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# $L$-shell x-ray production cross sections of Ni, Cu, Ge, As, Rb, Sr, Y, Zr, and Pd by (0.25—2.5)-MeV protons

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$L$-shell x-ray production cross sections by ${}^1_1H^+$ ions are reported. The data are compared to the first Born approximation (plane-wave Born approximation for direct ionization and Oppenheimer-Brinkman-Kramers approximation for electron capture) and to the ECPSSR (energy-loss and Coulomb-deflection effects, perturbed stationary-state approximation with relativistic correction) theory. The energy of the protons ranged from 0.25 to 2.5 MeV in steps of 0.25 MeV. The targets used in these measurements were ${}_{28}$Ni, ${}_{29}$Cu, ${}_{32}$Ge, ${}_{33}$As, ${}_{37}$Rb, ${}_{38}$Sr, ${}_{39}$Y, ${}_{40}$Zr, and ${}_{46}$Pd. The first Born theory generally agrees with the data found in the literature at high energies and overpredicts them below 1.5 MeV. The ECPSSR predictions are in better agreement with experimental cross sections. At 0.25 MeV our data, however, are underestimated by this theory and tend to agree with the first Born approximation.

## I. INTRODUCTION

Over the years, inner-shell ionization due to charged-particle excitation by protons has been studied extensively. The two primary processes responsible for inner-shell ionization are direct ionization (DI) to the target continuum and electron capture (EC) to the projectile ion. DI plus EC can be predicted in the first Born [plane-wave Born approximation[1] (PWBA) for DI and Oppenheimer-Brinkman-Kramers-Nikolaev approximation[2] (OBKN) for EC] approximation and by the ECPSSR approach.[3] The ECPSSR goes beyond first Born by including the effects of energy loss (E), Coulomb deflection (C), and relativistic effects (R) in the perturbed stationary-state (PSS) theory.

The measurements reported here are for the $L$-shell x rays whose energies are below 3 keV. A search of the literature shows that most of the published results for $L$-shell-ionization cross sections are for heavy elements whose $L$-shell binding energies are significantly greater than 3 keV. In what follows, we survey the low-energy $L$-shell measurements that have been made to date with protons and that overlap with our data in the range of the chosen target atoms ($28 \leq Z_2 \leq 46$). All of the published and the present results suffered from a number of experimental difficulties including (a) large experimental uncertainty in the detector efficiency below 3 keV, (b) contaminant x rays from the $K$ shell of light elements that exist as impurities on the carbon backings, and (c) for the target elements with atomic number $Z_2 < 47$, the $L$-subshell transitions are closely spaced in energy, making it difficult to resolve individual subshell transitions and hence to extract individual subshell cross sections.

First comprehensive surveys for $L$-shell x-ray production and ionization cross sections for protons were done by Brandt and Lapicki[4] and, as well for other projectiles published through 1974, by Hardt and Watson.[5] These surveys showed that for $28 \leq Z_2 \leq 46$ measurements were available for the following targets only (with protons of 0.026 to 5.0 MeV): ${}_{29}$Cu, ${}_{36}$Kr, ${}_{40}$Zr, and ${}_{42}$Mo.[6–10] Later Gray[11] extended the tables of Ref. 5 with data through 1977, and included measurements by Chaturvedi *et al.*[12] on ${}_{46}$Pd from 3 to 12 MeV and by Milazzo and Riccobono[13] for ${}_{37}$Rb and ${}_{39}$Y bombarded by 0.95-MeV protons. Since then additional measurements have been reported for targets of ${}_{28}$Ni, ${}_{35}$Br, ${}_{38}$Sr, ${}_{39}$Y, ${}_{42}$Mo, ${}_{45}$Rh, and ${}_{46}$Pd by protons in the energy range of 0.07–39.34 MeV.[14–20] The latest compilation of cross sections for $L$ shell is by Sokhi and Crumpton.[21]

The first attempts to compare theoretical results to experimental $L$-shell-ionization cross sections for low-$Z_2$ targets excited by protons were made by Jopson *et al.*[6] and Khan *et al.*[7] The PWBA overpredicted both the ${}_{40}$Zr and ${}_{42}$Mo data of Jopson *et al.*[6] (1962) and the copper data of Khan *et al.*[7] (1964—1966). Among the later studies[8–10] Winters *et al.*[10] (1973) saw excellent agreement between their data and the PWBA for targets of Kr in the 1.5—5.0-MeV range. More recent works on Pd (3—12 MeV);[12] Rb and Y (0.95 MeV);[13] Y (4—22 MeV);[14] Cu, Ga, Ge, As, Se, and Br (0.3—2.6 MeV);[16] and Y (2.92—39.34 MeV)[18] were compared to the PWBA,[1] the binary encounter approximation (BEA),[22] and precursors of the ECPSSR theory[3] for $L$-shell, i.e., an early approach that accounted for binding and Coulomb deflection effects[4] and a later CPSSR formulation.[23] All the theories generally predicted the trend of the data very well. The actual agreement ranges from fair to good, and is excellent in certain cases.

