Double-photoionization studies of Ne, O, and N from threshold to 280 eV

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The photoionization cross sections for double ionization of Ne by single photons have been measured from threshold to 280 eV. The results presented are in considerable variance with other published data. These cross sections are compared with those for N^{2+} and O^{2+}. It is found that the ratio of the oscillator strength for double ionization to the total photoionization oscillator strength for each of these atoms is proportional to $N-1$, where N is the total number of electrons in the atom.

I. INTRODUCTION

Studies of multiple photoionization of atoms by single photons have attracted considerable attention. A prime reason for this is because of the strong electron-correlation effects that are mainly responsible for multiple ionization. This is particularly true for direct multiple ionization, which is the only process that can occur in spectral regions where the incident photons have insufficient energy to release an inner-shell electron and thereby cause double ionization through an Auger process. Ne, O, and N are particularly suitable atoms in which to study this direct multiple-ionization process because their K-shell energies lie between 400 and 867 eV, far from their respective double-ionization thresholds.

Numerous measurements of the double-photoionization cross sections of Ne have been made.[1-5] But there is a wide discrepancy between the various results. Two calculations have been made[6,7] which also vary considerably between each other and the experimental data. The reason for such a large variation between the experimental data is not clear. However, because Ne belongs to the series $1s^22s^22p^x$ and we have already measured the double-ionization cross sections for oxygen and nitrogen,[8,9] both of that series, and because of the experimental disagreement in the value of the Ne^{2+} data, we decided to remeasure the cross sections and compare the results with those for N^{2+} and O^{2+}.

II. EXPERIMENT

The experiment was conducted on the MkII grasshopper grazing-incidence monochromator at the Synchrotron Radiation Center of the University of Wisconsin. The useful energy range covered was from 40 to 280 eV.

Details of the apparatus have been described previously.[8] Briefly, a gas jet of Ne intersected the radiation from the monochromator in a collision region that was maintained at a pressure of about 10^{-6} Torr. The ions were accelerated through about 800 V and were focused into a magnetic mass spectrometer. After mass analysis the ions were further accelerated through about 5000 V onto the first dynode of a Johnston electron multiplier and the count rate of ions/s was recorded. The incident photon beam was monitored with an aluminum photodiode allowing the simultaneous measurement of the ratio of ion counts to photodiode signal to be made. A complete scan of this ratio was made for Ne^+ from 40 to 280 eV. Then Ne^{2+} was selected, the focusing conditions maximized, and the scan repeated. A series of measurements were taken at different gas pressures, varying by about a factor of 6. A pressure effect was observed in the ratio. Thus, after averaging several runs at each pressure the data were extrapolated to zero pressure.

The absolute value of the double-ionization cross section σ^{2+} is given by

$$\sigma^{2+}=[\mathcal{N}^{2+}/(\mathcal{N}^{+}+\mathcal{N}^{2+})]\sigma_t \ , \tag{1}$$

where the term in the square brackets represents the branching ratio for Ne^{2+}, σ_t represents the total photoionization cross section for neon, $\mathcal{N}^+$ and $\mathcal{N}^{2+}$ represent the count rates per unit photon flux for the single- and double-charged neon ions, respectively. Although Ne^{3+} was observed, the amount was about 1% of the Ne^{2+} sign and thus could be neglected in Eq. (1). It is possible that the value of the branching ratio could be affected by a difference in the ion detection efficiency for singly and doubly charged ions. However, Peart and Harrison[10] have shown that when the ion velocities are greater than about 10^5 m/s, the detection efficiency of the Johnson electron multiplier changes very slowly with ion velocity and is essentially independent of the ionic charge. The ion velocities used in this work were greater than 10^5 m/s.

