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Probability for double photoionization of He and Ne

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The proportionality between the probability for producing a doubly charged ion by photon impact on a neutral atom and electron impact on a singly charged ion is discussed. An energy-dependent parameter is introduced that expands the proportionality from threshold to 500 eV for He and to 360 eV for Ne.

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Calculations and measurements of the production of doubly charged ions by photon impact on neutral atoms and of the ratio of double to single photoionization have been of considerable interest for many years [1–22]. The two major reasons for this interest are (a) the implication that electron-correlation processes should be taken into account in any calculation of the above ratio and (b) that most calculations predict that at "high" photon energies the ratio should reach a constant value.

We have recently shown [22] that over a considerable energy range the ratio of double to the total photoionization cross section (single plus double ionization) is proportional to the cross section σ_e^+ for electron impact ionization of an ion. We interpret this proportionality as experimental evidence that double photoionization proceeds first by the absorption of a photon by a single electron followed by the interaction of the photoelectron with the remaining orbital electrons. We present here an improved analysis of this result and show that the proportionality holds from the double-ionization threshold to 500 eV for He and from threshold to about 360 eV for Ne.

The probability for producing a doubly charged ion by photon impact is given by the ratio $\sigma_\gamma^{2+}/\sigma_\gamma(\text{abs})$, where σ_γ^{2+} is the cross section for double photoionization and $\sigma_\gamma(\text{abs})$ is the total absorption cross section and equals the sum of the partial cross sections for single and double photoionization. Although triple ionization occurs in Ne for photon energies in excess of 120 eV the cross section is an order of magnitude smaller than the double-ionization cross section [15] and will not be considered here.

Previously, we had shown [22] that over a limited energy range

$$\sigma_\gamma^{2+}/\sigma_\gamma(\text{abs}) \propto \sigma_e^+ , \tag{1}$$

and that the proportionality constant had a magnitude approximately equal to the cross-sectional area of the ion. Thus we can rewrite Eq. (1) as

$$\sigma_\gamma^{2+}/\sigma_\gamma(\text{abs}) = \sigma_e^+/a\sigma_e(\text{abs}) , \tag{2}$$

where a is a dimensionless constant, $\sigma_e(\text{abs}) = \pi R^2$, and R is the radius of the electron-ion interaction zone and is approximately equal to the ionic radius. The ratio $\sigma_e^+/\sigma_e(\text{abs})$ is then the probability for producing a doubly charged ion by electron impact on an ion.

Physically, the *effective* size of the interaction volume will depend on the energy of the incident electron. Because of the Coulomb attraction between the ion and incident electron, the electron will be pulled in toward the ionic target, as shown schematically in Fig. 1. Thus, we now have an *effective* radius b for the interaction zone, where b is the impact parameter for an electron of a given initial kinetic energy T_0, which can just enter the interaction zone of radius R. Therefore, $\sigma_e(\text{abs})$ becomes equal

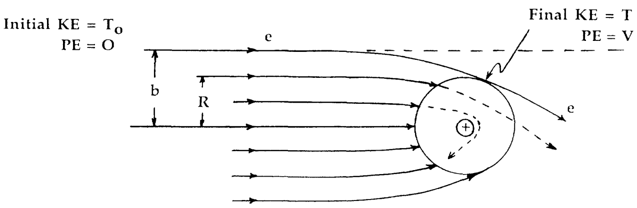

FIG. 1. Schematic diagram for electron impact on an ion showing the impact parameter b for an electron of initial kinetic energy T_0 that can just enter a zone of radius R.

to πb^2. From the conservation of energy and angular momentum we find that

$$b^2=R^2(1-V/T_0)\ , \tag{3}$$

where V is the potential energy of the electron at a distance R from the ionic nucleus. Thus, equating $\sigma_e(\text{abs})$ to πb^2, Eq. (2) becomes

$$\sigma_\gamma^{2+}/\sigma_\gamma(\text{tot})=\sigma_e^+/k(1-V/T_0)\ , \tag{4}$$

where k is a constant that is proportional to πR^2. The electron-impact ionization cross section of a single charged ion, σ_e^+, has been determined experimentally for several different ions [23–31]. To compare the electron and photon impact data we need to know the value of V. This is, of course, one of the difficulties encountered in a rigorous analysis of double photoionization. In comparing the photon- and electron-impact data we treat k and V as fitting parameters to obtain the best agreement. However, these parameters do have a physical significance as described above.

For Ne, we have taken $V=-43$ eV and $k=227$ Mb (1 Mb$=10^{-18}$ cm^2) in order to give the best fit to the photoionization data. The values for σ_e^+ were taken from the experimental data of Man, Smith, and Harrison [24]. The results, plotted as a function of the total energy carried off by the electrons, are shown in Fig. 2. There is an excellent fit between the two sets of data from threshold to 360 eV. Even out to 600 eV the deviation is only 20%. A reasonable fit could also be maintained by varying V between -41 and -47 eV provided the normalizing constant was varied from 233 to 217 Mb, respectively. The dotted curve results if we assume that $\sigma_e(\text{abs})$ is not dependent on the incident electron energy and is simply proportional to k. In this case k was chosen equal to 242 Mb simply to make the data agree with the solid-line curve at 600 eV. This illustrates the effect $\sigma_e(\text{abs})$ has on the ratio over the first few hundred electron volts. The dashed line represent the many-body perturbation calculation by Chang and Poe [11]. This calculation included several specific electron-correlation processes and is in reasonable agreement with experiment.

