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Bandwidth-induced reversal of asymmetry in optical-double-resonance amplitudes

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Optical-double-resonance measurements using ionization detection have been carried out in the $3S_{1/2}$-$3P_{1/2}$-$4D$ atomic-sodium system. Asymmetries observed in production of $4D$ atoms from the two components of the Stark-split $3P_{1/2}$ state are found to be controlled by the far, very weak wings of the 17-MHz full-width-at-half-maximum laser line which is used to drive the $3S_{1/2}$-$3P_{1/2}$ transition at detunings in the range 0–70 GHz. Suppression of the wings with a Fabry-Perot filter causes a pronounced reversal of the asymmetry.

I. INTRODUCTION

We report an experimental study of the effects of absorption of near-resonant 589-nm radiation by the $3S_{1/2}$-$3P_{1/2}$ system of atomic sodium irradiated by an intense pulsed laser field in a collision-free environment. We have used a double-optical-resonance technique to probe the $3P_{1/2}$ doublet produced by the ac Stark effect.

The phenomenon of ac Stark splitting is well known from early work in the radio frequency and microwave regimes.[1,2] The formal two-level theory of this effect was developed for the visible regime by Mollow[3] and confirmed in fluorescence experiments with well-stabilized lasers.[4] For a two-level system in a harmonic monochromatic field, each level splits into a doublet with separation

$$[(\Delta)^2+(\mu E/h)^2]^{1/2},$$

where Δ is the detuning of the laser from exact resonance and $(\mu E/h)$ is the frequency corresponding to the interaction energy of the atomic dipole (μ) in the electric field (amplitude E) of the laser radiation. Furthermore, when $\Delta \neq 0$ the center of each doublet is shifted by $\Delta/2$ in a direction compensating the detuning.

For a double-resonance (three-level) experiment in which a second laser is used as a very weak probe, the two-level picture for the strongly coupled states remains valid. In our case we probe the $3P_{1/2}$ doublet by coupling weakly to the $4D$ state, taking care that the Stark splitting of the $3P_{1/2}$ level is much larger than its hyperfine structure so that the $3S_{1/2}$-$3P_{1/2}$ system adheres reasonably well to two-level behavior. $4D$ atoms produced by the probe laser are subsequently photoionized by the intense 589-nm radiation. Measurement of the $4D$ ionization rate as a function of probe laser frequency thus maps out a two-peaked line with splitting equal to the $3P_{1/2}$ doublet splitting and peak heights dependent on the relative probability of reaching the $4D$ state via the two different components of the $3P_{1/2}$ state. The double-resonance method is illustrated by an energy-level schematic in Fig. 1.

We are primarily interested in the ratio of the two ionization peak heights and how this ratio changes with detuning of the intense laser. The monochromatic radiation model predicts an interesting limiting-case behavior. For laser intensities which vary adiabatically, the intermediate $(3P_{1/2})$ state is populated *only* by second-order radiative processes, in the absence of collisions.[5] As indicated in Fig. 2(a), one of the components of the detected signal corresponds to this type of excitation of the intermediate level, followed by direct excitation to the $4D$ state. The other component [Fig. 2(b)] can be identified as direct two-photon excitation of the $4D$ state. In the mono-

