, the simultaneous x-ray and scattered charge particle detection technique was employed. A detailed experimental set-up has been described earlier (Braziewicz *et al* 1984) so in this paper we only discuss the apparatus briefly.

The collimated beam of 3 mm diameter was directed onto the target tilted at an angle 45° relative to the beam direction. The L x-rays, emitted from a target, passed a 25 μm metallized Mylar chamber window, 10 mm air gap and 25 μm thick beryllium detector window before reaching a Si(Li) detector (FWHM 220 eV at 6.4 keV) positioned at 90° to the beam direction.

The total detector efficiency of the spectrometer, consisting of the intrinsic efficiency of the Si(Li) detector, the detector solid angle and the x-ray attenuation factor for absorption between the target and the intrinsic region of detector, was determined by the method described in detail by Pajek *et al* (1989). The total detector efficiency was experimentally determined from the K x-rays emitted from calibrating targets of light elements ($Z = 13$–29) bombarded by 3.0 MeV protons. In addition, the radioisotopic

${}^{241}Am$, ${}^{152}Eu$ and ${}^{133}Ba$ sources were used to check such a procedure. Finally, the total efficiency curve was obtained by fitting the detector parameters and in the x-ray region of interest ($4 \leqslant E_x \leqslant 10$ keV) (figure 1) was obtained with uncertainty ±6%.

The beam current was monitored by charge collection on the target and on the Faraday cup.

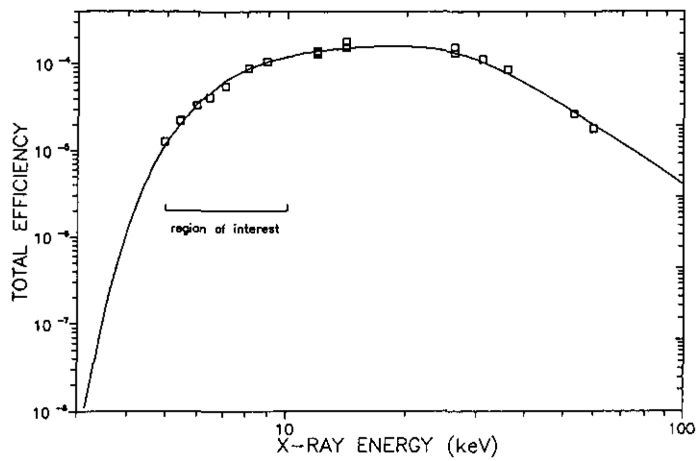

Figure 1. Experimental (□) and fitted (——) efficiency of the Si(Li) detector efficiency used in the present work plotted against photon energy.

The thicknesses of the targets were measured by the Rutherford backscattering technique. The surface barrier detector was placed at 150° relative to the beam axis and its solid angle was 3.84×10^{-4} sr.

The L x-rays emitted from the elements examined (bombarded by ${}^7_3Li^{+2}$ ions of energies 4.2, 5.6 MeV and by ${}^7_3Li^{+3}$ ions of energies 7.2, 9.8, 14.0 16.8 and 19.6 MeV) were collected by a multichannel analyser. The fitting code ACTIV (Zlokazov 1982), used for x-ray spectra analysis, removed the background and resolved the individual L x-ray transitions.

Using the procedure described in detail earlier (Braziewicz *et al* 1984) the L x-ray production cross sections for $L_{\alpha_{1,2}}$, $L_{\beta_{1,3,5}}$, L_{γ_1}, $L_{\gamma_{2,3}}$ and L_l transitions were calculated from the measured intensities. This procedure assumes an isotropic emission of x-rays emitted from the target and also takes into account the projectile energy loss and x-ray absorption in the target. In our case, the values of the correction factors associated with these effects were less than 3% for Tb and Er and less than 8% for Eu.

The influence of anisotropic emission of the L_3 x-rays caused by collisionally induced L_3-subshell alignment on the data measured have been estimated using the results of Sarkadi and Mukoyama (1984). This effect increases the x-ray production cross sections by less than 3% when the projectile energies are lowest and decreases by less than 1% when the projectile energies are highest.

