Inner-shell ionization of silver by 100–400-keV electrons and positrons

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$K\alpha$ and total L x rays resulting from K- and L-shell ionization in silver by electron and positron impact at 100, 150, 200, 300, and 400 keV have been measured using a Si(Li) x-ray detector. Within an experimental uncertainty $\pm$ 10% the $K\alpha$ x-ray production cross section by electrons, $\sigma_{K\alpha}^{-}$, and that by positrons, $\sigma_{K\alpha}^{+}$, were found to be the same in the impact-energy region higher than 200 keV, but in the region below this energy it was discovered that the ratio $\sigma_{K\alpha}^{-}/\sigma_{K\alpha}^{+}$ increases as the impact energy decreases. As for the L x-ray production, no difference between electron and positron impact was found over the whole energy region studied in the present work, although the experimental uncertainty was rather large. Some discussion is included concerning the implications of the present experimental results.

I. INTRODUCTION

At present there is a large amount of experimental and theoretical investigation into electron-impact ionization of atoms over a very wide energy range, from several tens of electron volts to several hundred MeV.[1] However, only a few works on ionization by positron impact have been reported.[2-5]

It is of great interest to compare inner-shell ionization by electron impact with that by positron impact, especially in the impact-energy region near the shell-electron binding energy, since it is to be expected that some information on effects of the electron exchange and the Coulomb deflection would be obtained. The basic difference exists between the $e^- - e^-$ interaction (Møller[6]) and the $e^+ - e^-$ interaction (Bhabha[7]). The electron-electron exchange effect is included in the former, while the so-called positron-electron "exchange" through virtual annihilation and recreation of a new pair is included in the latter. Moreover, the contribution from the Coulomb interaction of incident particles with the target nucleus is different in both cases owing to the attractive and repulsive forces for electrons and positrons, respectively.

Kolbenstvedt[8] developed a theoretical treatment of K-shell ionization by impact of relativistic electrons, using the impact-parameter method and dividing the total cross section into two terms, close and distant collisions. Since his treatment appeared applicable to positron impact, Schiebel *et al.*[5] and Tawara[9] improved it and calculated the cross section of the K-shell ionization for positron impact, by replacing the Møller cross section by the Bhabha cross section in the close collision term. In their calculations a correction for the atomic Coulomb field is taken into consideration in a classical way. Recently, Hock[10] attempted to refine their calculations by correcting the close collisions for the initial and final Coulomb interactions, as will be discussed in Sec. III.

Experimental studies on inner-shell ionization by positron impact are very scarce. Flammersfeld and his collaborators[2,3] measured first the absolute K-shell ionization cross sections for elements with Z higher than forty, Zr, Ag, Sn, W, Au, and Pb, by impact of electrons and positrons in the energy range of 0.1–1.44 MeV using a β-ray spectrometer as a source of monoenergetic projectile beams and a NaI(Tl) detector for x-rays in an x – e^- and x – e^+ coincidence arrangement. Down to the lowest energy no systematic difference was found in the corresponding measured cross sections for electrons and positrons within experimental uncertainties between 10 to 20%. Berényi and his coworkers[4] determined the ionisation cross sections by positron impact for the K shell of Ni, Y, and Ag at 670 and 490 keV, and for the L shell of Yb, Ta, Au, and Pb at 490 keV, by adopting a technique similar to that of the Göttingen group (Flammersfeld *et al.*). Comparing their measured values with those obtained by electrons[11] with corresponding impact energies, they found no difference between them within errors of about 30%. These results by the Göttingen group and the Debrecen group imply that both the electron exchage effect and the Coulomb distortion effect are small in the impact energy region concerned. In the fully relativistic range (10–20 MeV) Schiebel *et al.*[5] reported ratios of K- and L-shell x-ray yields by electron and positron impact on Au with an accuracy of better than ±1.6%. Based on the assumption that there is no difference between the L x-ray production cross sections by electron and positron impact, they showed that the K-shell x-ray production cross section by electrons is about 2% larger than that by positrons. Their results seem to be in agreement with their calculations, which modify the Kolbenstvedt theory and introduce a correction factor for the deflection of the projectile trajectories in the Coulomb field of the nucleus. Moreover, they suggested that a larger difference in electron and positron impact is to be expected near the threshold energy, since the Coulomb deflection effect is much enhanced there.

