early work since the He-cell data showed large systematic shifts, then attributed to the relativistic shift in $g_S(e)$ due to heating of the electrons. After the subsequent work[9] on $g_J(^4\mathrm{He}, 2^3S_1)/g_J(^1H, 1^2S_{1/2})$ and that by Deloche *et al.*[10] on He afterglow, we believe this heating interpretation to be correct. No such effect was observed in the cells using tritium for the source of electrons. Furthermore, by the admitting of H_2 gas to one of the He discharge cells, the systematic shift in R was removed. This preliminary work is now in agreement with the result reported in this Letter, although none of the previous data are included in the present determination of R.

In conclusion, a new level of experimental precision has been attained for the ratio $g_J(\mathrm{H})/g_S(e)$. Agreement with theory is obtained which, for the first time, confirms mass-independent corrections to order α^3 as well as the leading Breit term.

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Collisional Line Broadening Using Laser Excitation and Ionization

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A laser excitation and ionization process is used to measure Cs-Ar interaction forces at long range. With energy densities of 1 J/cm^2, nonlinear excitation persists as far out as 70 Å at one atmosphere of Ar. This method provides extreme sensitivity (even single absorption events can be measured) which allows absolute measurements on the very far wing where absorption or fluorescence becomes vanishingly small.

We report on a photoionization method which allows absolute measurements of line broadening on the very far wing where absorption and fluorescence become extremely small.[1] The method involves the conversion of essentially every absorption event of a colliding system to an ion pair in a two-photon ionization process. Thus, we avoid measurements of small numbers of absorbed or emitted photons in favor of the much more sensitive detection of free electrons. Previously we used this scheme, "resonance ionization spectroscopy" (RIS), in measuring absolute populations and lifetimes of excited states[2,3] following charged-particle excitation of He gas. Moreover, we recently used the RIS method in achieving detection and identification of single atoms[4,5] by utilizing the two-photon ionization process in a proportional counter.

Previously, line broadening by foreign gases was studied by absorption[6] and fluorescence.[7] In these studies, high-temperature absorption tubes are required in order to induce appreciable absorption of fluorescence. Dense samples introduce not only self-broadening but also dimer absorption. Moreover, high-pressure buffer gases can make three-body collisions wash out the satellite structure otherwise observable in the two-body collisions. The extra sensitivity achieved in the ionization method, however, will allow measurements at the very far wing of optically thin samples where the higher density problems are eliminated, and this promises excellent resolution of the satellites.

The present studies are also of particular inter-

est to single-atom detection in gas proportional counters since noble gases are frequently used in the counting-gas mixtures.[3,4] Specifically, the broadening and shift of the atomic line by the foreign gas will reduce the selectivity through overlap of nearby levels of some other atomic species, especially where isotope discrimination is required.

In this experiment, we have studied the two-photon ionization of Cs-Ar mixtures with the broadened Cs($7p$) as an intermediate state. The two-photon ionization line shapes and thus the absorption line shapes were measured as a function of laser field intensity. From the absorption line profile, the form and coefficient of the interaction forces between the colliding atoms were found. Also, the present measurements yield photoionization cross sections of the $5d$ excited state of cesium. The study of the cesium $5d$ state is of interest in view of the theoretical prediction of nonsmooth wavelength dependence of the cross section.[8] Our measurement will be the first test of the theory. From this measurement we derive also the radiative decay rate from $7p$ to $5d$, using a rate-equation model of the two-photon ionization of Cs. Photoionization of the $5d$ state is appreciable for laser pulses on the order of 1-μsec duration; because of this, considerable error may have been made on some previous measurements of the $7p$ cross sections.[9]

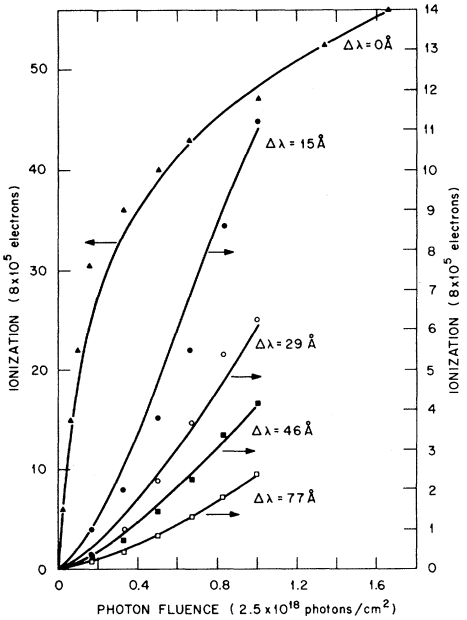

FIG. 1. Ionization signal as a function of photon fluence for the indicated detuning in argon gas at 760 Torr.

Pulses of 1 μsec and 3 cm^{-1} full width at half-maximum were used, and the ionization was collected with nickel-plated electrodes. See Ref. 4 for experimental details. The density of Cs ($\sim 10^9/cm^3$), governed by transport processes, was measured by saturating the two-photon ionization process. The beam profile was studied by using pinholes of various sizes and a photodiode, and was focused to the middle of the collection plate by a lens of 50-cm focal length.

