Two-photon absorption from a field with random telegraph frequency

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We have measured the absorption spectrum of a two-photon transition (3*S* to 5*S* in Na) for the case where the exciting laser has random telegraph frequency fluctuations. Good agreement was found between the experimental data and predictions for some features based on a theory due to Mollow [Phys. Rev. **175**, 1555 (1968)]. Comparison is made with two-photon absorption spectra measured with a laser having Gaussian frequency fluctuations.

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I. INTRODUCTION

Two-photon absorption (TPA) is perhaps the simplest of nonlinear optical processes and is thus a good starting point for experimental studies of the effect of laser noise on nonlinear processes. The fluctuations of the laser influence the response of a nonlinear system in a way that cannot be reconstructed just by adding the responses due to each Fourier component of the laser. Therefore laser noise must be present in its entirety if the full behavior of the system is to be determined.

Our approach to this problem is to excite a two-photon resonance with a partially coherent laser which is made by imposing electronic noise on a stabilized laser. We can determine the statistics of the noise and thus also of the laser field. In this paper we describe the use of a laser whose frequency noise is described by a random telegraph (RT) process, which has the general property that the stochastic variable (in this case the frequency) jumps discontinuously between two well-defined values at random times. The number of jumps n in a time interval T_0 is governed by the Poisson distribution $P(n)=\lambda^n e^{-\lambda}/n!$, where λ is the mean number of jumps in T_0. Such noise is added to a laser by first generating a random telegraph voltage, then imposing it on the laser frequency by use of an acousto-optic modulator (AOM). This complements previous TPA experiments where other models of the noise were appropriate: phase diffusion [1] and real Gaussian [2]. In the phase-diffusing field the phase fluctuation is an Ornstein-Uhlenbeck process, and its time derivative is the frequency fluctuation. This frequency is a stochastic process with a very different probability distribution (Gaussian) than the bimodal distribution of the RT frequency that we discuss in this paper. The real Gaussian field has amplitude fluctuations with a Gaussian distribution but has no phase or frequency fluctuations. A closely related experiment was performed with second-harmonic generation in a microwave system [3], but this experiment is the first that we are aware of with random telegraph noise in the optical regime. A more important difference is that in Ref. [3] the atomic decay time was much shorter than the field correlation time, whereas in our experiment they were of similar magnitude.

Two-photon absorption is of considerable spectroscopic importance, being used, for example, in measuring Lamb shifts. In these experiments heterodyne measurements of the instantaneous frequency of a pulsed laser have recently been reported [4]. The pulsed lasers used in some of these experiments have a frequency chirp that gives spectral broadening which results in shifts in the position and changes in the shape of the observed spectrum. In a laser pulse with a frequency chirp, the electric field is clearly nonstationary in the statistical sense. On the other hand, the spectral broadening in the case that we have concentrated on arises from a stationary stochastic process, and shifts of the TPA resonance are not expected, but the shape of the resonance does show interesting behavior, dependent on the exact statistical nature of the frequency noise.

Before describing our experiment, a brief review of what the theory predicts is in order, and this will occupy Sec. II. Then we will describe the experimental apparatus and procedure in Sec. III, followed in Sec. IV by presentation and discussion of the results.

II. THEORY

We adopt a plane wave description of the electromagnetic radiation field;

$$E(\mathbf{r},t)=E_0\exp[i\mathbf{k}\cdot\mathbf{r}-i\omega_L t-i\phi(t)]\ , \tag{1}$$

where $\mathbf{k}$ is the wave vector, ω_L is the center frequency of the laser, and $\phi(t)$ is the time-dependent phase which is used to describe the frequency fluctuations. Note that the instantaneous frequency $\omega(t)=\dot{\phi}(t)+\omega_L$. The coherence of light is properly specified by an infinite number of electric-field correlation functions. For many purposes the lowest-order nonzero correlation function $\langle E^*(\mathbf{r}_1,t_1)E(\mathbf{r}_2,t_2)\rangle$ provides a sufficient description of