The present work was attempted in order to obtain valuable information on electron exchange and Coulomb distortion of the projectile waves due to the target nucleus, especially near the threshold energy where the ionization theory has not yet been well established. By the use of a β-ray spectrometer and a Si(Li) x-ray detector, we have measured the $K\alpha$ x-ray and total L x-ray yields for 100–400-keV electrons and positrons in the case of $_{47}$Ag with the K-shell binding energy $B_K = 25.5$ keV and the average L-shell binding energy $B_L = 3.5$ keV. The impact energies of the present work correspond to about $4B_K$–$16B_K$ and $29B_L - 114B_L$. Since our experiment was focused on the study of the differences between electron and positron impact, the relative x-ray production cross sections have been measured, and the ratios of the cross sections by electrons to that by positrons have been evaluated. The present relative measurement was free from serious uncertainties caused by evaluation of the x-ray detection efficiency *in situ* and of the target thickness. Several annihilation processes with inner-shell electrons were discussed, in so far as they may be competing processes in enhancing x-ray yields in the positron impact. We report here the details of our experiment and compare the results with the theoretical calculations based on the Kolbenstvedt theory.[8] A brief account of the present work has been previously reported.[12]

II. EXPERIMENTAL

Monoenergetic electron and positron beams were obtained by the use of a sector-type double-focusing β-ray spectrometer[13] mounted with 12-mCi ^{137}Cs and 10-mCi ^{90}Sr(+ ^{90}Y) as electron sources and 10-mCi ^{22}Na as a positron source. Using these sources 100-, 150-, 200-, 300-, and 400-keV beams were achieved. The energy calibration of these beams was carried out using K- and L-conversion lines of ^{109}Cd, ^{113}Sn, and ^{137}Cs, which lie in the energy range from 63 to 656 keV. The resolution in momentum full width at half maximum (FWHM) was found to be 1.0%.

The experimental arrangement in the target chamber is shown in Fig. 1. A self-supporting pure (99.97%) silver target of 30×27 mm^2 in

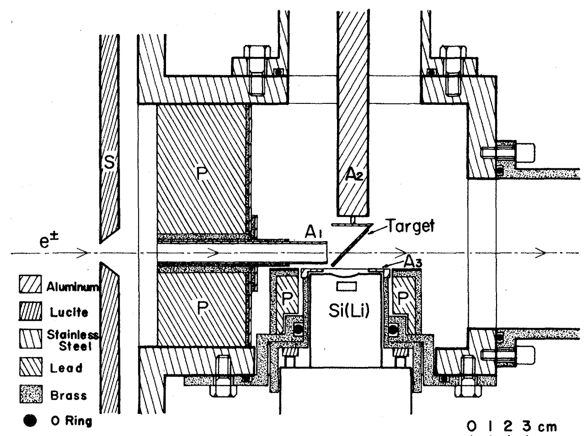

FIG. 1. Experimental arrangement in the target chamber: S, slit; P, lead shields; A_1, 10-mm-diam by 93-mm-long aluminum collimator; A_2, aluminum target holder; A_3, aluminum cap-ring with a 220-μm-thick beryllium window.

size and 0.52 mg/cm^2 in thickness was mounted at an angle of 45° with respect to the incident beam direction. The Ag K and L x-rays emitted by the process to be studied were observed by a Si(Li) detector (80×5 mm^2) with a 25-μm-thick Be window, which was placed just below the target but beyond a Be diaphram of 220-μm thickness. The energy resolution (FWHM) of this detector was found to be 280 eV for 5.9-keV Mn K x rays. The target chamber was designed as carefully as possible so as to reduce unfavorable effects of the annihilation rays emitted from the slits and of other x rays emitted from surrounding materials.

