ac Stark effect in multiphoton ionization

S. E. Moody* and Melissa Lambropoulos†
Joint Institute for Laboratory Astrophysics, National Bureau of Standards and University of Colorado, Boulder, Colorado 80309
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Multiphoton ionization of atomic sodium has been studied experimentally for near resonance between the radiation field and several bound-bound transitions of the atom. It has been found that the atom-field interaction must be treated in a highly nonlinear way to describe successfully the observations, even when the absolute spectral power densities are relatively modest. Rabi frequencies as high as 10 GHz are found to be directly observable with simple flashlamp-pumped tunable dye lasers.

A number of workers have recently shown that the spectral properties of the interaction between a quantum mechanical system and an intense resonant radiation field are considerably more complicated than classical resonance fluorescence.[1] In particular, the resonant scattering of intense monochromatic radiation by a two-level quantum-mechanical system resolves itself into three distinct spectral components due to an ac Stark effect when the incident intensity is high enough that stimulated processes dominate the spontaneous relaxation of the system. Theorists have now arrived at a consistent picture of the details of this emission, including the widths, separations, and relative intensities of the various components.[2,3] Experimental work is in general agreement with theory.[4-6] We have found that this same phenomenon must be considered to explain the results of our experiments concerning multiphoton ionization.

Figure 1 shows the situation that we have studied experimentally. Sodium in an atomic beam interacts with the radiation from two flashlamp pumped tunable dye lasers that are focused into the beam. One dye laser (which will be referred to as the "first" laser) is tuned into near resonance with the unperturbed transition frequency $3s^2S_{1/2}$ to $3p^2P_{1/2}$. A second dye laser connects $3p^2P_{1/2}$ to $5p^2P_{3/2}$ by means of a pure electric quadrupole transition.[7] Ionization from the latter state can occur with absorption of a photon from either laser, since they are of comparable intensity, and since the ionization cross section is a relatively weak function of photon frequency in this energy range. Both lasers are linearly polarized along the same axis. The atomic-beam density is sufficiently low that collisions play no role in the physics of the ionization process.

Ions and electrons resulting from the multiphoton process are collected from all angles by a relatively weak (40 V/cm) electric field and the sodium ions are swept to a particle multiplier, which measures the total ion yield on a pulse by pulse basis. The experiment consists of measuring the ionization rate as a function of frequency and intensity of the two dye lasers. Sodium diatomic ions are rejected from the integrated signal on the basis of their longer flight time from the interaction region to the detector, and hence do not contaminate the results. There is a very small contribution to the Na^+ signal from processes beginning with Na_2, the details of which are currently being investigated. This background is negligible for the cases shown here.

The characteristics of the lasers are as follows: Pulses are approximately 700 nsec in length, and the two lasers have their pulses synchronized in time with an accuracy of approximately 50 nsec. The spectrum of the lasers is contained in an envelope approximately 2 GHz FWHM. Intensities are of order 10^6 W/cm^2 at the interaction region. More specific values of the intensity will be given as the data are presented. The laser intensities are measured on a shot by shot basis to allow random variations to be removed from the data.

The physics of the strongly coupled two-state

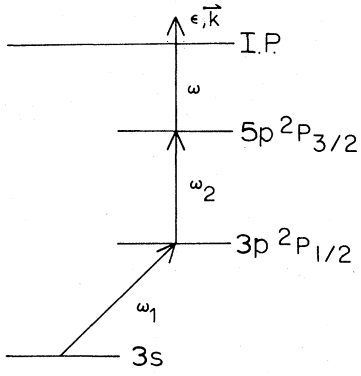

FIG. 1. States of atomic sodium involved in the experimental process.

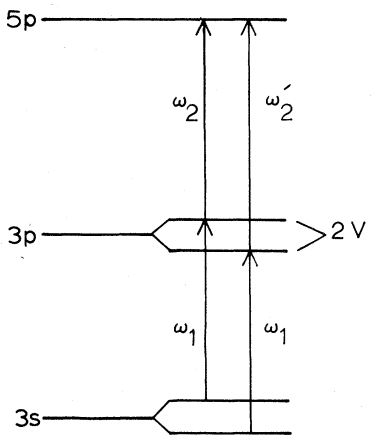

FIG. 2. Splitting model of strongly coupled states.

