

## INELASTIC ENERGY LOSS AND X-RAY EMISSION IN LARGE-ANGLE ATOMIC COLLISIONS\*

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Unusually large inelastic energy losses have been measured for single collisions of argon ions with argon atoms and of iodine ions with xenon atoms using ion energies from 0.5 to 6 MeV and scattering angles from 4 to 20 deg. These losses, as high as 28 keV, are attributed in part to the production of inner-shell vacancies. The measurement of the energies of x rays from these collisions support this hypothesis.

When heavy ions collide at energies greater than a few keV, their outer shells of electrons interpenetrate. During such a process, electrons in these shells may become excited, thus transferring some of the collision's kinetic energy to potential energy. A measurement of this inelastic energy loss  $Q$  is thus a measurement of the excitation energy transferred to the two atoms by the collision. Nearly all experiments concerning the collisions of heavy ions with heavy atoms investigate only the excitation of the one or two outermost electron shells, and these collisions usually result in inelastic losses of less than 2 keV. The purpose of the present experiment was to study the effects which arise when the innermost electron shells of heavy ions interpenetrate. Projectiles having energies up to 6 MeV were used and the study was limited to those collisions in which the incident ions were deflected through large (4-20 deg) angles. It was found, for example, that 1.5-MeV  $\text{Ar}^+$ -Ar collisions, which force the argon  $K$  shells to interpenetrate, produce a sudden increase in  $Q$ . This increase can be attributed to the collisional production of a  $K$ -shell vacancy. For the  $\text{I}^{n+}$ -Xe collisions, some  $Q$  values of nearly 30 keV were observed, and again, much of this loss must be due to the production of  $L$ - and  $M$ -shell vacancies during the collision.

A 3-MV tandem Van de Graaff accelerator provided  $\text{Ar}^+$  ions with energies from 0.5 to 1.5 MeV,  $\text{I}^{2+}$  ions at 3 MeV, and  $\text{I}^{3+}$  ions at 6 MeV. The coincidence method<sup>1</sup> was used to determine  $\bar{Q}$ , the average inelastic energy loss, for single large-angle collisions of these ions with target atoms. These data were taken using several combinations of incident ion energies and projectile scattering angles. Noncoincident measurements of the energies of the x-rays from these same collision combinations were made using a propor-

tional counter sensitive to x-rays having energies from 1 to 15 keV.

The distance of closest approach<sup>2</sup> between the two nuclei is a convenient parameter against which to plot  $\bar{Q}$ , and Fig. 1(a) shows such a plot for the  $\text{Ar}^+$ -Ar collisions. Contours are shown for several incident ion energies, with the larger distances of closest approach for each contour corresponding to the less violent, small-angle collisions. Figure 1(a) shows the lower energy data of Kessel and Everhart.<sup>1</sup> In the latter work, the data for 25-keV collisions display a discontinuity at about 0.24 Å where  $\bar{Q}$  is multiple valued. This distance of closest approach corresponds to an overlapping of the  $L$  shells of the two atoms, and Fano and Lichten<sup>3</sup> were the first to attribute this discontinuity to the formation of  $L$ -shell vacancies during the collision. Rudd and co-workers substantiated this idea by detecting the resulting Auger electrons.<sup>4</sup> The increase in  $\bar{Q}$  at about 0.10 Å is probably due to the ejection of additional Auger electrons from the  $M$  shells. For distances of closest approach between 0.10 and 0.02 Å there is only a gradual increase of  $\bar{Q}$ . The highest energy data of Pivovar *et al.*<sup>5</sup> extend into this region and indicate an average charge state of about 8 for the scattered argon ions for distances of closest approach from 0.1 to 0.03 Å. The present data show that between 0.02 and 0.01 Å, however,  $\bar{Q}$  increases by about 3000 eV. The approximate radius of the  $K$  shell is 0.03 Å, and thus this rise in  $\bar{Q}$  is probably due to the production of a  $K$ -shell vacancy in one of the argon atoms. The fluorescence yield for such a vacancy<sup>6</sup> is about 0.1 and therefore these vacancies will usually be filled by Auger transitions. For  $K$ -LL Auger decay of this vacancy, the resulting electrons would have energies of about 2.7 keV and should be easily observable. One would also ex-

