Coulomb field causes an increase in the cross sections of the ionization and of the dissociation.

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Electron Shake-Off in Neon and Argon as a Function of Energy of the Impact Electron*

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Abundances of satellite lines relative to diagram lines in the *K*-Auger spectrum of neon and *L*-Auger spectrum of argon are measured as a function of the energy of impact electrons. The ratios of satellite to diagram lines are found to be independent of the energy except near the threshold for initial ionization, where the ratios decrease sharply. These ratios are taken to be evidence of the extent of initial double-to-single ionization, i.e., the *KL* to *K* ionization in neon and *LM*-to-*L* ionization in argon; and the results are discussed in terms of electron shake-off and the sudden approximation.

INTRODUCTION

With the help of high-resolution electron spectroscopy, satellite lines have been observed and identified in the Auger spectra of the rare gases.[1] These extra lines arise when Auger transitions take place in atoms having a doubly-ionized configuration. For example, when neon is bombarded with electrons whose energies are the order of several kilovolts, *K* electrons can be ejected with a subsequent readjustment to the *K* vacancy by a *K-LL* Auger process. In addition, there also occurs the possibility of a double electron ejection involving a vacancy in both *K* and *L* shells. The subsequent *KL-LLL* Auger process produces satellite lines that are slightly altered in energy from the normal lines formed from the singly ionized species. These satellite lines offer an excellent opportunity to study the probability for initial double ionization. In this paper we present results on double ionization in rare gases as a function of energy of the bombarding electron, based on our observations of the Auger satellite lines. Specifically, we have examined the probability for simultaneous *KL* ionization in neon and *LM* ionization in argon.

It has been suggested[2,3] that the mechanism for double ionization leading to the observed Auger satellite lines is due to electron shake-off. A similar conclusion has also been reached in accounting for some of the x-ray satellites.[4] If the process for ejecting the inner-shell electron is rapid enough (that is, if the velocity of both the impact electron and the ejected inner-shell electron $\gg$ the orbital velocity of the outer-shell electron) then the sudden approximation is valid, and the probability for electron shake-off of an outer-shell electron can be computed solely from the initial- and final-state wave functions without knowledge of the specific mechanism for forming the initial inner-shell vacancy. Since the probability for electron shake-off is independent of the energy of the impact electron in the region where the sudden approximation is valid, but is strongly dependent on the energy where the sudden approximation fails, a study of the ratio of double to single ionization as a function of the energy of the bombarding electron should be of great help in testing the implications of the sudden-approximation model.

EXPERIMENTAL PROCEDURES AND RESULTS

Our experiment in brief was to produce inner-shell vacancies in neon and argon, by bombarding the gases with electrons from an electron gun. The subsequent Auger electrons formed from the atomic readjustment were allowed to pass through the entrance slit into a newly constructed electron spectrometer,[5] employing double focusing electrostatic plates and built for measuring electrons with an energy resolution of better than 0.1% full width at half-maximum (FWHM). We have confined our attention in neon to the most intense satellite lines arising from the Auger transition $KL_{II,III}$-$L_{II,III}L_{II,III}L_{II,III}(^2D)$, where we have indicated the initial and final states by the configuration of the vacant orbitals and the term value of the final state using the L-S coupling scheme. We have compared this satellite line to one of the diagram Auger lines representing single ionization in the initial state, viz., K-$L_IL_{II,III}(^3P)$ and, in some cases, K-$L_{II,III}L_{II,III}(^1D)$. In our study on argon we compared the strong satellite lines at 194.4 and 196.3 eV to the L_{III} -$M_{II,III}M_{II,III}(^1S)$ diagram line. The identification of the satellite and diagram Auger lines in neon and argon has been made by Mehlhorn from experimental binding energies and optical data.[1] Measurements were made with impact electrons of energies from 925 eV to 7.2 keV in the case of Ne and from 300 eV to 3.0 keV in the case of Ar. The bombarding electrons have an energy spread of about 1 eV. The gas pressure in the target chamber was normally at about 10 μ and in the spectrometer 1.5×10^{-5} Torr. Some data were taken at various pressures and the ratio of satellite to diagram Auger lines was found independent of pressure. In Fig. 1 are given examples of two runs on neon and argon. The correct shape and magnitude of the background was checked by making a sweep over wider portions of the spectrum. In Figs. 2(a) and 2(b) are given the ratios of the satellite line to the diagram line which may be interpreted in the case of neon to represent the probability for simultaneous ionization in the K and $L_{II,III}$ shells relative to single K ionization; and in the case of argon to represent the probability for ionization in both the $L_{II,III}$ and $M_{II,III}$ shells relative to single L_{III} ionization. The error bars include uncertainties in the background correction and small fluctuations in the linewidth as well as counting statistics. We see from Fig. 2 that the probability for double ionization relative to single ionization is indeed independent of the energy of the impact electron at high energies. However, for neon at lower energy the probability rises slightly and then drops suddenly at about 1500 eV. The data on argon show the same general behavior except that the rise is less noticeable and the drop-off occurs at about 600 eV.