The absolute number of the incident particles impinging on the target foil was measured for each selected energy by a 32-mm diam × 5-mm thick plastic scintillator placed at about 5 cm behind the target positron but with the target removed. Positioning of the target was examined to be reproducible within 0.7% as regards the effective solid angle subtended by the detector to the target. Typical energy spectra of the incident electrons and positrons (300 keV) are shown in Fig. 2. In the positron spectrum, as shown in the figure, a small tail is appreciable in the higher-energy region extending to energies about 400 keV higher than the peak position. This small higher-energy spectrum is caused by the Compton recoil electrons produced by the 511-keV annihilation rays, which were in turn produced by incident positrons stopped in the plastic scintillator. Taking account of this fact, for the total number of incident positrons on the target foil we adopted the total counts integrated over the

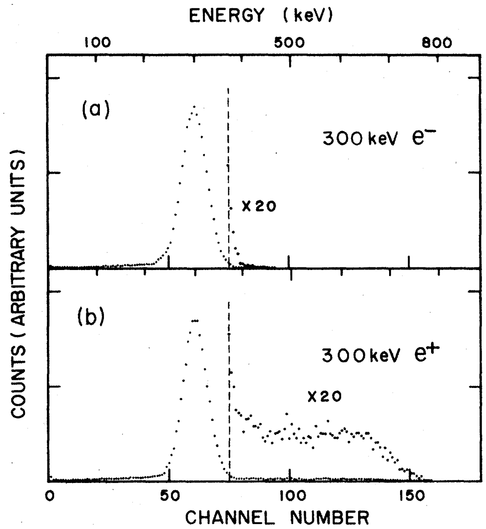

FIG. 2. Typical spectra of incident electrons and positrons observed with a plastic scintillation detector. Spectra (a) and (b) are obtained with 300-keV electrons and positrons, respectively.

energy region until about 400 keV higher than the peak positions for all the incident energies studied. The number of the incident particles on the target-per-unit time thus measured varied from 1.9×10^3 to 7.6×10^3/sec, depending on the impact energy and the source used. During long-run x-ray measurements the plastic detector was used for monitoring the particles passing through the target, but it was placed about 30 cm from the target position to reduce as much as possible the undesirable effects of the annihilation rays emitted from this detector. In each set of measurements it was confirmed that the intensity of the incident beam did not vary by more than ±2.0%.

When positrons are used as the projectiles, in the vicinity of the target foil there may be an abundance of Compton-scattered photons of the 511-keV annihilation rays originating from the surrounding materials of the target such as the collimator and walls of the target chamber. For this reason, in the case of positron impact the Ag x rays produced by impact ionization may be contaminated by those of another origin, i.e., by Ag x rays caused by the photoelectric effect of scattered γ rays with degraded energies around the K-absorption edge (25.5 keV) and/or L-absorption edge (~3.5 keV). In order to estimate the contribution by contamination, a Lucite plate of 50×50 mm^2 and 5-mm thickness was placed just in front of the target. The thickness of this plate was such that all incident positrons were stopped in it. In this case, since the target was irradiated by only photons, including both annihilation rays from the plate and scattered rays from the surroundings, the Ag x rays emitted from the target foil were caused by photoionization. Moreover, by inserting such a Lucite plate, a contribution to the x-ray yield from the bremsstrahlung by projectiles, if it exists, can also be determined. The background intensity with the plate was much stronger than that without it. For this reason, observed x-ray peak areas with and without the plate had to be compared with each other by normalizing them to the areas of the background under the peaks in the respective observed spectra. By this procedure it was confirmed that photoelectric production of K x rays was negligibly small, since no K x-ray peak could be observed with the Lucite plate. As to the L x-ray yield, however, the contribution from the same origin was found to be between 3.6–9.3% of the L x-ray yields for 100–400-keV positron impact. These results are reasonable because the atomic photoelectric cross section for L shell at L-absorption edge is about 26 times that for K shell at K-absorption edge.[14] This trend depends, of course, on the present geometrical arrangement of the target, the Si(Li) detector, and the walls of the target chamber. When we evaluated the L x-ray yield, the contribution estimated by this procedure was subtracted from the observed spectrum by taking account of the background intensities with and without the Lucite plate. It is also noted that by inserting the Lucite plate in the case of electron impact we could confirm photoproduction of x rays by the bremsstrahlung being negligibly small in the present work.

