Diode-laser-noise-based spectroscopy of allowed and crossover resonances

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This paper analyses the extraction of spectroscopic information on an atom via use of field fluctuations in a diode laser, wherein the statistics of diode-laser radiation are modeled by a phase-diffusion process. Using Monte Carlo methods, we solve density-matrix equations for a three-level V system, driven by this fluctuating field, tuned approximately to the two allowed transitions. The model is very general and allows us to incorporate arbitrary field strengths and bandwidths. We suggest two different signal detection schemes, each of which provides insight into different aspects of the energy-level structure in an atomic sample. Specifically, if intensity fluctuations in the field radiated from the sample are spectrally analyzed, via a homodyne technique, one can isolate contributions that are linear and quadratic in the radiated field, and these reveal resonances at the allowed transition frequencies of the atom. If we employ direct detection instead of the homodyne method, then the crossover transition is also revealed in the signal. The strengths of these resonances are very sensitive to the bandwidth of the field fluctuations, as well as the details of the field statistics. We also discuss the role of Stark shifts at higher intensities.

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I. INTRODUCTION

Diode lasers are becoming increasingly popular as spectroscopic tools, primarily due to their low cost and compact size. Now it is routinely possible to have single-mode diode lasers with a wide tunability range, which has increased their utility in nonlinear optics and spectroscopy. Continuous-wave (cw) diode lasers, when operated far above threshold, are characterized by a very stable amplitude, but have large phase fluctuations. In fact, the field power spectrum of these lasers may have a full width at half maximum (FWHM) of 5–20 MHz, but the spectrum is accompanied by very long tails which may extend up to nearly 1 GHz. In other words, there is sufficient spectral density in the tails of the spectrum to excite an atomic transition. One can then envision using these lasers to excite a transition by tuning the laser approximately in the vicinity of the transition frequency, since the power in the wings of the spectrum can still resonantly excite the atom. This feature of diode lasers was exploited in the experiments of Yabuzaki and co-workers [1], where they demonstrated that the field fluctuations can be useful in extracting spectroscopic information on atoms and molecules. Subsequent experiments by Fairchild and co-workers [2] have further illustrated the utility of diode-laser noise in spectroscopy.

The study of atom interactions with noisy fields has a long history [3–5]. Renewed interest in this subject has been motivated by the technique of incoherent spectroscopy [6] and the recent work of Zoller, Cooper, and co-workers [7], who have theoretically addressed the issue of employing fluctuating fields in spectroscopy, with the goal of using the sensitivity of atom response to different field statistics as an indicator of the field statistics themselves. Some of the predictions made by these authors were experimentally verified by Smith and co-workers [8]. The dependence of atomic observables on details of field statistics has also been studied in the context of four-wave mixing in two-level atoms [9]. While the focus of most of the earlier works that dealt with stochastic fields in atom-field interactions was on comparing atom response in fluctuating fields with that in monochromatic fields, or on investigating atom response in fields with different statistics (i.e., amplitude noise versus frequency noise), the experiments of Yabuzaki *et al.* [1] and of Fairchild *et al.* [2] started a new line of investigation, where diode-laser noise, in conjunction with electronic spectrum analyzers, was recognized as a powerful spectroscopic tool. The central idea is that one starts with a field which has a stable amplitude but large phase fluctuations, as found naturally in diode lasers. When such a field propagates through an atomic medium, the atomic resonances convert phase fluctuations into amplitude fluctuations, and thus the transmitted field has excess intensity fluctuations, compared to the input field. Since these excess fluctuations arise from the presence of atomic resonances, the frequency content of these fluctuations must contain information about the atomic lines. The experiments of Refs. [2] and [3] where theoretically modeled in Ref. [10].

