mann, C. Leroy, L. Palffy, A. Possoz, and A. Maio, Phys. Rev. Lett. 40, 302 (1978).

[8]K. Sugimoto, I. Tanihata, and J. Göring, Phys. Rev. Lett. 34, 1533 (1975); see also footnote 5 in Ref. 7.

[9]H. Brändle, L. Grenacs, J. Lang, L. Ph. Roesch, V. L. Telegdi, P. Truttmann, A. Weis, and A. Zehnder, Phys. Rev. Lett. 40, 306 (1978).

[10]K. Sugimoto, T. Minamisono, Y. Nojiri, and Y. Masuda, to be published.

[11]This well-known device [cf., e.g., E. Rutherford *et al.*, *Radiations from Radioactive Substances* (Cambridge Univ. Press, Cambridge, 1930)] was also used in Ref. 10.

[12]We used the same device in a repetition of the α-measurement (to be published).

[13]The main change was an additional thin counter in each telescope.

[14]Canberra Analog Stabilizer Model 1520.

[15]D. E. Alburger and A. M. Nathan, Phys. Rev. C 17, 280 (1978).

[16]Marita *et al.*, Ref. 1; see also W.-Y. P. Hwang and H. Primakoff, Phys. Rev. C 16, 397 (1977).

Observation of New Satellites in the Cs-Ar System Using Resonance Ionization Spectroscopy

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The absorption line shape of Cs-Ar system is recorded using two-photon ionization of the system with Cs($7P$) as an intermediate state. New satellite structures in the wings of Cs($7P$) are observed which were not resolved in previous absorption measurements. Also the absolute absorption cross section in the blue wing is measured.

Recently, we suggested[1] and reported[2] on a two-photon ionization method for collisional line broadening. The resonance ionization spectroscopy scheme (RIS) results in the conversion of all absorption events to ion pairs in a two-photon ionization process. Conversion of absorption events to ion pairs increases the sensitivity of the detection since it is easier to detect a small number of electrons than a small number of photons in absorption or fluorescence. The increased sensitivity achieved in the ionization method, over the traditional absorption[3] and fluorescence methods,[4] allows measurements at the far wing of optically thin samples and with low pressure of buffer gases where self-broadening, dimer absorption, and three-body collisions are essentially eliminated. The present Letter reports on the demonstration of such sensitivity and the potential of this method. We have resolved satellite structure in the wings of Cs($7P$) broadened by Ar which was not observed in the absorption measurements previously taken.[3] A recent calculation predicts the presence of the structure[5]; however, the positions do not agree completely with this experiment. Also the absolute absorption cross section in the blue wing is measured. The extra sensitivity achieved in this method also allowed the detection and identification of single atoms of Cs in the presence of other species.[6]

Collisional line broadening is traditionally studied by absorption[3] or fluorescence.[4] Recently a variation of the traditional fluorescence method was developed.[7] The system is excited in the wing, and the collisional redistribution of scattered light is studied as a function of the detuning of the incident light. Optical collision cross sections are then derived.

In the present measurements, a mixture of Cs and Ar at room temperature is photoionized by absorption of two photons from a 1-μsec pulse from a dye laser tuned to 455 nm. The collisionally broadened Cs($7P$) states serve as an intermediate state of the process. Low Cs density (10^9/cm^3) is used which is seven orders of magnitude less than what was used in the previous absorption studies. The details of the experimental setup were given in Ref. 6. The laser wavelength was tuned in 1–2-Å increments across both the $7P_{3/2}$ and $7P_{1/2}$ fine structure levels. At each setting, the dependence of the two-photon ionization yield on the pulse energy was recorded. The pulse magnitude ranged over two orders of magnitude (5×10^{16}–5×10^{18} photons/cm^2). At each pulse energy the two-photon ionization line shape can be easily plotted.

Figure 1 shows the combined line shape of both

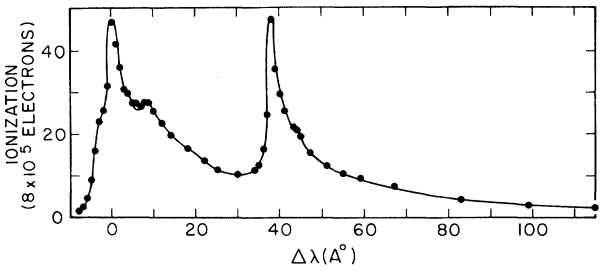

FIG. 1. Two-photon ionization line shape with $Cs(7P_{3/2})$ and $Cs(7P_{1/2})$ as intermediate states, at photon intensity 2.5×10^{18} photons/cm^2 and 1 atm of Ar buffer gas.

