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Gold L X-Ray Production by 0.5–30-MeV Protons*

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The characteristic L x-rays of Au, produced by 0.5–30-MeV proton beams on a thin Au target have been observed with a Si(Li) detector. Total L x-ray production cross sections derived from these measurements are in good agreement with presently available theoretical calculations. However, theoretical predictions of $L\alpha/L\beta$ and $L\alpha/L\gamma$ ratios and centroid energies of the $L\beta$ and $L\gamma$ lines as functions of proton energy show that a consistent quantum-mechanical treatment of L-shell ionization gives considerably better agreement with experimental results than the semiclassical or classical theories of inner-shell ionization.

I. INTRODUCTION

There is extensive experimental work on K-shell ionization by protons ranging in energy from 20 keV to 160 MeV over the whole Periodic Table. In general, the plane-wave Born approximation (PWBA), presented most completely by Merzbacher and Lewis,[1] which uses nonrelativistic hydrogenic wave functions, predicts K-shell ionization cross sections σ_K that are in good agreement with experimental values for σ_K at proton energies considerably above the binding energy. At lower proton energies the PWBA results usually lie above the experimental values of σ_K. This is considered to be due to nuclear repulsion effects. Bang and Hansteen[2] have treated the repulsion effects in a semiclassical approximation (SCA) by allowing the incoming projectile to follow a classical hyperbolic trajectory, which permits an impact-parameter description of the ionization process, while still providing a quantum-mechanical treatment of the atom. The SCA theory gives improved agreement with experimental measurements of σ_K at low projectile energies. Recently, a classical binary-encounter (BEA) description of K-shell ionization has been proposed by Garcia,[3] which includes nuclear repulsion effects. This is observed to give good agreement with experimental results over a broad proton energy range.[4] The experimental cross sections are generally derived from x-ray yield measurements, which required correction for the fluorescence yield. Presently there are reliable values[5] of ω_K for $Z \geq 10$, while for $Z < 10$, the situation is not nearly so clear.

The experimental measurements of L-shell ionization cross sections σ_L are not nearly so extensive, most measurements being for high-Z elements with low-energy protons (< 4 MeV). Recent measurements[6] with 2–30-MeV protons on Ag indicated that experimental values for σ_L fell consistently higher than either PWBA or BEA predictions (possibly because of inaccuracy in the value of the mean fluorescence yield $\bar{\omega}_L$ used to compare the L x-ray production cross section σ_{LX} with theoretical values of σ_L), although the observed energy dependence of σ_L was in better

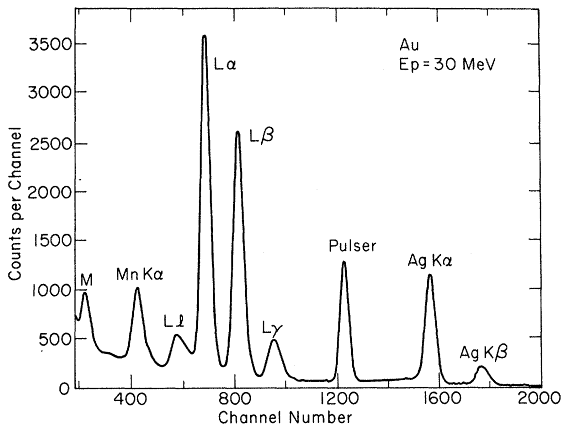

FIG. 1. Smoothed pulse-height distribution obtained at $E_p = 30$ MeV. Au L and M x-ray peaks, as well as peaks due to ^{109}Cd, ^{55}Fe, and the pulser, are shown.

agreement with the PWBA predictions. The present work presents the results of absolute $L\alpha$, β, γ x-ray production cross-section measurements for 0.5–30-MeV protons on a thin Au target. There are at present reasonably accurate experimental values[7] of ω_1, ω_2, and ω_3 and f_{12}, f_{13}, and f_{23} (Koster–Kronig yields) for Au, as well as recent theoretical values,[8] which permit one to compare the theoretical predictions of σ_{LX} with our experimental measurements.

