Two-Photon Absorption from a Phase-Diffusing Laser Field

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We report the first quantitative measurements of the effect of a phase-diffusing laser field on a nonlinear optical interaction. Using a nearly Lorentzian laser power spectrum, we have measured the spectral linewidth of an unsaturated two-photon absorption process. We find that the measured width scales as four times the laser width, in agreement with the theoretical predictions of Mollow.

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In 1968, Mollow[1] presented second-order perturbation-theory calculations concerning the simultaneous absorption of two photons from a constant-amplitude single-mode laser field undergoing random (Gaussian) frequency fluctuations. Agarwal[2] later generalized these calculations to the case of *n*-photon absorption. This model of the laser field is known as the phase-diffusion model, because the statistical behavior of the phase, $\phi(t)$, is analogous to that of the position of a particle undergoing Brownian motion (the random-walk process). Specifically, Mollow and Agarwal made use of the lowest-order correlation function of the field frequency $[\dot{\phi}(t)=d\phi/dt]$

$$\langle \dot{\phi}(t)\dot{\phi}(t+\tau)\rangle = 2b\delta(\tau), \qquad (1)$$

corresponding to a Lorentzian laser power spectrum with full width at half maximum (FWHM) $2b$. The angle brackets $\langle\,\rangle$ denote an ensemble averaging process, and $\delta(t)$ is the Dirac delta function. The predicted absorption profiles have some interesting properties which are not easily recognized intuitively. One of these is the prediction that the Lorentzian absorption profile has a FWHM that is wider by the factor n^2 than the Lorentzian spectral profile of the laser beam. The absorption width is given in terms of the transition frequency, equal to n times the laser frequency. We report an experimental test of this prediction for the two-photon case. This study is the first rigorous experimental investigation of the role of random optical frequency fluctuations on a nonlinear optical process. This work was done in the weak-field limit where saturation effects can be ignored.

For this study, we have developed a method of imposing well-characterized frequency fluctuations onto a highly stabilized cw single-mode laser beam using acousto-optic and electro-optic modulators.[3] To conform to the theoretical model, an essential requirement of the experiment is that the frequency fluctuations be a true Gaussian process. We have used the nearly white-noise output of a shot-noise diode, for which we have carefully verified the Gaussian properties. The Gaussian voltage distribution is converted to a Gaussian frequency distribution by standard frequency and phase-modulation techniques. The laser power spectrum is tailored through careful balancing of the several modulating elements. The unmodulated laser beam has rms frequency fluctuations of approximately 150 kHz, ensuring that the externally imposed fluctuations are dominant.

Since the δ-correlated fluctuations of Eq. (1) are somewhat unphysical (they lead to a truly Lorentzian laser power spectrum which has an unrealistic level of power in the far wings), the correlation function

$$\langle \dot{\phi}(t)\dot{\phi}(t+\tau)\rangle = b\beta e^{-\beta|\tau|} \qquad (2)$$

has been used in several recent theoretical works.[4,5] The laser power spectrum resulting from this modified frequency correlation function approaches a Lorentzian profile as β is made large ($\beta >> b$). At the other limit ($\beta << b$) the spectrum is Gaussian.[6] The parameter β corresponds to a limit on the rapidity with which frequency fluctuations may occur. The parameter b is the same as in Eq. (1). This modified correlation function is built into our synthesized laser power spectrum by introducing a low-pass filter with an *RC* rolloff on the output of the shot-noise diode.

We have used this technique to investigate the effect of such fluctuations on the absorption profile of a two-photon Doppler-free transition of atomic sodium, $3S(F=2)\rightarrow 5S(F=2)$, for nearly Lorentzian laser power spectra having widths (FWHM) ranging up to 14 MHz and laser line shapes controlled to 1 GHz from line center. The essential difference between our experimental setup and those of earlier two-photon Doppler-free exper-

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iments[7] is in our ability to control the frequency fluctuations. The laser beam, tuned to 602 nm, was passed through a sodium absorption cell ($\sim{}^{9}$ atoms cm^{-3}) and then retroreflected so that an atom in the interaction region may absorb one photon from the incident laser beam and one from the reflected beam. The 330-nm radiation resulting from radiative decay of the $5S$ state by way of the $4P$ state, emitted at right angles to the laser beams, was detected by a photomultiplier. The Doppler-free signal was seen as a narrow spike superposed on a very low-level Doppler-broadened background. In our work, the linearly polarized 70-mW laser beams were of ~ 1.3 mm diam to limit broadening due to the transit time of atoms through the laser beam. Minimum-absorption profile widths of approximately 4 MHz were obtained which were due to the natural lifetime of the $5S$ state (80 ns) modified somewhat by collisions with background gas.