In the present work, $L$-shell x-ray production cross sections have been measured for (0.25—2.5)-MeV ${}^1H^+$ ions incident on thin solid targets (thickness in $\mu g/cm^2$ in parentheses) of ${}_{28}$Ni (6.4), ${}_{29}$Cu (19), ${}_{32}$Ge (28), ${}_{33}$As (12),

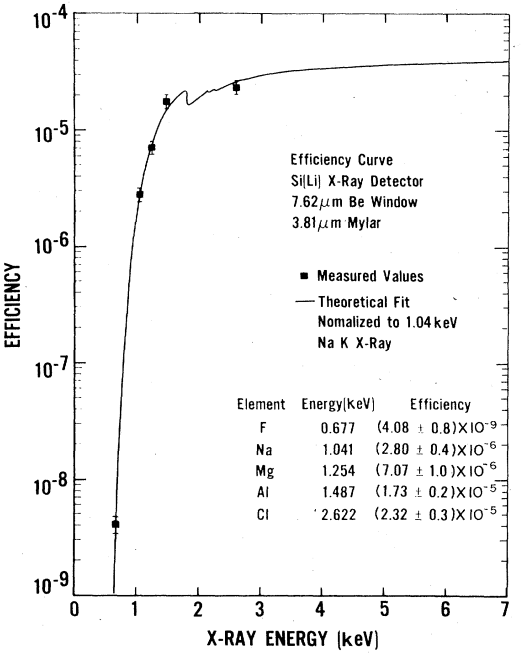

FIG. 1. Efficiency of the Si(Li) x-ray detector vs the x-ray energy.

$_{37}$Rb (29), $_{38}$Sr (11), $_{39}$Y (20), $_{40}$Zr (17), and $_{46}$Pd (11). For comparison, the predictions of first Born and ECPSSR theories for the $L$-shell ionization cross sections are converted to x-ray production cross sections using single-hole fluorescence yields and Coster-Kronig yields.[24]

## II. EXPERIMENTAL PROCEDURE AND DATA ANALYSIS

Ion beams of $^1H^+$ were obtained from the 2.5-MeV Van de Graaff accelerator at North Texas State University. Thin targets of $_{28}$Ni, $_{29}$Cu, $_{32}$Ge, $_{33}$As, $_{37}$Rb, $_{38}$Sr, $_{39}$Y, $_{40}$Zr, and $_{46}$Pd were prepared by vacuum evaporation and deposition technique. The (10—20)-$\mu$g/cm$^2$ carbon foils used as backing material for these targets were screened to make sure there was a minimal presence of low-$Z$ contaminants. The elemental layer then deposited on the carbon was thin enough so that the energy loss sustained by the projectile ion in passing through the target was negligible, but at the same time the layer was thick enough to provide x-ray peaks that were clearly above background. Further details for this technique for making contaminant-free targets are given in Ref. 25.

Simultaneous measurement of the yield of the $L$-shell x rays and the yield of the scattered particles during ion-target interaction was used to determine the $L$-shell x-ray production cross section. The Si(Li) x-ray detector and the Si surface-barrier particle detector were positioned at 90° and 150° to the incident beam, respectively. The target was positioned at 45° to the incident beam direction. Particulars of the experimental geometry and data analysis have been discussed in Ref. 26.

The efficiency of the Si(Li) detector was determined as described by Mehta *et al.*[26] Figure 1 shows the efficiency versus energy of the x rays for the present experiment. The absolute uncertainty in the x-ray cross sections reported here ranges from 13% to 26%, which is primarily due to uncertainty in the (i) background subtraction and polynomial fitting (2—13%) and (ii) efficiency of the Si(Li) detector at the $L$ x-ray energies (9—19%). The largest uncertainty due to efficiency occurs at low $L$-shell x-ray energies for nickel and copper due to the steepness of the fitted curve. The accuracy of the efficiency curve for other elements may be seriously affected by x-ray absorption edges in the (1—3)-keV range.

