Experimental study of absorption and gain by two-level atoms in a time-delayed non-Markovian optical field

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We have measured the absorption from a weak time-delayed probe field by a two-level atomic system [Na $3s_{1/2}(F=2,m_F=2)\rightarrow 3p_{3/2}(F=3,m_F=3)$] saturated by a phase-diffusing pump field. The pump and probe, derived from the same artificially-noise-modulated phase-diffusing laser beam, are frequency degenerate and resonant with the atomic transition. The probe is a time-delayed replica of the pump. Our results, carried out in a regime where the field bandwidth and the Rabi frequency are comparable, provide confirmation of recent theoretical predictions [K. Gheri, M. A. M. Marte, and P. Zoller, J. Opt. Soc. Am. B **5**, 1559 (1991)] for the atomic response to the non-Markovian composite of a pump and time-delayed probe, valid for arbitrary bandwidths and intensities. Amplification of the probe field is observed for delays comparable to the lifetime of the upper level and again for very long delays, if the bandwidth is less than the decay rate of the upper state.

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I. INTRODUCTION

The atomic response to Markovian stochastic optical fields is now largely well understood. Methods exist to calculate the first-order moments of the atomic density-matrix elements for a wide variety of field statistics, including phase-diffusing fields [1–5], phase-diffusing fields with colored noise [2,6,9], chaotic fields [5–9], real Gaussian fields [8,9], and phase jump fields [3,10,11]. These methods are valid for arbitrary fields strengths and bandwidths and properly account for all orders of electric-field correlations. It is generally straightforward to extend these methods to include products of density elements, useful for calculating observables such as the variance of fluorescence intensity or the intensity of four-wave-mixing signals [3,4,13,14]. In these works, colored noise models, which incorporate a memory into the process and are non-Markovian, can be reduced to Markovian processes by an extension of the number of dimensions in the problem [2].

In what follows we consider a two-level atom interacting with a single broadband laser pulse of length τ_p, Rabi-frequency Ω, and correlation time τ_c. If a cw laser is employed, τ_p is replaced with the lifetime of the upper state κ^{-1}. There are three primary regimes of interest in the atomic response to broadband fields which we refer to as (i) weak-field regime $\Omega\tau_c \ll 1$ and $\Omega^2\tau_c\tau_p \ll 1$, (ii) depletion regime $\Omega\tau_c \ll 1$ and $\Omega^2\tau_c\tau_p \geq 1$, and (iii) strong-field regime $\Omega\tau_c \geq 1$ and $\Omega^2\tau_c\tau_p \geq 1$. This paper deals with the response of two-level atoms to time-delayed *non-Markovian* fields in the strong-field regime.

To understand the meaning of these conditions it is helpful to consider the broadband laser pulse as a train of narrow noise pulses of width τ_c. The change in the angle of the Bloch vector after each noise pulse is then $\Omega\tau_c$. Since each noise pulse is uncorrelated with the next, subsequent pulses perturb the Bloch vector in the fashion of a random walk and the net displacement of the vector grows as the square root of the number $(N=\tau_p/\tau_c)$ of noise pulses. Hence, the second condition for the weak-field regime rewritten as $\Omega\tau_c\sqrt{\tau_p/\tau_c} \ll 1$ implies there is no population depletion of the lower state during the laser pulse τ_p. The condition $\Omega\tau_c \ll 1$ indicates no population transfer occurs for each narrow noise pulse. Since we are considering broadband fields $(\tau_c \ll \tau_p)$ the second condition for weak fields guarantees the first. In the weak-field regime, a perturbative solution of the atomic density matrix suffices, and the expression for signal intensity generally reduces to integrals over the field correlation functions, which are, in principle, known. In the depletion regime, population transfer can accumulate only over many successive noise pulses. Since the atomic response is small over a time scale given by τ_c, atomic variables can be decorrelated from field variables, simplifying the derivation of ensemble-averaged stochastic Bloch equations. A system of rate equations results. In strong fields, significant population transfer can occur in each fluctuation of the field. For strong fields, the atomic density elements obey a set of stochastic differential equations that must be averaged without any decorrelation approximations.