The values of σ_t have been tabulated by Marr and West.[11] However, we have found that their values are systematically 10–18% higher, in this spectral region, than our unpublished data and that of Watson.[12] We have initiated a program to remeasure the cross sections of the rare gases to an accuracy of 1% or 2%.[13] To date our measurements agree with our more extensive unpublished data. Because of this, and the fact that our data agree with Watson's overlapping data, we have used his values to extend our data to about 233 eV and Henke's[14] compilation to cover the remaining spectral region. Thus, in the present paper we use our own compilation to determine σ_t in Eq. (1). The branching ratio σ^{2+}/σ_t is also obtained from Eq. (1).

TABLE I. Total and double-ionization cross sections of neon.

$h\nu$ (eV)	λ (Å)	σ_t (Mb)	σ^{2+} (Mb)	$h\nu$ (eV)	λ (Å)	σ_t (Mb)	σ^{2+} (Mb)
62.528	198.288	6.57	0	130	95.4	2.40	0.270
65.5	190.7	6.34	0.044	140	88.6	2.08	0.247
70	177.1	5.88	0.087	150	82.7	1.80	0.221
75	165.3	5.50	0.118	160	77.5	1.55	0.194
80	155.0	5.12	0.149	170	72.9	1.35	0.169
85	145.9	4.76	0.183	180	68.9	1.20	0.147
90	137.8	4.44	0.219	190	65.3	1.05	0.125
95	130.5	4.15	0.246	200	62.0	0.93	0.106
100	124.0	3.85	0.266	220	56.4	0.75	0.077
105	118.1	3.57	0.277	240	51.7	0.61	0.056
110	112.7	3.30	0.284	260	47.7	0.52	0.043
115	107.8	3.05	0.285	280	44.3	0.44	0.031
120	103.3	2.81	0.283				

III. RESULTS AND DISCUSSION

Our measured cross sections for producing Ne^{2+} are shown in Fig. 1 (solid data points) and are tabulated in Table I. We have included, in Table I, the values of the total cross sections used in this work to convert the branching ratios to cross sections. Also shown in Fig. 1 are the various experimental and theoretical results. In order to make a meaningful comparison with the present data we have adjusted the published experimental data to reflect the same Ne total cross section as used here. As can be seen from Fig. 1 the variation in cross section between the different groups is more than 50%. Different techniques were used in several of the experiments. Wight and Van der Wiel[4] simulated the photoionization process by using the well-established method of measuring the energy loss of small-angle-scattered high-energy electrons in coincidence with the ions produced. Carlson[1] used filtered x rays from a conventional x-ray tube, whereas in the present work and that of Schmidt *et al.*[3] and Holland *et al.*,[5] synchrotron radiation was used as the ionizing source. In all of these studies mass spectrometers were used to identify the individual ions except in our earlier work[2] (open circle data points) where a double-ionization chamber technique was used. This method simply measured the total charge produced, from which we can derive the branching ratio for Ne^{2+} because no Ne^{3+} is produced in this photon energy region. Also a discrete line light source was used, which was free from the problems of scattered light and second-order spectra because these effects were easily identified and corrected. The biggest problem in using synchrotron radiation is the difficulty in correcting data for the presence of scattered light and higher-order spectra. Details of our correction method are given in Ref. 8. The very good agreement between our present results and our earlier ion chamber data gives us confidence in the accuracy of our data, which we estimate to be about ±10%. This error limit does not include the possibility of some unknown systematic error. The calculations by Chang and Poe[6] are in very good overall agreement with the present results whereas the calculations by Carter and Kelly[7] (dipole velocity) tend to be much lower on the rising portion of the curve and too high at higher photon energies. Although no definitive conclusions can be made as to the best values for the double-ionization cross section of Ne^{2+}, we believe that our data may represent an upper limit, whereas those of Schmidt *et al.*[3] may represent a lower limit. The basis for this statement lies in the comparison of the double-photoionization cross sections for Ne, O, and N. Our measurements for these cross sections are shown in Fig. 2 along with the theoretical results of Carter and Kelly[15] for C^{2+}. The sequence formed by these curves suggests that the peak value for the Ne^{2+} cross section is probably greater than about 0.22 Mb. The dashed line, indicating the position of F^{2+}, is purely speculative. However, when we measure the oscillator strengths for the direct double ionization of Ne, O, and N we find that the ratio of the direct double-ionization oscillator strength to the total absorption oscillator strength is proportional to $N-1$, where N is the total number of electrons in a given atom. Because the total absorption oscillator strength is simply equal to N, then