In order to obtain the final results with good statistics, x-ray data were accumulated for about two weeks or more in each measurement run for selected impact energies of electrons and positrons. The stability of the whole system, crucial to the success of the experiment, was checked every 12 h during long-run measurements.

III. RESULTS AND DISCUSSION

When we estimate the x-ray production from the target by the impact-ionization of positrons, we must examine the possible competing processes which may enhance the x-ray yield, viz., several annihilation processes of positrons in flight with strongly bound electrons which result in the creation of the inner-shell vacancy followed by emission of K or L x rays. Of these processes, single-quantum annihilation (SQA),[15-18] radiationless annihilation (ZQA),[19] and two-quantum annihilation (K-TQA)[20] are known; all of these processes have some probability to produce K and/or L x rays. These processes are more probable for the K

shell than for outer shells, as the recoil momentum must be taken up by the nucleus concerned. According to calculations of the total SQA cross sections by Johnson *et al.*,[15] which are in good agreement with the experimental results,[16,18] K-shell SQA cross section for Ag is 0.048 barns as a maximum value at 190-keV incident energy. The contribution of L-shell electons was calculated by Broda and Johnson[17]: about 13% of the K-shell cross section in nearly the same energy region studied in the present work. As for ZQA and K-TQA, available information is quite limited, since for each process only one experimental work has so far been performed, and this in our laboratory.[19,20] Nevertheless, from our experimental results on these processes it is reasonable to assert that the cross section of K x-ray production by ZQA is less than 0.01 barns and that by K-TQA is of the order of 0.1 barns for Ag as an upper limit and positrons with incident energies involved in the present work. Reflecting these very small probabilities for production of K x rays by SQA, ZQA, and K-TQA in comparison with the present impact-ionization process of about 50–60 barns,[21] contributions from these three annihilation processes can be ignored in the present work. However, it is emphasized that the contributions from these annihilation processes are sizable in the case of high-Z elements and several-hundred-keV impact-energy range, since the cross sections of the processes increase strongly with Z; e.g., the measured SQA cross section, which increases roughly with Z^5, is 0.6 ± 0.2 barns at 300 keV for $_{79}$Au (Ref. 18.)

Typical observed spectra of the x rays are shown in Fig. 3. Because of the limited resolution of the Si(Li) detector used, $K\alpha$ x-ray peak cannot be resolved into $K\alpha_1$ and $K\alpha_2$ lines and L x-ray peak into $L\alpha$, $L\beta$, . . . lines. The $K\beta$ x-ray peak is also observed, but with poor statistics, as shown in the figure. The background is due to the bremsstrahlung in the case of electron impact, while in the positron experiment it is due to the bremsstrahlung plus more abundant annihilation rays. The peak areas were evaluated by a least-squares fitting to the background counts on both sides of the peak. Here, the background for the $K\alpha$ x-ray peak was assumed to be linear, while for the L x-ray peak it was assumed to be polynomial. The errors were estimated by including the background-subtraction uncertainty and statistical error. In order to check and improve our estimation of peak area a nonlinear least-squares fitting to the $K\alpha$ x-ray peaks was also performed, assuming several peak shapes to be Gaussian with an exponential tail on the low-energy side. It was confirmed that the evaluated peak areas in both methods were in good agreement. However, since the errors obtained from the nonlinear fitting technique were not improved noticeably in comparison with the simple background subtraction, we adopted the subtraction method to evaluate the peak areas and their errors.