The atomic response to non-Markovian fields is much less well understood. This is primarily because the complete hierarchy of conditional probabilities must be known in order to describe a non-Markovian process. In contrast, for a Markovian process only the lowest-order conditional probability and probability density are required since all others can be constructed from them [12,13]. As mentioned, some non-Markovian processes can be made Markovian by extension to higher dimensions. In particular, if the governing differential equations of the system can be rewritten as an expanded but *finite* system of first-order differential equations that depend on a finite number of independent, δ-function correlated stochastic variables (i.e., Langevin equations), then the system can be reduced to a Markov process [12,13]. To illustrate the problem for time-delayed driving fields, consider a process that depends on a stochastic variable $\epsilon(t)$ and its time-delayed replica $\epsilon(t-\tau)$. One might be led to consider a Taylor expansion of $\epsilon(t-\tau)$ about t,

$$\epsilon(t-\tau)=\sum_{n=0}^{\infty}\frac{(-\tau)^n}{n!}\frac{d^n\epsilon(t)}{dt^n},$$

and define the new variables $\nu_n(t)=d\nu_{n-1}(t)/dt,\ldots,\nu_0(t)=d\epsilon(t)/dt$. This reduces the original systems to an *infinite* system of first-order equations. Hence, the system is fundamentally non-Markovian and resistant to solution by standard methods.

The problem of atomic response to multiple time-delayed non-Markovian fields has been attacked in earnest in the last decade. Early studies focused on weak fields where both $\Omega\tau_c \ll 1$ and $\Omega^2\tau_c\tau_p \ll 1$ [15–22]. Under weak-field conditions, the atomic system can be thought of as undergoing free precession during the delay time τ between a noise pulse from the first laser field and its correlated twin on the second laser field. The total effect is then a sum over all of the correlated noise pulse pairs. Expressions for observables reduce to integrals over the composite field correlations, which depend on the delay. Experiments and calculations in this regime have demonstrated these techniques to be practical for the measurement of fast transverse [16–19] and longitudinal [20] relaxation rates as well as inhomogeneous dephasing rates [22].

Some experiments in the weak-field regime have been reported with cw lasers [16,42]. In the work of Elliott *et al.* [42], phase-diffusing statistics were imposed on a frequency stabilized laser (as in this work) and the effects of field correlations were studied in a two-photon transition in sodium. The emission linewidth was measured as a function of the time delay between the counterpropagating fields. The line shapes could, again, be related to integrals over the field correlations (fourth order in the case of a two-photon transition) due to the weak exciting fields.

In the depletion regime one or more of the fields is strong, $\Omega^2\tau_c\tau_p \sim 1$, and significant depletion of the lower state can occur over the laser-pulse duration τ_p. However, the condition $\Omega\tau_c \ll 1$ still holds so the depletion must be accumulated over many noise pulses. In this regime there can be significant diffusion of the Bloch vector over the delay interval and so the assumption of free precession fails. This leads to a degradation of the population grating [23,24] and reduction of signal with increasing τ. The effect of the time delay on the field correlation function $\langle E(t)E^*(t')\rangle$ is to add peaks located at $\pm\tau$. If no limit is set on τ this prevents decorrelation of the atomic variables and field variables. In the work of Tchénio *et al.* [25,26], and independently Friedberg and Hartmann [29], it was shown that the averaged atomic density elements can be expanded in a hierarchy of couplings between the field and atomic variables. Under the conditions for the depletion regime, this series can be resummed giving the system evolution in terms of propagators. The method has also been applied to the product of density-matrix elements [27], required for the calculation of coherent transient signals.

If the delay between the fields is small, $\Omega^2\tau_c\tau \ll 1$, the decorrelation of atomic variables and driving field again becomes valid and rate equations for atomic variables are obtained. Despite the severe restriction on τ it has been shown by Tchénio *et al.* that, in the limit $\Omega^2\tau_c\tau_p=\infty$ for one field with the other field weak, coherent transient signals as a function of τ contain a direct dependence on τ_c [28]. In a detailed study, Finkelstein and Berman have extended these results to include finite field strength, pulses with unequal and equal amplitudes, and homogeneous and inhomogeneous broadening mechanisms [30]. In addition to a dependence on τ_c, they find that the coherent transient signals depend on a time scale given by $\tau_c/(\Omega^2\tau_c\tau_p)^{1/2}$ which can be much less than τ_c. This effect has been found to be much more prominent in three-level systems [31].

No general methods are known for calculating the response of atoms to time delayed non-Markovian fields in the strong-field regime. However, Monte Carlo techniques have proven a powerful method for numerical simulation of atomic response to non-Markovian fields [32]. In this method, a large sample of stochastic realizations of the atomic density-matrix elements are obtained by time-step integration of their governing differential equations over stochastic realizations of the input field. Observables are constructed from the resulting realizations of the stochastic density elements and averaged. The time delay is accounted for in the construction of the stochastic realizations of the input field.