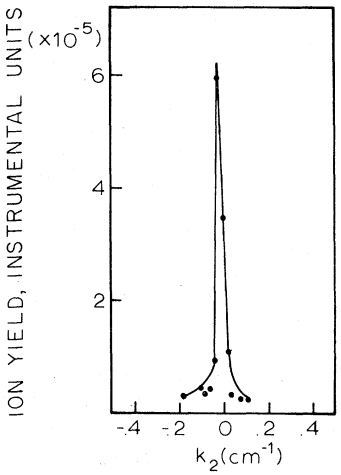

FIG. 3. Scan at relative intensity 1 (see text). Ion signal units result from normalization for atomic beam density and for laser power. Laser power normalization assumes the lowest-order intensity dependence, hence the apparent decrease in ion yield with increased power.

system can be most easily summarized as illustrated in Fig. 2. The two atomic states $3s$ and $3p$, when considered together with the possible states of the near resonant radiation field, are split into pairs of states with spacing characterized by the Rabi frequency:

$$\omega_R = [\delta^2 + (\mu E/\hbar)^2]^{1/2}, \quad (1)$$

where δ is the detuning of the first laser from the unperturbed resonance transition frequency, E is the magnitude of the incident electric field, and μ is the dipole matrix element of the resonance transition. ω_R characterizes the rate at which the system periodically moves between the states. It is this oscillatory behavior of the atomic wave function, frequently called the Rabi nutation, which is responsible for the "sidebands" found in the atomic spectrum when the field intensity is large. An equivalent description can be made in terms of the eigenstates of the coupled field-atom system, which are frequently referred to as "dressed states."[3]

This description assumes that the applied radiation field dominates the behavior of the atom, and therefore that spontaneous emission is weak by comparison. The spontaneous emission spectrum consists of four components, two of which are degenerate, resulting in the triply featured character of the resonance fluorescence. Conversely, if the two-state system is observed by inducing a weak transition to some third unperturbed state (in this case $5p$), then two "resonant" frequencies will be found for this probe transition.[3,8-11] This latter case clearly requires that the probe transition be sufficiently weak that the equations of motion of the strongly coupled lower two levels are only negligibly changed by the probe. In our experiment this condition has been ensured by the use of a pure electric quadrupole transition as the probe, which has a transition moment which is 10^5 times weaker than that which couples the lower two states.[7]

The experimental results shown here consist of tuning the first laser to resonance, as indicated by maximum fluorescence from the atomic beam (at low laser intensity). The second laser is then scanned through the $3p_{1/2}$-$5p_{3/2}$ resonance, and the ionization rate is measured as a function of its frequency.

Figures 3–6 show the results of four such scans at various intensities of the first laser. The intensities are given in instrumental units. Our

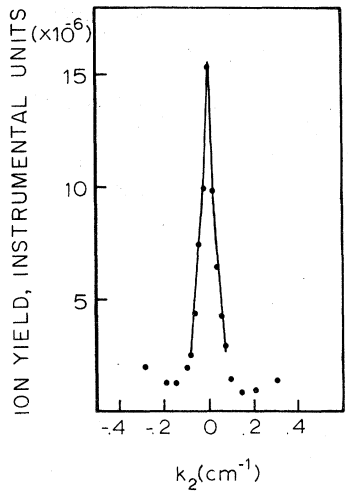

FIG. 4. Scan at relative intensity 6.5 (see text).

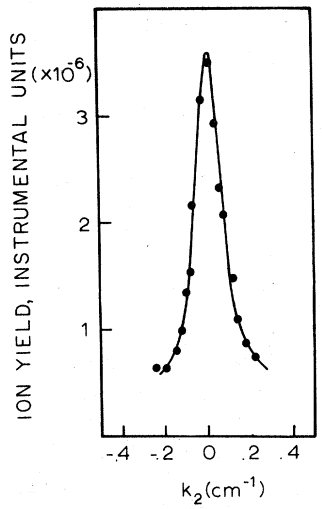

FIG. 5. Scan at relative intensity 25 (see text).

best estimate of the actual absolute intensity at the center of the interaction region is that 100 instrumental units correspond to a flux of 1.5 MW/cm^2. This estimate is believed to be reliable to within a factor of 2. Variation of the laser intensity is accomplished with neutral density filters, rather than by changes in the laser operating conditions, in order to avoid systematic variations in the spatial, temporal, or spectral output characteristics.

At the lowest intensity, the scan shows a line shape that is completely consistent with the laser linewidth. As the intensity is increased, the width of the observed resonance becomes distinctly

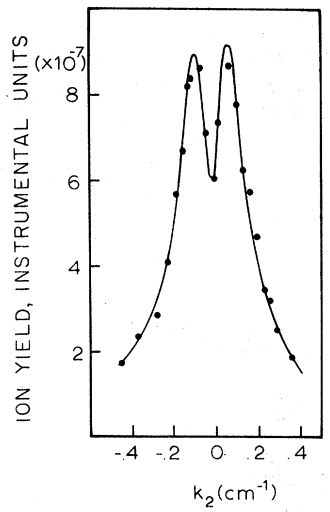

FIG. 6. Scan at relative intensity 150, corresponding to approximately 2.25 MW/cm^2 at the interaction region.