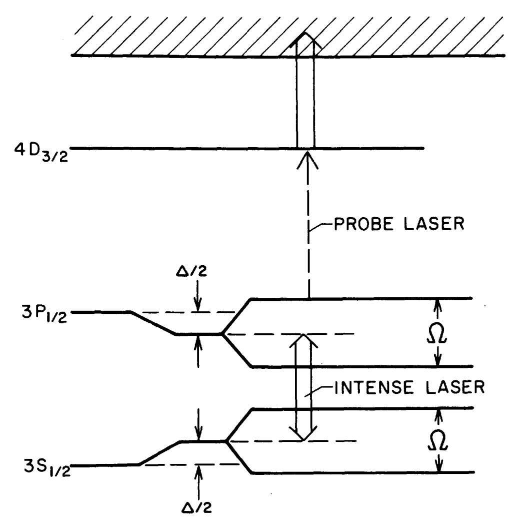

FIG. 1. Schematic representation of the dynamic Stark effect in three-photon ionization of sodium. The doublet splitting Ω of the $3S_{1/2}$ and $3P_{1/2}$ states is determined by the detuning, Δ, and electric field amplitude E of the intense laser according to $\Omega=[(\mu E/h)^2+\Delta^2]^{1/2}$. A measurement of ion signal as a function of probe laser frequency yields a two-peaked line as a result of the ac Stark splitting of the $3P_{1/2}$ level.

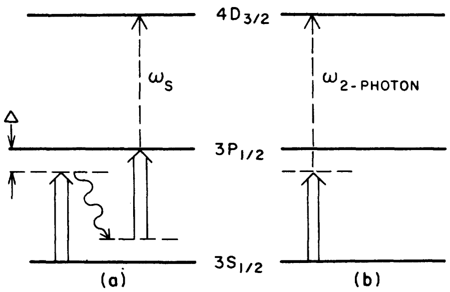

FIG. 2. Channels for reaching the $4D$ state under purely monochromatic excitation: (a) a second-order sequential process observed when the probe frequency is ω_S; (b) a direct two-photon process observed when the probe frequency is $\omega_{2\text{-photon}}$.

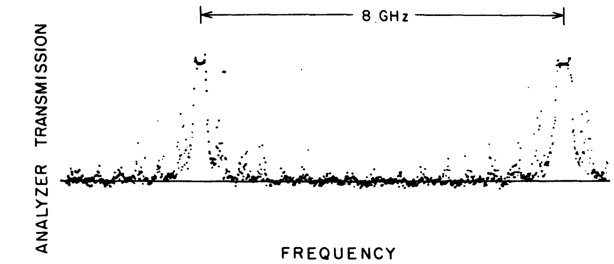

FIG. 3. Spectrum analysis of the intense laser revealing the discrete sidebands produced by amplitude modulation. The two large saturated peaks are due to the central intense component of the spectrum and are separated by the free spectral range of the analyzer.

chromatic model, the latter dominates for all nonzero detunings, the ratio of two-photon to the higher-order sequential process going as Δ^2 in the limit of large detuning. We shall refer to the asymmetry of the ionization line shape as "normal" when the two-photon component dominates and "reversed" when the sequential component dominates.

We note at the outset that previous double-resonance observations in this laboratory yielded results which could *not* be understood in terms of the monochromatic theory. In those experiments,[6,7] which were carried out in sodium with broad-band (1–2 GHz) flashlamp-pumped dye lasers, it was observed that the asymmetry of the ionization signal was reversed whenever the detuning of the intense laser was less than about three laser linewidths and normal for detunings larger than three linewidths. It was suggested by Georges and Lambropoulos[7,8] that the reversed asymmetries observed at the smaller detunings could be attributed to the finite bandwidth of the intense laser. The origin of this effect is related to the problem of nonadiabatic excitation in resonance fluorescence experiments as discussed, for example, in Ref. 5. It was the original objective of the present study to observe asymmetries in the same $3S_{1/2}$-$3P_{1/2}$ system of atomic sodium using a new intense laser having a very narrow bandwidth.