The L_1-, L_2- and L_3-subshell ionization cross sections were extracted from the production cross sections for $L_{\alpha_{1,2}}$, $L_{\gamma_{1,5}}$ and $L_{\gamma_{2,3}}$ lines using: (i) the well known relationship of Tawara *et al* (1974), (ii) the fluorescence (ω_1, ω_2 and ω_3) and Coster-Kronig (f_{12}, f_{13} and f_{23}) yields from Chen *et al* (1981), (iii) the theoretical relative x-ray emission rates from Scofield (1974).

The numerical values quoted in this paper have uncertainties which come from the following sources: detection of a peak area of less than 5%; beam current measurements ~1.5%; target thickness ~5%; total efficiency of detection system ~6%; the accuracy of ion energy was less than 10 keV. Finally, the uncertainty of the ionization cross sections was estimated for the L_1 subshell as 10–20%, for the L_2 subshell as 10–20% and for the L_3 subshell as 5–15%.

3. Results and discussion

The L_i-subshell ionization cross sections of Eu, Tb and Er induced by incident ${}^{7}_{3}Li$ projectiles are summarized in table 1.

Table 1. L-subshell ionization cross sections (in barn) for targets bombarded by Li ions.

	Eu			Tb		
E (MeV)	$L-1$	$L-2$	$L-3$	$L-1$	$L-2$	$L-3$
19.60	0.280E+04	0.265E+04	0.865E+04	0.177E+04	0.178E+04	0.561E+04
	14%	11%	11%	19%	12%	11%
16.80	0.195E+04	0.187E+04	0.607E+04	0.110E+04	0.133E+04	0.459E+04
	12%	11%	11%	18%	15%	14%
14.00	0.120E+04	0.153E+04	0.477E+04	0.740E+03	0.111E+04	0.354E+04
	13%	12%	13%	19%	16%	15%
9.80	0.334E+03	0.759E+03	0.251E+04	0.192E+03	0.501E+03	0.161E+04
	13%	14%	12%	19%	15%	13%
7.20	0.108E+03	0.403E+03	0.123E+04	0.803E+02	0.231E+03	0.781E+03
	16%	16%	15%	13%	12%	8%
5.60	0.631E+02	0.188E+03	0.644E+03	0.502E+02	0.124E+03	0.413E+03
	14%	14%	15%	14%	10%	7%
4.20	0.396E+02	0.781E+02	0.236E+03	0.357E+02	0.414E+02	0.154E+03
	13%	14%	13%	14%	12%	9%

	Er		
E (MeV)	$L-1$	$L-2$	$L-3$
19.60	0.108E+04	0.111E+04	0.393E+04
	17%	15%	14%
16.80	0.816E+03	0.101E+04	0.321E+04
	18%	14%	13%
14.00	0.397E+03	0.690E+03	0.210E+04
	13%	14%	13%
9.80	0.101E+03	0.311E+03	0.967E+03
	13%	15%	11%
7.20	0.596E+02	0.150E+03	0.495E+03
	14%	14%	11%
5.60	0.377E+02	0.829E+02	0.259E+03
	12%	12%	9%
4.20	0.251E+02	0.324E+02	0.942E+02
	14%	14%	10%

Up to first order, in ionization processes the charge state of the projectile has only some influence on its trajectory. In the SCA model this (classical) trajectory is completely determined by the screened Coulomb potential between projectile and target. Usually it is approximated by a point Coulomb potential, but the effect of screening by the electronic clouds has also been thoroughly investigated in the literature. Jakob *et al* (1982) have shown that, for kinematical conditions very similar to those discussed in this paper, this screening effect is quite small, being less than 5% in the most unfavourable situation. Moreover it is clear that the difference in the two different charge states ($^{7}_{3}Li^{+2}$ and $^{7}_{3}Li^{+3}$) will lead to only a small change in the screening potential, and thus the difference between the total ionization cross sections produced by the two charge states will be less than a few per cent and therefore negligible here.

The dependence of measured L_i-subshell ionization cross sections on projectile energy is presented graphically in figures 2–4. On these figures we also marked predictions of the ECPSSR model (full curve). This model is essentially a PWBA theory whose range of validity has been extended by several improvements, i.e. the perturbed stationary states (PSS) formalism has been applied to account for the increase of the binding energy and polarization of the wavefunction of the target electron in the field of the projectile, the energy loss (E) of the incoming projectile in the process of atom ionization, the Coulomb deflection (C) of the projectile in the vicinity of the target nucleus, the relativistic effects (R) of the L_i-subshell electrons of the target and is indicated in our figures as ECPSSR-SA.