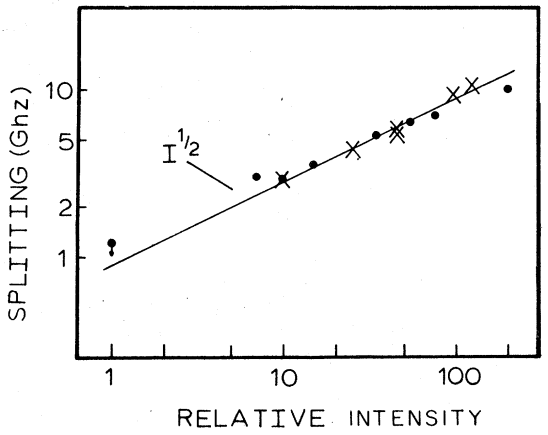

FIG. 7. Observed splitting as a function of laser intensity. Intensity units are the same as in Figs. 3–6.

larger, and the ionization rate increases only weakly with increased intensity. At the highest available intensity, the splitting into a two component feature is unambiguous. No other experimental parameters were varied in making these four scans.

Figure 7 shows the measured splitting as a function of laser intensity. Data are included from measurements made on two separate days in order to indicate the consistency of the observed splittings. The solid line represents an $I^{1/2}$ dependence, as predicted by Eq. (1) and is scaled to fit the data. The absolute magnitude of the splitting is more difficult to predict because of the uncertainties inherent in the spatial, temporal, and spectral characteristics of the laser output. Nonetheless, an estimate has been made using the information available, and is found to agree with the observed results to within 30%, well within the uncertainties inherent in making the estimate.

The shape of the ionization frequency dependence was also measured for slight detunings of the first laser from the unperturbed resonance frequency. The first laser was successively set to a number of wavelengths about the $3s$-$3p$ resonance, and the second laser was scanned through the $3p$-$5p$ resonance. Each of these scans becomes a single curve in a family of curves which was arranged according to the frequency of the first laser. Figure 8 shows these data, where ionization yield is plotted vertically, while the two laser frequencies determine the coordinates in the horizontal plane. It is clear that the symmetry of the observed line shape varies strongly with detuning of the first laser.

Figure 9 shows the magnitude of this asymmetry as a function of detuning. The parameter plotted is the ratio of heights of the two components as

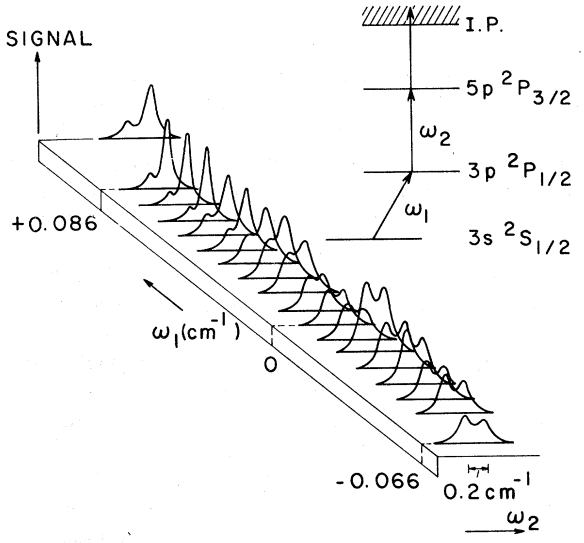

FIG. 8. Ionization rate vs both laser frequencies.

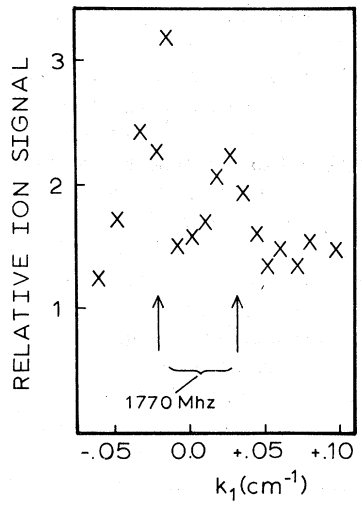

FIG. 10. Ionization yield versus detuning of laser 1. Frequency scale is the same as in Fig. 9. The ground-state hyperfine interval is indicated as being almost resolved.

determined by least squares fitting of a double-peaked line shape to the data. The solid line shown in Fig. 9 results from the three-level model of Khodovoi and Bonch-Bruevich, with the only adjustable parameter the center frequency.[10] It is assumed here that the detuning is small enough that the $3s$-$3p$ transition is totally saturated by the tail of the laser line, which is relatively broad on this scale. Because the experimental data involve not just three levels, but a whole host of hyperfine and magnetic sublevels, the agreement between the simple theory and the experimental data is somewhat surprising.