II. LASER CHARACTERISTICS AND EXPERIMENTAL DETAILS

The laser used in the present experiment is a unique Nd:YAG-pumped dye amplifier system which amplifies the output of a commercial cw single-mode laser stabilized to ~1 MHz. This system produces up to 20 kW of diffraction-limited radiation in 60-nsec pulses with a FWHM linewidth of 17 ± 4 MHz.[9] In addition to the intense narrow-band output, the laser produces a broad low-level background consisting of discrete sideband peaks (which are produced by amplitude modulation of the central narrow-band component) superimposed on a broad structureless component which originates in fluorescence in the first stage of the three-stage amplifier. Amplitude modulation of the output occurs due to mode beating and consequent amplitude modulation in the Nd:YAG pump laser. This leads to modulation of the gain in the dye amplifier stages, and therefore to modulation of the output pulse. As shown in Fig. 3, analysis of the amplified output with an 8-GHz confocal spectrum analyzer reveals a sequence of discrete sidebands spaced at intervals of approximately 0.5 GHz on either side of the central peak. The sideband peaks are sensitive to operating conditions in the pump laser and are not strictly reproducible from day to day. They are typically a factor of 50–200 smaller than the intense central peak. The fluorescence background, whose intensity may be inferred from the level of the analyzer transmission signal between the discrete peaks at high electronic gain, lies about four orders of magnitude below the central peak and typically contains 3% of the total laser power in its ~0.10-nm bandwidth.

In our experiment this laser, which couples the $3S_{1/2}$ and $3P_{1/2}$ states, is focused into the center of a diffuse sodium beam with a 1-m focal length lens, yielding intensities at the interaction region on the order of 5 MW/cm^2 in a focal spot approximately 250 μm in diameter. A weak, unfocused laser beam from a synchronously pumped Hänsch-type oscillator (bandwidth ≈5 GHz) is superimposed on the beam of the intense laser to serve as the probe in the double-resonance excitation, coupling either component of the Stark-split $3P_{1/2}$ doublet to the $4D$ state. This probe laser is scanned 150 GHz across the $3P_{1/2}$-$4D$ transition using a step-motor-driven intracavity etalon synchronized with a piezoelectric driven grating which is the end

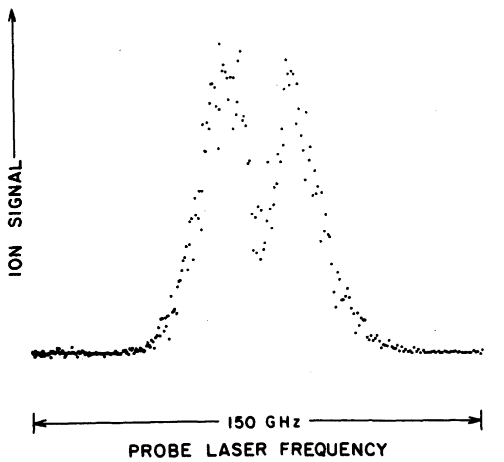

FIG. 4. Na^+ signal as a function of probe laser frequency showing the ac Stark splitting near resonance. The laser power is 2 kW. Each point represents the ionization yield of a single laser shot.

reflector of the cavity. Na^+ ions produced by photoionization of $4D$ atoms by the intense laser are collected in a weak electric field and subsequently detected with an electron multiplier. At the laser intensities used the $3P_{1/2}$-$4D$ step depends linearly on the probe laser intensity while the $4D$ ionization probability is of order unity. Molecular ions produced by multiphoton ionization of Na_2 molecules in the beam do not contaminate the data since they can be discriminated on the basis of a longer time of flight to the detector.

The Na^+ ion signal and both laser intensities are monitored on every laser shot and fed to a minicomputer which controls data acquisition and performs on-line data analysis. The computer is programmed to reject data points for which either laser intensity falls outside of a preset window. A plot of ion signal versus probe laser frequency is produced simultaneously with the accumulation of the data. The recorded ion signal represents an average over a predetermined number of data points at each probe frequency.

