obtained in a Lamb-shift source under the same conditions. It was assumed that  $\frac{1}{2}$  of the metastable atoms would be quenched and that the remaining metastable atoms would obtain complete nuclear polarization. This can be done, for example, by a scheme which was proposed by Sona<sup>13</sup> and verified experimentally by Clegg *et al.*<sup>14</sup> It was further assumed that in the process the metastable atoms which were quenched would be completely polarized in the opposite direction. The background and the atoms which were initially in the ground state were assumed to be completely unpolarized. The calculation at a proton energy of 0.7 keV indicates that

<sup>†</sup>Work supported in part by the U. S. Atomic Energy Commission.

- <sup>1</sup>W. Haeberli, Ann. Rev. Nucl. Sci. <u>17</u>, 406 (1967). <sup>2</sup>B. L. Donnally, T. Clapp, W. Sawyer, and M. Schultz,
- Phys. Rev. Letters <u>12</u>, 502 (1964).
- <sup>3</sup>I. A. Sellin and L. Granoff, Phys. Letters  $\underline{25A}$ , 484 (1967).

 ${}^{4}B.$  L. Donnally and W. Sawyer, Phys. Rev. Letters 15, 439 (1965).

<sup>5</sup>H. Brückmann, D. Finken, and L. Friedrich, Phys. Letters 29B, 223 (1969).

<sup>6</sup>B. Donnally and W. Sawyer, in *Proceedings of the* Sixth International Conference on the Physics of Electrons and Atomic Collisions: Abstracts of Papers (MIT a polarization of 0.7 can be expected.

The large values of Q and  $I_m$  indicate that the iodine charge-exchange reaction can be effectively used for production of positive polarized ions.

## ACKNOWLEDGMENTS

The author wishes to thank Professor W. Haeberli and Dr. L. W. Anderson for their helpful comments during the course of this experiment, and for their assistance in writing this paper. In addition, the author is indebted to M. H. Christensen for his assistance in taking data and maintaining the apparatus.

- Press, Cambridge, Mass., 1969), p. 488.
- <sup>7</sup>J. G. Lodge, J. Phys. B <u>2</u>, 322 (1969).
- <sup>8</sup>J. B. Hasted, *Physics of Atomic Collisions* (Butterworths, London, 1964), p. 440.
  - <sup>9</sup>H. O. Pritchard, Chem. Rev. <u>52</u>, 529 (1953).
  - <sup>10</sup>W. B. Person, J. Chem. Phys. <u>38</u>, 109 (1963).
- <sup>11</sup>W. E. Lamb, Jr. and R. C. Retherford, Phys. Rev. <u>79</u>, 549 (1950).
- <sup>12</sup>A. S. Schlachter, D. H. Loyd, P. J. Bjorkholm,
- L. W. Anderson, and W. Haeberli, Phys. Rev. <u>174</u>, 201 (1968).
  - <sup>13</sup>P. G. Sona, Energia Nucl. <u>14</u>, 295 (1967).
- <sup>14</sup>T. B. Clegg, G. R. Plattner, and W. Haeberli, Nucl. Instr. Methods <u>62</u>, 343 (1968).

PHYSICAL REVIEW A

#### VOLUME 2. NUMBER 5

NOVEMBER 1970

## Multiple Ionization of Iodine in 1.5- to 12.0-MeV I<sup>m+</sup>-Xe Collisions\*

Quentin C. Kessel<sup>†‡</sup>

Robert J. Van de Graaff Laboratory, High Voltage Engineering Corporation, Burlington, Massachusetts 01803

(Received 7 May 1970)

The results of an experiment measuring the charge states of 1.5-, 3.0-, 6.0-, and 12.0- MeV iodine ions scattered through angles between  $2.5^{\circ}$  and  $8.0^{\circ}$  by single collisions with xenon atoms are reported. The charge states measured range from 9 to 27, and the charge-state distributions showed average values from 11 to 23. The variation of these average values with the collision's distance of closest approach shows the influence of the *M* and *L* shells of the colliding ions.