Figure 3 shows an expansion of the threshold region between 0 and 120 eV. Again the dotted curve results if we assume $\sigma_e(\text{abs})$ is not dependent on the incident electron energy. But this time k was chosen equal to 370 Mb for the best fit to the experimental data near threshold. We can see the dramatic improvement (solid line) when Eq. (4) is used to obtain the best fit. The experimental data between 0 and 10 eV are consistently higher than the electron-impact curve. However, this is caused by an increase in the value of σ_γ^{2+} produced by numerous doubly excited neutral states of Ne that autoionize into the doubly ionized continuum [32,33].

For He, we have compared the photoionization data to the experimental electron-impact data of Peart, Walton, and Dolder [23] for σ_e^+. In the range $\varepsilon=0$–200 eV we find that the best fit to the photoionization data occurs when we consider $\sigma_e(\text{abs})$ to be constant and use a single normalizing constant $k=96.2$ Mb. This result is shown in Figs. 4 and 5 (long-dashed curve). However, above 200 eV an energy-dependent value of $\sigma_e(\text{abs})$ gives a best fit to our data with $V=-20$ eV and $k=87$ Mb (solid line). In either case the results of Eq. (4) satisfactorily reproduce the energy dependence of the ratio $\sigma_\gamma^{2+}/\sigma_\gamma(\text{abs})$ between 0 and 500 eV. Figure 4 shows the experimental data of several other groups [12–15,34]. The overall scatter in the data is less than ±8%. The short-dashed curve represents the theoretical data of Carter and Kelly [16], who used the many-body perturbation theory. Considering the experimental errors in both the photoionization and electron impact data we see a remarkable agreement between all sets of data. We note that the Carter and

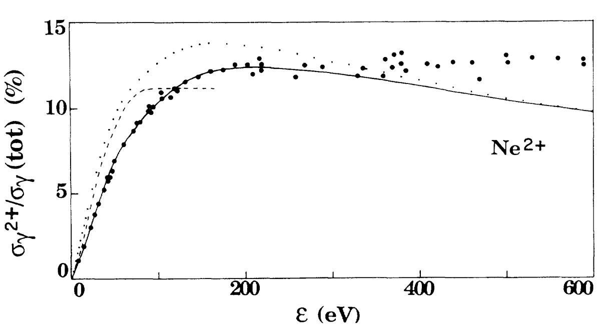

FIG. 2. The photoionization probability for producing Ne^{2+} plotted as a function of the total kinetic energy of the released electrons. ●, present data; ——, predicted probability for $k=227$ Mb and $V=-43$ eV; · · · ·, predicted probability for $\sigma_e(\text{abs})$ constant and $k=242$ Mb; — — —(theory), Ref. [11].

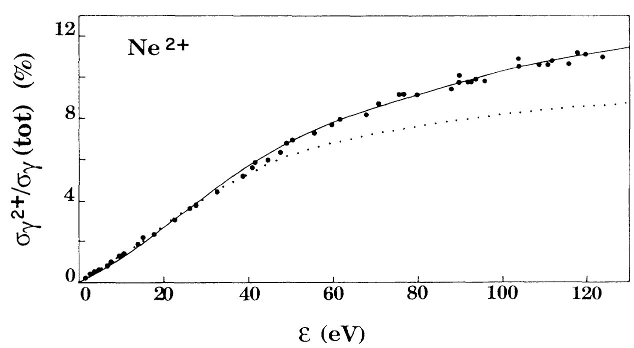

FIG. 3. The photoionization probability for producing Ne^{2+} plotted as a function of the total kinetic energy of the released electrons. The experimental data points represent the present data. The solid curve is the predicted probability for the parameters $k=227$ Mb and $V=-43$ eV; · · · ·, predicted probability for $\sigma_e(\text{abs})$ constant and $k=370$ Mb.

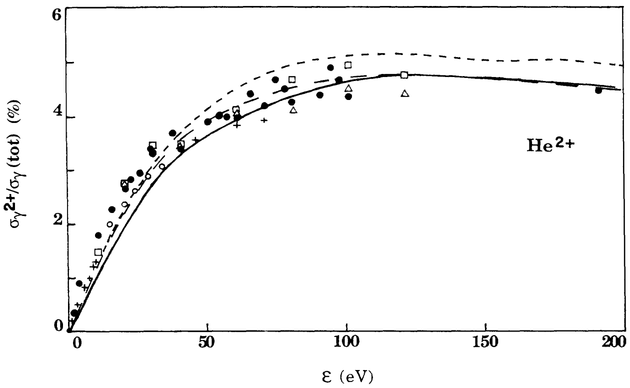

FIG. 4. The photoionization probability for producing He^{2+} plotted as a function of the total kinetic energy of the released electrons. ●, present data; +, Refs. [12,13]; □, Ref. [14]; △, Ref. [15]; ○, Ref. [34]; — — —(theory), Ref. [16]; —— —— ——, predicted probability for σ_e(abs) constant and $k=96.2$ Mb; ——, predicted probability for $k=87$ Mb and $V=-20$ eV.