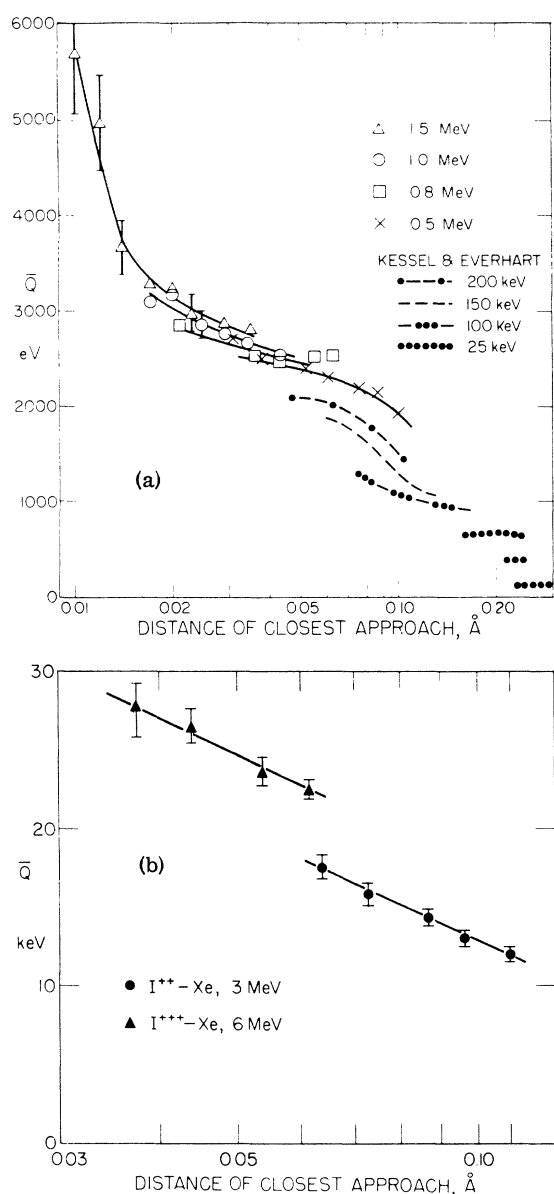


FIG. 1. The average inelastic energy loss  $\bar{Q}$  is plotted versus the distance of closest approach with the incident ion energy as a parameter. For a given contour, the smaller distances of closest approach correspond to the larger angle collisions. (a) Argon ion-atom collisions. Also shown are the data of Kessel and Everhart (see Ref. 1). (b) Collisions between iodine ions and xenon atoms.

pect x rays having energies of about 2.9 keV to result from about 10% of these vacancies. X rays in this energy range have been observed and will be discussed later.

Figure 1(b) shows  $\bar{Q}$  plotted versus the distance of closest approach for the I-Xe combination. Previous measurements for  $\text{Ne}^+-\text{Ne}$ ,<sup>7</sup>  $\text{Ar}^+-\text{Ar}$ ,<sup>1</sup>

Table I. The average x-ray energies measured for several collision combinations.

Incident ion energy (MeV)	Type of collision	X-ray energy (keV)
1.5	$\text{Ar}^+-\text{Ar}$	$2.8 \pm 0.2$
1.5	$\text{H}^+-\text{Ar}$	$2.8 \pm 0.1$
3.0	$\text{H}^+-\text{Ar}$	$2.8 \pm 0.1$
3.0	$\text{H}^+-\text{Xe}$	$4.2 \pm 0.1$
3.0	$\text{I}^{2+}-\text{Xe}$	$4.0 \pm 0.1$
6.0	$\text{I}^{3+}-\text{Xe}$	$4.0 \pm 0.1$
10.0	$\text{I}^{4+}-\text{Xe}$	$4.0 \pm 0.1$

and  $\text{Kr}^+-\text{Kr}$ <sup>8</sup> collisions have seldom measured energy losses greater than 2000 eV; in the present experiment, the highest loss (for 6-MeV  $\text{I}^{3+}$  ions scattering to 10 deg) is nearly 30 000 eV. For I and Xe, the *L*-shell radii are approximately 0.06 Å and the *M*-shell radii are approximately 0.17 Å. The  $\text{I}^{n+}-\text{Xe}$  collisions of Fig. 1(b) do force these inner two shells to interpenetrate, and the average x-ray absorption edges of these *L* and *M* shells are approximately 5.1 and 0.9 keV, respectively. Vacancies must be produced in these shells in order to account for the large  $\bar{Q}$  values of Fig. 1(b). The x-ray fluorescence yields are not accurately known for these shells, but it may be assumed that significant numbers of both x rays and Auger electrons will be produced.