II. EXPERIMENTAL

The experiment required use of three accelerators – the 4-MV Van de Graaff (0.5–3.0-MeV protons), the FN Tandem (2–15 MeV) and Cyclograaff (17–30 MeV), all at Triangle Universities Nuclear Labortory, Durham, N.C. Beams of 1–10 nA were used to bombard a thin target of Au evaporated onto a 20 μg/cm^2 C foil backing. The Au target thickness was measured initially with an optical interferometer to be $1.20 \pm 0.50 \times 10^{-6}$ cm thick. A later measurement using a 6-MeV α-particle beam and observing Rutherford scattering at 30° (lab) provided a more accurate value of $1.50 \pm 0.05 \times 10^{-6}$ cm. The same target and target chamber were used for all the measurements. Further details on the experimental arrangement are available from previous work.[6,9]

The Si(Li) detector used for these measurements, which had a resolution of 540 eV full width at half-maximum (FWHM) at 5.90 keV, was clearly able to resolve the Au $L\alpha$, $L\beta$, and $L\gamma$ lines at ~9.7, 11.5, and 13.5 keV with relative detection efficiencies of 0.84, 0.92, and 0.96, respectively (see Fig. 1). The Ll line showed some evidence of contamination by Cu and Zn K x rays from the target chamber and consequently was not analyzed. The contribution of this line together with the unresolved Lt and Ls lines to the total x-ray production cross section was less than 2% and was neglected. The intensities of the x-ray lines were determined using a Gauss fit program, capable of fitting one or two Gaussians to a peak, with a choice of isotropic, linear, or quadratic backgrounds. Energy calibration for all x-ray spectra was provided by a linear calibration derived from well-known x rays from ^{55}Fe and ^{109}Cd sources.[10] The calibration peaks were acquired simultaneously with the proton induced Au L x rays.

III. RESULTS AND DISCUSSION

A. Total L X-Ray Production Cross Sections (σ_{LX})

The inability of the Si(Li) detector to resolve the various subshell contributions to the $L\beta$ and $L\gamma$ lines (see Fig. 2) does not permit one to derive reliably accurate ionization cross sections for the individual L subshells. On the other hand, it is quite easy to calculate σ_{LX} using published fluorescence yields, Koster–Kronig fractions,[7] and theoretical values of σ_{L1}, σ_{L2}, and σ_{L3}. In Fig. 3 are presented the experimental results for σ_{LX} along with the BEA,[11] SCA,[12] PWBA (nonrelativistic),[13] and PWBA (relativistic)[14] theoretical predictions (experimental values for σ_{LX} are given in Table I). The observed $L\alpha$, β, γ intensities $N\alpha$, β, γ were converted to yields for each of the L x rays using

$$Y_{\alpha,\beta,\gamma} = N\alpha, \beta, \gamma / A\epsilon\phi t \Delta\Omega, \tag{1}$$

where A is the correction for x-ray absorption in

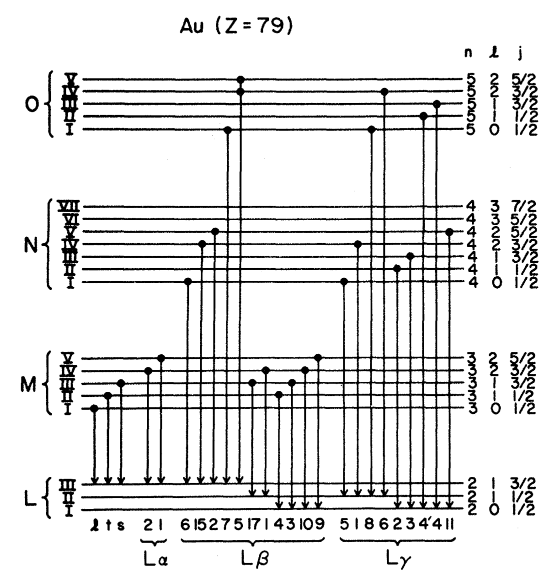

FIG. 2. Spectroscopic diagram for the major radiative transitions that comprise the characteristic Au L x-ray spectrum.