$7P_{3/2}$ and $7P_{1/2}$ levels for pulses of 2.5×10^{18} photons/cm^2. The line shape shows that the blue wing falls off more rapidly than the red wing which indicates that the interaction potential of the excited-state Cs* + Ar is more attractive than the ground-state Cs + Ar. When the excited-state potential is more attractive than the ground-state potential, then inelastic collisions occur at the red wing, and resonance absorption takes place which results in an extended red wing. For the same case, the contribution on the blue wing is only due to elastic dephasing collisions, and thus no extended wing is expected. Figure 1 shows that 10% absorption takes place even as far out as 80 Å from the line center at 1 atm of Ar buffer gas.

The line shape given in Fig. 1 indicates the presence of three satellites. One of these occurs at 19.6 cm^{-1} (4 Å) in the blue wing of $7P_{3/2}$. The second occurs at 41 cm^{-1} (8.6 Å) in the red wing of $7P_{3/2}$. The third occurs at 212.6 cm^{-1} from the unshifted $7P_{3/2}$ fine-structure level or 31.6 cm^{-1} (6.6 Å) in the red wing of $7P_{1/2}$. In previous absorption measurements[3] the red satellite of $7P_{3/2}$ was partially resolved and it agrees with the position measured in this experiment. However, the other two satellites were not observed in those absorption measurements. Recently potential curves of those high excited states of Cs perturbed by Ar, Xe, and He were calculated by Cuvellier *et al.*[5] They pointed out the importance of coupling between neighboring levels in such calculations. The coupling with neighboring levels gives rise to structure in the potential energy curves. The experimental study of inelastic collisional process involving Cs(7*P*) and Cs(6*D*) and the ground state of rare-gas atoms at thermal energy leads to disagreement with the calculations based on potential curves derived without coupling between 7*P* and neighboring states of Cs.[5] However, satisfactory explanation of the main features of the transfer between 7*P* and 6*D* of Cs is obtained based on the modified potential curves.[5] Also, the importance of the coupling with the neighboring levels has been shown experimentally by Gallagher with Tl–noble-gas systems.[8]

TABLE I. Experimental and calculated values, in units of cm^{-1}, of the positions of the satellites in the difference potential of Cs(7*P*) + Ar and Cs(6*S*) + Ar.

Calculation [a]	Experiment [b]	Experiment [c]
−23.9	−19.6	
0 ($7P_{3/2}$)	0	0
6.6	41	41
150.0		
179		
181 ($7P_{1/2}$)	181	181
186		
197.9	212.6	

[a] Results from Ref. 5.
[b] From present experiment.
[c] From Ref. 3.

Because our present measurement gives the direct absorption line shape of the broadened excited states, it can serve as a direct test of the importance of such coupling between neighboring excited states. Our resolution of the three satellites, in fact, shows that such coupling may be necessary to understand the measurements. In Table I, we compare the predicted positions of the structure by the above calculation and our present measurement. Although the calculation predicts structure, the agreement with the experiment is not complete. The calculation seems to be in fair agreement with the measured blue-wing satellite of $7P_{3/2}$. The calculation was also carried out for Xe and He perturbing gases, and it was found that the importance of such coupling depends on the perturber as well as on the energy separation. The most pronounced structure is predicted to be in the case of Cs-Xe. The effect in Cs-Ar is found to be more pronounced than the case of Cs-He. More measurements are to be taken with the present method in order to test the calculation over a wide range of size of the rare-gas atoms.

The cross section of absorption on the blue wing derived from the blue-wing data of $7P_{3/2}$ will not obey a power law because of the presence of the satellite on its blue wing caused by the coupling to the other states. To see this, we plot in Fig.

2 the square of the detuning on the blue wing of $7P_{3/2}$ versus the $\frac{3}{2}$ power of the corresponding detuning on the red wing of $7P_{1/2}$ which gives the same ionization yield. Note that far on the wing, the relation is linear. Deviation from the linear dependence indicates the deviation from the power laws. This also indicates the presence of satellites. From the slope of the linear portion, and using the measured cross section in the red wing $[4\times 10^{-17}(\Delta\lambda)^{-3/2}]$ from Ref. 2, we find that the far blue-wing absorption cross section is $\sigma_B = 4\times 10^{-18}/(\Delta\lambda)^2$ where σ_B is in square centimeters and $\Delta\lambda$ is in angstroms. The intercept of the linear portion yields an estimate of the position of the satellite mentioned above, namely 4 Å.