With a given synthesized laser power spectrum of FWHM $\Delta\nu$, the laser frequency was scanned over a range of about $10\Delta\nu$ in 256 steps. A minicomputer was used to control the laser frequency scan and record the number of 330-nm fluorescence photons detected for each frequency step of the scan. The maximum count rate was approximately 10 kHz, and between 1 and 500 counts were accumulated in each channel. We simultaneously monitored and recorded the intensity of the laser beam as well as frequency markers from a near-confocal Fabry-Perot interferometer with mirrors spaced by 45 cm. The laser power spectrum was monitored by spectrum analysis of the output of a fast avalanche photodiode on which the modulated and unmodulated laser beams were mixed.

Absorption profiles for the two-photon process were obtained for Lorentzian laser power spectra for widths ranging from 1 to 14 MHz ($\beta/2\pi = 100$ MHz and $0.5 \text{ MHz} < b/2\pi < 7$ MHz). We have generalized the predictions of Mollow to include fluctuations which follow the exponential correlation function [Eq. (2)], and find that in this regime the δ-correlated [Eq. (1)] and exponential correlation functions lead to essentially identical predictions for the absorption profile.

We have averaged the widths measured from about ten profiles obtained for each given laser linewidth. In Fig. 1 we have plotted the absorption widths obtained in this way against the width of the laser power spectrum. These measurements may be compared with Mollow's predictions, represented by the solid line of slope 4 plotted in Fig. 1. The intercept is the only adjustable parameter and was chosen to accommodate the measured absorption width in the absence of the imposed frequency fluctations. The agreement between experimental results and the theoretical line is quite good.

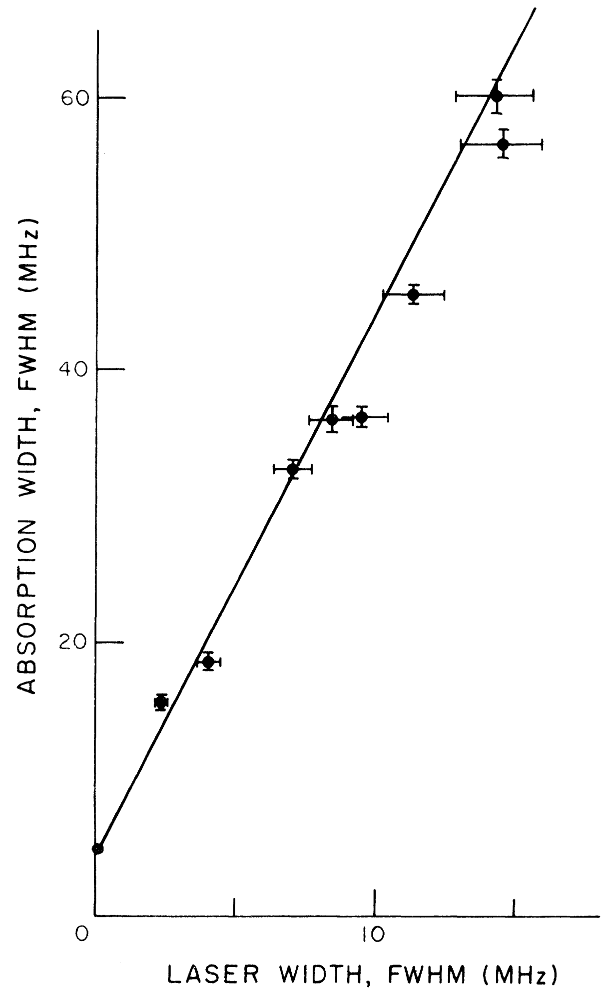

FIG. 1. Measured absorption widths (FWHM) are plotted against FWHM of the Lorentzian laser field. Vertical bars represent one standard deviation of the mean from about ten measurements of the absorption profile. Horizontal bars represent a calibration uncertainty of the laser linewidth determination. The predictions of Mollow (Ref. 1) are shown by the solid line.

This experiment is the first rigorous test of the effect on a nonlinear optical transition of the laser field undergoing phase diffusion. We are also studying other correlation effects in the two-photon process which will be reported in detail elsewhere. In light of the results presented here, it would be very interesting to investigate other closely related processes to which the phase-diffusion model has been applied theoretically. These include three-photon absorption[2] and two-photon resonant, three-photon ionization.[5,8]

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