Recently, Gheri, Marte, and Zoller [33] (hereafter referred to as GMZ) have proposed a method to calculate the atomic response to time-delayed optical fields that is valid for arbitrary values of the Rabi frequency and field bandwidth τ_c^{-1}, but requires the time-delayed field (probe) to be weak relative to the strong field (pump). The theory considers the perturbative effects of the probe using a propagator method, where the propagator is derived from the optical Bloch equations under the action of the stochastic pump alone. The method was applied to the case of probe absorption due to atoms saturated by the strong pump. This configuration is very similar to the case of absorption with monochromatic fields, considered by Mollow [34], and experimentally verified by Wu *et al.* [35]. As is well known, amplification of the probe field was predicted and observed in these works. Later, Lezama *et al.* demonstrated lasing in a similar configuration, which was attributed to inversion in the dressed-state basis even though there was no inversion in the atomic states [36]. For the pump on resonance, Mollow [34] predicted amplification of the probe field for pump-probe detunings δ that satisfy $\kappa^4/4\Omega^2<\delta^2<\Omega^2$, where κ is the radiative decay rate for the upper state. In particular, no gain is expected for degenerate pump and probe. In the work of GMZ, gain *is* predicted in the strong-field regime for degenerate fields ($\delta=0$) provided there is a time delay between the phase-diffusing pump and probe of either $\tau\sim0.8\kappa^{-1}$ or $\tau\geq5\kappa^{-1}$, the latter case describing uncorrelated fields. Both phase noise and a relative delay are required in order to observe gain. In strong fields, the field bandwidth $2b$ satisfies $2b \sim \Omega$ and the pump must saturate the transition. Currently, the method of GMZ is the only one valid for the calculation of atomic response to non-Markovian fields in this regime. Gain is not predicted for the broadband limit corresponding to the weak-field regime. Gain is predicted for the depletion regime, but the overall absorption is too small for it to be readily observable.

Our experiment is designed to provide a rigorous test of the work of GMZ. We have used the method of Elliott and Smith [37], utilizing a system of extra-cavity electro-optics to impose phase-diffusing statistics on the output of a single-mode, frequency stabilized ring dye laser. Absorption of a probe laser, time-delayed relative to a saturating pump, has been measured as a function of both the timed delay τ and the laser bandwidth $2b$.

In the next section we discuss the method used to modify the results of GMZ for the effects of Doppler broadening and variation of the Rabi frequency over the atoms in the interaction region. In Sec. III the method of the experiment is discussed and finally, the results and conclusions are presented in Sec. IV. A simple physical explanation of the existence of gain is also presented.

II. REVIEW OF THE THEORY

In this section we will review the problem considered by GMZ and discuss corrections for Doppler broadening and laser field profiles. A two-level system is assumed. The atoms are assumed to be driven by an electric field of the form

$$E(t,\mathbf{x})=\xi(t)e^{-i\omega t-i\mathbf{k}_1\cdot\mathbf{x}}+\lambda\xi(t-\tau)e^{-i\omega t-i\mathbf{k}_2\cdot\mathbf{x}}+\text{c.c.} \quad (1)$$

with the complex amplitude

$$\xi(t)=\tfrac{1}{2}\xi_0 e^{-i\varphi(t)} .$$

In these expressions, λ is a small expansion parameter, $\varphi(t)$ is the Gaussian fluctuating phase, and τ is the time delay. GMZ also consider a detuning δ between the pump and probe. In this work we take $\delta=0$. The phase is assumed to follow pure phase-diffusing statistics giving an autocorrelation function for the instantaneous frequency

$$\langle \dot\varphi(t)\dot\varphi(t+\tau)\rangle = 2b\delta(\tau) , \quad (2)$$

which indicates a white-noise or δ-correlated process. In what follows, $\Omega=\mu\xi_0/\hbar$ is a Rabi frequency associated with the pump, with μ the usual dipole moment, and $\Omega'=\lambda\Omega$ is the Rabi frequency of the probe. GMZ have calculated the absorption of the probe field

$$W'=\left\langle -i\frac{\Omega'^*}{2}e^{i\varphi(t-\tau)}\delta\alpha_+(t)\right\rangle+\text{c.c.} , \quad (3)$$

which describes the work done by the probe on the induced polarization [34], and where $\delta\alpha_+(t)$ are the off-diagonal atomic density-matrix elements, perturbed by the action of the probe, that multiply the phase factors $e^{-i\omega t-i\mathbf{k}_2\cdot\mathbf{x}}$ in the complete expression for the polarization induced by the composite field. The brackets indicate an average over the fluctuating phase. The matrix element $\delta\alpha_+(t)$ depends on Ω and the detuning from the atomic transition frequency $\Delta=\omega_0-\omega$, giving $W'=W'(\Omega,\Delta)$. When the atoms fill a spatial volume of finite extent, Ω becomes a function of position. The absorption is proportional to the probe intensity so the average over different Rabi frequencies is accomplished by multiplying $W'(\Omega(\mathbf{x}),\Delta)$ by the Gaussian intensity profile of the probe and averaging over the area