Figure 4 shows a typical result for the production of Na^+ as a function of probe laser frequency when the intense laser is resonant with, or within 1 GHz of, one of the hyperfine components of the unperturbed $3S_{1/2}$-$3P_{1/2}$ transition. The frequency

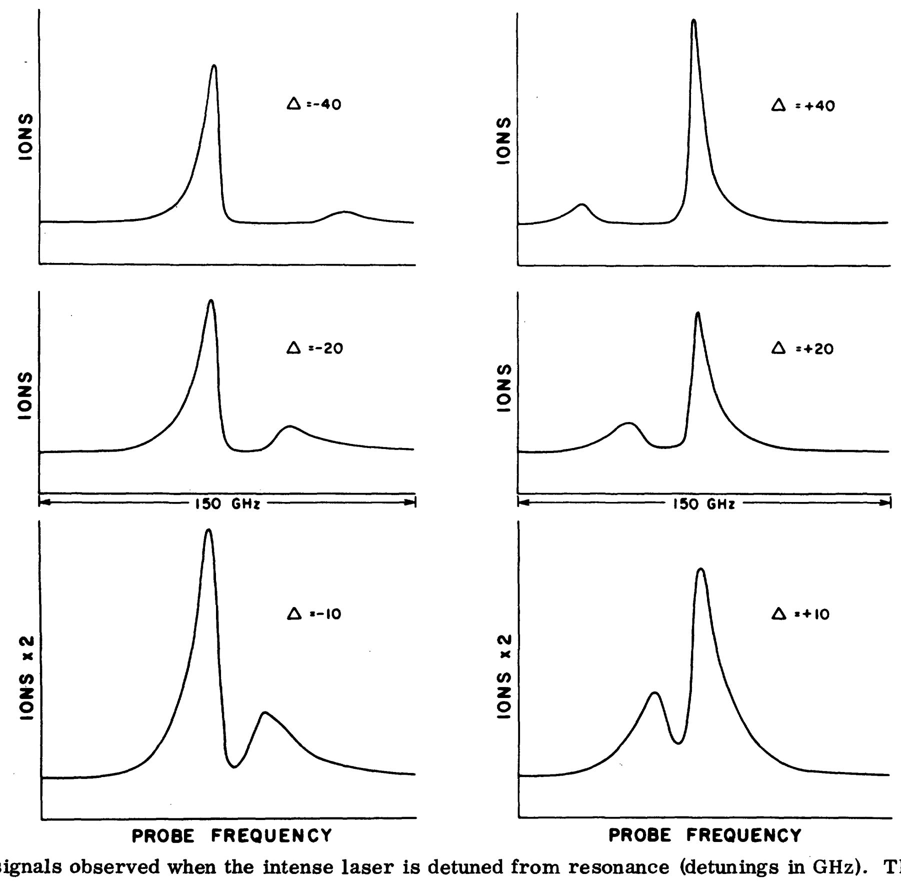

FIG. 5. Ion signals observed when the intense laser is detuned from resonance (detunings in GHz). The asymmetry of the doublet is reversed in all cases. Individual data points have been replaced by a smoothed curve.

calibration is made by observing fluorescence from a downstream portion of the atom beam which is irradiated by a small fraction of the cw laser output. The observed Stark-split peaks are approximately symmetric, as is expected very near resonance, and separated at the peak (in this instance) by roughly 25 GHz. It should be pointed out that the shape of the ionization curve, in particular its breadth, is to a large degree determined by experimental geometry. Since the intensity of the strong laser varies in space and in time, excited atoms produced at different positions (and at different times) in the interaction volume are characterized by different Stark splittings. Thus the outer extremes of the ionization curve represent ions produced at the center of the focused laser spot where the intensity (and hence the Stark splitting) is greatest.[10] The largest contributions to the ion signal come from regions of lower intensity (but larger volume) slightly off the axis of the laser beam. While these geometrical considerations lead to an altered shape of the ionization curve as compared with the ideal constant intensity case, they do not, however, influence the comparative peak heights, which is the primary concern of the present experiment.

III. INITIAL RESULTS

We present in Fig. 5 a series of results obtained in the manner described above when the intense laser is detuned from resonance. For a negative detuning of the intense laser, the "two-photon" peak of the ion signal occurs at a higher probe laser frequency than the "sequential" peak. For positive detunings the reverse is true. It is apparent from the figure that the sequential peak dominates in every instance; that is, we observe the asymmetry to be *reversed* at all detunings for which ion signals have been obtained. These measurements range to detunings as large as 70 GHz, which is approximately 4000 times the width of the central intense peak in the laser spectrum. This contrasts sharply with the results of the previous experiments, in which reversed asymmetries were observed only at relatively small detunings. These results also contradicted our original expectation that use of the new laser system would eliminate or reduce the range of reversed asymmetries.