As is seen from figures 2–4, most of the ionization cross sections, especially for L_1 and L_2 subshells, are above ECPSSR-SA predictions, i.e. this theory underestimates the experimental data. A similar tendency has been observed recently by Vigilante *et al*

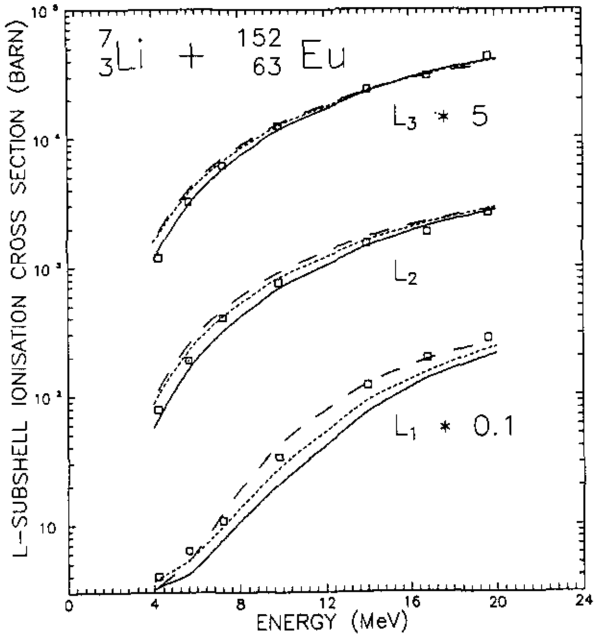

Figure 2. L-subshell ionization cross section for the europium target plotted against lithium energy: □, experimental data; ——, the prediction of ECPSSR-SA; – – –, ECPSSR-UA; — —, SCA models.

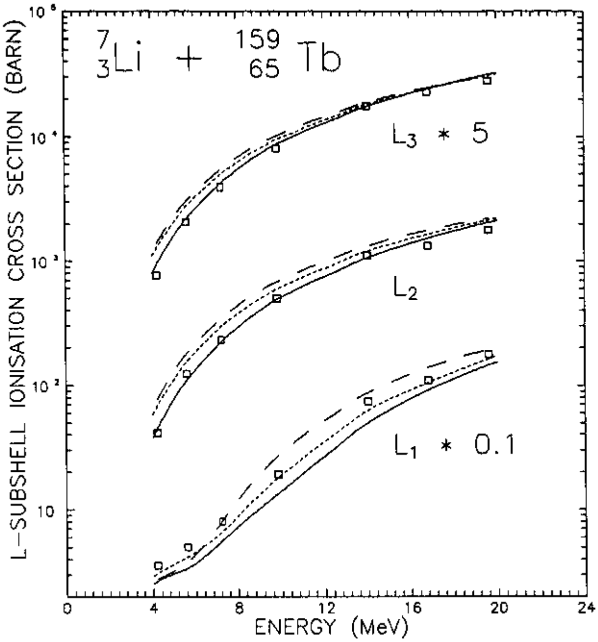

Figure 3. L-subshell ionization cross section for the terbium target plotted against lithium energy: □, experimental data; ——, the prediction of ECPSSR-SA; - - -, ECPSSR-UA; — —, SCA models.

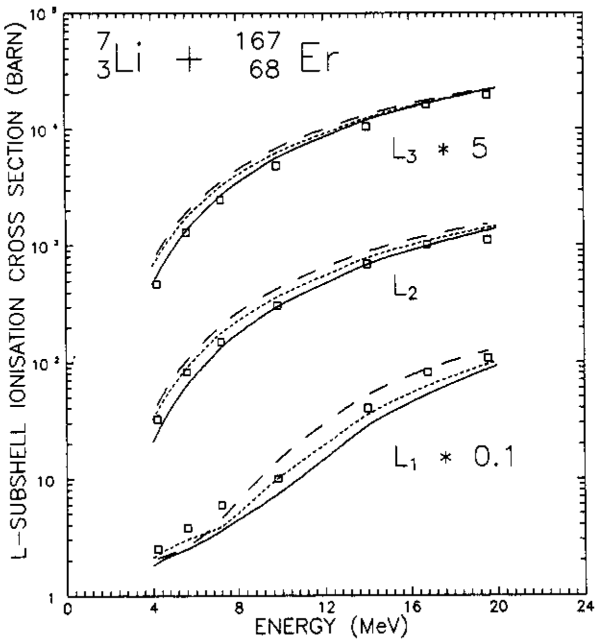

Figure 4. L-subshell ionization cross section for the erbium target plotted against lithium energy: □, experimental data; ——, the prediction of ECPSSR-SA; - - -, ECPSSR-UA; — —, SCA models.

