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Effect of Multiple Ionization on the Fluorescence Yield of Ne \dagger

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K Auger-electron and *K* x-ray yields of Ne have been measured simultaneously in 30-MeV O $^{7+}$ collisions with Ne. Both spectra are dominated by satellite transitions. The Ne *K*-shell fluorescence yield is 2.4 times larger than found in identical measurements using 5-MeV incident protons.

Atomic inner-shell vacancy production in heavy-ion collisions has been a subject of increasing interest in the past several years.^{1,2} Collisions involving heavy ions with energies of the order of MeV/amu have been previously studied exclusively via x-ray measurements.³ A common feature of all measurements in this energy range is that the x rays are found to be shifted to higher energies.⁴ The shifts are the result of the multiple ionization created in the heavy-ion collisions. The spectra are dominated by satellite transitions,⁵ each having a slightly different energy; when observed in low resolution, the spectra appear as broad, shifted lines.

This observation has created considerable speculation^{1,6} as to what extent the effective fluorescence yields in heavy-ion collisions are different from those characteristic of single vacancies.⁷ Fluorescence yields are needed to compare experimental x-ray production cross sections with theoretical cross sections for inner-shell ionization and, together with x-ray and Auger-electron energy shifts, make possible a precise specification of the ion states produced in heavy-ion collisions. In this Letter we report a simultaneous measurement of the Ne *K* x-ray and Auger-electron spectra in 30-MeV O $^{7+}$ collisions⁸ with Ne gas. By comparing the fluorescence yield for this collision with identical measurements using 5-MeV incident protons, it is shown that the ef-

fective fluorescence yield can increase substantially with the degree of ionization.

The incident beam was produced in the University of Washington's model FN tandem Van de Graaff accelerator. The beam was collimated by two 3-mm apertures, 30 cm apart, at the entrance of a differentially pumped gas scattering chamber and was collected in a 120-cm Faraday cup. Typical proton and oxygen beam intensities were 1 μ A. The target gas pressures were typically 10 $^{-2}$ Torr measured with a LN trapped McLeod gauge and were accurate to $\pm 10\%$. Separate diffusion pumps maintained the beam line and electron analyzer chamber at 5 $\times 10^{-6}$ Torr. The base pressure of the system was 2 $\times 10^{-7}$ Torr.

The x rays were detected at 90 $^\circ$ to the beam direction with a gas-flow proportional counter using P-10 gas at 1 atm. The effective solid angle of the detector was 0.28 cm sr with an energy resolution of 40% full width at half-maximum (FWHM) at 850 eV. The x-ray detector entrance window was 6 μ m Mylar coated on the detector side with 1600 \AA of Al. The detector was shielded from the first beam collimator and Faraday cup by 15 cm of lead. A typical x-ray spectrum is shown in Fig. 1. The single-*K*-vacancy, 2*p*-1*s* x-ray energy in Ne is 850 eV.

The efficiency of the proportional counter is assumed to be identically the transmission of the entrance foil. For 5-MeV proton excitation, the

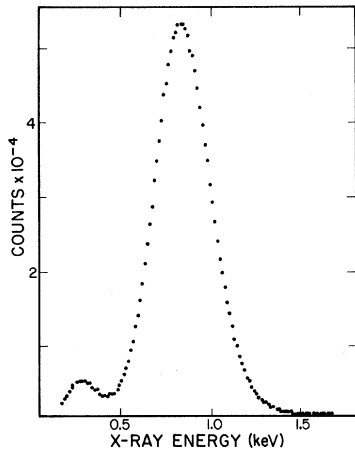


FIG. 1. Ne K x-ray spectrum accumulated during an Auger-electron energy scan. The spectra appear identical for proton or oxygen-ion excitation. The ~ 50 -eV shift discussed in the text is not discernible because of the poor resolution. The lower-energy peak is due to C K x ray (280 eV) produced by secondary excitation of the colloidal-graphite coating of the scattering-chamber wall.

foil transmission for the Ne K x rays was measured to be $(1.63 \pm 0.11)\%$. This value is consistent with the theoretical value for 850-eV photons determined from a mass weighted, \ln - \ln interpolation of the experimental absorption coefficient data⁹ for C, H, O, and Al. For the O^{7+} excitation, the measured Ne K x-ray transmission was $(3.36 \pm 0.23)\%$ for identical foils and geometry. These measurements were each made for a fixed number of accumulated Auger electrons. From the energy dependence of the absorption coeffi-

cients, these results imply that the Ne K x rays in the O^{7+} collisions have a mean energy of 50 ± 15 eV higher than in the proton collisions.