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the field because this gives the power spectrum and the intensity; the power spectrum is given by its (temporal) Fourier transform. (In what follows we will only be concerned with the temporal coherence, so the r dependence of the field will be omitted.) Knowledge of at least some of the higher-order correlation functions is necessary to describe nonlinear processes. Using perturbation theory, Mollow [5] showed that the two-photon absorption spectrum as a function of ω_L, the center frequency of the laser, depends on the Fourier integral of a fourth-order correlation function

$$W_2(\omega_L)=|g(\omega_L)|^2\int_{-\infty}^{\infty}\langle E^*(-t)E^*(-t)E(t)E(t)\rangle \times e^{i2\omega_f t-\kappa|t|}dt \ . \quad (2)$$

The energy difference between the upper and lower energy levels is $\hbar\omega_f$, and the quantity $g(\omega_L)$ incorporates the atom-field coupling. Provided there are no intermediate atomic energy levels too close to half the energy spacing between the initial and final levels, $g(\omega_L)$ is to a good approximation constant over the relevant range of laser frequencies. This was the case in our experiment. The atomic damping is incorporated in κ, which is the decay rate of the off-diagonal density-matrix elements [6]. Actually, Mollow took κ to be the natural width of the upper level, assuming the lower level to be a ground state and assuming that there are no other dephasing processes in the system. In our experiment we had a certain amount of collisional broadening, which is an off-diagonal broadening process and dominates the natural width of the transition. This also affects $W_2(\omega_L)$, and we incorporated it into κ. In practice this means that κ is the width of the TPA spectrum when there is no laser noise present.

Equation (2) was subsequently generalized [7,8] to n-photon absorption, giving the result that for RT noise the form of the equation for the n-photon absorption spectrum is the same as that for the laser spectrum, except that where the size of the random phase or frequency jump enters in the field correlation function of the integral, it is multiplied by a factor of n. For TPA, this means that $W_2(\omega_L)$ can be found by multiplying the size of the frequency jump by 2 wherever it appears in the equation for the field correlation function. For RT frequency noise, $\langle E^*(t+\tau)E(t)\rangle$ is given by [9]

$$\langle E^*(t+\tau)E(t)\rangle=\frac{1}{2}\left\{\left[\frac{1}{T\lambda}+1\right]e^{-[1/T-\lambda]|\tau|} - \left[\frac{1}{T\lambda}-1\right]e^{-[1/T+\lambda]|\tau|}\right\} \times E_0^2 e^{-i\omega_L\tau} , \quad (3)$$

where $\lambda^2=T^{-2}-\Omega^2$. The total size of the frequency jump is 2Ω. T is the mean time between frequency jumps, or dwell time. The stochastic frequency is stationary, so

$$\langle E^*(-t)E^*(-t)E(t)E(t)\rangle=\langle E^*(\tau)E^*(\tau)E(0)E(0)\rangle ,$$

where $\tau=-2t$. Then we get

$$\langle E^*(\tau)E^*(\tau)E(0)E(0)\rangle = \frac{1}{2}\left\{\left[\frac{1}{T\Lambda}+1\right]e^{-[1/T-\Lambda]|\tau|} - \left[\frac{1}{T\Lambda}-1\right]e^{-[1/T+\Lambda]|\tau|}\right\}E_0^4 e^{-i2\omega_L\tau} , \quad (4)$$

where $\Lambda^2=T^{-2}-b^2$, and $b=2\Omega$. The respective spectra are calculated from these functions. For the laser spectrum, they are

$$S_L(\omega)=\frac{8\pi\Omega^2|E_0|^2/T}{(\omega-\omega_L)^4+(\omega-\omega_L)^2(4/T^2-2\Omega^2)+\Omega^4} , \quad (5)$$

and for the two-photon absorption spectrum, taking into account the width of the upper state,

$$W_2(\omega)=W_0\frac{T^2\kappa^3+8T\kappa^2+4T(4+\kappa)b^2+16\kappa+4\kappa T^2(\omega-2\omega_L)^2}{T^2(\omega-2\omega_L)^4+(\omega-2\omega_L)^2[4-2T^2b^2+2T\kappa+T^2\kappa^2/2]+[Tb^2+\kappa+T\kappa^2/4]^2} . \quad (6)$$