Figure 4 displays measured $K\alpha$ x-ray production cross sections, expressed by a ratio of N_x/N_p, where N_x is the observed Ag $K\alpha$ x-ray yield and N_p is the total number of impact electrons or positrons during the net duration of measurement. In the region of impact energy higher than 200 keV, i.e., about eight times K-shell binding energy (25.5 keV), measured values with electron and positron impact lie very close to each other. From the experiments by Webster

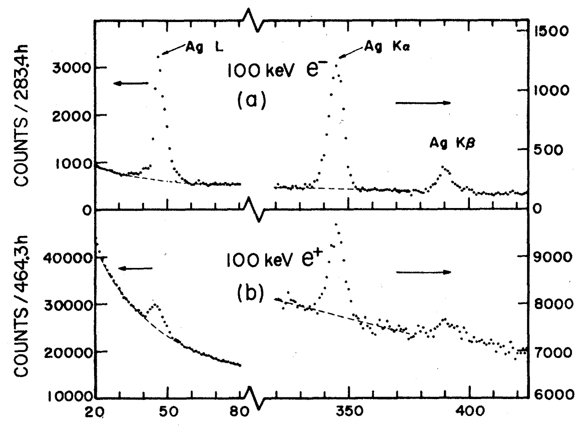

FIG. 3. Typical spectra of Ag x rays observed with a Si(Li) detector. Spectra (a) and (b) are obtained with 100-keV electrons and positrons, respectively. The broken curves indicate the most reasonable backgrounds determined by the least-squares fitting technique.

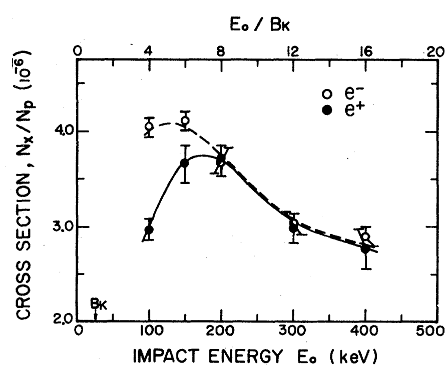

FIG. 4. $K\alpha$ x-ray production cross sections by electrons (open circle) and positrons (closed circle), expressed by a ratio of N_x/N_p, where N_x is the observed Ag $K\alpha$ x-ray yield and N_p is the total number of impact electrons or positrons during the net duration of measurement. The broken and solid curves are drawn to guide the eye.

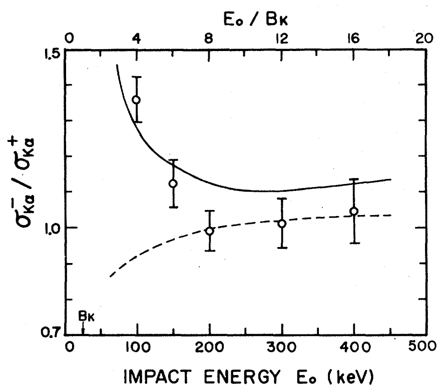

FIG. 5. Measured ratio $\sigma^-_{K\alpha}/\sigma^+_{K\alpha}$ of the cross section for $K\alpha$ x-ray production by electrons to that by positrons. The broken curve shows our estimation without Coulomb deflection within the framework of the Kolbenstvedt theory. The solid curve represents the theoretical prediction by Hock (Ref. 10), taking account of Coulomb deflection.

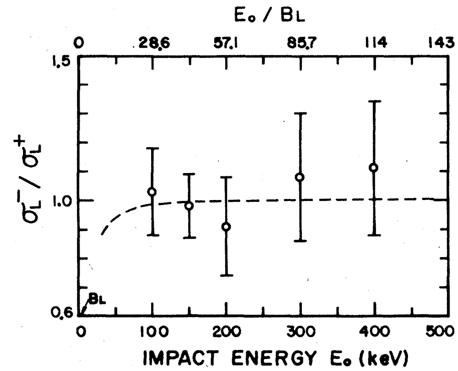

FIG. 6. Measured ratio σ^-_L/σ^+_L of the cross section for total L x-ray production by electrons to that by positrons. The broken curve shows our estimation similar to that in Fig. 5.

et al.[22] and Davis *et al.*[23] it has been found that the K-shell ionization cross section by electron impact becomes maximum at about 100 keV. This trend is also confirmed by the present work. However, in the case of positron impact, we have found that the impact energy which yields the maximum cross section shifts to the higher-energy side by roughly 80 keV.