Electrons emitted at 90° to the beam direction were energy analyzed with a cylindrical-mirror electrostatic analyzer and detected with a channel electron multiplier. Full details of the analyzer have been presented elsewhere.¹⁰ The effective solid angle of the analyzer was 3.89×10^{-4} cm sr with an energy resolution of 1.4% FWHM. An electron gun was used to determine the energy calibration of the analyzer. Magnetic fields in the region of the target gas and analyzer were reduced to ≤ 15 mG with three pairs of mutually perpendicular Helmholtz coils, each 1.5 m in diameter.

Electron spectra from the proton and oxygen collisions are shown in Fig. 2. Each datum point represents the accumulation of a fixed number of Ne K x rays. The proton-excited spectrum has three noticeable features: the highest peak at 805 eV corresponding to $K-L_{23}L_{23}$, a small peak at 770 eV, and a shoulder at ~ 750 eV corresponding to $K-L_1L_{23}$ and $K-L_1L_1$, respectively. These identifications are based on earlier, high-resolution studies^{11,12} of the Ne K - LL spectrum. References 11 and 12 include studies of the Ne Auger satellite spectra following electron and low-energy proton bombardment. The satellites in these cases involve, at most, one additional L vacancy.

The oxygen-excited spectrum is quite different, consisting of a broad peak centered at 735 eV. Transitions with intensities above the background occur at energies as low as 650 eV and as high

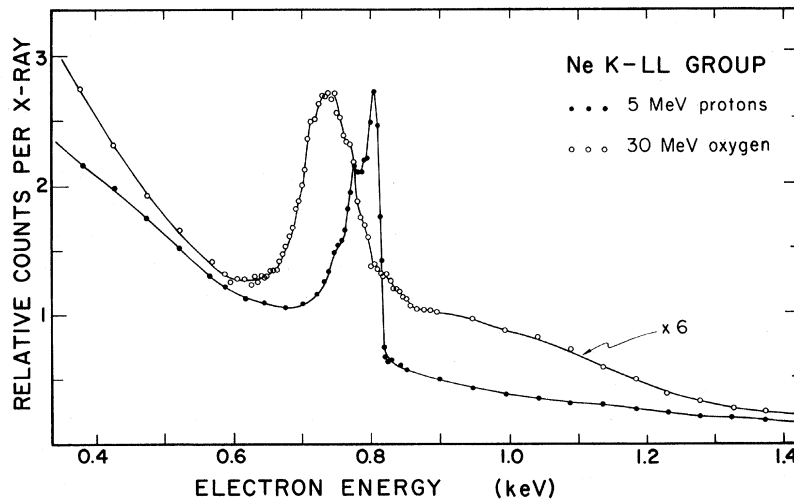


FIG. 2. Relative electron yields per Ne K x ray in proton and oxygen collisions with Ne gas observed at 90° . The energy resolution is 1.4% FWHM.

as 850 eV. Though individual lines are not resolved, we can definitely conclude that this spectrum is dominated by satellite transitions. This spectrum is similar to the results of Edwards and Rudd¹² for 200-keV Ne⁺+Ne measured at 160°. The oxygen-excited group is centered ~15 eV lower in energy and is broader—we note that the expected “Doppler” broadening effects³ are different in the two measurements. For 30-MeV O⁷⁺+Ne, observed at 90°, we estimate a line broadening of ~2 eV; whereas in the 200-keV Ne⁺+Ne at 160°, there is an expected line broadening of ~11 eV.

Ne Auger and x-ray energies as a function of charge state have been calculated by Larkins.¹³ His results, combined with a simple statistical weighting procedure¹⁴ to estimate relative intensities, would imply that the dominant Auger transitions we observe involve three or four additional vacancies. The inferred x-ray energy shift, however, would imply five or six. These conclusions do not take into account the measured fluorescence yield.

