Laser Triple-Quantum Photoionization of Cesium*

R. A. Fox,† R. M. Kogan, and E. J. Robinson
Physics Department, New York University, Bronx, New York 10453
(Received 5 April 1971)

We report observations of the ionization of atomic cesium by the simultaneous absorption of three ruby-laser quanta. Our results appear to be in qualitative agreement with theory, and indicate that circularly polarized ruby light produces ionization about twice as efficiently as does linearly polarized light.

The invention of the high-power laser has made the observation of nonlinear optical processes possible, and many studies, especially in solids, have been performed. Free atomic systems, however, offer the best chance for making comparisons between experiment and detailed theories, and a number of publications discussing multiple-quantum atomic transitions have also appeared.[1-11] The present Letter describes observations of the triple-quantum ionization of cesium by ruby light.

The transition rate for a three-photon process is given by

$$W = \delta_3 F^3, \tag{1}$$

where F is the photon flux per unit area and δ_3 is a coefficient containing the perturbation sum over intermediate states. (See Ref. 5, for example.)

In our experiment, a collimated oven beam of cesium, detected with virtually unit efficiency by surface ionization, is cross fired by light from a Q-switched ruby laser. Charge produced by the interaction between laser light and atoms is focused onto the first dynode of an electron multiplier, whose output constitutes the signal for the experiment.

The charge arriving at the anode of the multiplier is given by

$$Q = eNfA, \tag{2}$$

where N is the number of photoionization events, f the collection efficiency, and A the multiplier gain. The number of ion-electron piars is given by

$$N = H V n \delta_3 \int F^3 dt. \tag{3}$$

Here, n is the atomic number density in the interaction volume V and H is a factor correcting Eq. (1) for the multimode character of the laser beam (see Ref. 9).

In order to determine the flux-cubed integral, it is necessary to measure the shape of the laser pulse, in addition to its total energy. This is accomplished with a fast photodiode (ITT Industrial Labs F4000) communicating with a traveling-wave oscilloscope (Tektronix 519).

Figures 1(a) and 1(b) are typical oscilloscope

(a)
(b)
(c)

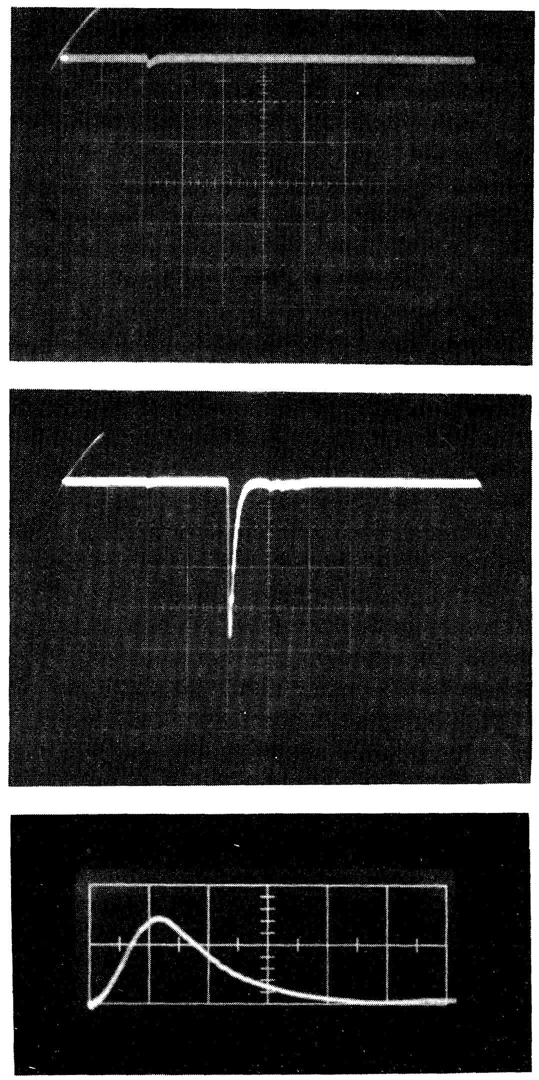

FIG. 1. (a) Multiplier output, atomic beam off. Scale: 20 mV/cm, 1 μsec/cm. (b) Photoelectric signal. Scale: 20 mV/cm, 1 μsec/cm. (c) Laser pulse. Scale: 50 MW/cm, 20 nsec/cm.

traces of the multiplier output pulse with the atomic beam off and on, respectively. Figure 1(c) is a laser pulse. The data for the experiment are contained on such photographs.

Our data reduction involves a plot of multiplier signal against flux-cubed integral. This is fitted quite well by a straight line, and demonstrates that we are indeed dealing with a three-photon process. Although our knowledge of the calibration factors in Eqs. (2) and (3) is not yet sufficient to distinguish between Bebb's theoretical value[5] of $\delta_3 = 6\times10^{-78}$ cm^6 sec^2 and Robinson's[12] $\delta_3 = 3\times10^{-77}$ cm^6 sec^2, reasonable estimates indicate at least order-of-magnitude agreement with theory. Relative transition rates for different states of polarization are, however, independent of the instrumental functions, if we collect the ions. We report that $\delta_{3c} = (2.15\pm0.4)\delta_{3l}$, where δ_{3c} and δ_{3l} are the respective transition coefficients for circularly and linearly polarized light.