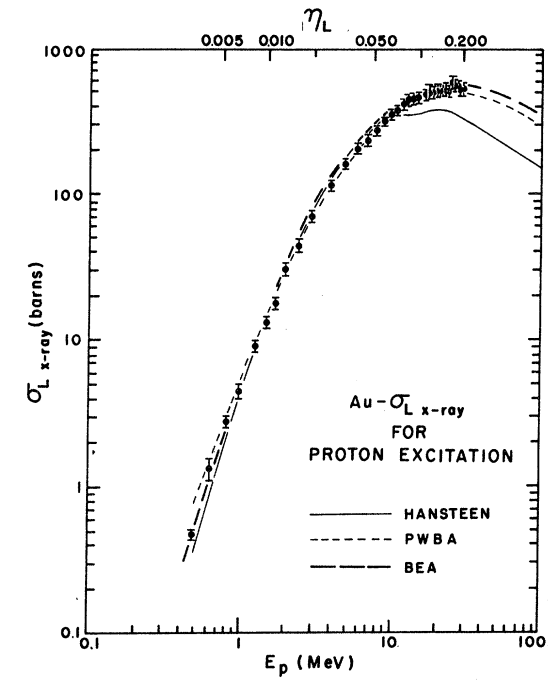

FIG. 3. Total Au L x-ray production cross sections for proton energies 0.5–30 MeV compared with available theoretical predictions.

the target, Mylar window, and air path, ϵ is the energy-dependent photopeak detection efficiency of the Si(Li) detector, ϕ is the number of protons incident on the target, t is the target thickness, and $\Delta\Omega$ is the solid angle subtended by the Si(Li) detector. The absolute x-ray production cross section σ_{LX} is then just

$$\sigma_{LX} = 4\pi(Y\alpha + Y\beta + Y\gamma) \tag{2}$$

assuming isotropy for L x-ray emission. This has been checked for Au L x rays in an experiment which was not able to resolve the $L\alpha$, $L\beta$, and $L\gamma$ lines.[15] More recently,[16] it was established that for $E_p = 2$ MeV, $L\alpha$, $L\beta$, and $L\gamma$ x rays are isotropic to within 3%.

Over the whole energy range covered in this experiment the nonrelativistic PWBA calculations agree best with experiment, although all theoretical predictions are quite close to the experimental results (see Fig. 3). However, the agreement at the lowest proton energies is probably fortuitous since the Coulomb deflection and relativistic effects will tend to cancel one another here.

B. $L\alpha/L\beta$ and $L\alpha/L\gamma$ Ratios

As pointed out in an earlier work[17] the intensity ratios $L\alpha/L\beta$ and $L\alpha/L\gamma$ can be calculated theoretically using ratios of subshell ionization cross sections. These ratios are expected to provide at least partial cancellation of Coulomb deflection corrections to σ_{Li}, as well as to diminish the effect of distortion of the L-electron wave functions by the incident proton. They were calculated from the following equations for the relative intensities:

$$I_{L\alpha} = \{[n_1(f_{13} + f_{12}f_{23}) + n_2 f_{23} + n_3]\,\omega_3\}\, F_{3\alpha}\,, \tag{3}$$

$$I_{L\beta} = n_1\omega_1 F_{1\beta} + (n_1 f_{12} + n_2)\,\omega_2 F_{2\beta} + [n_1(f_{13} + f_{12}f_{23}) + n_2 f_{23} + n_3]\,\omega_3 F_{3\beta}\,, \tag{4}$$

$$I_{L\gamma} = n_1\omega_1 F_{1\gamma} + (n_1 f_{12} + n_2)\,\omega_2 F_{2\gamma}\,, \tag{5}$$

where $n_3 = 1$, $n_1 = \sigma_{L1}/\sigma_{L3}$, and $n_2 = \sigma_{L2}/\sigma_{L3}$ and where σ_{Li} is the theoretical ionization cross section for the ith subshell.