## I. INTRODUCTION

Nearly all experiments concerning the collisions of heavy ions with heavy atoms investigate only excitations of one or two of the outermost electron shells. The Coulomb repulsion between atoms having high atomic number is too large to be overcome by the low-energy accelerators customarily used for atomic-collision research. In the present study, which used a 3-MeV tandem Van de Graaff accelerator, iodine ions having energies from 1.5 to 12.0 MeV were passed through thin targets of xenon. Effects of collisions involving not only the outer Oand N shells, but also the inner M and L shells were investigated by observing those iodine ions that were scattered through angles between 2.5 and 8.0 deg by single collisions.

Previous measurements in this laboratory have shown that such highly interpenetrating  $I^{n*}$ -Xe collisions do excite energy levels related to the L shells of these atoms.<sup>1</sup> The inelastic energy losses resulting from these collisions, nearly 30 keV in some cases, suggest that these collisions result in highly ionized ions. The purpose of this experiment was to measure these ionization states for the scattered iodine ions. Similar differential measurements have been made using lighter ions (including He, Ne, Ar, and Kr) at lower energies in Everhart's laboratory at the University of Connecticut<sup>2,3</sup> and in Fedorenko's laboratory at the Ioffe Physical-Technical Institute.<sup>4</sup> Higher-energy investigations of the charge states of scattered ions have been made by Pivovar and co-workers at the Khar' kov Physical-Technical Institute<sup>5-7</sup> using ion energies up to 1.8 MeV.

## **II. EXPERIMENTAL METHOD**

The differential scattering chamber used in this experiment has been described in detail.<sup>8</sup> The incident ion beam is collimated by two apertures and passed into a differentially pumped target-gas chamber. A second collimator samples those iodine ions that are scattered through  $\theta$  deg by single collisions with xenon atoms. The charge of these scattered ions is determined by electrostatic analysis, and the number of ions having charge state m,  $N_m$ , are counted. The probability  $P_m$  of the beam scattered through the angle  $\theta$  being found to have charge state m is given by

$$P_m = N_m / \sum_m N_m \quad . \tag{1}$$

At each of four energies, 1.5, 3.0, 6.0, and 12.0 MeV, the values of  $P_m$  were determined for a number of scattering angles between 2.5 and 8 deg. For each combination of incident-ion energy and scattering angle, the average charge of the scattered ions  $\overline{m}$  was also determined. The value of  $\overline{m}$ , determined from the corresponding values of  $P_m$ , is given by

$$\overline{m} = \sum_{m} m P_{m}.$$
 (2)

## III. EXPERIMENTAL RESULTS

The measured values of  $P_m$  are plotted in Fig. 1 for the four incident-ion energies investigated. The ordinate shows the values of  $P_m$  corresponding to each value of  $\theta$  plotted along the abscissa. Qualitatively these curves are similar to those found for heavy-ion scattering at lower energies.<sup>2-7</sup> For example, consider the 1.5-MeV data in Fig. 1(a): As the scattering angle increases, corresponding to a greater interpenetration of the electron shells, the degree of ionization of the scattered ions also increases. Only 1% of the 1.5-MeV ions scattered to 4 deg are 16 times ionized, while nearly 20% of those ions scattered to 8 deg have this charge.

Figures 1(a) - 1(d) together show the effect of increasing the collision energy. The additional interpenetration of the electron shells at the higher energies results in further ionization. While the predominant charge state for 4 deg scattering at 1.5 MeV is about 12, at 3.0 MeV it is 17, at 6.0 MeV it is 20, and at 12.0 MeV it is 23. The increase found for the 6.0- and 12.0-MeV data may, in part, be due to the higher initial ionization states of the incident ions. However, the 1.5- and 3.0-MeV data



FIG. 1. Probabilities  $P_m$  for the scattered iodine ions having charge *m* after collision as a function of the scattering angle  $\theta$ : (a)  $I^{2*}$ -Xe, 1.5 MeV; (b)  $I^{2*}$ -Xe, 3.0 MeV; (c)  $I^{3*}$ -Xe, 6.0 MeV; and (d)  $I^{5*}$ -Xe, 12.0 MeV.

show, as do much of the lower-energy data,  $^2$  that the effect is due primarily to the higher collision velocities and the greater electron-shell interpenetration associated with these higher-energy collisions. It is noteworthy that more than 20 electrons are often removed from the iodine ion by a single collision.