It may seem strange, at first, that the total cross section for the interaction of light with an unpolarized target should be a function of the state of polarization of the bombarding radiations. There do not appear to be any references to this interesting effect in the literature. We explain our result by noting that an nth-order transition proceeds via intermediate states. During the first $n-1$ transitions, the polarization of the radiation is partially transferred to the atoms, and the nth interaction is between polarized quanta and polarized atoms. These effects will be examined in detail in a future paper.

*Research supported by the U. S. Office of Naval Research under Contract No. NOOO-14-67-A-0467-002.

†This work constitutes a portion of a thesis to be submitted by R. A. Fox to the Graduate School of Arts and Science of New York University in partial fulfillment of the requirements for the Ph.D.

[1]S. Geltman, Phys. Lett. 4, 168 (1963), and 19, 616 (1965).

[2]A. Gold and H. B. Bebb, Phys. Rev. Lett. 14, 60 (1965).

[3]H. B. Bebb and A. Gold, Phys. Rev. 143, 1 (1966).

[4]H. B. Bebb, Phys. Rev. 149, 25 (1966).

[5]H. B. Bebb, Phys. Rev. 153, 23 (1967).

[6]W. Zernik, Phys. Rev. 135, A51 (1964).

[7]W. Zernik and R. W. Klopfenstein, J. Math. Phys. 6, 262 (1965).

[8]G. S. Voronov and N. B. Delone, Zh. Eksp. Teor. Fiz., Pis'ma Red. 1, 66 (1965) [JETP Lett. 1, 66 (1965)].

[9]J. L. Hall, E. J. Robinson, and L. M. Branscomb, Phys. Rev. Lett. 14, 1013 (1965).

[10]E. J. Robinson and S. Geltman, Phys. Rev. 153, 4 (1967).

[11]J. L. Hall, IEEE J. Quant. Electron. 2, 361 (1966).

[12]E. J. Robinson, unpublished. The method used in this calculation is similar to that reported by Robinson and Geltman in Ref. 10.

Promotion of L-Shell Electrons to Higher Bound States in Ar^+-Ar Collisions*

R. Del Boca and H. C. Hayden
Department of Physics, University of Connecticut, Storrs, Connecticut 06268

and

G. M. Thomson
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332
(Received 19 April 1971)

The $Ar^+ + Ar$ collision has been studied for evidence that an L-shell electron may be promoted to a higher *bound* level. An ion-ion coincidence apparatus was used on events where the incident energy T_0 was 25 keV and the scattering angle θ was 16°. We successfully sought those inner-shell vacancy-producing collisions in which no purely outer-shell autoionization processes took place and in which only one electron was ejected per vacancy. For such an event to occur the inner-shell electron must be promoted to a bound state. This effect can be accounted for using the promotion model of Fano and Lichten and provides additional evidence of the model's validity.

Several studies of both the inelastic energy loss[1-3] Q and the ejected-electron energy spectrum[4-6] have been done on the violent ion-atom collision $Ar^+ + Ar \to Ar^{+m} + Ar^{+n} + (m+n-1)e^-$, abbreviated (1 0, m n). The letters m and n represent the scattered and recoil charges, respectively. When the product of the incident energy T_0 and the scattering angle θ is near 400 keV deg

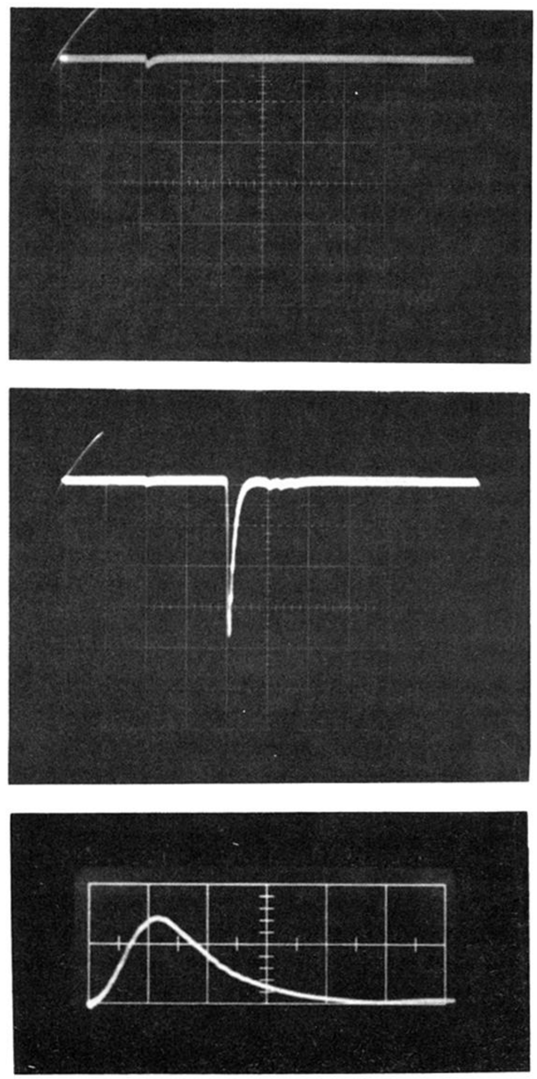
(a)
(b)
(c)

FIG. 1. (a) Multiplier output, atomic beam off. Scale: 20 mV/cm, 1 μsec/cm. (b) Photoelectric signal. Scale: 20 mV/cm, 1 μsec/cm. (c) Laser pulse. Scale: 50 MW/cm, 20 nsec/cm.