Quantities such as $F_{3\beta}$ are the fraction of radia-

TABLE I. Gold L x-ray production cross sections and intensity ratios.

E_p (MeV)	σ_{LX} (barns)[a]	$L\alpha/L\beta$[b]	$L\alpha/L\gamma$[b]
0.50	0.45	1.69 ± 0.06	12.0 ± 0.7
0.64	1.21	1.70 ± 0.03	11.7 ± 0.6
0.83	2.78	1.79 ± 0.03	12.8 ± 0.6
1.00	4.52	1.78 ± 0.03	12.7 ± 0.6
1.28	9.05	1.72 ± 0.03	12.2 ± 0.6
1.50	13.6	1.69 ± 0.03	11.3 ± 0.5
1.75	17.7	1.66 ± 0.03	10.9 ± 0.4
2.00	28.4	1.61 ± 0.03	10.7 ± 0.5
2.50	43.5	1.57 ± 0.03	9.9 ± 0.4
3.00	60.9	1.57 ± 0.03	9.6 ± 0.4
4.0	114	1.51 ± 0.05	8.8 ± 0.5
5.0	162	1.47 ± 0.05	8.3 ± 0.5
6.0	209	1.47 ± 0.05	8.2 ± 0.5
7.0	238	1.44 ± 0.05	7.9 ± 0.4
8.0	274	1.44 ± 0.05	7.8 ± 0.4
9.0	315	1.43 ± 0.05	7.4 ± 0.4
10.0	354	1.44 ± 0.05	7.8 ± 0.4
11.0	387	1.41 ± 0.04	7.6 ± 0.4
12.0	417	1.45 ± 0.04	7.6 ± 0.4
13.0	444	1.45 ± 0.04	7.6 ± 0.4
14.0	454	1.46 ± 0.04	7.7 ± 0.4
15.0	459	1.43 ± 0.04	7.5 ± 0.4
17.0	488	1.44 ± 0.04	7.6 ± 0.4
18.0	504	1.43 ± 0.04	7.4 ± 0.4
19.0	494	1.46 ± 0.04	7.4 ± 0.4
20.0	506	1.46 ± 0.04	7.6 ± 0.4
21.0	507	1.41 ± 0.03	7.6 ± 0.4
22.0	500	1.40 ± 0.03	7.3 ± 0.4
23.0	532	1.39 ± 0.03	7.5 ± 0.4
24.0	500	1.40 ± 0.03	7.0 ± 0.4
25.0	586	1.43 ± 0.03	7.7 ± 0.4
26.0	557	1.46 ± 0.03	7.6 ± 0.4
27.0	548	1.47 ± 0.03	7.6 ± 0.4
28.0	546	1.43 ± 0.03	7.5 ± 0.4
29.0	513	1.41 ± 0.03	7.4 ± 0.4
30.0	516	1.43 ± 0.03	7.2 ± 0.4

[a]Errors: 8.7% for 0.5–15 MeV, 9.4% for 17–30 MeV.
[b]0.05–3.0-MeV values from Ref. 17.