In order to study the phenomenon more precisely, we set out to determine the measured ratio of the cross section for the $K\alpha$ x-ray production by electrons to that by positrons, $\sigma^-_{K\alpha}/\sigma^+_{K\alpha}$, as a function of the impact energy, and our results are shown in Fig. 5. In the energy range higher than 200 keV measured ratios agree with unity within and experimental error ±10%, but below this range the ratio goes up with decrease in energy: 1.12 ± 0.07 at 150 keV and 1.36 ± 0.06 at 100 keV. The similar ratio σ^-_L/σ^+_L obtained for L x-ray production is presented in Fig. 6. Unfortunately, the errors are relatively large owing to the high background under the L x-ray peak in the positron-impact case, as shown in Fig. 3(b). Owing to these uncertainties, differences in the L x-ray production, if they exist, are not appreciable over the whole range.

It is noted, however, that in the impact-energy range of 0.1–1.44 MeV, σ^-_K and σ^+_K as measured with several elements including Ag by Flammersfeld and his coworkers,[2,3] agree with each other within errors of 10 to 20%. Their results seem to support the treatment of the plane-wave Born approximation (PWBA), based on the assumption that both the exchange effect between incident electron and atomic electron and the Coulomb-deflection effect due to the target nucleus are small in this energy range. It is, however, well known that their measured values for electron impact are in clear disagreement with those obtained by other workers.[21] Here, as viewed from the experimental side, we wish to emphasize that experimental determination of some of the factors necessary for evaluation of absolute cross sections is very difficult.

In the framework of the theory by Kolbenstvedt,[8] we have attempted to calculate these ratios by adopting a treatment similar to that used by Schiebel *et al.*[5] and Tawara,[9] but without including the correction for the Coulomb deflection. The total L x-ray production cross sections have been calculated in the usual fashion[24] by using the photoelectric cross section for the L shell provided by Hall[25] (instead of that for the K shell in the Kolbenstvedt theory) and by the use of the atomic parameters, i.e., fluorescence and Coster-Kronig yields of L subshells, taken from the work of McGuire[26] and relative radiative branching fractions from the calculations of Scofield.[27] Our results are shown by broken curves in Figs. 5 and 6.

In Fig. 5 the calculated curve is in good agreement with our experimental values at 200, 300, and 400 keV. In the energy region lower than 200 keV, however, the curve goes downward owing to the exchange effect incorporated in the Møller cross section and to a negligible contribution from the virtual annihilation terms in the Bhabha cross section. This tendency is strongly contrary to the behavior of our data. In order to explain our data, Hock[10] has recently attempted to calculate the σ^-_K/σ^+_K by taking account of the correction for the Coulomb distortion. He corrected the close collisions for the initial- and final-state Coulomb interactions, at least in the low-energy region. In his treatment, for the ejected electron-nucleus distortion, he used the ratio of the nonrelativistic Coulomb-wave atomic matrix element to that for plane wave. Further, for the projectile-nucleus Coulomb distortion, he used the nonrelativistic Coulomb-wave bremsstrahlung cross sections, different for electrons and positrons. His results, as given by a solid curve in Fig. 5, show agreement with the general trend of our experimental results. This fact strongly suggests that the effect of the Coulomb deflection plays an important role in the impact-energy range lower than several times K-shell binding energy. As for the experimental results of the L x-ray yields, the measured ratios σ_L^-/σ_L^+, although with large errors, seem to agree with our calculated prediction, as shown in Fig. 6.

Through the present work of the K and L x-ray production by electrons and positrons in the case of Ag, we conclude as follows. Within the experimental uncertainty of about 10% the K-shell ionization cross sections are the same for electrons and positrons in the energy range from 200 to 400 keV. This result provides an experimental support for PWBA treatment in this energy range. In the energy region lower than 200 keV, it has been found experimentally for the first time that the K-shell ionization cross section by electrons is larger than that by positrons. This result strongly indicates that the effect of the Coulomb deflection due to the target nucleus is very pronounced there in comparison with the effect of electron exchange. As for L x-ray yield no appreciable difference between the experimental data with electrons and positrons was found over the whole impact energy range studied.