More recently, Jyotsna, Agarwal, and Vemuri [11] presented a simple analytical frame work, based on a linear-response theory, to understand the use of noisy fields for spectroscopy. This work relied on the fact that a fluctuating field would induce a fluctuating polarization in the atomic medium, which in turn would give rise to a fluctuating radiated field. By spectrally analyzing the fluctuations in the radiated field, these authors showed that one can obtain infor-

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mation about the energy-level structure of atoms. Reference [11] suggested detection of the radiated field by homodyning it with the incident field [12] and examining the power spectrum of the resulting intensity-intensity correlation function. By including a phase shifter in the detection apparatus, it was shown that one can isolate contributions to the intensity-intensity correlation function that are linear and quadratic in the radiated field. While the quadratic contribution was shown to lead to Lorentzian shaped resonances, the linear contribution exhibited dispersive shaped resonances. In the case of the V system modeled in Ref. [11], there were always two resonances in the spectrum, corresponding to the two allowed transitions.

There are several open questions that need to be addressed, and so in this paper we extend the work of Ref. [11] to consider additional aspects of diode-laser-based fluctuation spectroscopy that are relevant to experiments. Briefly, the experimental scheme we have in mind is the following: a fluctuating field, derived from a diode laser, is used to irradiate an atomic sample. The transmitted field, after propagation through a vapor cell, is homodyned with the incident field, and the resulting photocurrent from a photodiode spectrally analyzed (with a radio frequency spectrum analyzer). This spectrum constitutes our signal, and we have shown in Ref. [11] that using a homodyne scheme, one can isolate some important contributions to the signal. Some of the issues we address are as follows. (i) While the work of Ref. [11], based on a linear-response theory, was valid for weak driving fields, it is important to analyze the influence of strong, saturating fields. (ii) The signal in Ref. [11] revealed only the allowed transitions, but not the ones that are dipole forbidden. However, for fluctuation spectroscopy to become an effective tool, it is necessary to explore methods that would highlight all transitions in the spectrum, including the unallowed ones. (iii) Reference [11] dealt with fields with a Lorentzian field spectrum, but made no attempt to delineate the effect of field statistics on the resulting signal. While the assumptions of phase-diffusion statistics, and hence a Lorentzian field spectrum, are quite adequate for a diode laser, it would be interesting to investigate the effects of non-Lorentzian line shapes on the signal, as well as the impact of using fields with amplitude fluctuations.

The atom-field system of interest to us is the three-level V system in Fig. 1, driven by a stochastic field that is tuned approximately to the two allowed transitions. We derive the equations describing the time evolution of the relevant density-matrix equations, solve the equations via a Monte Carlo procedure [13], calculate the spectrum of the intensity-intensity correlation function of the radiated field, and show the presence of resonances that correspond to the various atomic transitions. The Monte Carlo method provides maximum flexibility in allowing us to investigate an arbitrary parameter range for the field strengths and bandwidths. A detailed study of the effect of the noise parameters of the field is reported, and finally we also study the effect that an amplitude fluctuating field has on noise spectroscopy. The organization of the rest of the paper is as follows. In Sec. II is the theoretical model, consisting of the density-matrix equations, that describes the three-level V system driven by a fluctuating field. This section also contains a brief discussion on phase-diffusing fields and chaotic fields. Section III deals with the results of our calculations, specifically, the spectrum obtained from the homodyne process. The linear and quadratic contributions to the spectrum, as well as the spectrum via direct detection, are presented for a wide range of atom and field parameters. The paper concludes with a brief summary, in Sec. IV, of the major results and their significance.

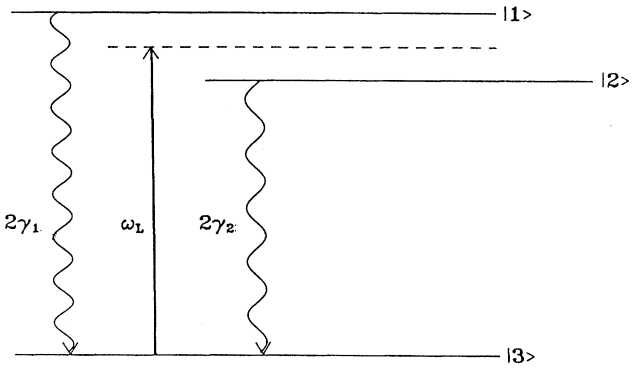

FIG. 1. Schematic representation of a three-level V system with ground state $|3\rangle$ and two excited states $|1\rangle$ and $|2\rangle$. The spontaneous decay rates from $|1\rangle$ to $|3\rangle$ and $|2\rangle$ to $|3\rangle$ are $2\gamma_1$ and $2\gamma_2$, respectively. The transition from $|1\rangle$ to $|2\rangle$ is not allowed. ω_L is the central frequency of the exciting field.

