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Observation of collisional narrowing in a two-photon transition in a three-level system

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We report the observation of collision-induced narrowing in the two-photon 6*S*-12*S* transition in Cs perturbed by argon using resonance ionization spectroscopy. The narrowing occurs at 25–50-Torr Ar buffer gas pressure. It is explained via the competition between collision-induced single- and two-photon processes.

Collisional effects in multiphoton processes in the nonimpact regime have only recently become a subject of theoretical and experimental work.[1–5] This is to be contrasted to the fact that collisional effects in two-photon transitions in three-level systems in the impact regime have received extensive studies. Recent activity in the nonimpact regime has included theoretical analysis of collisionally aided radiative excitation in a three-level system in the weak-field limit.[1] It was found that collision-induced coherent phase-interference effects can give rise to oscillatory structure in the total absorption cross section as a function of the relative speed. The effect of a nonresonant collisional state in two-photon transitions in a three-level system was also theoretically analyzed.[2] It was found that the intermediate state causes a shift in the two-photon transition and adds a component which falls off as $(\Delta\lambda)^{-1/2}$ to the dominant line shape which falls off as $(\Delta\lambda)^{-3/2}$.

Two-photon laser-induced radiative collisions were recently analyzed.[3] Both the two-photon–one-collision and the two-photon–two-collision cases were treated. In the latter case a new intensity-induced collisional shift was found which makes the two-photon line shape highly sensitive to the intensity and might make the line shape symmetric.

Experimentally, collisional effects in two-photon transitions in a three-level system have recently received attention too. Collision-induced two-photon excitation of *S*-*P* transitions was observed.[4] Two-photon laser-induced collisional transfer between Ba and Tl atoms was also observed recently.[5]

In this paper we report the observation of collisional narrowing of the *S*-*S* two-photon transition of Cs perturbed by Ar. We believe this effect is due to competition between collision-induced single- and two-photon processes. The detection scheme we used in this experiment, resonance ionization spectroscopy, is similar to the scheme we used in previous measurements.[6,7]

The second harmonic of an Nd-YAG (yttrium aluminum garnet) pulsed laser at 532 nm is used to pump a dye laser amplifier system. The dye laser pulses (0.1-Å bandwidth and 10-ns duration) are focused into an ionization chamber equipped with plane electrodes for charge detection. Both the ionization signal and the intensity of the radiation for each pulse are stored in a two-dimensional analyzer.

Figure 1 shows the line shape of the process showing the presence of a blue satellite at $\Delta\lambda = -1.5$ Å. Figure 2 shows the dependence of the width of the

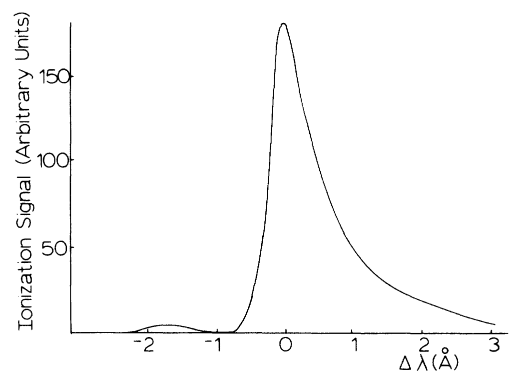

FIG. 1. Line shape of the two-photon 6*S*-12*S* taken at Ar buffer gas pressure 1.5 Torr, and at photon flux $4\times10^{18}/\mathrm{cm}^2$.

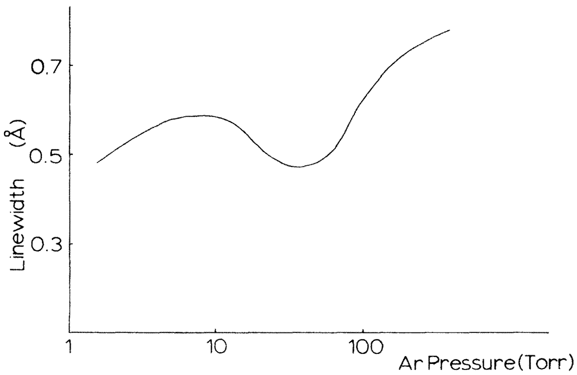

FIG. 2. Dependence of the width of the two-photon line shape on Ar pressure.

resonance on the Ar buffer gas pressure. It indicates that the width narrows in the 25–50-Torr region, then broadens again.