Figure 1 shows the intensity dependence of the ionization yield at some representative detunings, and Fig. 2 shows the line shape of the two-photon ionization process. In the rest of the Letter we give an analysis of these experimental results.

Because the lifetimes of the $7p$ fine-structure states are about 100 nsec and the exciting pulse duration is about 1 μsec, it is necessary to consider in the analysis of the two-photon ionization process some low-lying levels that can be radiatively populated. In the case of Cs, all the lower excited states can be ionized by the exciting pulse at 455-nm wavelength. At 760 Torr of Ar the $7p$ fine-structure substates are completely mixed. This agrees with this measurement since we get the same two-photon ionization yield when the laser resonates with the isolated $7p_{1/2}$ or $7p_{3/2}$. Therefore, weighted averages of cross sections and decay rates are to be used or measured. The rate from $7p$ to $5d$ is 1.2×10^6 sec^{-1},[10] and a competing radiative channel is a decay through the $7s$ state with a rate equal to $\sim 4.5\times10^6$ sec^{-1}. Both the $5d$ and $7s$ levels decay to the ground state $6s$ via the $6p$ which decays to $6s$ very rapidly (3×10^7 sec^{-1}). The $7s$ state decays to the $6p$ state fairly rapidly ($\sim 10^7$ sec^{-1}), while the $5d$ state decays to it at about 10^6 sec^{-1}.

Since the cross section of $7s$ is calculated[11] to

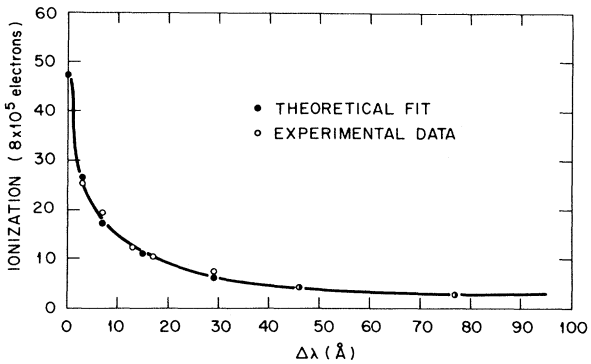

FIG. 2. Ionization signal as a function of detuning at a fixed photon fluence (2.5×10^{18} cm^2) for argon gas at 760 Torr.

be small ($< 10^{-19}$ cm^2) and its lifetime is only about 57 nsec, we predict no measurable contribution to the ionization yield from it. Although the estimated[11] $6p$ cross section is large (1.1×10^{-17} cm^2), its lifetime is only 30 nsec; thus, its contribution to the ionization will not be as large as that of the $5d$ state but will be comparable to that of $7p$. A simplified way of describing the combined yield from $7p$ and $6p$ is by using fast decay of the $7s$ and $6p$ states and comparing with the pulse length and decay rate of $7p$ in order to prove that for most of the pulse the decay of $6p$ is rate limited by the decay of $7p$. In this case, one can show that the two populations can be lumped together and their ionization described by an effective cross section $\bar{\sigma} = \sigma_{1I} + \gamma_{12}\sigma_{4I}/\gamma_{40}$, where γ_{12} is the decay rate from $7p$ to $7s$, γ_{40} is the inverse of the lifetime of $6p$, and σ_{4I} and σ_{1I} are the photoionization cross sections from $6p$ and $7p$, respectively. This form of $\bar{\sigma}$ fails at high power but only when the ionization of $7p$ is saturated and the variation is unimportant. Therefore, we describe the two-photon ionization process by a rate-equation formalism involving only three discrete levels: the $[7p, 6p]$, the $5d$, and the ground state (Fig. 3). At the far wing one can show either from statistical broadening arguments or from a dynamic treatment of the interaction[1,12] between the atoms that the cross section for photon absorption to the intermediate state with C_6/R^6 difference potential is $\sigma(\delta) = (\pi/6)N_F(C_6/\hbar)^{1/2}\lambda_0^2\gamma\delta^{-3/2}$, where N_F is the Ar density, λ_0 is the laser wavelength, γ is the spontaneous rate of decay from the excited state to the lower state, R is the separation between Cs and Ar atoms, and $\delta = C_6/\hbar R^6$.

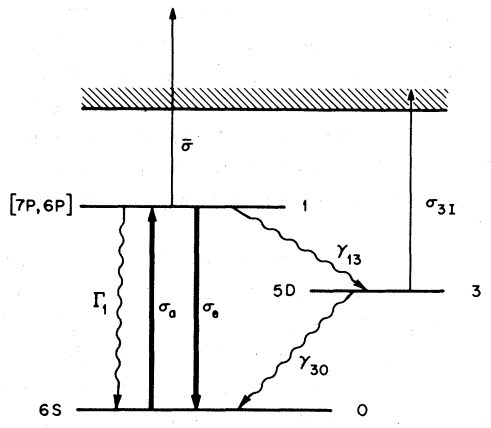

FIG. 3. A simplified model of the two-photon ionization process with effective decay rates and cross sections.