II. THEORETICAL MODEL

As previously stated, the atomic system of interest to us is the three-level V system shown in Fig. 1. The two excited states $|1\rangle$ and $|2\rangle$ decay into state $|3\rangle$ with radiative widths of $2\gamma_1$ and $2\gamma_2$, respectively. The $|1\rangle\leftrightarrow|2\rangle$ transition is dipole forbidden. A stochastic driving field, derived from a diode laser, drives the atom and couples the $|1\rangle\leftrightarrow|3\rangle$ and $|2\rangle\leftrightarrow|3\rangle$ transitions. The semiclassical Hamiltonian for this atom-field system is given by

$$\frac{H}{\hbar}=\omega_{13}|1\rangle\langle 1|+\omega_{23}|2\rangle\langle 2|+\alpha(t)e^{-i\omega_l t}|1\rangle\langle 3| + \alpha(t)e^{-i\omega_l t}|2\rangle\langle 3|+\text{H.c.}, \tag{2.1}$$

where $\alpha(t)$ represents contributions from the stochastic field (which includes the dipole moment transition matrix element) at frequency ω_l, and ω_{13} and ω_{23} are the atomic transition frequencies for the $|1\rangle\leftrightarrow|3\rangle$ and $|2\rangle\leftrightarrow|3\rangle$ transitions, respectively. We now have to make a choice regarding the statistics of the stochastic field. The output of a diode laser is characterized by a stable amplitude, but has large phase fluctuations. Further, the dominant noise mechanism in these lasers is spontaneous emission noise, which would indicate a Lorentzian field power spectrum, unlike dye or Ti:sapphire lasers which are dominated by technical noise and hence have Gaussian field spectra. The statistics of cw diode-laser radiation are hence well-described by a phase-diffusion model (PDM) [14], which is known to mimic the output of single-mode lasers that are operated well above threshold. This implies that $\alpha(t)$ can be written as

$$\alpha(t)=\Omega e^{-i\phi(t)}, \tag{2.2}$$

where Ω is the Rabi frequency of the field and $\phi(t)$ is its random phase. The stochastic frequency $\mu(t)$ is given by

$$\frac{d\phi}{dt}=\mu(t), \tag{2.3}$$

where $\mu(t)$ is assumed to be a Gaussian-Markovian random process with zero mean and a correlation function given by

$$\langle\mu(t)\mu(t')\rangle=b\beta e^{-\beta|t-t'|}. \tag{2.4}$$

In Eq. (2.4), b is the strength of the frequency fluctuations, and physically describes the spectral density within the field power spectrum, while β is the bandwidth of the fluctuations. One can identify two distinct regimes based on the relative values of b and β. When $\beta\gg b$, the correlation function in Eq. (2.4) reduces to a δ-correlated function, i.e.,

$$\langle\mu(t)\mu(t')\rangle=2b\,\delta(t-t'). \tag{2.5}$$

In this case, the field has a Lorentzian spectral profile with a FWHM given by $2b$. This is the situation most appropriate for describing the radiation from diode lasers. In the opposite limit, when $b\gg\beta$, one obtains a Gaussian field line shape, with a FWHM related to the product $b\beta$. This situation is appropriate for lasers where the dominant noise mechanism is technical or pump noise, instead of quantum noise.