At the line center the excitation of 12*S* can proceed via the atomic intermediate state 6*P* and 7*P* as one channel (see Fig. 3). The rate of an atomic two-photon excitation, *R*, from an initial state 0 to a final state *f* by a light source of one mode of frequency ω is calculated using second-order perturbation theory. For the present case the electric dipole oscillator strengths of all 6*S*-6*P* and 6*S*-7*P* are ~ 0.77 and $\sim 1.3\times10^{-2}$, respectively. The electric dipole oscillator strenghts of all 6*P*-12*S*, 7*P*-12*S*, and all the other allowed transitions from 12*S* to the lower levels were calculated using the Hartree-Fock approximation.[8] The results are $\sim \frac{1}{3}\times10^{-3}$, 3×10^{-3}, 6.8×10^{-3}, 1.7×10^{-2}, 5×10^{-2}, and 0.5 for 12*S*-6*P*, 7*P*, 8*P*, 9*P*, 10*P*, and 11*P*, respectively. These rates give 0.6 μs for the lifetime of the 12*S*. Moreover, at the peak of the transition, that is at $2\hbar\omega=(E_f-E_0)$, the cross section for two-photon excitation to the 12*S* is calculated to be 5.9×10^{-42} cm^{4}s.

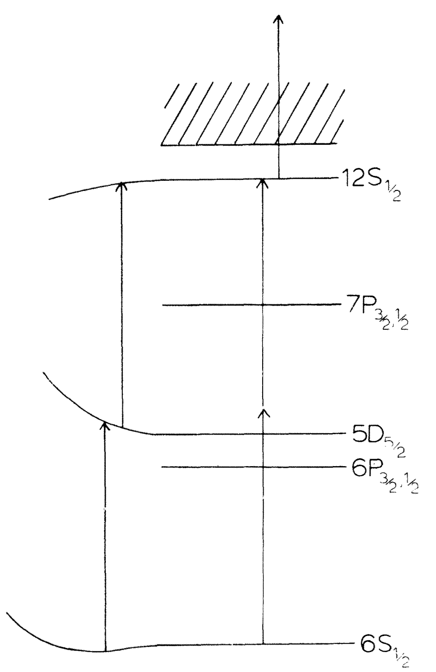

FIG. 3. Partial energy-level diagram of the states that may contribute to the two-photon process.

Another process that can take place at the line center or at the wing of the lines involves collisional states. In the presence of the Ar buffer gas the atomic levels are shifted as a function of the interatomic separation. Moreover, the perturbations of the wing of forbidden lines caused by collisions mix the wave functions and hence induce dipole oscillator strengths. In the present process mixing in the wing of the electric quadrupole transition 6*S*-5*D* is important. Calculations of the adiabatic potential curves of the Cs-Ar system indicate that the potential of Cs(5*D* $\frac{5}{2}$)-Ar($m=\frac{1}{2}$) state is repulsive at large internuclear separation, while the states Cs(5*D* $\frac{5}{2}$)-Ar($m=\frac{5}{2},\frac{3}{2}$) and Cs(5*D* $\frac{3}{2}$)-Ar($m=\frac{3}{2},\frac{1}{2}$) are attractive at large internuclear separation and repulsive only at small internuclear separations.[9] Contrary to the fact that these calculations of the Cs–rare-gas system are widely believed to be inaccurate, recent experiments have shown that they are more accurate than is widely known.[4] Since the 5*D* state is less than halfway to the 12*S*, the Cs(5*D* $\frac{5}{2}$)-Ar($m=\frac{1}{2}$) can provide an intermediate state in the form of an induced crossing at internuclear separation equal to 10.5 a.u. Similarly, mixing in the wing of the dipole forbidden transition 5*D*-12*S* causes an induced dipole oscillator strength which results in absorption at finite internuclear separations.