X rays from these heavy-ion collisions were detected by placing a proportional counter in the target gas chamber with the counter's 0.05-mm-thick Be window parallel to and about 1 cm from the beam path. The pulses from the proportional counter were amplified and accumulated in a 1024-channel pulse-height analyzer. There existed a predominant peak, approximately Gaussian in shape, for each energy spectrum obtained. These peaks were fitted by Gaussian-shaped curves, and the average energy  $E$  of each spectrum is given in Table I. Figure 2 shows these fitted Gaussians for the 3-MeV data. Their half-widths at  $1/e$  of their heights are approximately 0.5 keV for the argon targets and 0.7 keV for the xenon targets. No attempt was made to determine the relative contributions of the detector width and the natural widths to the observed widths. It must be assumed that each peak may contain many unresolved lines. The 2.8-keV x rays from the argon collisions show that *K*-shell vacancies are being produced when either protons or argon ions are used for projectiles. Similarly, the data in Table I show that the  $\text{H}^+-\text{Xe}$

collisions produce  $L$ -shell vacancies in the xenon targets. X rays associated with  $L$ -shell vacancies in iodine and xenon have approximately the same energies, and the present experiment did not determine their relative contributions to the 4.0-keV x-ray peaks shown in the table.

The cross sections for producing these vacancies must be roughly proportional to the cross sectional areas presented by the shells involved because the vacancies are produced only after the electron shells are forced to interpenetrate. Similar findings have been reported by Der *et al.*<sup>9</sup> in connection with the cross sections for x-ray production in carbon foils by heavy atoms. The actual mechanism by which heavy-ion collisions do produce inner-shell vacancies is uncertain. With light projectiles the Born approximation is often used to calculate the probability of the projectile removing a  $K$  or an  $L$  electron by direct collision.<sup>10</sup> The low relative velocities of heavy-ion collisions and the distortion of wave functions by these collisions preclude the usefulness of this approach. Collective oscillations of electron shells have been proposed by Amusia,<sup>11</sup> but to date, a model making use of the level crossings of diabatic molecular orbitals proposed by Fano and Lichten<sup>3</sup> has been more successful. It has predicted the positions of level crossings at lower energies as well as the Auger electron energies that might be expected from resulting vacancies. This model predicts  $K$ -shell level crossings for the  $\text{Ar}^+-\text{Ar}$  collisions for the internuclear distances of about 0.01 Å shown in Fig. 1(a); however, if the  $L$  shells are assumed full, the model does not allow for the production of a  $K$ -shell vacancy at these level crossings.

The x-ray measurements presented here are "total" with respect to ion-atom collision and are not differential as are the coincidence measurements of Fig. 2. The measurement of an x-ray's energy by itself does not indicate if it came from a gentle collision having a large impact parameter or a violent, large-angle collision having a small impact parameter. The 2.8-keV x rays from 1.5-MeV  $\text{Ar}^+-\text{Ar}$  collisions, however, are attributable to the relatively few collisions (less than 1%) that result in scattering angles of greater than 10 deg. The reason for this is that 1.5-MeV collisions with scattering angles of less than 10 deg (corresponding to distances of closest approach of greater than 0.02 Å in Fig. 1) have average energy losses of less than 3 keV, and such collisions are unlikely to produce 2.8-keV x rays. An experiment detecting x rays and scattered

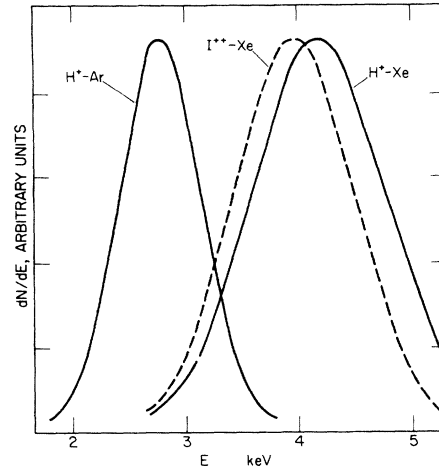


FIG. 2. X-ray spectra for the following 3-MeV collisions:  $\text{H}^+-\text{Ar}$ ,  $\text{I}^{++}-\text{Xe}$ , and  $\text{H}^+-\text{Xe}$ .

particles in coincidence will resolve this ambiguity and aid in the interpretation of these excitations.

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<sup>2</sup>This calculation for the distance of closest approach assumes the validity of a screened Coulomb potential. See, for example, E. Everhart, G. Stone, and R. J. Carbone, *Phys. Rev.* **99**, 1287 (1955).