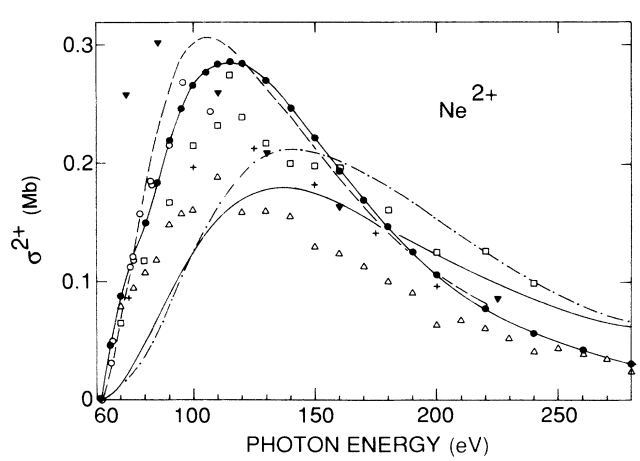

FIG. 1. Double-photoionization cross sections of Ne as a function of photon energy. Experiment: ●, present work; ▼, Ref. 1; ○, Ref. 2; +, Ref. 3; □, Ref. 4; △, Ref. 5; theory: — — —, Ref. 6; —·—·—· and ——, length and velocity approximation, respectively, Ref. 7.

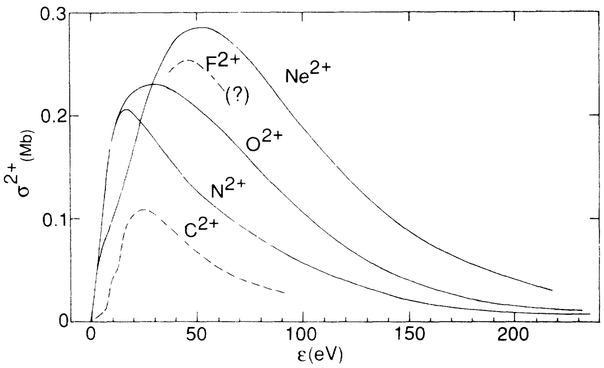

FIG. 2. Comparison of the double-photoionization cross sections for the sequence $1s^2 2s^2 2p^x$. Ne^{2+}, present data; F^{2+}, speculated position; O^{2+}, Ref. 8; N^{2+}, Ref. 9; C^{2+}, Ref. 15.

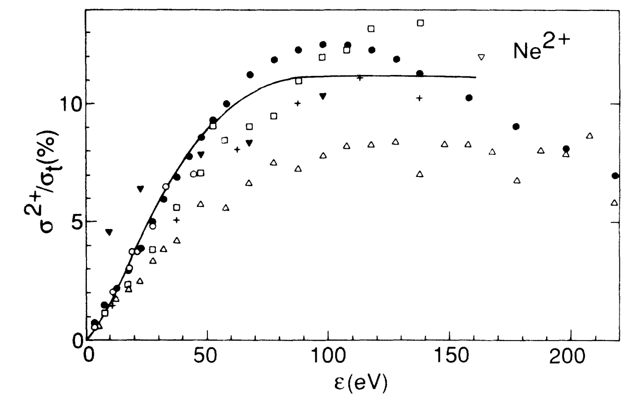

FIG 3. Variation of the Ne^{2+} branching ratio as a function of the total electron energy released. The solid line represents the calculated values by Chang and Poe (Ref. 6). ●, present data; ▼, Ref. 1; ○, Ref. 2; +, Ref. 3; □, Ref. 4; △, Ref. 5.

the double-ionization oscillator strengths are proportional to $N(N-1)$.