From our previous experiments, the absorption cross section due to the induced oscillator strength at the first induced crossing between 6*S*-5*D* is measured to be 3×10^{-20} cm^2 at 100 Torr of Ar buffer gas pressure.[4] The adiabatic potential of Cs(12*S*)-Ar has not been calculated and there is no calculation of the induced oscillator strength. However, we expect the induced oscillator strength to be no larger than 10^{-2}, which is the oscillator strength of the allowed transition 5*D*-11*P*. In a previous measurement the induced oscillator strength in the wing of 6*S*-7*S* was found to be 10^{-2}, which is as large as the oscillator strength to the nearby allowed transition 7*P*.[10]

The absorption rate, $K(\lambda)$, is given by the following quasistatic formula[11]:

$$\frac{K(\lambda)}{NN_f}=\frac{\lambda^4}{2c}g_u/g_l A R^2\frac{dR}{d\lambda}\exp(-\Delta V_l/kT) \ ,$$

where *A* is the transition probability, ΔV_l is the energy of the lower state at R with respect to the infinite internuclear separation, g_μ and g_l are the statistical weights of the upper and lower state, respectively, N and N_f are the densities of the absorbing and the perturber atoms, respectively, R is the internuclear separation at which the absorption takes place, $\lambda = hc(V_u - V_l)^{-1}$ is the absorption wavelength, and V_u and V_l are the energies of the upper and lower states. Since at 10.5 a.u., the Cs(6S)-Ar potential with respect to the infinite internuclear separation ΔV_l is -44 cm^{-1}, then the Boltzmann factor at room temperature is 1.2. The corresponding Boltzmann factor of the second transition is ~ 0.5 since the energy of Cs(5D)-Ar at 10.5 a.u. with respect to the infinite internuclear separation energy is 164 cm^{-1}. The slopes $dR/d\lambda$ at the induced crossings at the first and at the second transitions are almost equal since the potential of Cs(6S)-Ar is almost flat, and Cs(12S)-Ar is expected to have a small slope at large internuclear separations. Hence the absorption coefficient at the second transition is of the same order but perhaps smaller than the absorption coefficient at the first transition.

The excitation in the wing of the line can also proceed in the presence of collisions via the nonresonant collisional state Cs(6P)-Ar. The result indicates that the two-photon absorption mainly depends on the slope of the difference potentials of the ground state Cs(6S)-Ar and the final state Cs(12S)-Ar at the two-photon induced crossing. This slope $dR/d\lambda$ is expected to be large compared to the slope at the single-photon crossings discussed above because both states have flat potentials at large internuclear separations. Moreover, the Boltzmann factor in this case is nearly unity. Also the absorption probability is proportional to the two-photon coupling A_1A_2/Δ where A_1 and A_2 are the transition probabilities of the transitions between Cs(6S)-Ar and Cs(6P)-Ar and between Cs(6P)-Ar and Cs(12S)-Ar, and Δ is the detuning from Cs(6P)-Ar.

The process at the wing is then described in terms of a rate equation formalism of three closely coupled states: the ground state 6S, the intermediate collisional state 5D, and the final state 12S with populations n_0, n_1, and n_2, respectively. The populations n_0 and n_1, and n_1 and n_2 are coupled via the single-photon cross sections σ_1 and σ_2, respectively, with rates $\sigma_1 I$ and $\sigma_2 I$ where $\sigma_i = N_f\alpha_i(\Delta\lambda)$ and α_i is a function of the detuning of the difference interatomic potential. The populations n_0 and n_2 are coupled via the two-photon cross section $\sigma = N_f\alpha(\Delta\lambda)$ with a corresponding excitation rate σI^2. At the line center, the two-photon collision-induced cross section σ is replaced by the isolated atomic two-photon cross section σ_0.

At a given intensity of radiation the population of the 12S in the wing at low pressures proceeds mainly through the direct two-photon excitation. As the pressure rises, this rate rises linearly. At some intermediate pressure, the crossing at the one-photon process in the wing of Cs(6S)-Cs(5D) becomes important, and hence competes with direct excitation. Until excitation from Cs(5D)-Ar to Cs(12S)-Ar via the second crossing becomes appreciable, the rate to Cs(12S)-Ar will first slow down and then drop. When excitation at the second crossing becomes appreciable, then the overall rate to 12S in the wing will start rising with pressure. This yield at the line center, however, is independent of pressure at low pressure, starts falling as the excitation via the first crossing becomes important, and finally starts rising as the excitation via the second crossing becomes appreciable. Since the line center pressure dependence starts out constant whereas the wing dependence starts our linear, we expect the line center yield to go through a minimum at a lower pressure then the wing yield. Because of this the normalized line-shape width goes through a minimum.