Figure 2(a) plots the average scattered charge  $\overline{m}$ versus the scattering angle  $\theta$ , for each energy. This displays some features of the ionization that were not evident in Fig. 1. The 1.5-MeV data show a relatively rapid rise in  $\overline{m}$  for scattering between 4 and 7 deg. The 3.0-MeV data show a similar rise between 2.5 and 4 deg. A rise, but not so pronounced, is also evident in the 6.0-MeV data; however, the 12.0-MeV data show only a slight increase in  $\overline{m}$ . It is illuminating to plot these same data versus the collision's distance of closest approach  $r_0$ . It is possible to calculate  $r_0$  for each combination of incident-ion energy and scattering angle, <sup>9</sup> and in Fig. 2(b) the measured values of  $\overline{m}$  are plotted versus their corresponding values of  $r_0$ . For each energy the larger values of  $r_0$  correspond to the smallerangle collisions, and the smaller values of  $r_0$  correspond to the more violent larger-angle collisions.

The width and asymmetry of the distributions of charge states around each value of the average charge  $\overline{m}$  may also be calculated. The distribution width or standard deviation  $\sigma$  of the percentages from the average charge  $\overline{m}$  may be defined by

$$\sigma^2 = \sum_m (\bar{i} - i)^2 P_m. \tag{3}$$

The asymmetry or skewness  $\gamma$  of such a distribution may be defined by

$$\gamma = \sum_{m} (\vec{i} - i)^3 P_m / \sigma^3. \tag{4}$$

Figure 3 shows the values of  $\sigma$  and  $\gamma$  associated with each m. The width  $\sigma$  increases gradually, reaching a maximum of nearly 2.0 for scattered ions having an average charge of about 20. For higher values of  $\overline{m}$ ,  $\sigma$  appears to decrease. On the other hand,  $\gamma$ is close to zero for values of  $\overline{m}$  from 11 to 20. This corresponds to a nearly symmetric charge-state distribution. However,  $\gamma$  does show a definite trend toward negative values for values of  $\overline{m}$  greater than 20. A negative value of  $\gamma$  corresponds to an asymmetric distribution having the higher charge states bunched near  $\overline{m}$  and the lower charge states tailing off less rapidly on the other side of m. The ionization of 25 electrons represents the complete removal of the O and N shells of the iodine atom. The smaller values for  $\sigma$  and the negative values of  $\gamma$  for values of  $\overline{m}$  approaching 25 probably reflect the difficulty of removing additional electrons.

#### IV. DISCUSSION

These data represent an extension of earlier differential-scattering investigations.<sup>2-7</sup> The present



FIG. 2. (a) Average charge  $\overline{m}$  for the scattered iodine ions is shown as a function of the scattering angle  $\theta$ for each of four incident-ion energies. (b) The average charge  $\overline{m}$  is plotted versus the corresponding distance of closest approach  $r_0$ . The horizontal bracket indicates the range of  $r_0$  approximately equal to the diameters of the L shells of iodine.

experiment combines the use of higher acceleration energies and a heavier ion-atom combination than have been reported by other investigators. As a result, the ionization states here are higher than those previously observed by this technique; however, the general dependence of the ionization on incident-ion energy and scattering angle is the same as that found using lower collision energies.

The dependence of  $\overline{m}$  on  $r_0$ , shown in Fig. 2(b), suggests that the degree of ionization is strongly dependent upon the shell structure of the colliding ions. For a distance of closest approach corresponding to the L shells of the two ions beginning to interpenetrate (the horizontal bracket in Fig. 2 indicates the range of  $r_0$  approximately equal to the diameters of the L shells of iodine),  $\overline{m}$  increases rapidly. Experiments detecting x rays from these and similar collisions<sup>1,10</sup> suggest that this increase in  $\overline{m}$  may be due to the production of a vacancy in the L shell of the iodine ion and the resulting ionization cascade. In this way, the ionization is similar in nature to that found when the L shells of two argon ions are forced to interpenetrate. <sup>1,2,7,11</sup> Figure 2(b) shows that for further interpenetration of the L shells,  $\overline{m}$ 