Since we will compare the results of a phase-diffusing field with one that has amplitude fluctuations, we discuss that model also. A chaotic field model (CFM) is often used to describe amplitude fluctuating fields [4,9] and this model mimics the output from a multimode or pulsed laser. In the CFM, $\alpha(t)=z(t)$ is the fluctuating complex process, with zero mean and an autocorrelation function given by

$$\langle z(t)z^*(t')\rangle=D\Gamma e^{-\Gamma|t-t'|}. \tag{2.6}$$

In Eq. (2.6), D is the strength of the fluctuations and Γ^{-1} is the correlation time of the noise. This field has a Lorentzian spectral profile with a FWHM given by 2Γ. The product $D\Gamma$ gives a measure of the variance of the Gaussian random process and is physically identified with the intensity of the field.

In a frame rotating at the fast optical frequency ω_l, the Hamiltonian in Eq. (2.1) becomes

$$\frac{H}{\hbar}=\Delta_1|1\rangle\langle 1|+\Delta_2|2\rangle\langle 2|+\alpha(t)|1\rangle\langle 3|+\alpha(t)|2\rangle\langle 3|+\text{c.c.}, \tag{2.7}$$

where $\Delta_1=\omega_{13}-\omega_l$ and $\Delta_2=\omega_{23}-\omega_l$. From Eq. (2.7), the following density-matrix equations are easily obtained:

$$\dot\rho_{11}=-2\gamma_1\rho_{11}-i\alpha(t)\rho_{31}+i\alpha^*(t)\rho_{13}, \tag{2.8a}$$

$$\dot\rho_{22}=-2\gamma_2\rho_{22}-i\alpha(t)\rho_{32}+i\alpha^*(t)\rho_{23}, \tag{2.8b}$$

$$\dot\rho_{12}=-(\gamma_1+\gamma_2)\rho_{12}-i(\Delta_1-\Delta_2)\rho_{12}-i\alpha(t)\rho_{32}+i\alpha^*(t)\rho_{13}, \tag{2.8c}$$

$$\dot\rho_{13}=-\gamma_1\rho_{13}-i\Delta_1\rho_{13}-i\alpha(t)\rho_{33}+i\alpha(t)\rho_{12}, \tag{2.8d}$$

$$\dot\rho_{23}=-\gamma_2\rho_{23}-i\Delta_2\rho_{23}-i\alpha(t)\rho_{33}+i\alpha(t)\rho_{21}. \tag{2.8e}$$

The density-matrix equations are Langevin equations where the stochastic field appears as multiplicative noise. The fluctuating driving field produces a fluctuating polarization in the medium, which in turn produces a fluctuating radiated field, and is given by the sum of the off-diagonal density-matrix elements ρ_{13} and ρ_{23}. The net polarization in the medium is proportional to the density of atoms. Our detection scheme is based on homodyning the radiated field with the incident field and so we write the total intensity at the photodetector as

$$I(t)\sim|\tilde\alpha(t)+\rho_{13}(t)+\rho_{23}(t)|^2. \tag{2.9}$$

We have now absorbed all the inessential scale factors such as atom density, dipole moment matrix elements, etc., in $\tilde\alpha(t)$. Thus, the two-time intensity correlation function $C(\tau)$ is given by

$$C(\tau)=\langle I(t)I(t+\tau)\rangle. \tag{2.10}$$

The final spectrum $P(\omega)$ that we wish to calculate is given by the Fourier transform of Eq. (2.10) and formally is

$$P(\omega)=\int d\tau C(\tau)e^{i\omega\tau}. \tag{2.11}$$

It is clear from Eqs. (2.9) and (2.10) that the intensity correlation function will have terms that are independent of the radiated field, and others that are first, second, third, and fourth order in the radiated field. It was established in Ref. [11] that by including a phase shifter in the homodyne process, one can isolate the linear and quadratic contributions in the signal. On the other hand, if direct detection [i.e., set $\tilde\alpha=0$ in (2.9)] instead of homodyne detection is employed, one would essentially measure the fourth-order contribution. In what follows, we will discuss these various orders separately.

