

tions based on the first Born approximation even though correct total cross sections have been calculated.

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<sup>1</sup>R. A. Mapleton, *Theory of Charge Exchange* (Wiley-Interscience, New York, 1972).

<sup>2</sup>E. Merzbacher and H. W. Lewis, in *Encyclopedia of Physics*, edited by S. Flügge (Springer, Berlin, 1958), Vol. 34, p. 166.

<sup>3</sup>M. Inokuti, *Rev. Mod. Phys.* **43**, 296 (1971).

<sup>4</sup>J. D. Jackson and H. Schiff, *Phys. Rev.* **89**, 359 (1953).

<sup>5</sup>P. M. Stier and C. F. Barnett, *Phys. Rev.* **103**, 896 (1956).

<sup>6</sup>A. M. Halpern and J. Law, *Phys. Rev. A* **12**, 1776 (1975).

<sup>7</sup>J. R. Oppenheimer, *Phys. Rev.* **31**, 349 (1928).

<sup>8</sup>H. C. Brinkman and H. A. Kramers, *Proc. Acad.*

*Sci. Amsterdam* **33**, 973 (1930).

<sup>9</sup>K. Omidvar, J. E. Golden, J. H. McGuire, and O. L. Weaver, *Phys. Rev. A* **13**, 500 (1976).

<sup>10</sup>J. R. Macdonald, S. M. Ferguson, L. D. Ellsworth, T. Chiao, and W. W. Eidson, in *Proceedings of the Seventh International Conference on the Physics of Electronic and Atomic Collisions*, edited by L. M. Branscomb *et al.* (North-Holland, Amsterdam, 1971), p. 516.

<sup>11</sup>J. R. Macdonald, C. L. Cocke, and W. W. Eidson, *Phys. Rev. Lett.* **32**, 648 (1974).

<sup>12</sup>R. R. Randall, J. A. Bednar, B. Curnutte, and C. L. Cocke, *Phys. Rev. A* **13**, 204 (1976).

<sup>13</sup>J. M. Hansteen and O. P. Mosebekk, *Nucl. Phys. A201*, 541 (1973); J. M. Hansteen, O. M. Johnsen, and L. Kocbach, to be published.

<sup>14</sup>E. Laegsgaard, J. U. Andersen, and L. C. Feldman, *Phys. Rev. Lett.* **29**, 1206 (1972).

<sup>15</sup>N. Bohr, *Kgl. Dan. Vidensk. Selsk., Mat.-Fys. Medd.* **18**, No. 8 (1948).

<sup>16</sup>D. R. Bates, *Proc. Roy. Soc., Ser. A* **247**, 294 (1958); P. J. Kramer, *Phys. Rev. A* **6**, 2125 (1972); R. H. Basssel and E. Gerjuoy, *Phys. Rev.* **117**, 749 (1960); T. B. Day, L. S. Rodberg, G. A. Snow, and J. Sucher, *Phys. Rev.* **123**, 1051 (1960); M. L. Goldberger and K. M. Watson, *Collision Theory* (Wiley, New York, 1964), p. 156.

## Ultraviolet Radiation Produced in Near-Threshold Ar + Ar Atomic Collisions\*

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First measurements are reported of the near-threshold behavior of the vacuum-ultraviolet emission cross section for Ar + Ar atomic collisions. The experimentally observed threshold is  $18 \pm 2$  eV. The absolute emission cross section is presented for center-of-mass energies from threshold to 150 eV. The monitored radiation results from the decay of the lowest excited resonance state of the Ar atom and individual substate contributions are determined over most of the energy range investigated.

Measurements of the absolute ultraviolet emission cross section for Ar + Ar collisions have been made from near threshold to 150 eV center-of-mass energy. These measurements represent the first experiment in which the emitted uv radiation is spectrally dispersed in the low-energy region. The bulk of the observed radiation is shown to emanate from the lowest excited state of the Ar atom, which forms a quartet of levels from the  $3p^5 4s$  configuration. Two of these levels are metastable and two decay optically. Photons from the decaying levels, at wavelengths of 104.8 and 106.7 nm, have been observed with absolutely calibrated detectors that collect radiation emitted at  $90^\circ$  relative to the neutral-atomic-beam axis.

No attempt has been made to distinguish direct collisional excitation of these levels from reactions feeding these levels via cascading processes.