increases less rapidly. Since the removal of 25 electrons corresponds to the ionization of all the electrons in both the O and N shells of iodine, the leveling off of the  $\overline{m}$  curves in Fig. 2 probably reflects the greater energy required for ionization of additional electrons that now must come from the M shell. The data in Fig. 3 substantiate this. Therefore, these data reflect the shell structure of iodine in two ways: First, when the L shells of the two ions interpenetrate, one or more vacancies are apparently produced in the L shell of the iodine ion, and this results in an increase in  $\overline{m}$ . Second, as would be expected, the electrons in the M shell prove more difficult to ionize than do the outer O- and Nshell electrons, and this is reflected in the chargestate distributions. Although Fig. 1 does show the removal of the 26th and 27th electrons from some ions, Figs. 2 and 3 indicate the difficulty of doing this.

#### ACKNOWLEDGMENTS

The author is grateful to Professor Edgar Everhart and Professor Arnold Russek for their interest in this investigation. The experiment could not have been undertaken without the enthusiastic support of Dr. Peter Rose and the High Voltage Engineering Corporation. Walter Powers ably supervised the accelerator operation and assisted in taking the data.

### APPENDIX

(a) *Scattering geometry*. The incident ion beam was collimated by two 0.76-mm-diam apertures



FIG. 3. Characteristics of the charge-state distributions shown as a function of the average charge  $\overline{m}$ : (a)  $\sigma$ , a measure of the distribution's asymmetry, obtained from Eq. (3); (b)  $\gamma$ , a measure of the distribution's asymmetry, obtained from Eq. (4).



FIG. 4. The dependence of  $\overline{m}$  upon the target-gas pressure is shown. The arrow indicates the pressure at which the rest of the data for this paper were obtained.

31 cm apart. After collision, the scattered ions were collimated by two apertures. The first of these, 1.0 cm from the scattering center, was a 0.23-mm slit mounted on a 1.15-mm-diam hole. The second, 8.65 cm from the scattering center, was a 0.44-mm slit mounted on a 1.52-mm-diam hole. These slits subtended an angle of 0.29 deg from the geometric scattering center in the plane of the apparatus.

(b) Single collision criteria. The electron capture and loss cross sections for these highly ionized atoms are unknown. It was necessary to determine an appropriate target-gas pressure for single collisions experimentally. Figure 4 shows the dependence of  $\overline{\overline{m}}$  on target-gas pressure for 4 deg scattering at 3.0 and 6.0 MeV. For pressures above  $10^{-3}$  Torr,  $\overline{m}$  drops rapidly. Below  $10^{-3}$  Torr,  $\overline{m}$  is constant, and it is evident that single collisions predominate. The data presented in this paper were obtained using a pressure of  $10^{-4}$  Torr to minimize the effects of multiple collisions. The path length 1 of the beam in the target gas was 2.0 cm, and the partial pressure due to residual impurities in the target-gas chamber was less than  $3 \times 10^{-6}$  Torr.

(c) *Charge-state analyzer and particle detector*. After collimation, the scattered iodine ions were



FIG. 5. A profile showing the resolution of the electrostatic analyzer.

passed through an electrostatic analyzer and into a silicon surface-barrier detector. The electrostatic analyzer is of a cylindrical design<sup>8</sup> with a 30cm radius. For charge separation the ion beam is deflected through 117 deg, and after analysis the ions are further collimated by a 6-mm slit before passing into the particle detector. Figure 5 demonstrates the analyzer's resolution.

\*This research was generously supported by the High Voltage Engineering Corporation.

<sup>†</sup>Present address: Department of Physics, University of Connecticut, Storrs, Conn. 06268.

<sup>‡</sup>On leave to the Institute of Physics, University of Aarhus, Aarhus, Denmark, for the 1970-71 academic year.

<sup>1</sup>Q. C. Kessel, P. H. Rose, and L. Grodzins, Phys. Rev. Letters <u>22</u>, 1031 (1969).

<sup>2</sup>E. N. Fuls, P. R. Jones, F. P. Ziemba, and E. Everhart, Phys. Rev. 107, 704 (1957).

<sup>3</sup>F. P. Ziemba, G. J. Lockwood, G. H. Morgan, and E. Everhart, Phys. Rev. 118, 1552 (1960).