## III. RESULTS AND DISCUSSION

Table I lists both the measured and the theoretical values of the total $L$-shell x-ray production cross section

TABLE I. $L$-shell x-ray production cross sections (in barns) for $^1H^+$ ions.

| Target element | | Proton energy (MeV)<br>0.25 | 0.50 | 0.75 | 1.0 | 1.25 | 1.5 | 1.75 | 2.0 | 2.25 | 2.5 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| $_{28}$Ni | Measured | 959 | 1630 | 2110 | 2490 | 2660 | 2680 | 3020 | 3070 | 3260 | 2770 |
| | Theory-ECPSSR | 500 | 1470 | 2220 | 2670 | 2960 | 3130 | 3220 | 3250 | 3260 | 3240 |
| $_{29}$Cu | Measured | 537 | 1207 | 1920 | 2360 | 2740 | 2740 | 3050 | 2910 | 3140 | 2760 |
| | Theory-ECPSSR | 422 | 1320 | 2030 | 2510 | 2820 | 3020 | 3140 | 3200 | 3220 | 3220 |
| $_{32}$Ge | Measured | 202 | 615 | 1370 | 1620 | 1810 | 2100 | 2280 | 2300 | 2570 | 2570 |
| | Theory-ECPSSR | 195 | 729 | 1250 | 1660 | 1970 | 2180 | 2330 | 2440 | 2500 | 2550 |
| $_{33}$As | Measured | 197 | 471 | 1060 | 1420 | 1640 | 1820 | 2010 | 2290 | 2270 | 2190 |
| | Theory-ECPSSR | 148 | 586 | 1040 | 1420 | 1700 | 1920 | 2070 | 2180 | 2250 | 2300 |
| $_{37}$Rb | Measured | 139 | 369 | 550 | 706 | 856 | 990 | 1200 | 1160 | 1570 | 1320 |
| | Theory-ECPSSR | 59.2 | 284 | 577 | 859 | 1100 | 1300 | 1470 | 1600 | 1700 | 1780 |
| $_{38}$Sr | Measured | 67.6 | 231 | 403 | 627 | 783 | 880 | 1070 | 1180 | 1550 | 1310 |
| | Theory-ECPSSR | 46.5 | 238 | 488 | 742 | 969 | 1160 | 1320 | 1450 | 1550 | 1780 |
| $_{39}$Y | Measured | 33.4 | 200 | 386 | 602 | 660 | 835 | 947 | 1040 | 1380 | 1210 |
| | Theory-ECPSSR | 36.6 | 191 | 411 | 639 | 848 | 1030 | 1180 | 1310 | 1410 | 1490 |
| $_{40}$Zr | Measured | 37.5 | 137 | 386 | 612 | 764 | 988 | 1145 | 1230 | 1440 | 1390 |
| | Theory-ECPSSR | 29.7 | 161 | 356 | 564 | 761 | 934 | 1081 | 1210 | 1310 | 1400 |
| $_{46}$Pd | Measured | 18.1 | 65.6 | 127 | 190 | 295 | 284 | 417 | 543 | 437 | 670 |
| | Theory-ECPSSR | 8.6 | 57.7 | 144 | 253 | 370 | 485 | 595 | 695 | 786 | 867 |

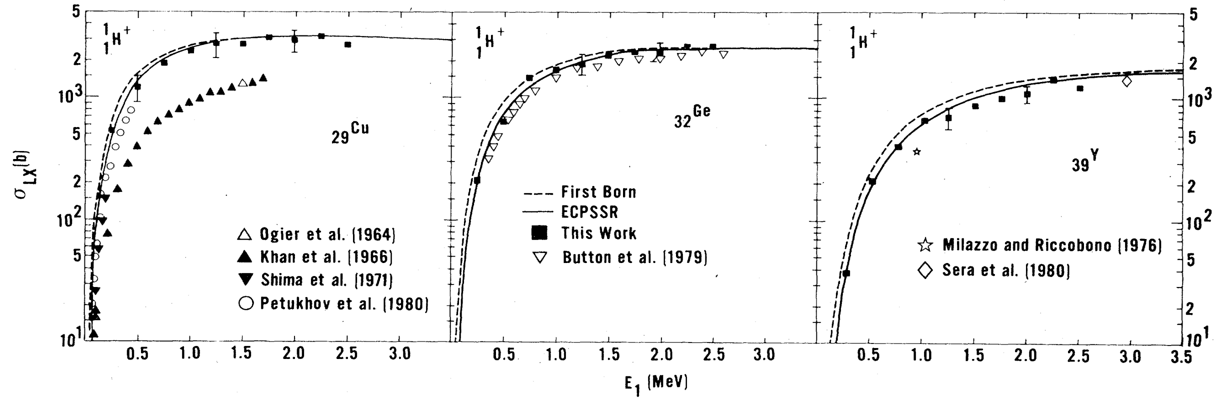

FIG. 2. $L$-shell x-ray production cross sections for protons incident on copper, germanium, and yttrium. Predictions of the first Born approximation (dashed curves) and ECPSSR (solid curves) theories are shown. Data are as reported in this work (solid squares) and from other sources (Refs. 7–9, 13, and 16–18); data of Khan *et al.* (Ref. 7) are for $L_3$ subshell only.