Since the lifetime of Cs($7P$) is of the order 100 ns and the excitation duration is 1 μsec, then appreciable population by radiative and collision-induced fluorescence processes of $5D$, $6P$, and $7S$ states takes place during the pulse duration. Moreover the photons at 455 nm are energetic enough to ionize all the Cs excited states. Therefore appreciable ionization may be expected from the lower excited states. Figure 3(a) shows the line-center ionization yield from both levels, while Fig. 3(b) shows an enlargement of the low-intensity parts of the curves. The curves show that for weak excitation ($< 10^{17}$ photons/cm^2), the ionization from the $7P_{3/2}$ rises more rapidly, but for intensities $> 10^{17}$ photons/cm^2, the ionization yield is identical. The normalized difference between the two-photon ionization yields from the line peaks of $7P_{3/2}$ and $7P_{1/2}$, $D = (I_{3/2} - I_{1/2})/I_{3/2}$, is measured. The difference D is constant for powers less than 5×10^{16} photons/cm^2. For such powers, the populations of $7P_{3/2}$ and $7P_{1/2}$ are found to be proportional to their statistical weights. For powers greater than 5×10^{16}, D starts decreasing and asymptotically reaches zero for power on the order of 10^{17} photons/cm^2. Note that the line-center ionization rises very quickly. On the order of 50% of the ionization is produced by the first 6% of the total power. The enlargement of the first few percent of the intensity dependence [Fig. 3(b)] shows a plateau, which is an indication of a saturation phenomenon with an associated cross section of the order of 5×10^{-17} cm^2. The second plateau occurs at powers of the order of 2×10^{18} photons/cm^2 which is associated with a cross section of the order of 3×10^{-18} cm^2. The large cross section is associated with ionization from the $5D$ state which is radiatively populated from the directly excited $7P$ state. The smaller cross section is associated with ionization from the $7P$. Radiative population of the $5D$ is appreciable for pulses of the order of 1-μs width, and therefore previous measurements of the photoionization of the $7P$ with pulses of the order 0.5 μs may have been in error, since they were analyzed with one ionization channel.[9]

In conclusion, the two-photon ionization process provides a sensitive method for monitoring the broadening process. The extra sensitivity achieved with this method allowed the study of

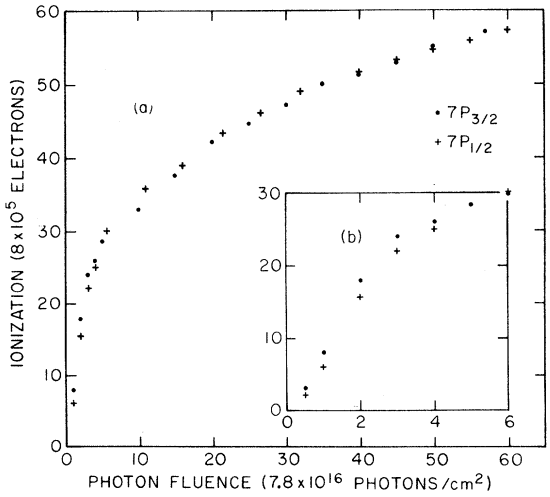

FIG. 2. (a) Ionization yield as a function of photon fluence when the laser is resonant with the line center of both Cs($7P_{3/2}$) and Cs($7P_{1/2}$). (b) Enlargement of low-intensity part of (a). Ar pressure is 1 atm.

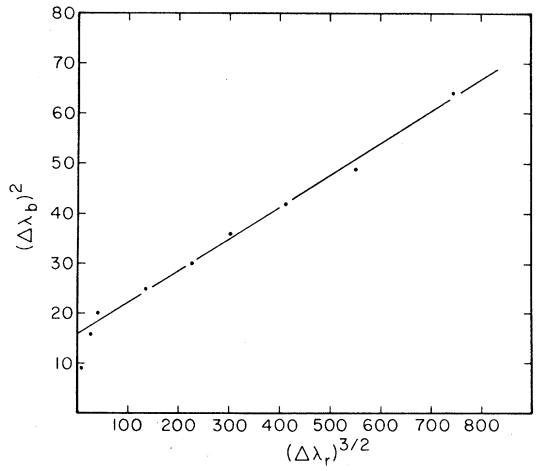

FIG. 3. The square of detuning on the blue wing of $7P_{3/2}$ ($\Delta\lambda_b$) vs the power $\frac{3}{2}$ of the corresponding detuning on the red wing of $7P_{1/2}$ ($\Delta\lambda_r$) which gives the same ionization yield. Both $\Delta\lambda_b$ and $\Delta\lambda_r$ are in units of angstroms.