In both Eqs. (5) and (6) the middle term of the denominator determines whether the corresponding spectrum is single or double peaked. If $4-2(bT)^2+2\kappa T+(\kappa T)^2/2\geq 0$, the TPA spectrum is single peaked, otherwise it is double peaked. If $\kappa=0$, a comparison of (5) and (6) shows that a single-peaked laser power spectrum giving a two-peaked TPA spectrum is possible, but not the other way round. If, however, $\kappa>0$, a two-peaked laser power spectrum with a single-peaked absorption spectrum also becomes possible. It is also interesting to note the presence of the term in $(\omega-\omega_L)^2$ in the numerator of Eq. (6). The atomic relaxation restores the ω^{-2} dependence of the wings of the absorption spectrum from the ω^{-4} dependence given by the frequency fluctuations.

III. EXPERIMENT

We performed an experiment in which the form for the TPA spectrum predicted in the Sec. II was checked. We used the $3S$ to $5S$ two-photon transition in sodium vapor (see Fig. 1) excited by a laser with random telegraph frequency noise. A dye laser operating with rhodamine $6G$ was used to produce the desired wavelength of 602 nm. The $3P$ level was not so close to the intermediate level of the absorption process as to be significantly populated directly by the laser. Yet it was close enough to significantly enhance the TPA probability and give an easily observed resonance. The experiment used the standard Doppler-free method with counterpropagating laser

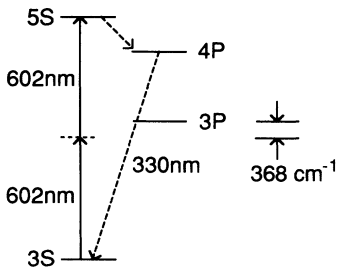

FIG. 1. The sodium $3S$-to-$5S$ two-photon transition and the decay channel through the $4P$ level.

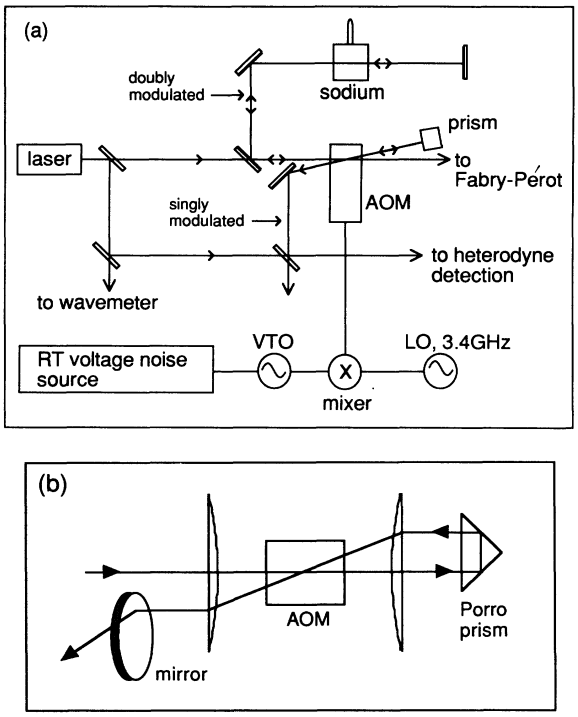

FIG. 2. (a) A schematic of our experimental setup. The diagram is a plan view, so the vertical separation of the beams to and from the prism is not shown. Abbreviations are AOM (acousto-optic modulator), VTO (varactor-tuned oscillator), and LO (local oscillator). (b) Side view of the double-pass arrangement in the AOM.

beams [10]. We used linearly polarized light which did not preclude TPA from a single propagation direction. Such absorption results in a Doppler-broadened pedestal on which the Doppler-free part is superimposed, but in our experiment this pedestal was obscured by the detection noise. Two Doppler-free peaks were observed corresponding to the $F=2$ to $F=2$ and $F=1$ to $F=1$ hyperfine transitions. We concentrated only on the larger $F=2$ to $F=2$ resonance for our measurements.