The mean fluorescence yield for proton excitation, $\bar{\omega}_p$, can be written

$$\bar{\omega}_p = \sigma_p^x / (\sigma_p^x + \sigma_p^e) \approx \sigma_p^x / \sigma_p^e.$$

σ_p^x and σ_p^e are the x-ray and Auger-electron production cross sections for proton excitation. The approximate equality is accurate to within ($\bar{\omega}_p \times 100$)%. The single-vacancy, K-shell fluorescence yield of Ne is ~0.018.⁷ Using the subscript “O” to denote oxygen-ion excitation, we can write

$$\frac{\bar{\omega}_O}{\bar{\omega}_p} = \frac{\sigma_O^x \sigma_p^e}{\sigma_p^x \sigma_O^e} = \frac{R_p(e/x) \epsilon_p(x)}{R_O(e/x) \epsilon_O(x)},$$

where $R(e/x)$ is the observed ratio of K Auger electrons to K x rays and $\epsilon(x)$ is the x-ray detector efficiency. The last equality assumes the geometries of both detector systems and the target gas pressure remain constant for both measurements. The latter condition provides for the cancelation of electron scattering losses in the target gas. $R(e/x)$ was determined by integrating the number of counts under the Auger peak weighted by (electron energy)⁻¹, the transmission function of the analyzer. We find that $R_p(e/x)/R_O(e/x) = 5.1 \pm 0.9$ and, as discussed above, $\epsilon_O(x)/\epsilon_p(x) = 2.1 \pm 0.2$ which yields $\bar{\omega}(30\text{-MeV O}^{7+}) = (2.4 \pm 0.5)\bar{\omega}(5\text{-MeV } p)$. Fluorescence yields for defect configurations can be estimated using the statistical scaling procedure¹⁴ mentioned above. This procedure predicts that no degree of $2p$ ionization alone can account for the observed increase

in $\bar{\omega}$. We find, however, that the initial configurations $(1s)^{-1}(2s)^{-n}(2p)^{-m}$ yield the following increases for (n,m) : 2.1 for (1,3), 2.8 for (1,4), 2.3 for (2,2), and 3.4 for (2,3).

Using Hartree-Fock-Slater wave functions, Bhalla¹⁵ has calculated the fluorescence yields for all defect configurations in Ne. From his results we find that our data on x-ray and electron-energy shifts and fluorescence yields are simultaneously consistent only with the configurations $m=4$ or 5 if $n=1$ and $m=2$ or 3 if $n=2$. His calculated increases are then 1.9, 2.7, 2.0, and 2.8, respectively.

This comparison assumes that the 5-MeV p excitation results in predominantly single-K-vacancy states. This assumption is supported by the proton-induced multiple-ionization studies of Stolterfoht^{1,16} (Auger electrons) and of Knudson, Burkhalter, and Nagel¹⁶ (x rays). It was found that multiple ionization decreases as the p velocity increases above the L-shell electron velocity. Cross sections for electron production in gases by protons up to 2 MeV have also been studied in detail by Toburen.^{1,17}

An accurate calculation of the multiple-defect spectrum is difficult. The relative intensities are sensitive to the coupling scheme and exchange approximation used and to the relative production probabilities for the various terms of the initial configurations. Calculations of the Ne KL-LLL spectrum have been carried out by Schmidt.¹⁸ Of the 21 resulting transitions, ~10 share the bulk of the intensity, and these lines are spread over a broad energy range. Indeed, a major conclusion of the present work is that the Auger groups from highly ionized target atoms can, in some cases, be observed above the continuous backgrounds in high-energy heavy-ion collisions.

High degrees of multiple ionization are not unique to high-energy collisions. Inelastic energy-loss studies of Kessel, McCaughey, and Everhart¹⁹ have found that in 200-keV Ne⁺+Ne close collisions can result in charge states of up to 4+. In the present measurements, by looking at K-shell transitions, we also restrict ourselves to impact parameters which are small compared to the L shell.

We have made similar measurements of relative fluorescence yields for the same projectiles and energies on Ar. Preliminary results show that for the oxygen collisions $\bar{\omega}_K$ increases by about 30% and $\bar{\omega}_L$ increases by about 10³!

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¹Current reviews of this field will appear in Inner Shell Ionization Phenomena and Future Applications, Proceedings of the Atlanta Conference, edited by R. W. Fink, S. T. Manson, J. M. Palms, and P. V. Rao (U. S. Atomic Energy Commission, Oak Ridge, to be published), available from the National Technical Information Service, Springfield, Va. 22151.