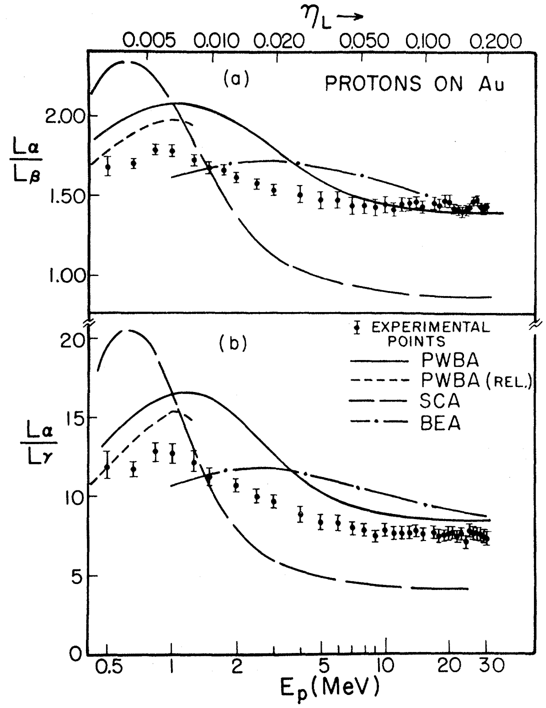

FIG. 4. Experimental intensity ratios $L\alpha/L\beta$ and $L\alpha/L\gamma$ for 0.5–30-MeV protons on Au compared with available theoretical predictions. The ratios for 0.5–3 MeV are from Ref. 17.

tive transitions in the $L\beta$ group which are associated with filling a hole in the $L3$ subshell, i.e.,

$$F_{3\beta} = \Gamma_{3\beta}/\Gamma_3 .$$

$\Gamma_{3\beta}$ is the sum of radiative widths for transitions which contribute to the $L\beta$ line that are associated with filling the hole in the $L3$ subshell. These radiative widths were taken from the calculations of Scofield.[18]

In Fig. 4, the results of the present experiment for the $L\alpha/L\beta$ and $L\alpha/L\gamma$ ratios are shown along with all available theoretical predictions (the ratios are also given in Table I). The observed peak in both experimental ratios lies at $E_p \approx 1$ MeV and is best reproduced by the PWBA calculations using relativistic hydrogenic wave functions,[14,17] although these calculations do not extend above 1.25 MeV. Over all, it is clear that PWBA calculations best predict the observed energy dependence of these ratios, and when the relativistic PWBA calculations are extended to higher projectile velocities, it is expected that these will offer further improvement in this agreement.

C. Centroid Shifts in $L\beta$ and $L\gamma$

The expected centroid shifts in $L\beta$ and $L\gamma$, which are composite lines with radiative contributions from more than one subshell, can also be calculated theoretically using the same subshell cross sections. The experimental centroid energies for $L\beta$ and $L\gamma$ are shown in Fig. 5, along with BEA, SCA, PWBA, and relativistic PWBA predictions of these centroids. These centroids are calculated from

$$\bar{E}_{L\beta} = \{n_1\omega_1 F_{1\beta}\bar{E}_{1\beta} + (n_1 f_{12} + n_2)\,\omega_2 F_{2\beta}\bar{E}_{2\beta} + [n_1(f_{13} + f_{12} f_{13}) + n_2 f_{23} + n_3]\,\omega_3 F_{3\beta}\bar{E}_{3\beta}\}/I_{L\beta}\,, \qquad (6)$$

$$\bar{E}_{L\gamma} = [n_1\omega_1 F_{1\gamma}\bar{E}_{1\gamma} + (n_1 f_{12} + n_2)\,\omega_2 F_{2\gamma}\bar{E}_{2\gamma}]/I_{L\gamma}\,, \qquad (7)$$

where $\bar{E}_{i\delta}$ is the centroid energy for contributions from the Li ($i = 1, 2, 3$) subshell to $L\delta$($\delta = \alpha, \beta, \gamma$). These are calculated from

$$\bar{E}_{i\delta} = \frac{\sum_j \Gamma_{i\delta_j} E_{i\delta_j}}{\sum_j \Gamma_{i\delta_j}}\,. \qquad (8)$$

$E_{i\delta_j}$ and $\Gamma_{i\delta_j}$ are the energies[19] and radiative widths,[18] respectively, of the components of $L\delta$ associated with radiative filling of the hole in the Li subshell. The classical BEA theory fails to predict the large centroid position variations (~100 eV) observed in $L\gamma$ (the predicted ~20 eV variations