(1990) for proton and helium bombardment. These authors have concluded that this discrepancy can be mainly attributed to the overestimation of the binding effect in the ECPSSR procedure. To avoid this inconsistency there are two approaches. Vigilante *et al* (1990) suggest very simply a 'united atom' model (designated here ECPSSR-UA) by putting the binding correction factor at a value taken at $\xi^{r}_{L_i}$(UA). Sarkadi and Mukoyama (1991) proposed a different way to account for this effect. They have constructed a procedure which provides a smooth transition between the two velocity ranges where the UA and SA approximation are applied.

We have repeated the ECPSSR calculation with the modified binding energy of the united atom and with the modified (for the UA approach) correction factors for relativistic and energy loss effects. Results of these recalculations are marked in figures 2–4 as short broken curves. The ECPSSR-UA modification caused only minor changes for the L_i-subshell ionization cross sections for colliding systems with reduced velocity $\xi^{r}_{L_i} \geqslant 0.9$. However, in the region of lower projectile velocities the changes are essential and depend on the value of the reduced velocity parameter.

As is seen from figures 2–4 the experimental L_1-subshell ionization data are systematically greater than the ECPSSR-SA predictions by about a factor ~ 1.5 and greater than the ECPSSR-UA predictions by about 1.1. In both cases the theoretical data qualitatively describe the dependence of experimental data on projectile energy.

In the case of the L_2 subshell the ECPSSR-SA model describes experimental points for the reduced velocity parameter $\xi^{r}_{L_2} \geqslant 0.6$ satisfactorily. For the lowest ion energy, the ECPSSR-SA model systematically overestimates the experimental data by up to ~ 1.3. In contrast the ECPSSR-UA overestimates experiment in the full examined energy region. In this case experimental data are situated between predictions of the ECPSSR-SA and the ECPSSR-UA approximation.

In the case of the L_3-subshell ionization the ECPSSR-SA theoretical predictions satisfactorily describe all experimental data in the full ion energy region examined. The disagreement factor of theoretical and experimental data lies between 0.85 and 1.1. On the other hand it is clearly visible in figures 2–4 that the ECPSSR-UA model predicts an incorrect energy dependence of the ionization cross section. The observed disagreement is a monotonic function of the $^{7}_{3}$Li ion energy.

In the last few years the SCA model has been used extensively as a description of the ion–atom collision process. Here we use the SCA version adapted by Trautmann and Rösel (1983) in which: (i) the projectile moves along a classical trajectory, so the effect of the Coulomb deflection is fully accounted for; (ii) the recoil of the target nucleus is taken into account; (iii) the ionized electron is fully described by hydrogen-like electronic relativistic wavefunctions; (iv) an effective charge of the target atom is calculated according to the recipe of Slater to account for screening effects; (v) the perturbation of the L_i-subshell electrons of the target atom by the projectile is simulated using 'united atom' values for binding energy; and (vi) coupling between the different subshells is not taken into account. Results of this SCA approximation are marked on figures 2–4 as long broken curves.

The SCA predictions for the L_1 subshell are systematically lower than the experimental data in the region of the radial node in the $2s_{1/2}$ wavefunction. Analogous disagreement, but not so clear, was observed in our recent paper (Braziewicz *et al* 1991), where we compared L-shell ionization cross section data of rare earth elements bombarded by hydrogen and helium ions. These results evidently show that the wavefunctions used in this model must be more carefully examined and a more realistic description of an ionized electron is necessary. An analogous conclusion was made by Trautmann and Kauer (1989) when they compared SCA calculations with K-shell ionization experimental data.

In the case of L_2- and L_3-subshell ionization used in this work, the SCA model predicts an inadequate energy dependence of ionization cross sections. The SCA predictions for the L_2 subshell systematically overestimate the experimental data by a factor of 0.75. In the case of L_3-subshell ionization, the disagreement is a monotonic function of the projectile energy; the experimental values lie below the SCA predictions at low projectile energies and are quite satisfactory with these predictions at high energies.