<sup>4</sup>D. M. Kaminker and N. V. Fedorenko, Zh. Tekhn. Fiz. 25, 2239 (1955).

<sup>5</sup>L. I. Pivovar, M. T. Novikov, and V. M. Tubaev, Zh. Eksperim. i Teor. Fiz. <u>46</u>, 471 (1964) [Soviet Phys. JETP <u>19</u>, 318 (1964)].

<sup>6</sup>L. I. Pivovar, M. T. Novikov, and A. S. Dolgov, Zh. Eksperim. i Teor. Fiz. <u>50</u>, 537 (1966) [Soviet Phys. JETP 23, 357 (1966)].

<sup>7</sup>L. I. Pivovar, G. A. Krivonosov, and V. M. Tubaev, Zh. Eksperim. i Teor. Fiz. <u>53</u>, 1872 (1967) [Soviet Phys. JETP <u>26</u>, 1066 (1968)].

<sup>8</sup>Q. C. Kessel, Rev. Sci. Instr. 40, 68 (1969).

<sup>9</sup>E. Everhart, G. Stone, and R. J. Carbone, Phys. Rev. <u>99</u>, 1287 (1955). This classical calculation makes use of a screened Coulomb potential. Extensive tables of this and other classical collision parameters have been prepared by Felton W. Bingham and are available as Document No. SC-RR-66-506, Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. (price, \$3.00) (unpublished).

<sup>10</sup>H. J. Stein, H. D. Lutz, P. H. Mokler, K. Sistemich, and P. Armbruster, Phys. Rev. Letters <u>24</u>, 701 (1970).

<sup>11</sup>Q. C. Kessel and E. Everhart, Phys. Rev. <u>146</u>, 16 (1966).

## PHYSICAL REVIEW A

VOLUME 2, NUMBER 5

NOVEMBER 1970

# Shift and Broadening of the <sup>87</sup>Rb 0-0 Line Due to Collisions with Krypton Buffer-Gas Atoms

Francis Hartmann\* and Francoise Hartmann-Boutron<sup>†</sup> Joint Institute for Laboratory Astrophysics, Boulder, Colorado 80302 (Received 20 April 1970)

The shift and broadening of the  ${}^{87}$ Rb 0-0 line (frequency 6.8 GHz) due to collisions with Kr atoms have been studied in the pressure range 0.2-10 Torr. The shift versus pressure has been found to be linear and equal to  $-542 \pm 10$  Hz/Torr. The linewidth results from the superposition of several effects which are analyzed, one of the most important being the relaxation due to the spin-orbit interaction experienced by Rb atoms while temporarily bound in van der Waals Rb-Kr molecules.

## I. INTRODUCTION

The interactions between two colliding atoms are currently the subject of many investigations. Direct observation of the internal degrees of freedom of the colliding partners can provide useful information complementing the results of scattering experiments. Study of the relaxation of polarized <sup>87</sup>Rb atoms in krypton buffer gas<sup>1</sup> has thus proved to be very sensitive to the presence of transient bound states in the Rb-Kr pair. In this connection one is led to wonder if the very narrow 0-0 hyperfine line of <sup>87</sup>Rb (unperturbed frequency<sup>2</sup> 6 834 682 614 Hz) should not be affected by the same phenomena.

Measurements, using standard optical pumping techniques, of the 0-0 linewidth at  $27 \degree C$  (Rb vapor

pressure  $3 \times 10^{-7}$  Torr) in the presence of various buffer gases (He, Ne, Ar, N<sub>2</sub>, CH<sub>4</sub>) have already been made, at pressures ranging from 1 to 400 Torr.<sup>3</sup> When only binary collisions between Rb and buffer-gas atoms take place, one expects the linewidth to be the sum of two terms: a collision-narrowed Doppler width, inversely proportional to buffer-gas pressure, and a contribution proportional to the buffer-gas pressure, due to disorienting or dephasing collisions.<sup>4</sup>

Whereas the experimental results agree well with such a prediction in the case of He and Ne, there exists in the cases of Ar,  $N_2$ , and  $CH_4$  a small extra linewidth which has been attributed by Bender and Cohen<sup>3</sup> to the formation of bound states.

In view of the relaxation experiments<sup>1</sup> which dem-