ac Stark Splitting in Resonant Multiphoton Ionization with Broadband Lasers

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An intense laser and a probe laser in near resonance with the sodium $3S_{1/2}\rightarrow 3P_{1/2}$ and $3P_{1/2}\rightarrow 4D_{3/2}$ transitions, respectively, produce a three-photon ionization current used to study the broadening and Rabi splitting of the $3P_{1/2}$ intermediate state as a function of probe-laser tuning. We find a narrow, intense-laser detuning region where the peak-heights ratio of the Rabi-split $3P_{1/2}$ doublet is reversed from that predicted for a monochromatic laser field. This is attributed to the finite (~ GHz) laser linewidth.

In view of the recent interest in various aspects of ac Stark splitting under strong laser fields,[1-7] this paper reports experimental results and their interpretation showing the effect of a finite laser bandwidth. Although some theoretical aspects of the effect of the bandwidth have been discussed in the literature[1-5]—especially in the context of resonance fluorescence—this seems to be its first experimental demonstration. The present experiment involves the observation of ac Stark splitting in double optical resonance in atomic Na vapor. Double optical resonance in alkali atoms has been studied by a number of authors who have predicted[8-13] or observed[14-16] effects that are optical analogs of effects first observed by Autler and Townes[17] in the microwave region.

In our work, the lower ($3S_{1/2}\rightarrow 3P_{1/2}$) transition in atomic sodium is driven by an intense near-resonant tunable laser field and the upper ($3P_{1/2}\rightarrow 4D_{3/2}$) transition by a nonsaturating laser field, also tunable. Under these circumstances, one can treat the interaction of the intense field with the two lower levels by two-level theory, which predicts[12] the splitting of each of the two levels into two components separated by $\Omega_{Ra}=[\Delta_a^2+(\mu E_a/\hbar)^2]^{1/2}$, where μ is the dipole matrix element connecting the two lower levels, E_a is the electric field of the intense laser, and Δ_a is the detuning of the laser frequency from the unperturbed transition frequency. The quantity Ω_{Ra} is the Rabi nutation frequency while $\omega_{Ra}=\mu E_a/\hbar$ is referred to as the on-resonance Rabi frequency. This model, in which the laser is assumed to be monochromatic, predicts that the center of each doublet will be pulled toward the detuned laser line by an amount $\Delta_a/2$. Scans of the weak laser serve as probes of this doublet structure of the intermediate ($3P$) state. The excitation of atoms into the upper ($4D_{3/2}$) level leads to observation of this doublet structure, either in the ionization from the $4D_{3/2}$ state by the intense laser, or in the optical fluorescence from this upper level. At zero detuning ($\Delta_a=0$) of the intense laser, the two peaks of the doublet are expected to be of equal height, while for $\Delta_a\neq 0$ the heights would be unequal (asymmetric peaks).

Whitley and Stroud[12] point out that, for detuning to frequencies lower than the unperturbed transition ($\Delta_a<0$), the upper component of the split intermediate state contains more of the unperturbed intermediate state ($3P$), while the lower component contains more of the ground state ($3S$). For large detuning, the dipole strengths for the probe transitions from the two components are not equal and strongly favor the upper component. However, the larger population of the lower component more than compensates, thus causing a pronounced asymmetry in favor of the lower component. Another way to understand this asymmetry qualitatively is to note that in the limit of large detuning, one expects the two-peak structure to reduce to a single peak representing a two-photon transition from the ground to the upper ($4D_{3/2}$) state. Energy conservation thus requires that the higher peak occur at $\Delta_b>0$ when $\Delta_a<0$, and vice versa, where Δ_b is the detuning of the probe laser. The asymmetry as described above is expected to occur when both the intense laser and probe laser are monochromatic. Hereafter, we refer to this as normal asymmetry.

These predictions are in general accord with observations of several authors, notably those of Bjorkholm and Liao,[16] who obtained quantitatively consistent splittings of approximately 1 or 2 GHz, in fields of about 750 W/cm^2 from a linearly polarized single-mode cw laser tuned within ± 2 GHz of the unperturbed $3S_{1/2} \rightarrow 3P_{1/2}$ transition. Picqué and Pinard,[15] on the other hand, using the $3S_{1/2} \rightarrow 3P_{3/2}$ transition, have reported observations that are still not completely understood.[18]

The three-level system of the present ionization experiment is the same system studied in fluorescence by Bjorkholm and Liao. However, we have worked at larger Rabi frequencies and at laser linewidths larger by more than an order of magnitude. Our laser linewidths, $\Delta\nu \sim 1$ GHz full width at half-maximum (FWHM) (~ 0.03 cm^{-1}), were thus large compared to the hyperfine separations of the intermediate state but the same order as that of the ground state (1.772 GHz). We used two synchronously fired flashlamp-pumped tunable dye lasers with 130-cm cavities. The collinear laser beams, linearly polarized along the same transverse axis, were focused at right angles into a collimated sodium beam. The sodium beam flux was about 10^{13} atoms/cm^2 sec with a 0.02-rad full width and a density of about 10^9 atoms/cm^3. Collisional transfer and radiative trapping effects could be ignored. The transverse velocity corresponded to a Doppler width of about 10 MHz. The duration of the laser pulses was about 700 nsec with a rise time of about 75 nsec. A full account of this and related experiments is in preparation.[19] The apparatus has been described in more detail by Moody and Lambropoulos.[20] The asymmetry reported in that paper, measured over a rather narrow range of Δ_a, was the reverse of what we called normal asymmetry above, thus raising a question as to the applicability of the three-level–system theory with monochromatic light.