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<sup>5</sup>L. I. Pivovarov, G. A. Krivonosov, and V. M. Tubaev, *Zh. Eksperim. i Teor. Fiz.* **53**, 1872 (1967) [translation: *Soviet Phys.-JETP* **26**, 1066 (1968)]; L. I. Pivovarov, M. T. Novikov, and V. M. Tubaev, *Zh. Eksperim. i Teor. Fiz.* **46**, 471 (1964) [translation: *Soviet Phys.-JETP* **19**, 318 (1964)].

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<sup>10</sup>E. Merzbacher and H. W. Lewis, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, Germany, 1958), Vol. 34, p. 166.

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### ELECTRON-IMPACT EXCITATION OF CARBON MONOXIDE NEAR THRESHOLD IN THE 1.5- TO 3-eV INCIDENT ENERGY RANGE\*

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A low-energy excitation process in CO at approximately 2.3 eV is investigated near threshold by a trapped-electron method. Structure has been observed in this excitation for the first time. An upper limit to the electron affinity of CO is established at  $-1.8 \pm 0.1$  eV.

Excitation of carbon monoxide by electron impact in the region 1.5-3 eV incident electron energy has been studied by a trapped-electron method in which a rather small well depth of approximately  $0.05 \pm 0.03$  V is maintained. A low-energy inelastic process peaking at about 2.3 eV is observed including structure as is shown in Fig. 1. This process has not been directly studied close to threshold before, and no structure has been observed near threshold by previous investigators. Schulz<sup>1</sup> has observed the phenomenon but not close to threshold. At least five "bumps" are clearly distinguishable with a spacing of approximately 0.2 eV. In the present work, it was found that increasing the well depth shifted the peak towards lower energy. At sufficiently large well depths where no vibrational structure could be observed in the  $\alpha^3\pi$  excitation (see below), this low-energy peak increased in magnitude and peaked at approximately 1.7 eV in agreement with the data of Schulz.

The type of device used in this experiment was a Tate-Smith ionization tube.<sup>2</sup> "Trapped-electron" spectra have been recorded in such a device.<sup>3</sup>

The estimation of the well depth is difficult and can hence lead to errors in the calibration of the electron energy. In the present experiment the well depth was kept sufficiently low to observe the vibrational fine structure of the  $\alpha^3\pi$  excitation. According to Herzberg<sup>4</sup> the  $v=0$  vibrational level for this state occurs at 6.01 eV. One method of calibrating the electron-energy axis is by assigning a value of 6.0 eV to this level. By extrapolating the electron beam current linearly to zero and establishing this intercept as zero energy, the  $v=0$  vibrational level of the  $\alpha^3\pi$  state occurs at 6.0 eV in agreement with the spectroscopic data of Herzberg<sup>4</sup> and the electron-impact

data of Brongersma and Oosterhoff.<sup>5</sup>

In Fig. 2 another method of energy calibration is shown. By increasing the voltage across the parallel plates, only negative ions can be collected at the collector plate.<sup>6</sup> The energy scale is calibrated by assuming the peak on the dissociative-attachment cross section occurs at 9.9 eV.<sup>7</sup> This mode of operation is much like that used by Rapp and Briglia.<sup>8</sup> In Fig. 2 the voltage across the plates is 6.2 V for the negative ions (open circles) which are taken at a sensitivity of 3.3 times the sensitivity of the trapped electrons. The voltage across the parallel plates for the trapped electrons is again zero. The peak at approximately 9.95 eV in the trapped-electron

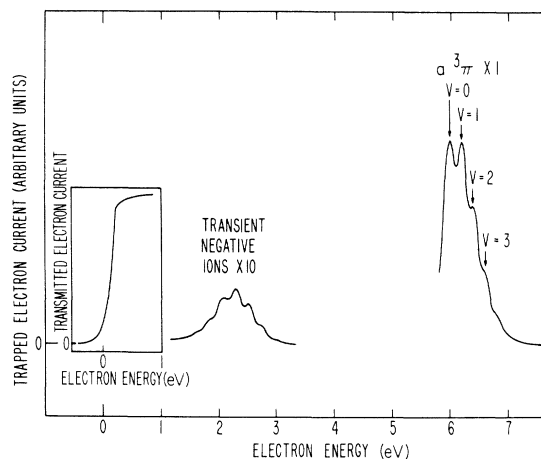


FIG. 1. Plot of trapped-electron current versus electron energy showing evidence of structure in transient negative-ion formation. Insert shows transmitted electron current. Calibration of electron-energy axis is obtained by linear extrapolation of transmitted electron current to zero, which places  $v=0$  vibrational level of  $\alpha^3\pi$  excitation at 6.0 eV.