$$\langle W'(\Delta)\rangle_r=\frac{\int_0^\infty W'(\Omega(r),\Delta)e^{-\alpha r^2}2\pi r\,dr}{\int_0^\infty e^{-\alpha r^2}2\pi r\,dr} \quad (4)$$

with

$$\alpha=\frac{4\ln(2)}{\delta_{\text{probe}}^2}$$

and

$$\Omega(r)=\Omega_0\exp\left[-\frac{2\ln(2)}{\delta_{\text{pump}}^2}r^2\right] ,$$

where δ_{pump} and δ_{probe} refer to the full width at half maximum (FWHM) of the intensity profiles of the beams. Note that $\Omega(r)$ is proportional to the square root of the pump intensity profile.

The Doppler average is a convolution over the Doppler velocity profile of the atoms. In this experiment, the pump laser is tuned to the center of the atomic resonance so a given velocity group sees a laser detuning of $\Delta=kv$, where v is the velocity component of the atoms in the direction of the k vector of the pump beam. The averaging over velocity groups reduces to a one-dimensional average of the form

$$\langle\langle W'\rangle_r\rangle_v=\frac{\int_{-\infty}^{+\infty}\langle W'(kv)\rangle_r e^{-av^2}dv}{\int_{-\infty}^{+\infty}e^{-av^2}dv} , \quad (5)$$

where again,

$$a=\frac{4\ln(2)}{\delta_v^2} ,$$

and δ_v is the width of the Doppler distribution of velocities. These integrals were approximated numerically with a midpoint rule using values of $W'(\Omega,\Delta)$ obtained by the methods of GMZ.

III. EXPERIMENT

The method of extra-cavity noise modulation (ECNM) used to produce the phase-diffusing field at the output of a cw ring dye laser has been reported in detail [37] and has been used in a number of experiments designed to study the nonlinear dependence of atomic response to phase-diffusing fields [38–42]. Here, we will only discuss the form of the phase-diffusing field produced with this system. The ECNM imposes Gaussian phase fluctuations $\varphi(t)$ and corresponding frequency fluctuations $\dot\varphi(t)$, which are described by the autocorrelation function

$$\langle \dot\varphi(t)\dot\varphi(t+\tau)\rangle = b\beta e^{-\beta|\tau|} .$$

For $\beta \rightarrow \infty$ this becomes pure phase diffusion given by Eq. (2). In these expressions β is the inverse of the correlation time for the frequency fluctuations and the variance of the frequency fluctuations is given by $b\beta$. When β^{-1} is much smaller than any other time scale in the system, a good approximation can be obtained by assuming a pure phase-diffusing field, i.e., δ-correlated frequency fluctuations. This is the case in this experiment where $\beta/2\pi = 100$ MHz throughout, and $\kappa/2\pi$, $\Omega/2\pi$, and $b/2\pi$ are 10 MHz, <30 MHZ, and <10 MHz, respectively. It has been shown [37] that the laser spectrum is Gaussian with FWHM given by $\sqrt{8b\beta \ln 2}$ for $\beta \ll b$, and Lorentzian with FWHM $=2b$ for $\beta \gg b$. The latter case applies here.

A schematic of the experiment is given in Fig. 1. The $3s_{1/2}(F=2, m_F=2) \rightarrow 3p_{3/2}(F=3, m_F=3)$ transition in atomic sodium was chosen for the two-level system. This transition has a convenient natural linewidth $\kappa/2\pi = 10$ MHz, with respect to the experimentally attainable values of the bandwidth b (up to ~ 10 MHz). Doppler broadening and collisions were reduced by the use of an atomic beam apparatus.

After passage through the ECNM system, the laser beam was split into a weak beam (probe) and strong beam (pump) with an uncoated beam splitter. The pump beam was passed through a fixed 3.26 m length of single-mode optical fiber which served primarily as a spatial filter. The fiber provided a time delay of $1\kappa^{-1}/3.26$ m. Time delays of $-7/8\kappa^{-1}$ to $5\kappa^{-1}$ were obtained by inserting varying lengths of fiber in the optical path of the probe. The use of optical fiber for the time delay provided high-quality intensity profiles for each beam at the fiber outputs. This made data collection tedious, but insured that signal variations, as the optical path length increased, were due to a time delay and not to degradation of the beam profiles and spatial overlap.