for $^1H^+$ ions. The absolute errors for the cross-section measurements range from 13% to 26% with the larger errors at lower energies and for lower-$Z_2$ targets. The $L$-shell ionization cross section calculated in the ECPSSR approximation was converted to an x-ray production cross section using the single-hole fluorescence yields and Coster-Kronig transition rates from Krause.[24]

Figures 2 and 3 present $\sigma_{LX}$, the total $L$-shell x-ray production cross section, in targets of $_{29}Cu$, $_{32}Ge$, $_{39}Y$, and $_{46}Pd$ versus the energy of the incident protons. $L$-shell x-ray production cross sections predicted by the first Born and the ECPSSR theories are shown as dashed and solid curves, respectively. Also shown are the measurements done for these four elements by others (Refs. 7–9, 12, 13, 16–18, and 20). In Fig. 2, earlier measurements from two decades ago (Refs. 7 and 8; upright triangles) for $_{29}Cu$ are smaller than present measurements by factors ranging from 2 to 4. This is not surprising in the case of the Khan *et al.*[7] cross sections as they were measured for $L_3$ ionization only; serious disagreement with the Ogier *et al.*[8] measurement remains, however, to be a puzzle. Our data (solid squares) follow the trend of the theories with increasing energy of the projectile. Good agreement is seen between the measurements and both the theories at all energies of the $^1H^+$ ion. Petukhov *et al.*[17] have extended the measurements at the low-energy range down to 0.07 MeV. Their data for copper, shown as open circles in Fig. 2, fall below the trend of ours in the overlapping region of 0.25–0.5 MeV, and are in excellent agreement with the predictions of the ECPSSR theory below 0.25 MeV, in the region where the first Born calculation overestimates the data by an order of magnitude. Earlier copper data of Shima *et al.*[9] (inverted triangles) lie, however, distinctly below the measurements of Ref. 17.

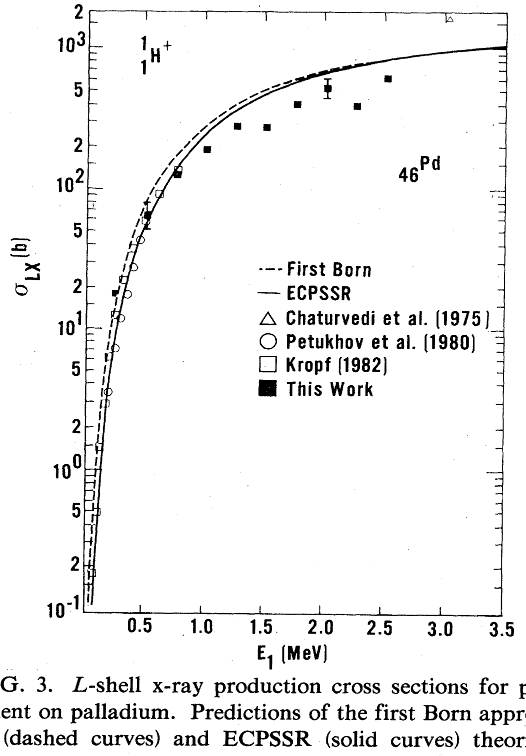

FIG. 3. $L$-shell x-ray production cross sections for protons incident on palladium. Predictions of the first Born approximation (dashed curves) and ECPSSR (solid curves) theories are shown. Data are as reported in this work (solid squares) and from other sources (Refs. 12, 17, and 20).

For the $_{32}Ge$ data, equally good agreement is seen between our measurements and the predictions of both theories above 1 MeV. Also plotted are data of Button *et al.*[16] which are slightly lower than our measurements but within experimental uncertainties. Button *et al.*,[16] as well as our data, favor the ECPSSR theory below 1 MeV. A similar observation can be made based on our data below 1 MeV for $_{39}Y$. At higher energies both theories overpredict the data somewhat. The measurement by Milazzo and Riccobono[13] at 0.95 MeV is $\frac{2}{3}$ of our interpolated cross section for this energy. The cross section determined by Sera *et al.*[18] at 2.92 MeV agrees clearly with an extrapolation of our highest-energy data.