optically thin samples and resolution of satellite structure which was not previously resolved in straight absorption measurements of thicker samples. The agreement with the theoretical prediction is marginal and more measurements need to be taken with other dephasing atoms in order to make more stringent test of the calculations. The absolute absorption cross section on the wing due to dephasing and inelastic collisions was measured.

The present method yields photoionization cross sections of short-lived excited states. The saturated ionization is necessary in measurements of the photoionization cross sections of short-lived excited states, because, in the linear regime, big errors can be introduced due to the radiative decay to the lower excited states.

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[1]M. H. Nayfeh, Phys. Rev. A 16, 927 (1977).

[2]M. H. Nayfeh, G. S. Hurst, M. G. Payne, and J. P. Young, Phys. Rev. Lett. 39, 604 (1977).

[3]C. L. Chen and A. V. Phelps, Phys. Rev. A 7, 470 (1973).

[4]R. E. M. Hedges, D. L. Drummond, and A. Gallagher, Phys. Rev. A 6, 1519 (1972); F. H. Mies, J. Chem. Phys. 48, 482 (1968); J. M. Farr and W. R. Hindmarsh, Phys. Lett. 27A, 512 (1968).

[5]J. Cuvellier, P. R. Fournier, F. Gourand, J. Pascale, and J. Berlande, Phys. Rev. A 11, 846 (1975); J. Pascale and J. Vandeplanque, J. Chem. Phys. 60, 2278 (1974), and unpublished.

[6]G. S. Hurst, M. H. Mayfeh, and J. P. Young, Appl. Phys. Lett. 30, 229 (1977), and Phys. Rev. A 15, 2283 (1977); G. S. Hurst, M. H. Nayfeh, J. P. Young, M. G. Payne, and L. W. Grossman, in *Springer Series in Optical Science*, edited by J. L. Hall and J. L. Carlsten (Springer-Verlag, New York, 1977), Vol. 7.

[7]J. L. Carlsten and A. Szöke, Phys. Rev. Lett. 36, 667 (1976), and J. Phys. B 9, L231 (1976); M. G. Raymer and J. L. Carlsten, Phys. Rev. Lett. 39, 1326 (1977); J. L. Carlsten, A. Szöke, and M. G. Raymer, Phys. Rev. A 15, 1029 (1977).

[8]B. Cheron, R. Scheps, and A. Gallagher, J. Chem. Phys. 65, 326 (1976).

[9]H. D. Zeman, in *International Symposium on Electron and Photon Interactions with Atoms*, edited by H. Kleinpoppen and M. R. C. McDowell (Plenum, New York, 1974), pp. 581–594.

Observation and Relaxation of the Two-Photon Echo in Na Vapor

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We have used a pair of simultaneously pumped dye lasers to produce in Na vapor the sum-frequency two-photon analog of the photon echo. Quantum-beat effects were observed. Relaxation of the $3^2S_{1/2}$-$4^2D_{3/2}$ superposition was measured.

We report the first observation of an optical two-photon echo and its application to the study of relaxation of the $3^2S_{1/2}$-$4^2D_{3/2}$ superposition state in atomic Na vapor. Although optical echo effects in two-photon transitions were discussed as early as 1968[1] and have been described by many authors,[2-5] it was only recently that echo experiments with two-photon character were reported. The first, a nuclear-spin-resonance experiment, was performed by Hatanaka and Hashi,[6] who, working on the ~10-MHz split ground-state levels of ^{27}Al in Al_2O_3, demonstrated the local rephasing character of the two-photon echo. Shortly thereafter Hu, Geschwind, and Jedju[7] used optical-frequency laser fields to create and probe via the two-photon "Raman" echo the coherence of the ~30-GHz magnetic-field-split donor-electron spin levels in a crystal of *n*-type CdS. In the present work, *all* transitions are in the optical regime. Utilizing the effective elimination of Doppler broadening, which is inherent in echo phenomena,[8,9] we have examined the foreign-gas-