The solution of the rate-equation model is carried out at the line center, the very far wing, and the intermediate region separately. Taking a Gaussian pulse shape of waist r, we get at the line center (for photon fluence $> 5\times10^{16}$ cm^{-2})

$$\frac{n_i}{4\pi r^2 N} = e_{in}(\alpha y_0) - e^{\alpha}[e_{in}(\alpha y_0 + \alpha) - e_{in}(\alpha) - e_{in}(\beta x y_0 + \beta x) + e_{in}(\beta x)] - e^{-x}[e_{in}(\beta x y_0) - e_{in}(x y_0)], \quad (1)$$

where n_I is the number of electrons produced per unit beam length, N is the Cs density, $\beta = 1 + \sigma_{3I}/2\bar{\sigma}$, $\alpha = \sigma_{3I}/2\bar{\sigma}(\gamma_{13}\tau)$, $y_0 = \bar{\sigma}F_0/\gamma_{13}$, $x = \gamma_{13}\tau$, τ is the pulse duration, F_0 is the number of photons per unit area per second at the beam center, and $e_{in}(z) \equiv \int_0^z (dz'/z')(1 - e^{-z'})$. We note that the time dependence of the population of the $5d$ state due to the decay of $7p$ was dealt with by introducing the proper total number at the midpoint of the laser pulse instead of continuously as the decay actually occurs. Numerical calculations taking into account the details of the time dependence showed, for the present parameters, less than 1% effect at line center and even less on the far wing.

At the far wing we get the following power dependence:

$$\frac{n_I(\Delta\lambda, F_0, R)}{n_I(\Delta\lambda, F_m, R)} = \left(\frac{F_0}{F_m}\right)\frac{1 - (\Gamma_1/\bar{\sigma}F_0)\ln(1 + \bar{\sigma}F_0/\Gamma)}{1 - (\Gamma_1/\bar{\sigma}F_0)\ln(1 + \bar{\sigma}F_m/\Gamma)}, \quad (2)$$

where $\Gamma_1 = \Gamma - \gamma_{13}$; Γ is the inverse of the lifetime of the $7p$ level; $n_I(\Delta\lambda, F_0, R)$ is the ionization yield at a particular detuning $\Delta\lambda$ and photon flux F_0; $n_I(\Delta\lambda, F_m, R)$ is the ionization yield at detuning $\Delta\lambda$ and at a photon flux F_m. Note that the normalization in Eq. (2) gives the power dependence of the far wing. Equation (2) is valid for $\Delta\lambda \geq 70$ Å if $F_0 = 2.5\times10^{24}/(\text{cm}^2\ \text{sec})$, i.e., $\sigma(\Delta\lambda)F_0 < 0.2$ and $\tau F_0 > 10^{17}/\text{cm}^2$. The effect of the decay of the $5d$ (γ_{30}) on the far wing was also found to be negligible except at the lowest power levels.

In the region between the line center and 70 Å on the red wing and for low powers, analytical expressions were derived. However, for high powers, numerical work was necessary in order to include the approximately Gaussian decrease in photon flux as the distance from beam center is increased. In the model the simultaneous absorption of both fine-structure levels of $7p$ was

taken into account. This effect is minimal at the line centers of both; however, on the far wings, when both absorptions are weak, it is sizable.

Figure 1 shows the power dependence of the ionization yield at the line center and at 15-, 29-, 46-, and 77-Å detunings from the isolated atom line. The power dependence predicted by the above theoretical model is also shown in Fig. 1 as solid lines. Figure 2 shows the two-photon ionization line shape at 2.5×10^{18} photons/cm^2 power level along with the theoretical fit (solid line). Both Figs. 1 and 2 show good agreement with the theoretical model. The analysis yields the following quantities for the cross sections and the decay rates: $\bar{\sigma}=5\times10^{-18}$ cm^2, $\sigma_{3l}=4\times10^{-17}$ cm^2, $\gamma_{13}=8\times10^5$ sec^{-1}, and $\sigma(\Delta\lambda)=4\times10^{-17}/(\Delta\lambda)^{3/2}$ cm^2 where $\Delta\lambda$ is in angstroms. A single-particle, central-field approximation theory[8] gives 2.8×10^{-7} cm^2 for the $5d$ and 4×10^{-18} cm^2 for $7p$ which agrees with the experiment. Agreement between the measured line shape and the model implies an R^{-6} potential in agreement with the expected dependence.

In conclusion, we have shown that in studies of line broadening, the ionization channel provides an extremely sensitive and simple way of monitoring absorption. Even on the far wing where absorption of fluorescence becomes vanishingly small, the ionization method can be used because single absorption events are detectable in proportional counters.[4,5] Therefore, dense and high-pressure samples are not a requirement, thus allowing the possibility of complete resolution of satellite structure on the far wing. These studies also provide accurate measurement of the potential parameters as we discussed above.

The above study indicates that in some cases selectivity in single-atom detection is reduced because of pressure broadening. However, low-pressure proportional counters, or even channeltrons working in a vacuum, can be used where extreme selectivity is essential. Pressure broadening can be put to advantage in studies of rare events where there is a lack of accuracy on the energy levels. Such a case arises in searches for free quarks attached to a nucleus.

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