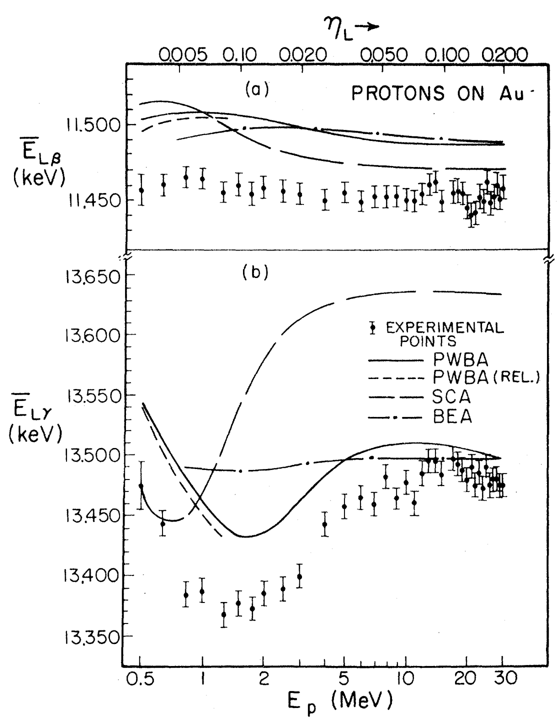

FIG. 5. $L\beta$ and $L\gamma$ centroid energies for 0.5–30-MeV protons on Au, compared with available theoretical predictions. The errors shown are the statistical errors in the peak position only. (These include calibration line errors. However, systematic errors from, e.g., amplifier nonlinearities, ADC nonlinearities, etc., are not included.)

in $\bar{E}_{L\beta}$ are just at the limits of detection with this detector) while the SCA predictions indicate ~200 eV variation over the energy range covered in this experiment and a minimum in $\bar{E}_{L\gamma}$ at $E_p \approx 800$ keV, much below the experimentally observed minimum at $E_p \approx 1.5$ MeV. Again the PWBA calculations are closest to experiment over the energy range covered, although, as in the case of the $L\alpha/L\beta$ and $L\alpha/L\gamma$ ratios, the agreement is not as good at the lowest energies. No shifts (< 10 eV) were observed in $L\alpha$ over the entire energy range. Although Eqs. (3)–(8) in principle would permit one to determine σ_{L1}, σ_{L2}, and σ_{L3} using the experimental $L\alpha/L\beta$, $L\alpha/L\gamma$, $\bar{E}_{L\beta}$, and $\bar{E}_{L\gamma}$ values, small systematic errors in analog-to-digital converter (ADC) or detection-system linearity of the order of 0.1%, as well as uncertainties in fluorescent and Koster–Kronig yields, have precluded extracting these subshell cross sections.

IV. CONCLUSIONS

The available theoretical predictions for inner-shell ionization cross sections, which range from the classical BEA to the first-order quantum-mechanical PWBA, all give rather good agreement with experimental values of σ_{LX}. Similarly, in the case of K-shell ionization there is also generally good agreement between theory and experiment, with the classical BEA theory in somewhat better agreement with experiment for projectile velocities at or below the root-mean-square (rms) velocity of the K-shell electron. However, the observed projectile energy dependence in the Au $L\alpha/L\beta$ and $L\alpha/L\gamma$ ratios, as well as the centroid shift in $L\gamma$, is poorly reproduced by the BEA theory. Part of this is probably because the values of σ_{L1}, etc., are scaled from calculated values[3] of σ_K for Mg, which means that we are using a theory employing K-shell electron velocity distributions to calculate L-subshell ionization cross sections.

It should be noted that the velocity distribution for the L-shell, averaged over all angular momenta[20] for $n=2$ gives just the expression used by Garcia for the K shell. This perhaps explains why the BEA predictions of σ_{LX} are in generally good agreement with experiment, while the $L\alpha/L\beta$, $L\alpha/L\gamma$, and $\bar{E}_{L\gamma}$ predictions are not.

Similar remarks hold true for the SCA results of Hansteen and Mosebekk, although here the predicted $L\alpha/L\beta$, $L\alpha/L\gamma$, and $\bar{E}_{L\gamma}$ values show far too much variation with projectile energy. Part of the reason for this large variation is the fact that σ_{L3} is not the largest subshell cross section. This is quite different from the results for all other theories, and is surprising in view of the fact that (a) $L3$ is the least tightly bound subshell and (b) it has four electrons while $L1$ and $L2$ both have only two electrons.