The energy scale is also plotted in terms of

$$f(E)=E_e/(E_o+E_i)\ , \qquad (1)$$

and also $f(E)=(E_e-E_i)/E_o\ ,\qquad (2)$

where E_e is the energy of the impact electron, E_i the binding energy of the inner-shell electron, and E_o the energy necessary to remove the outer electron in which an inner-shell vacancy has been formed. Equation (1) gives the ratio of impact energy to energy necessary for double ionization, a commonly used unit. Equation (2) gives the ratio of energy available for electron shake-off to the energy required for electron shake-off, a unit better suited for evaluating the breakdown in the sudden approximation. In Fig. 2(c) are also plotted data of Paratt[6] on the intensity of the $K\alpha_{3,4}$ line in titanium relative to the $K\alpha_1$ line, as a function of the electron impact energy. The $K\alpha_{3,4}$ satellite lines have been interpreted[7] as resulting from transitions in an atom having both K and L vacancies, while the $K\alpha_1$ results from the filling of a single K hole. All three studies show that at higher energies the relative amount of multiple ionization is independent of the impact electron, consistent with the sudden-approximation theory, and that the sudden approximation breaks down when the ratio in Eq. (2) is smaller than approximately 10. The slight rise in the curve for neon as one goes to lower impact energies, and less so with argon, may be due to electron correlation which has been studied by Åberg[8] in the case of He or to a small admixture of direct collision[9] which would be inversely dependent on the velocity of the impact and ejected electrons. Both these factors would appear to be smaller in the titanium experiment.

In Fig. 2(d) is plotted a theoretical calculation for simultaneous K and L vacancies in photoionization, based on noncorrelated wave functions,[10] together with experimental data on photoionization in neon.[11] Double ionization resulting from the photoelectric effect has the same general behavior