Photodiodes monitored portions of each beam at the outputs of the fibers, and the corresponding signals were processed and fed back to acousto-optic modulators (AOM), which actively stabilized the intensity of the beams. One AOM derived its error signal from the pump beam and also provided the lower end of the frequency fluctuations in the ECNM system. A separate AOM was used for the probe beam alone. Intensity stabilization was necessary to eliminate noise due to fluctuations in coupling into the fibers and also low-frequency intensity noise on the dye laser, which was at the 5% level. Stabilization also insured a constant laser power over the long period of data collection. Low-frequency intensity fluctuations served to wash out Rabi oscillations in the atomic response similar in effect to a variation of Rabi frequencies across the interaction region due to overlapping beam profiles. To reduce the latter effect, the waist (0.044 cm) of the probe beam at the interaction region was a factor of 5.16 smaller than the waist (0.227 cm) of the pump. By the methods of the previous section, the resulting intensity variation was predicted to have only a small effect. The peak intensity of the probe beam was measured to be about 1.5% that of the pump.

The probe beam was further split on a beam splitter with 30% reflecting surfaces into the actual probe beam and a stronger reference beam. Alignment was such that the reference beam traveled parallel to the probe, but did not pass through the atom beam. Signal extraction was accomplished by subtraction of the relative intensities on a photodiode subtraction circuit suggested by Hobbs [43] that uses active feedback to balance the photocurrents produced by the reference and probe beams. The extra noise cancellation of this circuit (~ -35 db over the detector bandwidth of ~ 200 kHz) was necessary since the maximum absorption was a factor of $\sim 10^{-4}$ smaller than the probe intensity.

A detail of the interaction chamber is given in Fig. 2. The single largest source of noise in the experiment was due to pump light scattered directly into the spatial mode defined by the probe, giving rise to interference effects on the probe detector. This worsened when there was significant overlap of the pump and probe beams on ele-

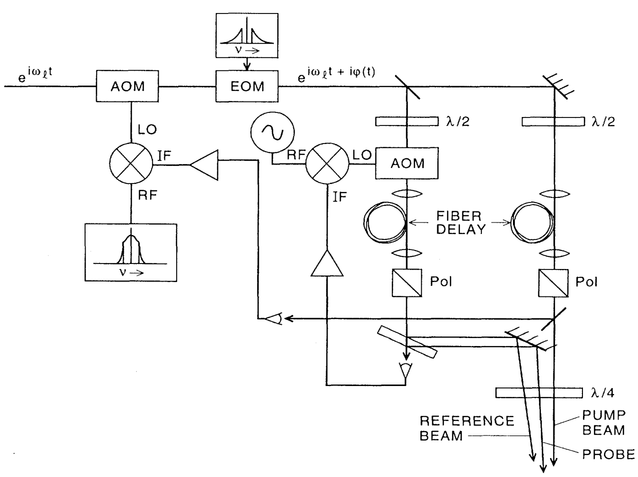

FIG. 1. Schematic of the experiment. AOM denotes an acousto-optic modulator and EOM denotes an electro-optic modulator. RF, LO, and IF denote the radio-frequency, local oscillator, and intermediate ports of a mixer.

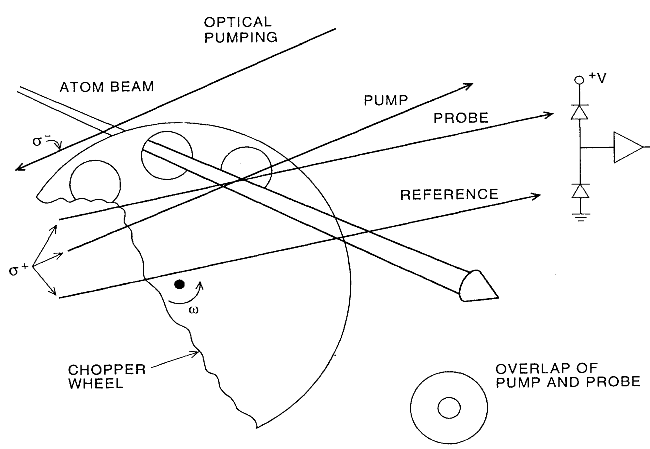

FIG. 2. Schematic of the interaction region. The reference beam travels parallel to the probe but does not intersect the atom beam.