The present work greatly extends the detuning range, confirming the reversal of the asymmetry near zero detuning, but showing that for large detuning it reverts to normal. Typical results are shown in Fig. 1 where ion current is plotted against probe-laser frequency. The change of the asymmetry from reversed to normal—with increased detuning—is clearly indicated in this figure which shows results for zero detuning and for several values of negative detuning. A similar pattern is observed for positive detunings as indicated in Fig. 2. For small detunings the behavior of the peak asymmetry is contrary to predictions of three-level theory with monochromatic fields.

This behavior can be explained theoretically if one considers the finite bandwidth of the intense laser. The complete analysis will be published elsewhere.[21] It is based on methods which, most recently, have been employed in the treatment of resonance fluorescence[1-5] as well as multiphoton ionization.[22,23] References to earlier work, especially as it pertains to three-level systems, can be found in Ref. 21. The reversed asymmetry, however, can be understood qualitatively in terms of the picture used earlier for monochromatic fields. Consider negative detunings for the moment and recall that the upper component of the split intermediate state ($3P_{1/2}$) contains more of this state than of the ground state. If the intense laser line is broad (com-

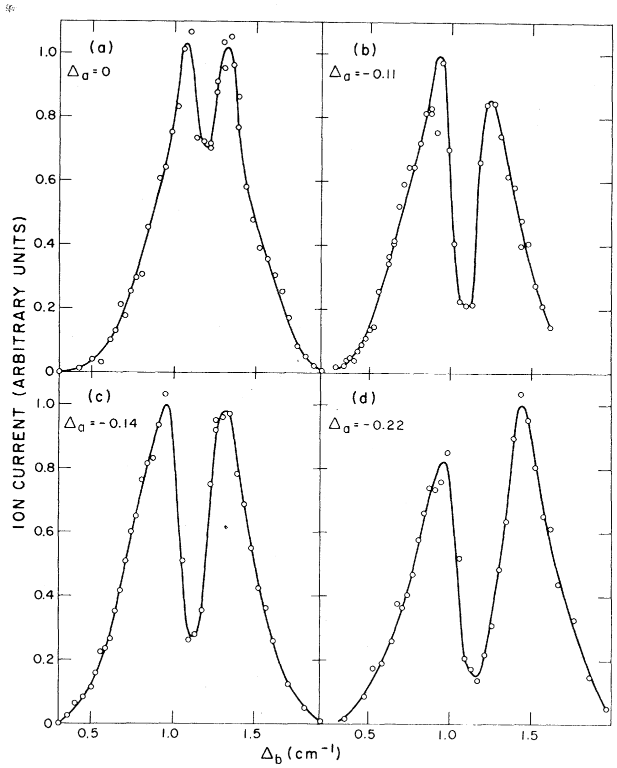

FIG. 1. Relative ionization current (Na^+) is plotted against frequency scan of the probe laser (a) for $\Delta_a = 0$, and (b), (c), (d) for three progressively greater negative detunings of the intense laser. Part (b) represents detuning in the region of reversed asymmetry, part (c) the point of finite detuning at which symmetry is again realized, and part (d) the region of normal asymmetry which persists for larger detunings. The abscissa represents displacement of the probe-laser line center from that corresponding to normal incidence on the 1-mm solid tuning etalon. Laser power densities are ~ 2 MW/cm^2 and $\sim 10^2$ W/cm^2. Each point represents the average ion yield of ten laser pulses

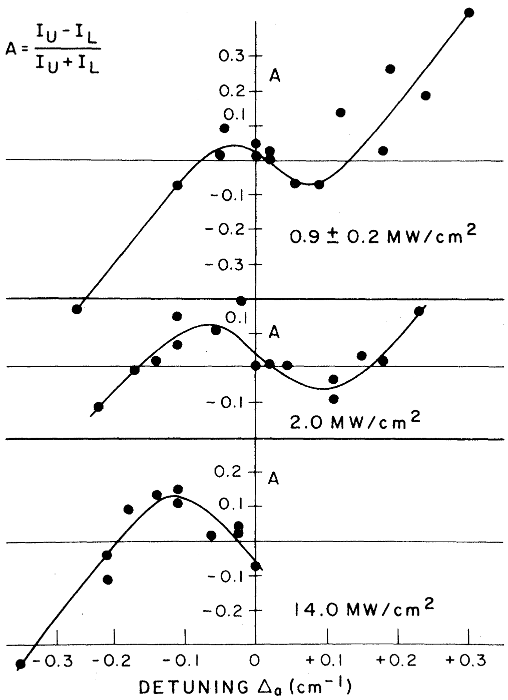

FIG. 2. Observed asymmetry (A) of the ac-Stark–split doublet is shown as a function of the intense-laser detuning for the three different power densities. Here A is defined in terms of peak intensities I_U and I_L corresponding to upper and lower components of the doublet.