²F. W. Saris, in *Proceedings of the Seventh International Conference on the Physics of Electronic and Atomic Collisions, Invited Talks and Progress Reports*, edited by T. R. Govers and F. J. de Heer (North-Holland, Amsterdam, 1972); M. E. Rudd, *ibid.*

³Electron emission in low-energy ion-atom collisions has been reviewed by G. N. Ogurtsov, *Rev. Mod. Phys.* **44**, 1 (1972); see also M. E. Rudd, Ref. 2, and M. E. Rudd and J. H. Macek, in *Case Studies in Atomic Collisions*, edited by E. W. McDaniel and M. R. C. McDowell (North-Holland, Amsterdam, to be published).

⁴See references contained in Ref. 2; H. D. Betz and H. W. Schnopper *et al.*, Ref. 1, p. 1348 and p. 1374; M. J. Saltmarsh, A. van der Woude, and C. A. Ludemann, *Phys. Rev. Lett.* **29**, 329 (1972).

⁵A satellite is a transition which takes place in the presence of additional vacancies or excitation—the initial state is other than the ground state of an atom singly ionized in an inner shell.

⁶F. W. Saris and D. Onderdelinden, *Physica (Utrecht)* **49**, 441 (1970); F. P. Larkins, *J. Phys. B: Proc. Phys. Soc., London* **4**, L29 (1971); R. J. Fortner *et al.*, *J. Phys. B: Proc. Phys. Soc., London* **5**, L73 (1972); L. H. Toburen and F. P. Larkins, *Phys. Rev. A* **6**, 2035 (1972); R. K. Cacak, Q. C. Kessel, and M. E. Rudd, *Phys. Rev. A* **2**, 1327 (1970).

⁷For a thorough review of the subject of fluorescence yields, see W. Bambynek, B. Crasemann, R. W. Fink, H.-U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. V. Rao, *Rev. Mod. Phys.* **44**, 716 (1972).

⁸The symbol "O⁷⁺" is herein used to denote an oxygen beam in charge-state equilibrium at a mean charge state of ~7+.

⁹B. L. Henke *et al.*, *Norelco Rep.* **14**, 112 (1967).

¹⁰J. S. Risley, *Rev. Sci. Instrum.* **43**, 95 (1972).

¹¹H. Körber and W. Mehlhorn, *Z. Phys.* **191**, 217 (1966); W. Mehlhorn and D. Stalherm, *ibid.* **217**, 294 (1968); M. O. Krause *et al.*, *Phys. Lett.* **31A**, 81 (1970); T. A. Carlson, W. E. Moddeman, and M. O. Krause, *Phys. Rev. A* **1**, 1406 (1970), and *J. Phys. (Paris)* **32**, C4-139 (1971).

¹²A. K. Edwards and M. E. Rudd, *Phys. Rev.* **170**, 140 (1968); A. K. Edwards, Ph.D. thesis, University of Nebraska, Lincoln, 1967 (unpublished).

¹³F. P. Larkins, *J. Phys. B: Proc. Phys. Soc., London* **4**, 14 (1970).

¹⁴E. J. McGuire, *Phys. Rev.* **185**, 1 (1969); F. P. Larkins, Ref. 6.

¹⁵C. P. Bhalla, private communication.

¹⁶N. Stolterfoht, *Z. Phys.* **248**, 92 (1971), and to be published; A. R. Knudson, P. G. Burkhalter, and D. J. Nagel, Ref. 1, p. 1675.

¹⁷L. H. Toburen, *Phys. Rev. A* **3**, 216 (1971), and **5**, 2482 (1972).

¹⁸V. Schmidt, Ref. 1, p. 548.

¹⁹Q. C. Kessel, M. P. McCaughey, and E. Everhart, *Phys. Rev.* **153**, 57 (1967).

Diffusion-Limited Destruction of Metastable States in He II[†]

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The variation with temperature of the destruction rate of metastable states in He II is accounted for in terms of their roton-limited diffusion. Rough estimates of the reaction distance at which collision-induced destruction occurs are consistent with a "bubble" model for the metastable systems.

Recently Keto, Stockton, and Fitzsimmons¹ reported measurements of the destruction rate of $a^3\Sigma_u^+$ He molecules produced by 160-keV electron bombardment of He II. The metastable concentration M was observed following electron beam cutoff, and its decay could be accounted for in terms of $a^3\Sigma_u^+ - a^3\Sigma_u^+$ encounters, assum-

ing destruction to predominate over diffusion, such that

$$dM/dt = -\alpha(T)M^2. \quad (1)$$

The experimental values of $\alpha(T)$ are shown in Fig. 1. While offering no general interpretation of its behavior, Keto, Stockton, and Fitzsim-