IV. EFFECTS OF FINITE LASER BANDWIDTH

We now digress briefly to summarize the ideas put forward by Georges and Lambropoulos[7,8] to account for the possibility of reversed asymmetries in double-resonance experiments. In their

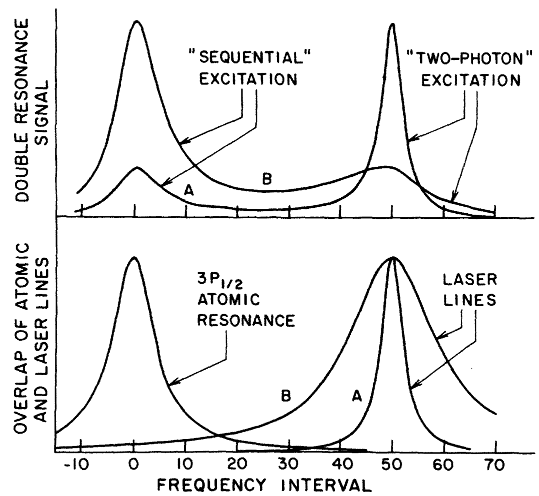

FIG. 6. Finite-bandwidth picture of asymmetry in Stark splitting by a detuned laser using Lorentzian functions. A scan of probe laser frequency across the $3P_{1/2}$-$4D$ transition produces a signal (upper traces) which varies as the product of atomic and intense laser line shapes (lower traces). The $3P$-$4D$ excitation probability is assumed constant. For Lorentzian line shapes, the "two-photon" excitation, occurring off resonance, dominates if the laser line is narrower than the atomic line (case A), while the asymmetry is reversed if the laser line is broader than the atomic line (case B).

model, which we illustrate qualitatively in Fig. 6, the laser line and the atomic resonance are described by Lorentzian functions and second-order radiative processes are neglected. The process described as "sequential excitation" in the monochromatic model is determined in this picture by the intensity of the wing of the laser line at exact resonance, while the "two-photon" rate is determined by the strength of the atomic resonance at the center frequency of the laser. As illustrated by the product functions in Fig. 6, *either* process may dominate depending on which Lorentzian is narrower. Thus the asymmetry is normal if the laser line is narrower than the atomic resonance and reversed if it is broader. For pure Lorentzian functions, this result holds true for arbitrarily large detunings. One also has the possibility that, for equal-width Lorentzians, the peaks would be of equal height for all detunings. As noted, the picture as we have described it neglects second-order radiative processes and also differs from detailed laser line-shape theory, according to which a laser mode line falls off more steeply than a Lorentzian several linewidths from center.[11] Theoretical papers exploring the consequences of this and other laser properties in optical double resonance have recently appeared in the literature.[12]

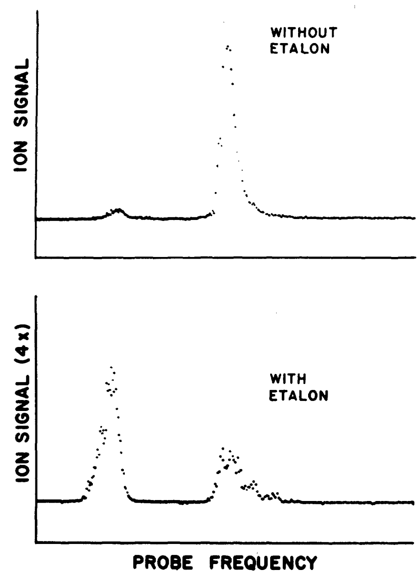

FIG. 7. Comparison of ac Stark-effect signals at a fixed detuning of +40 GHz demonstrating the change in asymmetry resulting from filtering the intense-laser background light with a Fabry-Perot etalon.