The PWBA calculations are clearly in better agreement with observed experimental trends in $L\alpha/L\beta$, $L\alpha/L\gamma$, and $\bar{E}_{L\gamma}$ than the BEA and SCA theories, even though PWBA predictions for σ_{LX} tend to deviate somewhat more from experimental results at lower energies. However, the Coulomb deflection and relativistic effects are strongest here so perhaps this is to be expected. Inclusion of relativistic wave functions in the PWBA calculations does improve agreement with experiment over the restricted range of these calculations. The effect of Coulomb-deflection corrections on the PWBA predictions of the subshell ionization cross sections is less certain. It seems clear though, that a consistent quantum-mechanical treatment of inner-shell ionization, even that used in the PWBA theory, provides better agreement with those experimental results that rely on subshell ionization cross sections than a classical or semiclassical treatment, even though the latter predict total L-shell ionization cross sections (or in this case, total L x-ray production cross sections) that are in as good agreement with experiment as the quantum-mechanical predictions.

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e^+-H Collisions by the Faddeev Approach

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The positron-hydrogen collision problem has been investigated by the Faddeev formalism as used by Sloan and Moore. The cross sections for the elastic scattering and the positronium formation have been calculated above the positronium-formation threshold for the incident positron energy up to 1.36 keV, and the results have been compared with those of other theoretical calculations. Appreciable differences have been found between the present results for the elastic-scattering cross sections and the corresponding results obtained by the first-order Born approximation at energies as high as the order of keV. The proton-positronium elastic-scattering cross sections are also reported. These cross sections which vanish in the first-order Born approximation are found to be significant at low incident proton energies.

I. INTRODUCTION

In view of the significant physical differences between positron-atom and electron-atom interactions, the study of positron-atom collisions deserves special attention. The scattering of the positron by a hydrogen atom has been extensively investigated theoretically. Below the positronium-formation threshold some refined calculations have been performed by Schwartz,[1] Kleinman *et al.*,[2] Drachman,[3] Perkins,[4] and recently by Kramer and Chen.[5] Above this threshold energy, Smith *et al.*[6] have calculated the elastic e^+-H-scattering cross sections by the close-coupling method. They have retained only the $1s$, $2s$, and $3s$ states of the hydrogen atom in their expansion and neglected the Ps formation. Burke *et al.*[7] have also investigated the same problem. In their close-coupling method they have, however, considered the $1s$, $2s$, and $2p$ states of the hydrogen atom. They have obtained the values of the cross sections which are less than those of Smith *et al.*[6] Recently Garibotti and Massaro[8] have applied the rational Padé approximants to calculate the cross sections for the elastic scattering of electrons and positrons by a hydrogen atom. They have also neglected the effects of the rearrangement channels in both cases. The importance of the Ps-formation channel even in the elastic scattering process has been emphasized by several authors.[9,10] Few theoretical calculations have so far been made on the e^+-H collision problem including the effect of the Ps formation, and the results of these calculations do not agree among themselves. Bransden and Jundi[11] have investigated the e^+-H collision problem in the close-coupling approximation taking the ground states of the hydrogen atom and the positronium in their eigenfunction expansion for the incident energy varying from 6.8 to 11.1 eV. They have considered the partial waves for $l=0$ and $l=1$ only. As they have concentrated on the low-energy region, they have also taken into account the effects of polarization in both the channels. Fels and Mittleman[12] have also considered the same problem, making allowance for the effect of polarization of the hydrogen atom and the positronium through phenomenological potentials. They have considered four partial waves ($l=0$, 1, 2, and 3) and obtained the values of the Ps-formation cross sections much smaller as compared to the first-Born-approximation (FBA) results of Massey and Mohr.[13]

The scattering of the positron by a hydrogen atom is a three-body problem, and this can be better investigated by the rigorous and elegant formulation of Faddeev.[14] Sinfailam and Chen[15] have used the Faddeev–Watson multiple-scatter-