The absorption of the laser light was measured by detecting light with a photomultiplier at a wavelength of 330 nm that came from the relaxation of the $5S$ through $4P$ levels as illustrated in Fig. 1. To stop stray laser light from reaching the photomultiplier, a colored glass filter was used, as well as a lens that imaged the active region in the cell onto a slit in front of the photomultiplier.

Figure 2(a) shows a schematic of the optical layout of the experiment. Note that the AOM is used in a double-pass configuration where the singly diffracted beam (first order) is vertically shifted by a Porro prism and redirected back through the AOM, as shown in more detail in Fig. 2(b). The first order of the doubly diffracted beam is then picked off by the top edge of a mirror sitting below the input beam coming from the laser. This technique is used because the frequency shift acquired by the singly diffracted beam is accompanied by an angular shift that depends on the frequency. The angular shift is cancelled and the frequency shift doubled by the double-pass technique. The relevant numbers for our experiment are the travel time of the light to the Porro prism from the AOM, the smallest value of T, and the transition time of the jump itself. These are 0.42, 45, and 20 ns, respectively. It is important that the first of these be much less than the other two to maintain a two-valued frequency telegraph. The shortest value of T is that for which we could reliably maintain Poissonian statistics, though some of the data that we consider are for smaller values of T than 45 ns. The transition time of the jump is discussed at the end of the next paragraph.

The details of the random modulation technique and apparatus are the subject of a separate paper [11], which also deals with the imposition of phase fluctuations, but some pertinent points are reviewed here. The RT voltage signal is derived from photon arrivals at a photocathode (of a second photomultiplier). The advantage that this method of deriving RT signals has over other methods based on pseudorandom binary sequence generators [12] is that there is no clock frequency to discretize the times between transitions and to produce dips in the noise spectrum at harmonics of the clock frequency; i.e., the jumps that result from photon arrivals are truly random provided care is taken with the photomultiplier count rate and detection efficiency. The count rate should be low enough that the dead time does not affect the statistics, and the detection efficiency should also be low so that the counting statistics are not influenced by the emission statistics of the thermal photon source. The photocurrent pulses were amplified and routed to a "divide by two" counter whose output was the RT voltage. To control the size of the frequency jumps, this was attenuated or amplified before being fed to a varactor-tuned oscillator (VTO). The one that we used operated at about 3.2 GHz, and a second oscillator (at 3.4 GHz) was necessary to downshift the frequency fluctuating signal to about 200 MHz, this being the center frequency of the AOM. We used a VTO because they are designed for particularly rapid frequency shifting. However, the main limiting factor in the speed of the frequency jump was the finite speed of the AOM, given by how fast a change in the acoustic frequency could propagate across the laser beam.

The laser was stabilized so that the dominant laser fluctuations were those imposed electronically. The strength of the residual rms frequency fluctuations varied but was of the order of a few hundred kHz. The natural width of

the transition $3S$ to $5S$ is 2 MHz (lifetime 80 ns), although residual gas in the cell pressure broadened the transition to about 3.0 MHz, measured in terms of the laser frequency scan, or 6.0 MHz in terms of the total energy absorbed by each atom. The total size of the frequency jump was varied from about 3 to 15 MHz.