In fluorescence measurements of the same ac Stark effects, it has been found that only a carefully constructed experiment which eliminates all but two levels from the interaction gives results that agree well with theory.[4-6] One unresolved difficulty is that both physical intuition and more sophisticated calculations predict an asymmetry parameter that varies in the opposite way from the one shown here, at least for larger detunings.[12] A full understanding of these results therefore awaits a theory of this process which can accommodate a much larger number of sublevels, and a driving field which is not monochromatic.[13]

Figure 10 shows a third way of expressing the data of Fig. 9. The area under each scan of the second laser is plotted as a function of the detuning of the first laser, giving the integrated ionization yield as a function of the first-laser frequency. The ground-state hyperfine interval seems to be just barely resolved, indicating the importance of including the hyperfine structure in a full explanation of the data.

Figure 11 shows the laser-power dependence

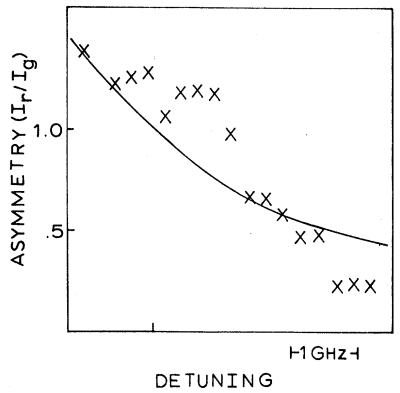

FIG. 9. Asymmetry of the split lineshape vs detuning of the strong coupling laser field.

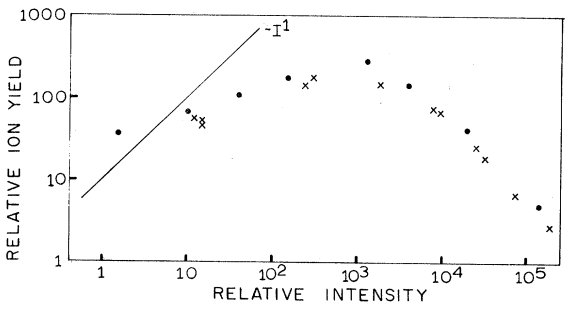

FIG. 11. Total ion yield as a function of intensity of laser 1. Intensity units are: 10^5 instrumental units $= 2 \times 10^6$ W/cm^2.

of the total ionization rate at fixed laser frequency. Both lasers are tuned for optimum double resonance at low intensity, and the ionization rate is measured as a function of the intensity of the laser. To lowest order, the ionization signal would be expected to increase somewhat faster than linearly, since the first laser is entirely responsible for inducing the $3s$-$3p$ transition, as well as contributing somewhat to the final ionization step. Instead, as the intensity is increased, the power dependence falls well below the linear prediction, and in fact at the highest intensities that were available, the ionization rate was found to actually decrease rather strongly with increasing power. This decrease presumably results from the loss of the double-resonance condition at high intensities, where the energy spectra of the atomic states are strongly modified by the incident field. Unfortunately, a quantitative description of this effect without making unrealistic assumptions has not been possible. The implications of this result for the scaling of laser-driven resonant processes in various applied spectroscopy situations should be clear.

The second caution which is pointed to by these results is the difficulty of performing pure three-photon ionization in the case of a resonant process (and by analogy, other multiphoton processes in various systems suffer the same problem). Instead, the probability of the exchange of many photons between the atom and field is rather high in realistic experimental situations, and can be expected to be significant. The result is that the complications of these multiphoton processes is very often much larger than was anticipated at first view. Perhaps the most general importance of these results, however, is the clear failure of any "stepwise" transition model in even qualitatively understanding the experimental data.[14] In the presence of laser intensities, it seems clear that even the simplest cases must be considered in terms of a unified theory of the atom-field interaction.

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*Staff Member, Time and Frequency Division, National Bureau of Standards.

†Present name and address: Melissa Reading, Department of Physics, University of California, Santa Barbara, Calif. 93106.

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[14]By "stepwise" we mean models which treat each pair of states independently by calculating a simple two-state transition rate.