We studied the pressure dependence of the satellite observed in the blue wing of the two-photon line (Fig. 4). As the Ar pressure increases from 1.5 to 50

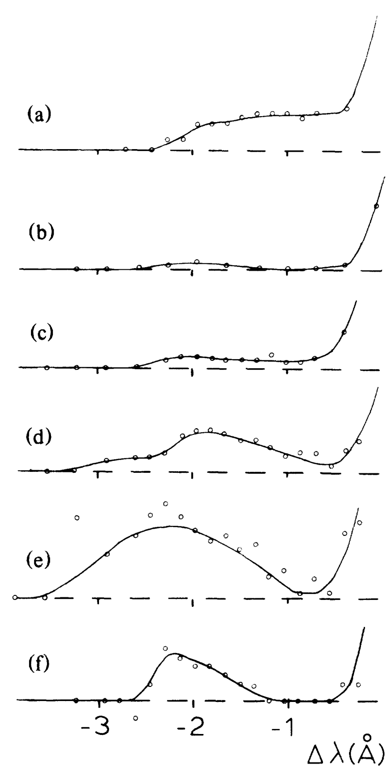

FIG. 4. Absolute line shape of the resolved satellite at photon flux 4×10^{18}/cm^2. (a), (b), (c), (d), (e), and (f) are taken at 100, 50, 25, 10, 5, and 1.5 Torr, respectively. The peak of the quadrupole transition is at $\Delta\lambda = 0$.

Torr the satellite grows and broadens, then it diminishes and narrows. As the pressure increases further, it appears in the 100-Torr range with near-flat shape.

To further substantiate these results we examined the transitions $6S$-$10S$ and $6S$-$13S$. In the latter the resonance occurs at $\lambda = 6658$ Å as compared to 6740 Å for the $6S$-$12S$ transition. Since this wavelength is shorter than the atomic resonance wavelength of the $5D_{5/2}$ state, the narrowing phenomenon is expected to play a role in this two-photon transition. Because of the shorter wavelength needed for resonating with $13S$, the single-photon collisional effect occurs at smaller internuclear separations than those of the $12S$. Because of the fact that the one-photon collision-induced cross section decreases as the internuclear separation at which resonance occurs decreases, we expect the pressure dependences of these two transitions to differ. In fact, we expect the phenomenon in the case of $6S$-$13S$ to occur at higher pressure than for the $6S$-$12S$ case. In the case of the $6S$-$10S$ transition, the resonance occurs at $\lambda = 7067$ Å. The radiation at this wavelength has photon energy equal to 1.76 eV which is less than the excitation energy of the $5D$ states. Moreover, the photon energy is sufficiently low such that single-photon crossing at the intermediate state cannot occur. As a result we expect the narrowing not to occur for this transition.

We made pressure studies of the $6S$-$13S$ transition over the range 0–150 Torr. The line shape was found to exhibit the narrowing phenomenon. The linewidth rose in the pressure range 0–50 Torr, and then decreased in the range 50–100 Torr. Beyond 100 Torr it started rising again. The minimum in the width occurred at 100 Torr. This is to be contrasted with the results of the $6S$-$12S$ transition where the minimum occurred at 55 Torr. We also took similar data for the transition $6S$-$10S$ over the pressure range 0–200 Torr and found that the line shape continued to widen monotonically. These results are in agreement with the above expectations and thus support the results of the study of the $6S$-$12S$ transition.

Finally, we emphasize that the final state in the present experiment is only $12S$ with no contribution from the nearby states such as $10D$ and $8F$. Our excitation of the $10D$ from the ground state using a two-photon process showed that its width at 760 Torr of Ar is 8 cm^{-1}. With 193-cm^{-1} separation between $10D$ and $12S$ we project negligible contribution from $10D$ in the pressure range used in the present experiment.

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