The TPA spectrum was measured for varying values of Ω, and varying dwell times T. The procedure was as follows; for a fixed value of T, ten TPA spectra were recorded for a particular Ω, along with transmission fringes of a spherical mirror Fabry-Pérot interferometer which calibrated the laser (center) frequency scan. TPA spectra with the random modulation turned off were recorded at the beginning and end of each group of ten spectra. These latter spectra enabled us to determine κ. The scan of the laser frequency was typically about 60 MHz. Dye lasers scan somewhat nonlinearly over such a small range, making the scan calibration particularly important. To get enough fringes within the 60 MHz, we exploited the off-axis (non-TEM_{00}) modes of the Fabry-Pérot interferometer (see Fig. 3). The $TEM_{n,m}$-to-$TEM_{n,m+1}$ mode spacing was measured by applying phase modulation sidebands to the laser, and adjusting the modulation frequency so that the sidebands matched the adjacent modes. This matching was easily observed with a second scanning Fabry-Pérot interferometer used as an optical spectrum analyzer. The modulation frequency and thus the mode spacing could then be read from a counter. Using the fringes we could linearize each laser scan and then form an average from the ten traces for a particular set of T and Ω. These averaged spectra are what we describe in Sec. IV.

The dwell time was measured independently of the TPA spectra by recording the distribution of frequency jumps. The size of the frequency jumps was measured from the laser power spectra, which we monitored by mixing some modulated and unmodulated light on a photodiode and viewing the beat signal on a radio frequency spectrum analyzer. To maximize the light available to the sodium cell (typically about 80 mW), we used the zeroth order of the second pass through the AOM, which was only partially modulated, for the purpose of monitoring the laser spectrum. Thus we measure directly $\Omega_s = \Omega/2$. The correctness of the relationship of this spectrum to that of the light reaching the sodium cell was checked in a separate measurement. Other diagnostics were performed on the laser; the single-frequency operation of the laser was monitored with a scanning Fabry-Pérot interferometer, and the wavelength was measured with a wave meter for coarse tuning purposes.

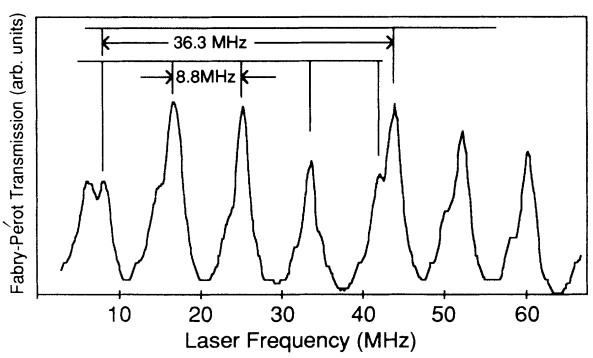

FIG. 3. Fabry-Pérot fringes used for frequency scan calibration.

IV. RESULTS AND DISCUSSION

Three typical sets of TPA data are shown in Fig. 4, along with the laser power spectrum used to obtain each. Data points are indicated by crosses. Overlaid on the TPA spectra are fits to the data using Eq. (6). These fits were used to obtain a value of b which could be compared to the expected value of 2Ω. The overall quality of the fits to the TPA data varied from excellent in the low-Ω limit, to poor at the largest values of Ω. Figure 4(a) corresponds to small Ω, Fig. 4(c) to intermediate Ω, and Fig. 4(e) to large Ω. In the large-Ω regime, where the TPA spectrum consisted of two well-resolved peaks, the determination of b can be made from the peak separation and so is unequivocal. For large Ω, Eq. (6) tended to predict peaks that were sharper and better resolved that those observed. Although in the fit shown in Fig. 4(e) the vertical scale was a poorly fitted quantity, a substantial shape change that could only come from allowing κ to be a free parameter is necessary to achieve a better fit. This is unjustified, though, since κ is an independently measured quantity. This problem with the shape remains unresolved, and we will return to it later in the paper after discussing the laser spectra.