The monoenergetic beam of Ar atoms is formed by near-resonant charge transfer of suitably prepared  $\text{Ar}^+$  ions.<sup>1</sup> The ion source, of electron-impact type, is operated at electron energies below the threshold for excited-state ion formation. The charge-transfer species  $\text{H}_2$  is employed for neutralizing  $\text{Ar}^+$  ions to ensure that the product neutral Ar atoms are in their ground electronic states at the lower collision energies. Thus for the  $\text{Ar}^+ + \text{H}_2$  reaction, sufficient energy in the center-of-mass system (11.6 eV) to excite a

product Ar atom can be obtained only for  $\text{Ar}^+$  laboratory energies above 240 eV. While the symmetric-resonant charge-transfer reaction (i.e.,  $\text{Ar}^+ + \text{Ar}$ ) would appear favorable, there is strong evidence that the product neutral beam contains metastable atoms. Generation of these metastable atoms by the resonant process, or by the process used here when high ion-source electron energies are employed, is the topic of another paper.<sup>2</sup>

The ground-state neutral-atom flux enters the target scattering cell (typically at  $10^{-4}$  Torr) where two detectors monitor the emitted uv radiation. The first, a broad-bandpass total-emission detector, is a channel electron multiplier (CEM) restricted to a determined viewing field and fronted by a thin (0.2-mm) LiF window. This detector has a bandpass between 104 and 150 nm. The second detector is a vacuum monochromator having peak efficiency between 100 and 125 nm (1-nm resolution) followed by a second CEM. Photon counting techniques are used throughout.

The quantum efficiencies of the CEM's are estimated to be known to within  $\pm 20\%$  uncertainty in the wavelength range of interest.<sup>3</sup> To check this assignment and other potential uncertainties, the total-emission detector was allowed to view Lyman- $\alpha$  ( $L_\alpha$ , 121.6 nm) radiation from the reaction



After a scan of the total-emission detector bandpass region with the monochromator to verify that no other radiation was present, the absolute  $L_\alpha$  emission cross section for this reaction was measured. Values obtained averaged 10% lower than those reported by Dunn, Geballe, and Pretzer<sup>4</sup> and are estimated to be uncertain by less than  $\pm 30\%$  (to be compared with an uncertainty of  $\pm 40\%$  claimed by Dunn, Geballe, and Pretzer). Absolute calibration of the monochromator detector is also accomplished with Reaction (1) by normalizing its CEM output to that obtained with the total-emission detector.<sup>5</sup> Manufacturer's data on the wavelength dependences of such detector parameters as CEM efficiency, grating reflectivity, etc., are then used to infer absolute detector sensitivities at the wavelengths of interest.

The cross section measured with the total emission detector is thus determined within an absolute uncertainty of  $\pm 30\%$  and a relative uncertainty (as a function of energy) of  $\pm 15\%$  except in the immediate threshold region where uncertainties increase to about twice the above values. Individual

line-emission cross sections are uncertain by about  $\pm 35\%$ .

The results on absolute uv emission cross section are shown in Fig. 1. The lower two curves depict the cross sections for the labeled line emissions from 30 to 150 eV, the sum of which is indicated by the dashed curve. The upper curve is the result obtained with the total-emission detector. This cross section should be comparable in magnitude to the dashed curve, provided that this detector's bandpass does not encompass other spectral emissions.<sup>6</sup>

An interesting feature of these data is the constant ratio between the cross sections for the substate emissions. The 106.7-nm to 104.8-nm intensity ratio remains at  $1.4 \pm 10\%$  over the energy range investigated.

The energy dependence of the total emission cross section in the near-threshold region is shown in Fig. 2. In the region between 40 and 60 eV, the cross-section energy dependence appears to be linear. An extrapolation of this linear portion intercepts the energy axis at about 34 eV. Below 40 eV, however, the cross section assumes a basically exponential dependence. Between 19 and 40 eV, the data points have been least-squares fitted by the expression

$$\sigma = 0.324(\Delta E)^{0.75} e^{0.167\Delta E}, \quad (2)$$

shown by the curve drawn through the data points. Here,  $\Delta E$  is the energy in excess of 18 eV, the

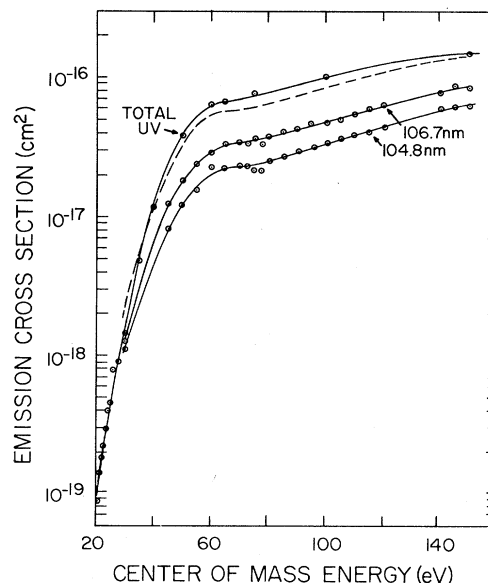


FIG. 1. uv emission cross sections for Ar + Ar collisions.