ments in the optical train, such as the entrance windows on the vacuum chamber, and required the pump and probe to converge at an angle of about 20 mrad. The plane of the beams was orthogonal to the propagation direction of the atomic beam. Upstream from the interaction region, the ground state of the atoms was prepared with a counterpropagating (to reduce scattered light) σ^+ polarized optical pumping beam [44]. The pumping was done with an *FM* sideband, which, by use of an offset locking technique, also served to lock the dye laser to the transition and control the detuning [41]. The atom beam was chopped at 130 Hz with a chopping wheel placed between the optical pumping and interaction regions. This provided for direct phase-sensitive detection of the absorption signal. The Earth's magnetic field was nulled to < 10 mG with a system of Helmholtz coils and a magnetic field of 0.5 G was applied in the direction of the k vector of the pump beam to preserve alignment of the magnetic sublevels. Efficiency of the optical pumping was demonstrated by the reduction of the $3s_{1/2}(F=2)\rightarrow 3p_{3/2}(F=2)$ weak-field absorption signal to a value less than the contribution of the wings of the $3s_{1/2}(F=2)\rightarrow 3p_{3/2}(F=3)$ transition.

The atomic beam emanated from an oven held to about 284 °C and had a collimation ratio of about 160:1. The transverse Doppler broadening was measured to be 0.95κ. From weak-field absorption measurements ($\sim 1\%$) we estimate $\sim 6\times 10^4$ atoms were under observation in the prepared ground state.

Data collection consisted of detecting the absorption signal with a lock-in amplifier set to a 10-s integration time. The value with the phase noise on was divided by the value with the noise off, compensating for drifts in the oven temperature and atom beam density over the measurement interval. This significantly reduced scatter but introduced a small systematic error since the laser with phase noise off is not purely monochromatic. Data points were taken for the time delays of $-7/8\kappa^{-1}$ to $5\kappa^{-1}$ in steps of $0.25\kappa^{-1}$. The value for b was obtained from heterodyne scans of the noise modulated laser with portions of the unmodulated laser used for a local oscillator. The power in the pump beam was monitored with a calibrated thermopile detector (with an estimated accuracy of about 5%) and used to calculate the Rabi frequency. Amplification of the probe beam resulted in a 180° phase shift of the absorption signal relative to the atom beam chopping frequency and was observed simply as a negative reading from the lock-in amplifier.

IV. RESULTS AND DISCUSSION

In Fig. 3 we show results of probe absorption as a function of the time delay τ (in units of κ^{-1}). The data are the crosses and the solid line is a theoretical prediction of GMZ using the experimental parameters. Typically, five data points were taken for each delay. The best fit occurred with a value of the Rabi frequency of $\Omega=2.62\kappa$ and $b=0.78\kappa$ which was well within the uncertainty for the Rabi frequency determined from measured pump power, and b determined from heterodyne scans. The theory was calculated with corrections for Doppler averaging using the measured Doppler width of 0.95κ and spatial intensity averaging using a pump-probe beam waist ratio of 5.16. At each delay, the measured absorption was divided by the absorption measured with the phase noise off. To account for this in the calculations, the absorption profile was divided by an absorption profile calculated for the estimated residual laser bandwidth of $2b=0.06\kappa$. This served to normalize the calculations in the same way the data were collected and set an overall scale factor. The profile calculated for the 60 kHz unmodulated laser linewidth varies slightly more than the scatter in our data. For a larger correction one should take into account the finite correlation time of the frequency fluctuations in the unmodulated dye laser. The Doppler and intensity averaging also affected the shape of the profile only slightly, but reduced the overall amplitude of the oscillation by about 30%.

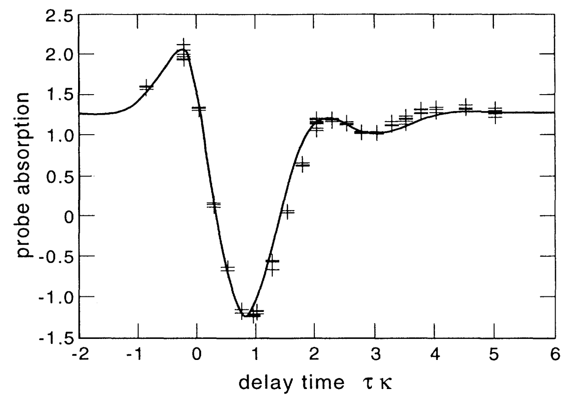

FIG. 3. Absorption vs time delay between the pump and probe. The horizontal axis is in units of lifetime of the upper states. The vertical axis is given as $W'(b=0.775\kappa)/W'(b=0.06\kappa)$. The value $b=0.06\kappa$ accounts for the estimated residual linewidth of the unmodulated laser (see text). The Rabi frequency is 2.62κ. The theory has been modified to include effects of Doppler broadening of 0.95κ and a ratio of pump to probe beam waist of 5.16. Data are shown as crosses.