The value of Ω was obtained from fitting Eq. (5) to the laser power spectrum data [Figs. 4(b), 4(d), and 4(f)]. In each of these graphs, two theoretical curves are shown with the data, which is labeled "1." The first of these, labeled "2," is the fit of Eq. (5) to the data, which of course were obtained from the singly diffracted (but double-passed) beam from the AOM. The amplitude of the random frequency jump thus measured, Ω_s, is half of that experienced by the sodium atoms. Note that the data are somewhat asymmetric; this arises because of the finite transition time of the random frequency jump. This results in the singly diffracted light being somewhat nonuniform in frequency across the beam. The spectrum is in fact symmetric at the center of this beam, and also across the doubly diffracted beam, but the observed symmetry in the singly diffracted beam is very sensitive to small misalignments of the optics involved in the laser spectrum measurement. We circumvented this problem by aligning the optics as best we could so that the asymmetry was evident only in the wings. Then we converted from the log scale in which form the data were supplied by the spectrum analyzer, to a linear scale, so that for the purpose of fitting the asymmetry in the wings was unimportant. The theoretical curves labeled "3" are the inferred spectra of the light reaching the sodium atoms after being twice diffracted by the AOM. In Fig. 4(f) the asymmetry in the laser spectrum shown is more obvious than in other cases, but this is unlikely to explain the poor fit in Fig. 4(e) since the asymmetry is not present on the doubly modulated beam. In each fit the value of T was obtained by independent measurement, as was the