The implication of the ideas of Georges and Lambropoulos for the experiment we are describing is clear: the reversed asymmetry we observe even at very large detunings is presumably due to the broad background by virtue of which the laser spectrum falls off much more slowly than a Lorentzian of width 10 MHz (the sodium $3P_{1/2}$ linewidth). A specific comparison illustrates the point more vividly: at a detuning of 40 GHz, for example, a 10-MHz Lorentzian laser line would overlap the center of the atomic resonance with an intensity on the order of 10^{-7} of the peak intensity. Our spectrum analysis, however, indicates that the background originating in fluorescence maintains the intensity at a level roughly 10^{-4} of the peak intensity out to 60 GHz from line center. Thus our observation of only reversed asymmetries appears to be consistent with the picture described above.

V. RESULTS OBTAINED WITH A MODIFIED LASER SPECTRUM

According to the discussion in the previous section, the laser intensity 40 GHz from line center would need to be reduced by three orders of magnitude in order for a normal asymmetry to be observed. Such a reduction can in fact be achieved if the laser output is filtered with a high-resolution Fabry-Perot etalon. We show in Fig. 7 the results of a measurement at 40-GHz detuning in which ion signals were obtained at fixed incident laser power with and without the use of an etalon

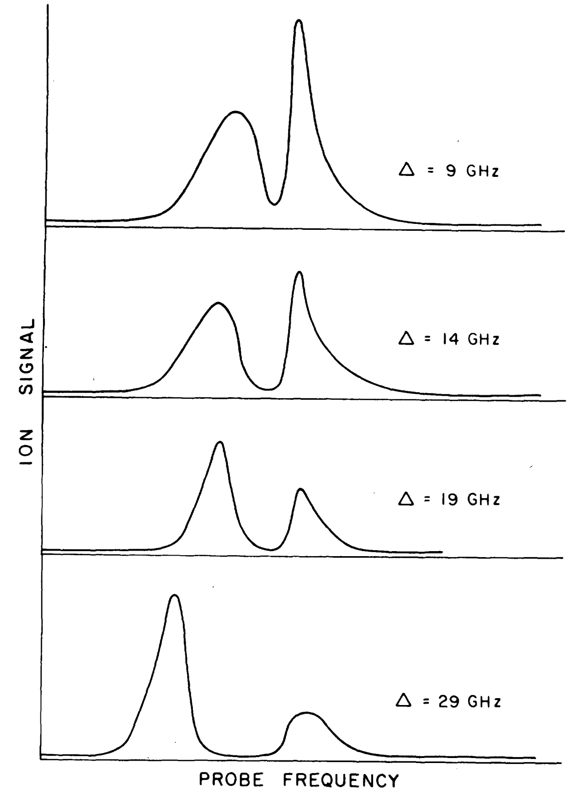

FIG. 8. Stark-effect signals at four different detunings when the intense laser is filtered. The asymmetry changes from reversed to normal at a detuning of 15 GHz.

tuned to transmit the center frequency. This etalon has a free spectral range of 90 GHz and a measured finesse of 60, which corresponds to a 1400-fold reduction of intensity 40 GHz from the center frequency.[13] It is clear from the figure that insertion of the etalon does indeed change the asymmetry of the ion signal from reversed to normal. This observation represents the most direct evidence to date in support of the idea that the asymmetry of the Stark-split doublet in optical double resonance is governed by the intensity in the far wings of the laser spectrum.