So, the SCA model using fixed united atom values for binding energy agrees well with the K-shell experimental data only for the case of proton and deuteron impact on heavy targets. For heavier projectiles and for L-shell binding energy effects become more important and to describe our ion–atom collision process by SCA more accurately, additional improvements must be introduced to simplify and to increase the quality of the numerical treatment. As was clearly shown in the case of K-shell ionization, only a model where the time dependence of the electronic state is included can give satisfactory agreement with accurate data.

Some additional information can be obtained when the 'relative cross sections', i.e. ratios of the substate cross sections, are compared to theoretical predictions. In figures 5 and 6 relative $\sigma_{L_1}/\sigma_{L_2}$ and $\sigma_{L_1}/\sigma_{L_3}$ values are presented together with the ECPSSR-SA (full curve), the ECPSSR-UA (broken curve) and SCA (dotted curve) predictions. Both the ECPSSR models predict dependence of the mentioned 'relative cross sections' on a reduced velocity parameter qualitatively and reproduce the $\sigma_{L_1}/\sigma_{L_2}$ and $\sigma_{L_1}/\sigma_{L_3}$ minima satisfactorily. On the other hand, the theoretical relative cross sections lie systematically about 30% lower than the experimental values although the ECPSSR-UA gives a somewhat better description for the $\sigma_{L_1}/\sigma_{L_3}$ ratio especially in the higher velocity region. The observed disagreement is mainly connected to the incorrect description of the L_1-subshell ionization by both ECPSSR models.

The SCA model predicts satisfactory $\sigma_{L_1}/\sigma_{L_2}$ and $\sigma_{L_1}/\sigma_{L_3}$ ratios only for a mean reduced velocity parameter $\xi \geq 0.8$, while for lower values the agreement is unsatisfac-

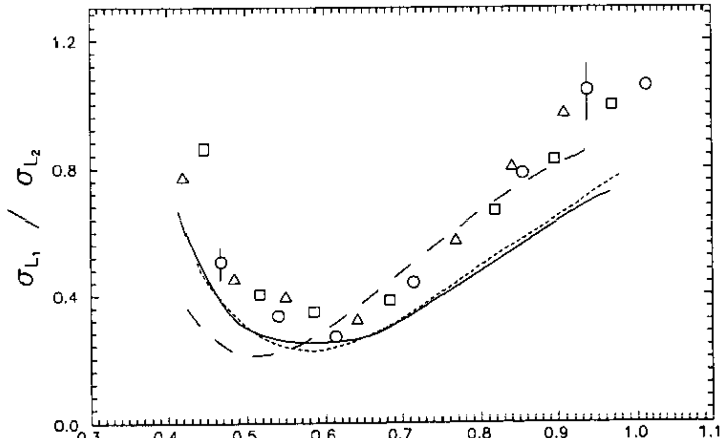

Figure 5. L-subshell ionization cross section ratio $\sigma_{L_1}/\sigma_{L_2}$. Experimental data for Eu (circles), Tb (squares) and Er (triangles) and theoretical predictions of ECPSSR-SA (full curve), ECPSSR-UA (short broken curve) and SCA (long broken curve).

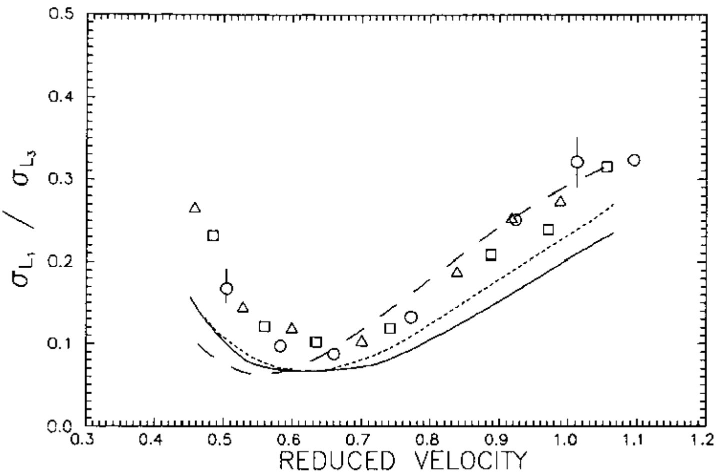

Figure 6. L-subshell ionization cross section ratio $\sigma L_1/\sigma_{L_3}$. Experimental data for Eu (circles), Tb (squares) and Er (triangles) and theoretical predictions of ECPSSR-SA (full curve), ECPSSR-UA (short broken curve) and SCA (long broken curve).