The agreement between the predictions of GMZ and our data is quite good, with most groups of points falling on the calculated curve. Amplification of the probe beam is observed for delays $0.25<\tau\kappa<1.25$. It is evident in Fig. 3 that the period of oscillation ~ 39 ns is closely associated with the time period of a Rabi oscillation. This has a simple physical explanation if we consider transients induced in the population difference (and coherence) between the two states, initiated by the phase noise on the pump. These transients oscillate at the Rabi frequency and damp out on the time scale of κ^{-1}. Amplification of the probe field occurs if the time-delayed fluctuations of the probe beam are timed to arrive just as the population has been momentarily inverted due to their twin fluctuations on the pump. We would like to emphasize that the atom is not undergoing free precession between correlated fluctuations on the pump and probe, but is being driven by the noisy pump. In the calculations, the transients are incorporated into the stochastic Green functions, which propagate the density-matrix elements in the presence of the pump. For pure phase-diffusing fields with $b \leq \Omega$, Rabi oscillations will occur. However, Rabi oscillations wash out in the broad-band limit, $b \gg \Omega$, characteristic of the weak field and depletion regimes. Hence, the observation of gain along with Rabi oscillations, predicted only in the strong field regime, provides the most rigorous test of the method of GMZ.

Some discrepancies are apparent in Fig. 3. The theory predicts the first minimum at somewhat smaller delays, the second minimum at slightly larger delays, and the dipwidth to be slightly narrower than observed. These differences cannot be explained by either Doppler, detuning, or spatial averaging effects. Detuning effects tend to narrow the dipwidth and decrease the period of oscillation since the transients, in general, oscillate at the generalized Rabi frequency $\overline{\Omega}=(\Omega^2+\Delta^2)^{1/2}$. Doppler and detuning effects are quite similar until the Doppler width or detuning becomes larger than κ. Spatial averaging over a larger overlap range can broaden the dip slightly, but we found the overall fit to be inconsistent with the data if the overlap differed from the measured amount enough to affect the dipwidth. An error in the calculated value for the speed of light in the fiber can cause a change of scale, bringing the first minimum into agreement, but cannot explain the observed discrepancy since the mismatch of the second minimum becomes worse.

A possible cause for these discrepancies is the lack of a phase-diffusing field with Lorentzian spectrum defined by Eqs. (1) and (2). As τ increases from zero, the higher-frequency components of the laser spectrum decorrelate first. Hence, we expect the profile at small τ to be more sensitive to the wings of the spectrum corresponding to faster frequency fluctuations. At larger delays, the profile is sensitive to both the wings and core of the laser spectrum. If the laser spectrum is not exactly Lorentzian, this can lead to systematic errors as the delay is varied. Band-shape discrepancies can occur due to small errors in the rf filters which shape the output of the shot-noise diodes used in the ECNM system, or they can result from a finite β for the frequency fluctuations. The value of β in this experiment is 10κ, giving $0.1\kappa^{-1}$ for the correlation time of the frequency fluctuations, which might be expected to affect our results.

In Fig. 4, we show plots of absorption as a function of b for τ of 0, 1, and $5\kappa^{-1}$. Corresponding calculations are shown as solid, dashed, and dotted lines, respectively. The parameters used in the calculation are the same as for Fig. 3, except that the Doppler broadening was increased to 1.35κ. A smaller amount of Doppler broadening predicts the gain for $\tau=5\kappa^{-1}$ to be at the same level as for $\tau=1\kappa^{-1}$. This increase of Doppler broadening probably resulted from laser-beam misalignment from the atom beam during these measurements.

Figure 4 demonstrates a marked difference in the absorption for different delays, even for moderate laser bandwidths. In particular, for $\tau=5\kappa^{-1}$, which corresponds to essentially uncorrelated fields, maximum gain is observed for a laser FWHM of only 0.4κ, differing by 150% from the value for correlated fields. This effect can

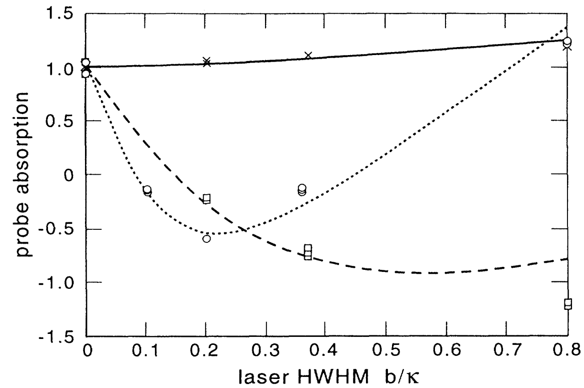

FIG. 4. Probe absorption vs b ($2b$ is the laser FWHM). Parameters are the same as for Fig. 3 except the Doppler width is increased to 1.35κ. The curves refer to calculations for $\tau=0$ (solid line), $\tau=1\kappa^{-1}$ (dashed line), and $\tau=5\kappa^{-1}$ (dotted line).