A series of measurements at different detunings utilizing the extracavity etalon produces results which have some of the same characteristics as found by Hogan *et al.*[7] (see Fig. 8). Normal asymmetries are observed for detunings greater than 15 GHz while detunings less than 15 GHz yield reversed asymmetries. Since the etalon reduces intensity by a factor of 360 and a 10-MHz Lorentzian is diminished by a factor of 2×10^6 15 GHz from the center frequency, the implied intensity of the *unfiltered* laser is $360/(2\times10^6)$ or about 2×10^{-4} of the peak intensity. This happens to be the approximate level of the fluorescence component of the laser spectrum. This probably indicates that the discrete sidebands in the spectrum, which have a relative intensity of 10^{-2} near the center frequency, fall to the level of the fluorescence pedestal 15 GHz from the center frequency. Thus the persisting reversed asymmetry between 0 and 15 GHz in Fig. 8 is likely due to the presence of the amplitude modulation sidebands.

Referring back to the result in Fig. 7, it is indeed surprising to find that so drastic a change in the observed ionization ratio is brought about by attenuating a frequency component in the laser spectrum which is only 10^{-4} of the peak intensity to begin with. A detailed, quantitative understanding of this and other experiments employing intense lasers to drive resonant transitions will require that laser line shapes be known with high precision over an intensity range encompassing many orders of magnitude. Alternatively, it is clear that such methods can provide a sensitive, if semiquantitative, characterization of the behavior of far wings of intense lasers.

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[1]I. I. Rabi, Phys. Rev. 49, 324 (1936); 51, 652 (1937).

[2]S. H. Autler and C. H. Townes, Phys. Rev. 78, 340 (1950); 100, 703 (1955).

[3]B. R. Mollow, Phys. Rev. 188, 1969 (1969); Phys. Rev. A 2, 76 (1970).

[4]F. Schuda, C. R. Stroud, and M. Hercher, J. Phys. B 7, L198 (1974); F. Y. Wu, R. E. Grove, and S. Ezekiel, Phys. Rev. Lett. 35, 1426 (1975); H. Walther, in *Laser Spectroscopy* (Springer, Berlin, 1975), pp. 358–369.

[5]E. Courtens and A. Szöke, Phys. Rev. A 15, 1588 (1977).

[6]S. E. Moody, Ph.D. thesis, University of Colorado, 1975 (unpublished); S. E. Moody and M. Lambropoulos, Phys. Rev. A 15, 1497 (1977).

[7]P. B. Hogan, S. J. Smith, A. T. Georges, and P. Lambropoulos, Phys. Rev. Lett. 41, 229 (1978).

[8](a) A. T. Georges and P. Lambropoulos, Phys. Rev. A 18, 587 (1978); (b) 20, 991 (1979).

[9]G. L. Eesley, M. D. Levenson, D. E. Nitz, and A. V. Smith, IEEE J. Quantum Electron. QE-16, 113 (1980).

[10]One may in fact infer the peak intensity from the known $3S_{1/2}$-$3P_{1/2}$ dipole matrix element and the extreme width of the ionization curve. This *in situ* measurement is especially useful in light of the difficulty associated with determining absolute intensities in focused laser spots.

[11]H. Haken, in *Encyclopedia of Physics*, edited by S. Flügge (Springer, Berlin, 1970), Vol. XXV/2c.

[12]See Ref. 8(b) and P. Zoller, Phys. Rev. A 20, 1019 (1979); A. T. Georges, P. Lambropoulos, and P. Zoller, Phys. Rev. Lett. 42, 1609 (1979); S. N. Dixit, P. Zoller, and P. Lambropoulos, Phys. Rev. A 21, 1289 (1980); P. Zoller and P. Lambropoulos, J. Phys. B 12, L547 (1979); K. I. Osman and S. Swain, *ibid.* 13, 2397 (1980); J. J. Yeh and J. H. Eberly, Phys. Rev. A (in press).

[13]If T_{max} represents the etalon transmission at the center frequency, the relative transmission at a frequency $\Delta\nu$ from the center frequency is given by $T/T_{max} = [1+(2F/\pi)^2\sin^2(\pi\Delta\nu/\nu_{FSR})]^{-1}$ where F is the finesse, ν_{FSR} is the free spectral range, and the etalon is used at normal incidence. See, for example, M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1959), Sec. 7.6.