tory. This model also does not give the correct position of the minima in the $\sigma_{L_1}/\sigma_{L_2}$ and $\sigma_{L_1}/\sigma_{L_3}$ ratios. This is generally related to the fact of an incorrect description of the L_1 subshell by SCA model, especially in the region of the nodal structure of $2s_{1/2}$ subshell.

The L_2/L_3 subshell cross section ratios are presented in figure 7. In the examined region of reduced velocity parameter ($0.4 \leq \xi \leq 1.1$) the experimental 'relative' values take a constant value ~0.31. In this case the theoretical values are not as representative qualitatively as quantitatively. The theoretically predicted inadequate shape is mainly caused by the incorrect theoretical description of the L_2- and L_3-subshell ionization especially in the low velocity region. Sarkadi and Mukoyama (1991) take into account the effect of the collisionally induced intrashell transitions by ${}^{3}He$ ion bombardment.

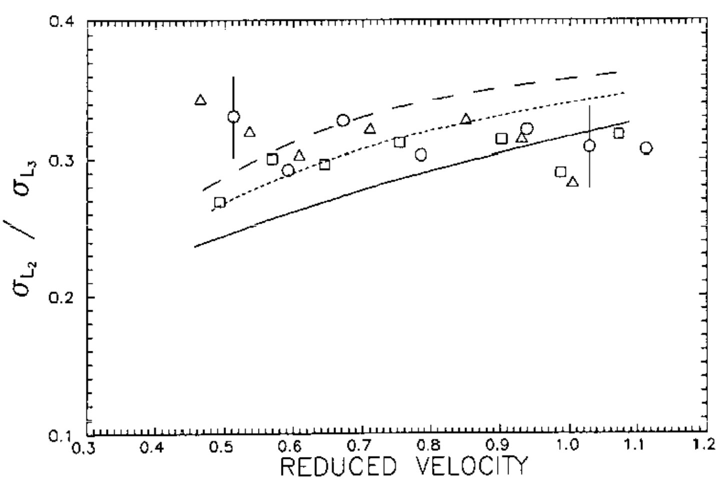

Figure 7. L-subshell ionization cross section ratio $\sigma_{L_2}/\sigma_{L_3}$. Experimental data for Eu (circles), Tb (squares) and Er (triangles) and theoretical predictions of ECPSSR-SA (full curve), ECPSSR-UA (short broken curve) and SCA (long broken curve).

In their case the L_i-subshell ionization modification is not essential for the L_1 and L_3 subshells, and for the L_2-subshell caused an increase in the theoretical values as a function of decreasing reduced velocity parameter.

4. Conclusion

In this paper we report on the L_i-subshell ionization cross section measurements of some rare earth elements by ${}^{7}_{3}Li$ ion bombardment. Results presented show that there are some essential deviations of theoretical ECPSSR and SCA data from experimental ones especially in the low energy region.

Theoretical and experimental disagreement is caused by two different reasons.

The first is connected to experimental problems. Investigations for still heavier projectiles become even more difficult to interpret due to the increasing importance of multiple vacancies in inner shells. This may be of considerable influence on the fluorescence yields.

The second reason is connected to the theoretical description of the ion–atom collision process. For large asymmetry between the collision partners the ionization is treated perturbatively using first-order theories such as PWBA or SCA. In most cases the states of the inner-shell electrons can be well represented by screened hydrogen-like wavefunctions. But even for light projectiles some attempts have been made to introduce a more realistic description of electronic states. It also seems that a further investigation of the Coulomb deflection, binding and polarization effects may improve the description of the atom ionization processes within the theories considered. The vacancy rearrangement effect must be taken into account, both in the ECPSSR and in the SCA models. The probability of such transitions is a strong function of the atomic number of the projectile. Because the coupled-states calculations are very time consuming, the parametrization proposed by Sarkadi and Mukoyama (1991) for helium bombardment should be provided for different ion–atom systems.

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