We obtained the direct double-ionization oscillator strength f^{2+} from the relation

$$f^{2+} \propto \int_0^{\lambda_0} (\sigma^{2+}/\lambda^2) d\lambda ,$$

where the limits of integration are from the double-ionization threshold wavelength λ_0 to zero wavelength. We made a linear extrapolation of our data from 44 Å (280 eV) to 0 Å, ignoring the production of double ionization caused by Auger processes. The results are given in Table II. The average value of the constant of proportionality between the oscillator strengths and $N(N-1)$ was 0.0036. If the relationship $f^{2+} \propto N(N-1)$ holds for C and F, we can predict that $f^{2+}(C)=0.11$ and $f^{2+}(F)=0.26$. Unfortunately, the energy range of the theoretical data for C^{2+} is insufficient to determine its oscillator strength.

The ratio of double to single photoionization is expected to reach an asymptotic value at high photon energies.[6,16] Unfortunately, these energies are not well defined. In their study of He, Amusia *et al.*[16] show that the ratio, after reaching a maximum value, decreases with increasing photon energy reaching a value about 30% above the asymptotic ratio at 700 eV. Whereas in Chang and Poe's calculations on Ne^{2+} the ratio reached a plateau at about 140 eV. However, their calculation did not continue above 220 eV. In Fig. 3 we show the variation of the branching ratio for Ne^{2+} as a function of ε (eV), the total electron energy released. The branching ratio for double ionization is simply the ratio σ^{2+}/σ_t where σ_t is the total photoionization cross section. Included in the figure are the various experimental data and the theoretical curve of Chang and Poe (solid line). The photon energy scale can be determined by adding the threshold energy of 62.5 eV to ε. The results of Holland *et al.*[5] are the only experimental data to show a plateau similar to the theoretical data. However, the magnitude of the plateau is about 27% lower than the calculated value. The data of Wight and Van der Wiel actually continue to increase with photon energy (not shown in Fig. 3) to their limit at ε = 180 eV. The present data agree very well with the calculated data both in magnitude and shape up to 120 eV, but then rapidly decreases. The data of Schmidt *et al.*[3] also start to decrease at about the same energy. This decrease from a maximum value may continue to some asymptotic limit not yet observed. Obviously, more

TABLE II. Relationship between the double-ionization oscillator strengths and the product $N(N-1)$.

Ion	f^{2+}	$f^{2+}/N(N-1)$
Ne^{2+}	0.330	0.003 67
O^{2-}	0.210	0.003 75
N^{2+}	0.144	0.003 43
		average 0.003 62

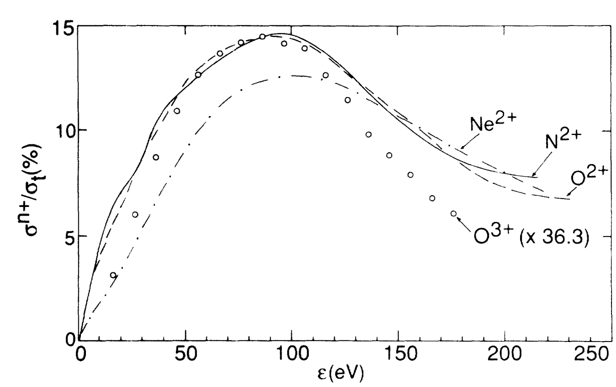

FIG. 4. Comparison of the branching ratios for double photoionization of Ne, O, and N as a function of the total electron energy released. Also included are the branching ratios for O^{3+} normalized to the maximum of the O^{2+} curve.

detailed studies of Ne^{2+} are required to clarify this point and the present discrepancies.

When we plot the branching ratios for Ne, O, and N together we note a great similarity in the curves. These results are shown in Fig. 4. The branching ratios for double ionization are all absolute. However, the ratio for O^{3+} was normalized to the O^{2+} data at 86 eV to illustrate the similarity in shape between all the curves. Analyses of these results are underway.

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