Kelly data, shown in Fig. 4, can be reproduced within ±3% by use of Eq. (4) when σ_e(abs) is constant and $k=90$ Mb.

The dotted curve in Fig. 5 represents the calculated results of Amusia *et al.* [10] and the short-dashed curve represents the calculations of Ishihara, Hina, and McGuire [20]. The single experimental data point at 2720 eV was obtained by Levin *et al.* [19]. Their error bar lies within the ±10% random and systematic errors for the measured values of σ_e^+. This is indicated by an error bar on the solid curve. The theoretical data of Ishihara, Hina, and McGuire predicts that the ratio of double to *single* photoionization reaches a near limiting value of 1.60%. Earlier calculations by Byron and Joachain [2] and by Aberg [5] both predicted asymptotic values of 1.66%, and a recent calculation by Dalgarno and Sadeghpour [35] predicts a ratio of 1.68%. The work

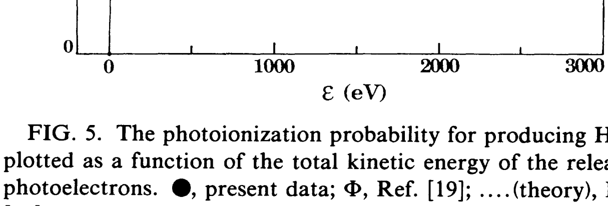

FIG. 5. The photoionization probability for producing He^{2+} plotted as a function of the total kinetic energy of the released photoelectrons. ●, present data; Φ, Ref. [19]; ....(theory), Ref. [10]; — — —(theory), Ref. [20]; —— —— ——, predicted probability for σ_e (abs) constant and $k=96.2$ Mb; ——, predicted probability for $k=87$ Mb and $V=-20$ eV (the error bar indicates a ±10% error in σ_e^+).

by Brown [6–8] and Amusia *et al.* predict limiting ratios of 2.0% and 2.33%, respectively. Recent additional data by Levin *et al.* [36] indicate the existence of a plateau between 3 and 5 keV with a ratio of approximately 1.7%. Certainly, the present data in conjunction with the Levin data are compatible with the existence of a limiting value of the ratio between 3 and 5 keV.

In the many-body perturbation calculations the three major electron-correlation processes considered are initial-state correlation, shake off, and electron-electron scattering. The latter correlation process can be expected to occur in both photon-neutral-atom and electron-ion impact experiments. In the case of He, initial-state correlations can occur only in the neutral atom. This is one fundamental difference between the photon- and electron-impact experiments. However, the good agreement between the two experiments suggests that for low-energy impact on He^+ (arbitrarily, say <500 eV) the interaction time allows the two electrons to couple in a way analogous to the initial-state correlations in neutral He. This is certainly true in the threshold region (Wannier law) where the energy dependence for the probability that two electrons can escape is proportional to $\varepsilon^{1.056}$ for either photon–neutral-atom or electron-ion impact on helium [37,38]. At higher energies the interaction time decreases and the coupling may be expected to decrease eventually leaving only electron-electron-scattering processes to produce He^{2+}. Thus, the ratio σ_e^+/σ_e(abs) could be expected to fall off more rapidly than $\sigma_\gamma^{2+}/\sigma_\gamma$(abs) at higher energies.

The limiting value of the photoionization ratio predicted by theory depends upon the single- and double-ionization cross sections having exactly the same energy dependence. Although various calculations [5,10,20,35] have predicted that at "high" energies the single- and double-ionization cross sections both vary as $E_\gamma^{-3.5}$ we have to be cautious because no exact calculation has been made at present. If we check the energy dependence of the published total cross sections of He between 2 and 3 keV we find that the theoretical data of Bell and Kingston [4] and of Veigele [39] give an exponent of ~ -3.26, whereas the compilation tables of Marr and West [40] give a value of -2.87. It is interesting to note that if we assume that the energy dependence of σ_γ^{2+} is, in fact, equal to $E_\gamma^{-3.5}$ then using the Marr and West data the ratio $\sigma_\gamma^{2+}/\sigma_\gamma$(abs) must have an energy dependence of $E_\gamma^{-0.63}$, which is very close to the value we obtain from our electron-impact model, namely, $E_\gamma^{-0.75}$ for photon energies between 800 eV and 4 keV. The good agreement may be fortuitous because experimental data for σ_γ(abs) are scarce and not too accurate in this energy region. More precise experimental data are needed to determine whether the ratio of double to single photoionization of He reaches a limiting value or continues to decline slowly.

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