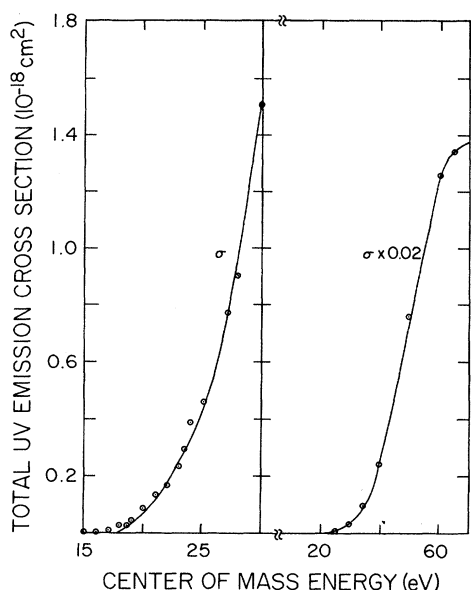


FIG. 2. Total uv emission cross section at low energies for Ar + Ar collisions.

experimental onset, to within  $\pm 2$  eV, for production of measurable amounts of radiation. This experimental onset corresponds to our minimum detectable cross section of about  $5 \times 10^{-21}$  cm<sup>2</sup>. The  $\pm 2$ -eV uncertainty limits reflect consideration of the initial neutral-atom-beam energy profile ( $\sim 1$  eV, full width at half-maximum), degradation of the average atom energy by elastic scattering within the target cell, small experimental energy offsets, and Doppler broadening<sup>7</sup> effects.

Two previous measurements of total uv emissions (including radiation extending to much shorter wavelengths) from Ar + Ar collisions are compared with the present results by the three upper curves in Fig. 3. The cross-section structure at about 70 eV is reproduced in all data, but the energy dependences are different below 50 eV. The present authors suggest that trace numbers of metastable Ar atoms in the neutral beams used in the earlier studies may contribute to the observed differences.<sup>11</sup>

Brenot *et al.*<sup>12</sup> have developed a model for outer-shell excitation in low-energy rare-gas collisions. Inelastic collisions are treated in terms of molecular state transitions occurring at crossings of simplified diabatic molecular potential energy curves. On the basis of the model, Brenot *et al.* suggest that one preferred crossing between the incident diabatic ground-state curve and a curve dissociating to two  $3p^5 4s$  excited Ar atoms leads to both ionized and excited-state collision

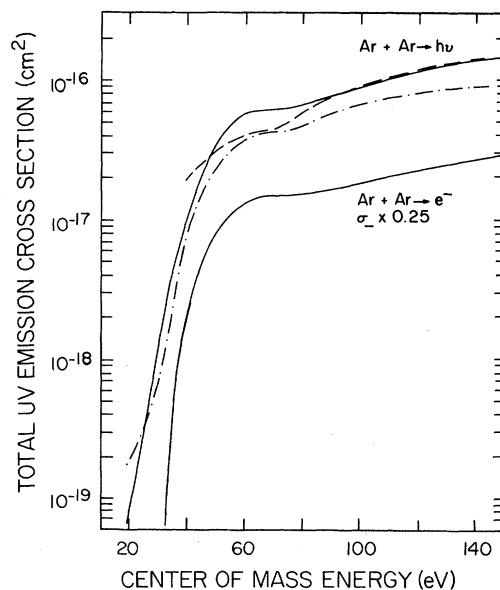


FIG. 3. Comparison of total uv emission cross sections and the ionization cross section for Ar + Ar collisions. The emission-cross-section curves are from the present work (—), Haugsjaa and Amme, Ref. 8 (---), and Kempter, Veith, and Zehnle, Ref. 9 (-·-·-). The ionization-cross-section curve is from Haugsjaa and Amme, Ref. 10.

products. Supportive of this hypothesis is the fact that the energy dependences of the reported emission cross section and that for ionization,<sup>10</sup> shown renormalized by the lower curve in Fig. 3, are remarkably similar at energies beyond the threshold region.