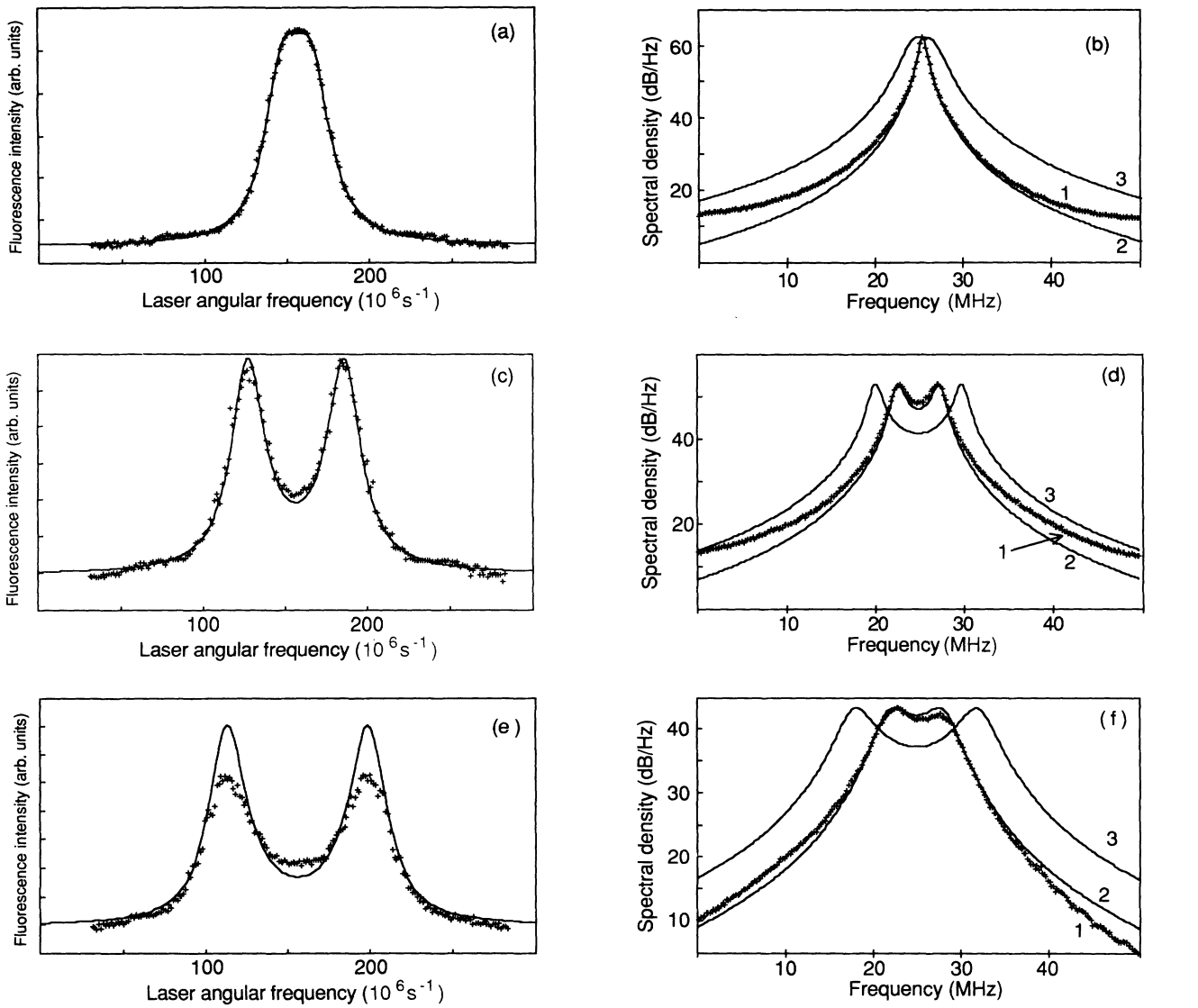

FIG. 4. Two-photon absorption spectra [(a) and (c)] and laser power spectra [(b) and (d)] with RT frequency. (a) and (b) $T=115$ ns, $\Omega_s=5.83\times10^6$ s^{-1}, $b=23.9\times10^6$ s^{-1}, and $\kappa=39.8\times10^6$ s^{-1}. (c) and (d) $T=240$ ns, $\Omega_s=15.1\times10^6$ s^{-1}, $b=60.7\times10^6$ s^{-1}, and $\kappa=41.8\times10^6$ s^{-1}. (e) and (f) $T=89$ ns, $\Omega_s=22.9\times10^6$ s^{-1}, $b=87.3\times10^6$ s^{-1}, and $\kappa=40.4\times10^6$ s^{-1}. In the laser spectra the IF bandwidth of the spectrum analyzer was 100 kHz.

value of κ for the TPA spectra.

The data obtained are summarized in Fig. 5, where the fitted values of b are plotted against the fitted values of Ω_s. Since this latter quantity is obtained from the singly diffracted laser beam, we expect that $b=4\Omega_s$. This is shown by the straight line of gradient equal to 4 on the graph. Representative error bars are shown; generally the uncertainties are smallest for intermediate values of b and Ω_s, where the shape of the spectra tends to be single peaked with a broad flat top, and the fits are excellent. The apparent trend of the data for $T=86$ ns (squares) to fall below the line at small Ω_s is possibly significant, although the size of the error bars is such as to make each data point consistent with $b=4\Omega_s$. However, the data shown as triangles consistently fall below the line $b=4\Omega_s$. These were obtained with $T=29.3$ ns. Generally we found that for $T<45$ ns the statistics of the RT process deviated significantly from Poissonian, though in this case, using the criterion mean equals variance, we found that the statistics for $T=29.3$ ns were quite close to Poissonian. A more likely explanation of the behavior of the data as T is reduced is that the rise time of the frequency jump ($\sim$20 ns) becomes comparable to T. This introduces a significant non-Markovian character to the stochastic process, and it is reasonable to expect that the simple theory of Sec. II would begin to break down. However non-Markovian effects, at least of this type, are not likely to explain the poor fits of the theory to the data for larger frequency jumps; the robustness of the agreement between the data and the line $b=4\Omega_s$ is noteworthy in this case. [The data of Fig. 4(e) give the point labeled "A" in Fig. 5.] This experiment suggests two directions