In Fig. 3 our measurements for palladium are in disagreement with the theory and the data of Petukhov *et al.*[17] and Kropf,[20] especially at 0.25 MeV where our data are by as much as a factor of 2 above the ECPSSR predictions. The measurement reported by Chaturvedi *et al.*[12] at 3 MeV is higher by a factor of 3 than the trend that our data predict. Again, at high energies both theories overpredict the data. Below 1 MeV the ECPSSR theory is in excellent agreement with all the data that we could find in the literature for $_{46}$Pd. This is remarkable in view of the sharply decreasing cross sections with decreasing energy of the hydrogen ion and larger experimental uncertainties in the low-energy range.

Figure 4 depicts $\sigma_{LX}$ versus target atomic number $Z_2$ at four different and equidistant energies of $^1H^+$. The measurements of Jopson *et al.*,[6] Khan *et al.*,[7] Milazzo and Riccobono,[13] Button *et al.*,[16] Petukhov *et al.*,[17] and Kropf[20] for protons are shown. For copper targets, the data of Khan *et al.*[7] (only $L_3$-subshell cross sections were measured) are in part understandably below the ECPSSR predictions and also our data by a factor of about 3; the ECPSSR calculations show that $\sigma_{LX}/\sigma_{L_3X}\simeq 1.5$. Button *et al.*[16] measurements are in good agreement with our data and ECPSSR predictions for $Z_2 \geq 32$. Milazzo and Riccobono's[13] data for $_{37}$Rb and $_{39}$Y lie well below (almost a factor of 2 for $Z_2=37$) our measurements; even correcting for the fact that they were obtained at 0.95 MeV, not exactly at 1 MeV. The cross section by Jopson *et al.*[6] for $_{42}$Mo, interpolated to 0.25 MeV, is by as much as a factor of 10 below our data. Our measurements at 1.75 and 2.5 MeV for $_{37}$Rb, $_{38}$Sr, and $_{39}$Y are somewhat lower than the ECPSSR predictions but they are in good agreement for other elements; except for current $_{46}$Pd data which are significantly overestimated by the theories at 1 MeV and above, and definitely underestimated by the ECPSSR approach at 0.25 MeV. In fact, our measurements at 0.25 MeV, contrary to other data for this energy, are in good agreement with the first Born results.

In conclusion, present measurements of the $L$-shell x-ray production cross sections are roughly consistent with the data reported by laboratories in the last decade. Earlier measurements differ generally by large factors from the recent measurements and thus seem less reliable. The ECPSSR shows better agreement with the measured data than the first Born approximation; this is particularly so at low proton energies. At 0.25 MeV, however, our data are almost twice as high as other[17,20] measurements as well as the ECPSSR results. This trend is reverse to the behavior of the early molybdenum $L$-shell cross sections from Ref. 6, which are 1 order of magnitude below our data. Considering the wide spread that exists between measured cross sections for $L$-shell x-ray production in relatively light target elements by low-velocity protons, the need for more experimental studies of such collisions becomes evident.

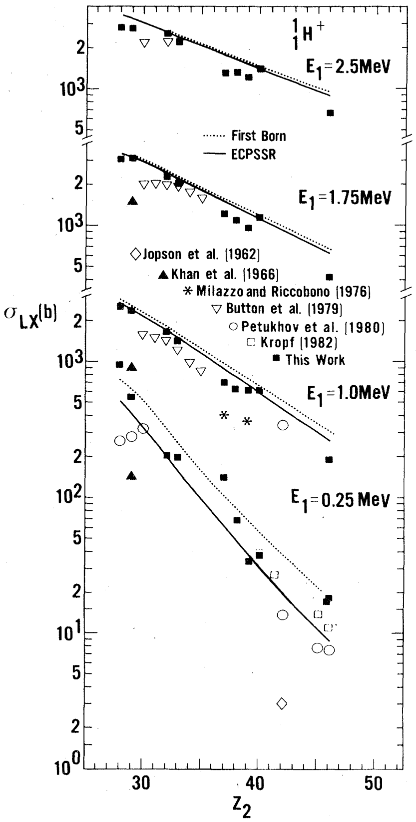

FIG. 4. $L$-shell x-ray production cross section for protons at energies of 0.25, 1.0, 1.75, and 2.5 MeV incident on various targets of atomic numbers $28 \leq Z_2 \leq 46$. Data, some of which are taken from Refs. 6, 7, 13, 16, 17, and 20, are compared to the first Born approximation (dashed curves) and ECPSSR theory (solid curves); data of Khan *et al.* (Ref. 7) are for copper $L_3$ subshell only.

## ACKNOWLEDGMENTS

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