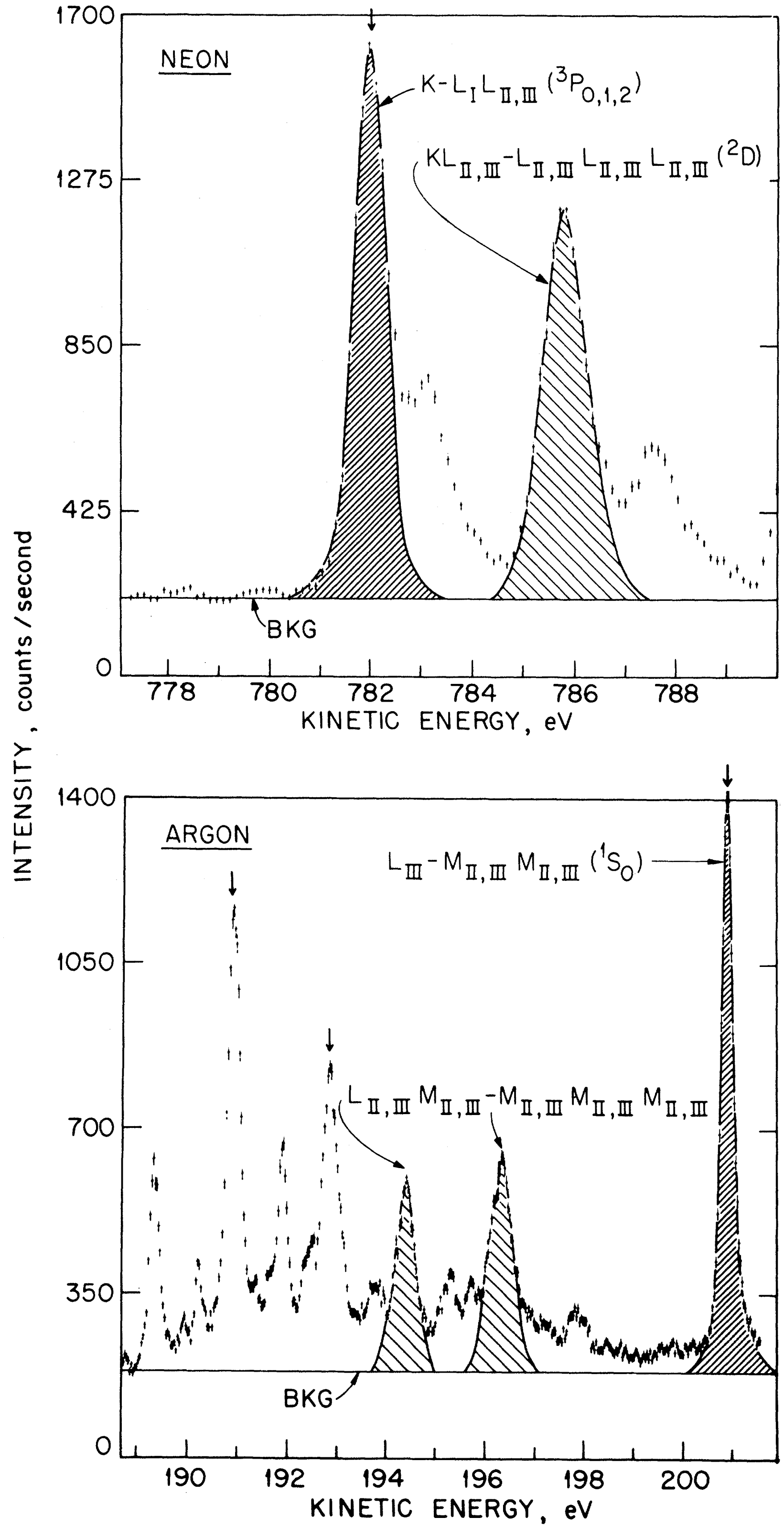

FIG. 1. Portions of the neon *K-LL* and argon *L-MM* Auger spectra. The position of diagram lines indicating initial single ionization is shown by arrows. The remaining structure is due to satellite lines arising from initial doubly ionized atoms. The peaks primarily used in the analysis are labeled and cross-hatched. The electron impact energy in these examples was 4000 eV for the neon and 1500 eV for the argon spectrum.