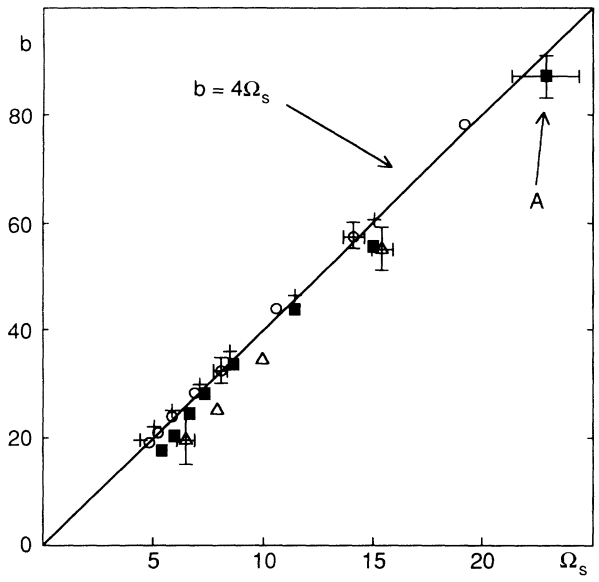

FIG. 5. Summary of fitted values of b vs Ω_s; crosses, $T=240$ ns, $\kappa=41.8\times10^6$ s^{-1}; circles, $T=115$ ns, $\kappa=39.8\times10^6$ s^{-1}; squares, $T=86$ ns, $\kappa=40.4\times10^6$ s^{-1}; triangles, $T=29.3$ ns, $\kappa=39.6\times10^6$ s^{-1}.

that could be pursued to resolve these discrepancies. The first would be to develop a model for the RT frequency that takes account of the finite rise times. The second would be a *direct* measurement of the frequency jump on the actual laser field to see if perhaps the cause does lie in the modulation process for large-frequency jump amplitudes–i.e., our characterization of the field rests on measurements of the laser power spectrum together with a knowledge (to some extent inferred) of the random noise process. The latter is perhaps the weak point in that it relies on our understanding of the modulation process.

That the two-photon absorption process accentuates the size of the laser-frequency jump admits of a fairly simple heuristic explanation; the peaks of the absorption spectrum correspond to one or the other of the two laser frequencies being two-photon resonant. Because it is a two-photon transition the center frequency of the laser must be tuned by twice the peak separation of the laser power spectrum. These data can be compared with TPA data obtained for the case of the phase-diffusing field [1], i.e., where the probability distribution of the frequency is Gaussian. In the limit of short correlation time of the laser noise, the TPA spectrum and the laser spectrum are both Lorentzian, but the TPA spectral width scales as four times that of the laser spectrum. This behavior is clearly similar to that observed here with RT frequency (see Fig. 5), but the heuristic explanation above breaks down when the correlation time in the phase-diffusing case gets longer and the scaling factor then becomes closer to 2. Although this is actually predicted by Mollow's theory, an intuitive explanation remains elusive which emphasizes the difficulties of forming a physical understanding of phenomena that depend on higher-order coherences.

In summary, we have measured two-photon absorption spectra with a laser beam which had random telegraph frequency fluctuations. The measurements confirm predictions that the TPA process accentuates the size of the frequency jump, and general agreement was found between the shape of the TPA spectra and that expected from the parameters of the laser noise. Fits of theoretical functions to the data gave good agreement in the regime of small-frequency jumps, but became progressively poorer as the size of the jump increased.

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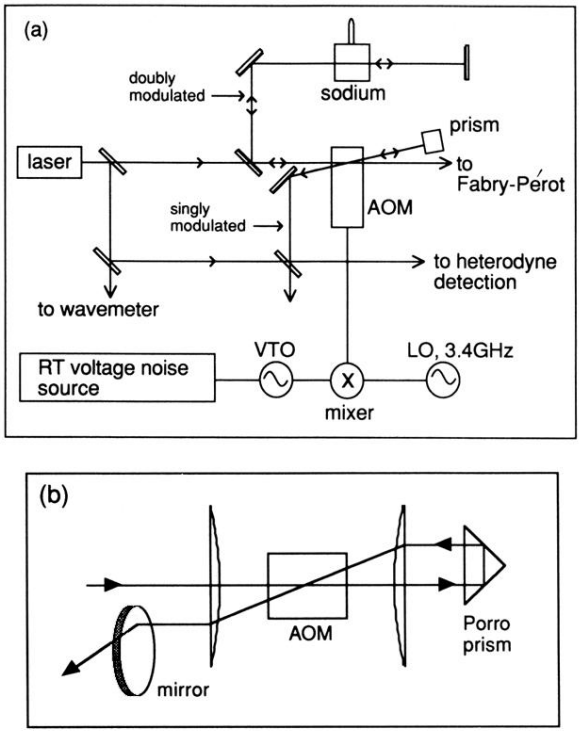

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