be of practical importance in experiments using beams from different lasers, or from the same laser if one beam experiences a significant time delay. The origin of gain for these time delays is different than that for transient induced gain at $\tau\sim 1\kappa^{-1}$ discussed previously. The effect of correlated frequency fluctuations is minimal, as seen in the curve corresponding to $\tau=0$. For uncorrelated fields, effects due to fluctuations in the relative pump-probe detuning δ dominate. An oversimplified but helpful picture views the probe absorption as undergoing excursions into areas of gain given by a typical absorption profile versus δ as predicted by Mollow [34]. In Fig. 5 we reproduce an absorption spectrum for a monochromatic pump on resonance, with $\Omega=2.6\kappa$. Since the pump and probe are derived from the same source, the time average $\langle \delta \rangle$ is zero, corresponding to a region of absorption in Fig. 5. For small fluctuations in δ (i.e., small laser bandwidths) the probe will sample mostly areas of gain in the absorption profile. Net gain will result as an average over the excursions. As the laser bandwidth is increased, more and

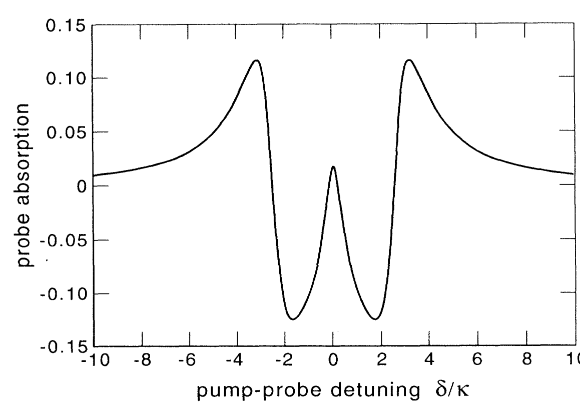

FIG. 5. Calculation of the Mollow absorption spectrum for a pump Rabi frequency of 2.62κ.

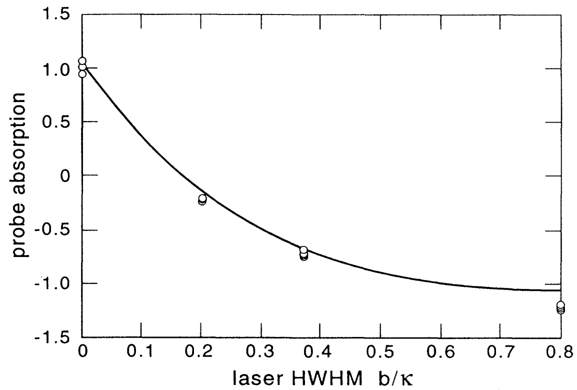

FIG. 6. Probe absorption vs b for data taken at $\tau=1\kappa^{-1}$ [measured minimum of absorption profile (see Fig. 3)], plotted with a calculation with $\tau=0.8\kappa^{-1}$ (predicted minimum).

more excursions into regions of absorption occur, and the net average becomes positive, resulting in net absorption. The actual picture is somewhat more complicated because the absorption spectrum itself shifts asymmetrically as the pump frequency (detuning) undergoes an excursion to one side of the atomic transition.

In Fig. 4, the data for $b=0.8\kappa$ and $\tau=1\kappa^{-1}$ fall well below the predicted curve. This discrepancy is consistent with the slight mismatch of minima in Fig. 3, discussed above. In Fig. 6 we show the same data taken at $\tau=1\kappa^{-1}$ plotted against calculations using $\tau=0.8\kappa^{-1}$, the position of the predicted minimum. The agreement is much better.

V. CONCLUSIONS

We have measured the absorption of a weak time-delayed phase-diffusing probe laser due to two-level atoms driven by a strong pump laser derived from the same noisy source. Our results are in excellent agreement with the predictions of Gheri, Marte, and Zoller [33] for atomic response to non-Markovian, time-delayed driving fields. We have observed amplification of the probe for time delays on the order of an atomic lifetime, which we attribute to transients in the population inversion induced by the noisy pump, and again for large time delays, which we attribute to fluctuations in the pump-probe detuning.

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