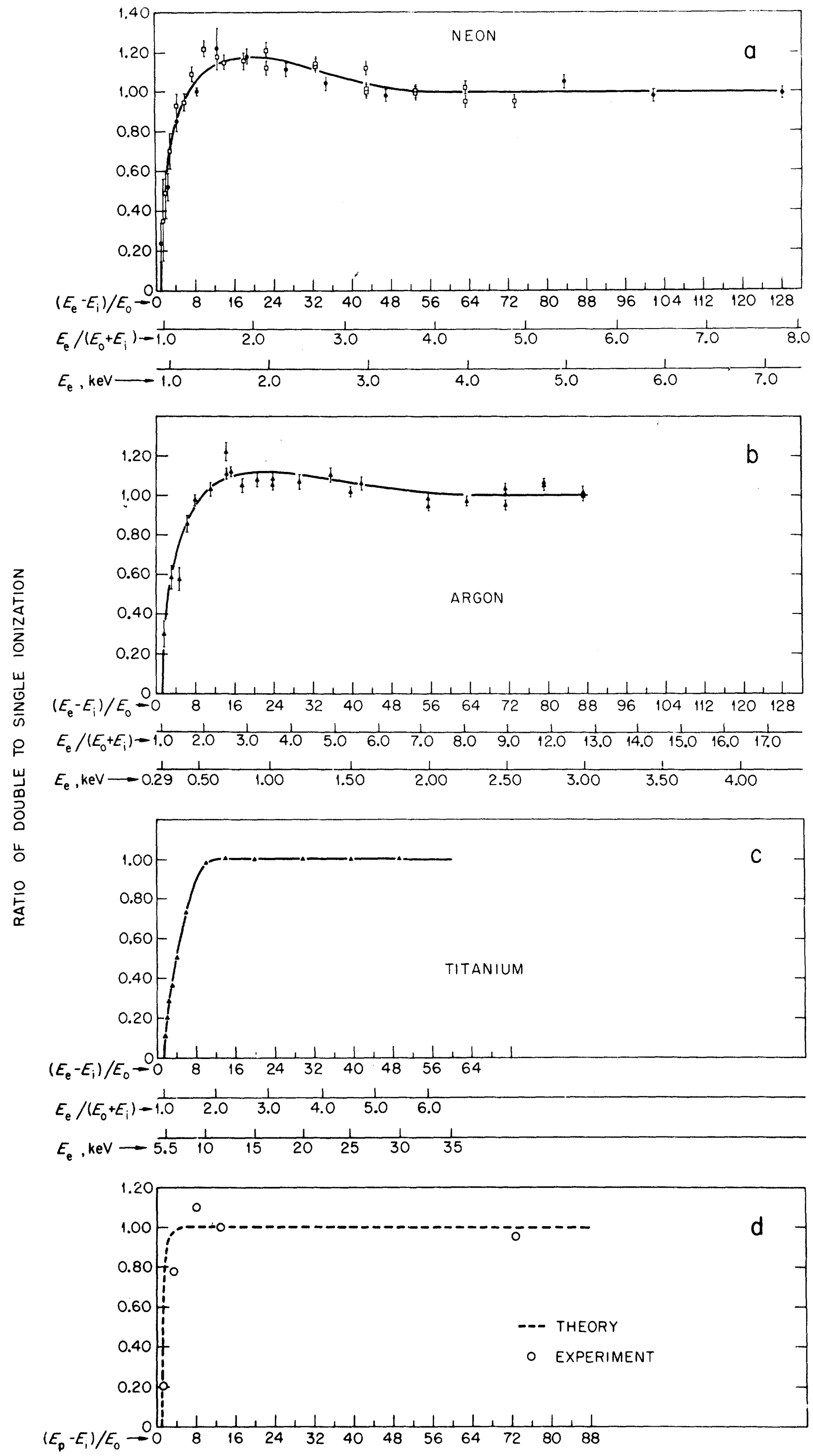

FIG. 2. Ratio of double to single ionization as a function of the energy of the impact electron. E_e is the energy of the impact electron, E_i is the binding energy of the inner-shell electron, E_o is the binding energy of the outer-shell electron where an inner-shell vacancy has been formed. E_i and E_o have been estimated from experimental atomic binding energies and eigenvalues from Hartree-Fock wave functions. (a) Data have been taken from the Auger spectrum of neon (cf. Fig. 1), with double ionization being referred to the $KL_{II,III}$-$L_{II,III}L_{II,III}L_{II,III}$ (^2D) transition, and single to the K-$L_{II,III}L_{II,III}$ (^1D) transition (solid circles) with the ratios normalized to 0.098, or to the K-$L_IL_{II,III}$ (^3P) transition (squares) with the ratios normalized to 0.87. (b) Data have been taken from the Auger spectra of argon (cf. Fig. 1) with double ionization referred to the $L_{II,III}M_{II,III}$-$M_{II,III}M_{II,III}M_{II,III}$ transitions and single ionization to the L_{III}-$M_{II,III}M_{II,III}$ (^1S) line, the ratios being normalized to 1.17. (c) Data have been taken from x-ray spectra of Ti where double ionization is taken from the $K\alpha_{3,4}$ satellite lines and single ionization from the $K\alpha_{1,2}$ lines, the ratios being normalized to 0.022, cf. Ref. 6. (d) Data related to the phenomenon where the initial ionization arises from the photoelectric effect. Theory has been determined for K, L photoionization in Al, the ratios being normalized to 0.013, cf. Ref. 10. Experimental data have been taken from the K, L photoionization of neon, the ratios being normalized to 0.188, cf. Ref. 11. E_p refers to the energy of the photon.

as electron impact except that the breakdown in the sudden approximation appears to occur at an energy about a factor of 2 lower. As pointed out previously, and as will be confirmed in detail in a subsequent publication,[3] simultaneous ionization in the outer shells as the result of forming an inner-shell vacancy is essentially identical for photoionization and for electron impact if $f(E)$ in Eq. (2)

>10, where E_e here can stand either for the initial energy of the photon or of the impact electron. Bombardment with ions, however, produces simultaneous ionization in the inner and outer shells that is entirely in variance with photoionization and electron impact.[12]

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Excitation of O_2^+ First Negative Bands by Electron Impact on O_2 †

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Absolute cross sections for the electron-impact excitation of the O_2^+ first negative bands were measured from threshold to 400 eV and extrapolated to 1000 eV by means of a Bethe-Oppenheimer relationship. The peak cross sections for the $(n+1, n)$ and $(n+2, n)$ sequences were found to be 5.30×10^{-18} cm$^2\pm10\%$ and 2.10×10^{-18} cm$^2\pm10\%$ at 100 eV, respectively. The cross sections for the (0, 0) and (1, 0) bands had a maximum value of 2.14×10^{-18} cm$^2\pm15\%$ and 4.33×10^{-18} cm$^2\pm15\%$, respectively. The ratio of the total ionization cross section of O_2 to the excitation cross sections of the first negative bands was nearly constant over the energy range 30–1000 eV; it had an average value of 64 for the (1, 0) band. The lifetime of the $v'=1$ level of the $b^4\Sigma_g^-$ state was found to be 1.19 μsec $\pm$ 5%.

I. INTRODUCTION

The first negative bands of O_2^+ appear prominently in the visible spectrum of an aurora,[1–3] where this system is excited principally by electron impact. So far few laboratory studies have been performed on the excitation of the first negative system in the single-collision domain.[4–7] Absolute excitation cross sections can be used together with rocket measurements to determine the primary O_2^+ ionization rate and the total electron flux in auroras.[8,9] We have therefore attempted to measure accurately (10–15%) the absolute cross sections for the most prominent first negative bands. Since most secondary electrons in an aurora have low energies, we have worked in the energy range from threshold to 1000 eV. The existing studies of the first negative system