It is hoped that experiments in the impact energies nearer to the threshold energy will be attempted so that additional information can be obtained. Experiments with heavy target elements would be of great interest; measurements using $_{79}$Au are now in progress. It is also hoped that more elaborate theoretical work will be done on the difference between electron and positron impact.

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[1]For a review see D. H. Madison and E. Merzbacher, in *Atomic Inner-Shell Processes*, edited by B. Crasemann (Academic, New York, 1975), Vol. 1, Chap. 1; C. J. Powell, Rev. Mod. Phys. 48, 33 (1976); S. Morita, in *Proceedings of the Second International Conference on Inner Shell Ionization Phenomena—Invited Papers, 1976, Freiburg, West Germany*, edited by W. Mehlhorn and R. Brenn (Universität Freiburg, Germany, 1976), p. 339.

[2]H. Hansen, H. Weigmann, and A. Flammersfeld, Nucl. Phys. 58, 241 (1964).

[3]H. Hansen and A. Flammersfeld, Nucl. Phys. 79, 135 (1966).

[4]S. A. H. Seif el Nasr, D. Berényi, and Gy. Bibok, Z. Phys. 271, 207 (1974).

[5]U. Schiebel, E. Bentz, A. Müller, E. Salzborn, and H. Tawara, Phys. Lett. 59A, 274 (1976).

[6]C. Møller, Ann. Physik 14, 531 (1932).

[7]H. J. Bhabha, Proc. R. Soc. London Ser. A 154, 195 (1936).

[8]H. Kolbenstvedt, J. Appl. Phys. 38, 4785 (1967); 46, 2771 (1975).

[9]H. Tawara, in *Proceedings of the 10th International Conference on the Physics of Electronic and Atomic Collisions, Paris, July 21–27, 1977*, edited by G. Watel (North-Holland, Amsterdam, 1978), p. 311.

[10]G. Hock, in *Abstracts of Contributed Papers of the XIth International Conference on the Physics of Electronics and Atomic Collisions, Kyoto, August 29–September 4, 1979*, edited by K. Takayanagi and N. Oda (The Society for Atomic Collision Research, Tokyo, Japan, 1979), p. 974.

[11]S. A. H. Seif el Nasr. D. Berényi, and Gy. Bibok, Z. Phys. 267, 169 (1974).

[12]S. Ito, K. Kubota, and S. Shimizu, Jap. J. Appl. Phys. 17, Suppl. 17–2, 338 (1978).

[13]H. Yamamoto, K. Takumi, and H. Ikegami, Nucl. Instrum. Methods 65, 253 (1968).

[14]J. H. Hubbell, At. Data 3, 241 (1971).

[15]W. R. Johnson, D. J. Buss, and C. O. Carroll, Phys. Rev. 135, A1232 (1964).

[16]H. Mazaki, M. Nishi, and S. Shimizu, Phys. Rev. 171, 408 (1968).

[17]K. W. Broda and W. R. Johnson, Phys. Rev. A 6, 1693 (1972).

[18]T. Mukoyama, H. Mazaki, and S. Shimizu, Phys. Rev. A 20, 82 (1979).

[19]S. Shimizu, T. Mukoyama, and Y. Nakayama, Phys. Rev. 173, 405 (1968).

[20]T. Nagatomo, Y. Nakayama, K. Morimoto, and S. Shimizu, Phys. Rev. Lett. 32, 1158 (1974).

[21]J. W. Motz and R. C. Placious, Phys. Rev. 136, A662 (1964); D. H. Rester and W. E. Dance, *ibid.* 152, 1 (1966).

[22]D. L. Webster, W. W. Hansen, and F. B. Duveneck, Phys. Rev. 43, 839 (1933).

[23]D. V. Davis, V. D. Mistry, and C. A. Quarles, Phys. Lett. 38A, 169 (1972).

[24]Y. K. Park, M. T. Smith, and W. Scholz, Phys. Rev. A 12, 1358 (1975).

[25]H. Hall, Rev. Mod. Phys. 8, 358 (1936).

[26]E. J. McGuire, Phys. Rev. A 3, 587 (1971).

[27]J. H. Scofield, Phys. Rev. 179, 9 (1969).