The position of this preferred crossing occurs near 31 eV, according to the model. This value is in agreement with the 32-eV threshold value claimed by Kempter, Veith, and Zehnle,<sup>9</sup> who obtain their value by a linear extrapolation to zero of their measured 795-nm ( $3p^5 4p - 3p^5 4s$ ) emission cross section. A similar value ( $\sim 34$  eV) is obtained, as mentioned earlier, by linearly extrapolating the straight-line portion of the present cross section to its zero intercept. A plot of the ionization-cross-section data<sup>10</sup> in a manner analogous to Fig. 2 also exhibits a straight-line region with a 32-eV intercept.

It is apparent, however, that these values are not characteristic of either the true or the experimental thresholds for the excitation (or ionization) process. The  $18 \pm 2$ -eV "minimum detectable threshold" for uv emission reported here is well below the preferred crossing value of 31 eV in the Brenot *et al.* model. In fact, our 18-eV value is even substantially below the lowest po-

tential-energy-curve crossing at 24.6 eV reported by Brenot *et al.* (This crossing leads to a  $3p^54p$  excited Ar atom which could, via a cascade process, contribute to the observed uv emission.) Furthermore, the true energy threshold must be only 11.6 eV, the excitation energy of the lowest  $3p^54s$  resonance level of the Ar atom.

The basically exponential energy dependence of the reported uv emission cross section, as given by Eq. (2), is perhaps suggestive of a quantum-mechanical barrier-penetration phenomenon, with transitions occurring at somewhat larger separations than the crossing radius. Of course, such transitions between the molecular potential energy curves at radii beyond the crossing vicinity remain energetically possible down to 11.6 eV.

If the interpretation that photons are being observed at energies below the crossing energy is correct, it points to the danger of establishing curve-crossing radii from threshold-energy determinations. If it is not correct, then it would appear that the crossing responsible for the observed uv emission has been improperly identified.

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trophysics, University of Colorado, Boulder, Colorado 80302.

<sup>1</sup>The basic apparatus is described by N. G. Utterback and G. H. Miller, *Rev. Sci. Instrum.* **32**, 1101 (1961).

<sup>2</sup>H. L. Rothwell, Jr., R. C. Amme, and B. Van Zyl, to be published.

<sup>3</sup>M. C. Johnson, *Rev. Sci. Instrum.* **40**, 311 (1969).

<sup>4</sup>G. H. Dunn, R. Geballe, and D. Pretzer, *Phys. Rev.* **128**, 220 (1962).

<sup>5</sup>The more limited field of view of this detector dictated corrections for the fraction of an ion or neutral beam passing within its viewing field.

<sup>6</sup>Spectral scans failed to identify any such emissions. The small differences in magnitude between these curves are probably due to uncertainties in accounting for various experimental sensitivity factors.

<sup>7</sup>P. J. Chantry, *J. Chem. Phys.* **55**, 2746 (1971).

<sup>8</sup>P. O. Haugsjaa and R. C. Amme, *Phys. Rev. Lett.* **23**, 633 (1969). The lower-limit cross-section values reported have been revised in light of proper evaluation of the detector solid angle and other factors entering these experiments.

<sup>9</sup>V. Kempter, F. Veith, and L. Zehnle, in *Proceedings of the Ninth International Conference on the Physics of Electronic and Atomic Collisions, Seattle, Washington, 1975*, edited by J. S. Risley and R. Geballe (Univ. of Washington Press, Seattle, Wash., 1975), p. 617.

<sup>10</sup>P. O. Haugsjaa and R. C. Amme, *J. Chem. Phys.* **52**, 4874 (1970).

<sup>11</sup>In the present experiment, it was found that a beam containing metastable atoms (Ref. 2) caused the measured cross sections to become increasingly too large at the lower collision energies.

<sup>12</sup>J. C. Brenot, D. Dhuicq, J. P. Gauyacq, J. Pomier, V. Sidis, M. Barat, and E. Pollack, *Phys. Rev. A* **11**, 1245 (1975).

## Structure of Sodium Rydberg States in Weak to Strong Electric Fields\*

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We present results of an experimental survey of the  $n = 15$  state of Na in electric fields from 0 to 4 kV/cm. Both linear and higher-order Stark effects play important roles. States with low  $|m|$  show marked differences from hydrogenic behavior, particularly in the vicinity of level crossings. Computed values for the energy levels are in good agreement with the data. The experimental method provides a simple technique for populating selected Stark or angular momentum levels of a Rydberg state.

A number of research groups have now developed techniques for creating and detecting atoms in pure Rydberg states,<sup>1</sup> opening the way to studies in such areas as electron-core interactions, tunneling and photoionization phenomena, corre-

lated electron motion, and collisions. For many applications it is essential to populate selected angular-momentum states or, in the case that an electric field mixes angular momentum, to populate selected Stark states.<sup>2</sup> We present here re-