pared to the natural width of the intermediate state) and its wing overlaps the intermediate state, it excites more atoms into that state than would a monochromatic laser. As a result, the population of the upper component is larger than it would be with a monochromatic laser, while that of the lower component is smaller. This causes the reversal of the asymmetry. When the laser is detuned further, so that the wing does not overlap with the intermediate level, the asymmetry will revert to normal, since the laser would appear relatively monochromatic to the atom (an important exception to this is noted below). For zero detunings the peaks would be symmetric.

On the basis of this picture, one would expect the behavior shown in Figs. 1 and 2. The range of reversed asymmetry will depend on the laser bandwidth and shape. For a laser profile with a long tail, the reversed asymmetry will persist longer than for a profile that drops off abruptly. In fact, if the laser profile is Lorentzian, the reversed asymmetry persists for arbitrarily large detunings. This is because, in the limit of large Δ_a, the two-photon nonresonant transition between $3S$ and $4D$ is also Lorentzian (as a function of Δ_a), and being weaker cannot overtake the double single-photon resonance $3S \to 3P \to 4D$ caused by the Lorentzian tail of the laser line overlapping $3P$.

The complete calculation encompassing the experimental conditions requires the time-dependent solution of the density-matrix equations, although limiting cases can be investigated in terms of approximate steady-state equations. Note that the presence of ionization in this problem precludes, in general, the existence of a steady state. Results of such calculations illustrating the above behavior have been obtained and will be published elsewhere.[21]

An approximate analytical result, which illustrates the qualitative aspects of the asymmetry, can be obtained if the ionization is weak. In that case, a steady-state approximation can be made, in the sense that the derivatives of the slowly varying parts of the density matrix elements are assumed to be nearly zero. Assuming Lorentzian line shapes for both lasers, one finds that the relative heights of the two peaks, to within some common factors, are given by

$$h_+ \propto \omega_{Rb}^2/(\Gamma_3 + \gamma_a + \gamma_b), \tag{1}$$

$$h_- \propto \frac{\omega_{Rb}^2}{\Gamma_2 + \Gamma_3 + \gamma_b}\left[\frac{1}{4}\,\frac{\omega_{Ra}^2}{\Delta_a^2}\left(1 + \frac{\gamma_a}{\Gamma_2}\right) + \frac{\gamma_a}{\Gamma_2}\right], \tag{2}$$

where Γ_2 and Γ_3 are the natural widths of the intermediate ($3P$) and upper third level ($4D$), respectively. ω_{Ra} and ω_{Rb} are the on-resonance Rabi frequencies for the transitions $3S \to 3P$ and $3P \to 4D$, respectively. These equations, which are valid for $\Delta_a^2 \gg \omega_{Ra}^2$ and $\omega_{Rb}^2 \ll \omega_{Ra}^2$, Γ_2^2, Γ_3^2, γ_a^2, γ_b^2, show that $h_+ > h_-$ if $\gamma_a < \Gamma_2$. This is in accordance with the monochromatic model ($\gamma_a = \gamma_b = 0$) for double resonance. If, however, $\gamma_a \geq \Gamma_2$, then $h_+ < h_-$, which means reversed asymmetry. Note, moreover, that it is the finite bandwidth (γ_a) of the intense laser— and not γ_b —that causes the reversal. These equations also confirm a point made earlier, namely that for Lorentzian line shapes the reversal persists for arbitrarily large detunings Δ_a. Equations (1) and (2) are obtained after considerable tedious mathematical manipulation and will be published in Ref. 21.

A number of other factors can influence the apparent widths of the observed peaks; for example, shot-to-shot variations in laser power and, hence, Rabi splitting. There are further theoretical reasons why amplitude fluctuations of the laser field are expected to cause substantial broadening of the peaks. The methods used in treating the field nonmonochromaticity are rigorously applicable only in the case of phase fluctuations.[21] As far as amplitude fluctuations are concerned—which surely were present in the experiment— the theoretical results could only be considered as approximate. Finally, the hyperfine structure should play a role in the broadening of the peaks. One way this comes about is through the superposition of splittings of distinct pairs of levels that are not resolved by the intense laser.

It will take considerable further work, experimental and theoretical, to disentangle all these effects. However, the basic effect discussed in this paper seems to emerge clearly. It is a fundamental feature of the behavior of a three-level system. A broadband intense laser will cause a reversed asymmetry.

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