The numerical procedure for solving Eq. (2.8) is based on a Monte Carlo approach, which is described in detail elsewhere and will not be reported here [13]. In essence, the technique consists of numerically simulating Gaussian distributed random numbers, with the desired statistics and correlation time. The first-order ordinary differential equations in Eq. (2.8) can be solved accurately by either a Runge-Kutta or Euler method with a sufficiently small step size. The quantities to be solved for are ρ_{13} and ρ_{23}. Once the steady-state values have been obtained, one can determine the intensity at the detector via Eq. (2.9), and hence the correlation function from Eq. (2.10). Finally a fast Fourier transform (FFT) routine would yield the desired spectrum, given by Eq. (2.11). However, calculating the correlation function as in Eq. (2.10) and then performing the FFT was very time consuming. To reduce the computational time and effort needed, we used the following theorem [15], which is true for two real functions $A(t)$ and $B(t)$:

$$\int d\tau\langle A(t)B(t+\tau)\rangle e^{i\omega\tau}=\langle A(\omega)B^*(\omega)\rangle, \tag{2.12}$$

where $A(\omega)$ and $B(\omega)$ are Fourier transforms of $A(t)$ and $B(t)$, respectively. This method avoids the necessity of calculating the correlation function first and then taking the

Fourier transform. For our purpose, $A(t)$ and $B(t)$ are simply the intensities as determined from Eq. (2.9). The final signal displayed in our results is an ensemble average over the field fluctuations, and was obtained by numerically averaging the signal over several thousand iterations, each with a different set of independent random numbers. This ensured that the results were not affected by small number statistics.

In the following, for brevity, we denote $[\rho_{13}(t)+\rho_{23}(t)]$ as $\rho(t)$, and the various contributions to $C(\tau)$ by $C_i(\tau)$, where $i=0,1,2,3,4$. The subscript i refers to the order of the radiated field that appears in the correlation function. Thus

$$C_0(\tau)=\langle|\tilde{\alpha}(t)|^2|\tilde{\alpha}(t+\tau)|^2\ \rangle, \tag{2.13a}$$

$$C_1(\tau)=\langle 2\ \mathrm{Re}[\tilde{\alpha}^*(t)\rho(t+\tau)]\rangle + \text{terms with } t \text{ and } t+\tau \text{ interchanged}, \tag{2.13b}$$

$$C_2(\tau)=\langle|\tilde{\alpha}(t)|^2|\rho(t+\tau)|^2\rangle + 4\langle \mathrm{Re}(\tilde{\alpha}^*(t)\rho(t))\mathrm{Re}(\tilde{\alpha}^*(t+\tau)\rho(t+\tau))\rangle + \text{terms with } t \text{ and } t+\tau \text{ interchanged}, \tag{2.13c}$$

$$C_3(\tau)=\langle 2|\rho(t)|^2\mathrm{Re}(\tilde{\alpha}^*(t+\tau)\rho(t+\tau))\rangle + \text{terms with } t \text{ and } t+\tau \text{ interchanged}, \tag{2.13d}$$

$$C_4(\tau)=\langle|\rho(t)|^2|\rho(t+\tau)|^2\rangle. \tag{2.13e}$$

The intensity-intensity correlation $C(\tau)$ is thus given by

$$C(\tau)=C_0(\tau)+C_1(\tau)+C_2(\tau)+C_3(\tau)+C_4(\tau). \tag{2.14}$$

Note that the third- and fourth-order terms were not discussed in Ref. [11], and as we will show in the next section, the fourth-order contribution contains significant spectroscopic information. For each of the $C_i(\tau)$, we denote the corresponding Fourier transforms by $P_i(\omega)$. Hence the total power spectrum is given by

$$P(\omega)=P_0(\omega)+P_1(\omega)+P_2(\omega)+P_3(\omega)+P_4(\omega) \tag{2.15}$$

and it is these power spectra that constitute our signal.

III. RESULTS

We now present the results for the homodyne spectrum, as defined by Eq. (2.11), when a diode-laser field propagates through an atomic sample. In particular, we discuss the behavior of the various orders of $P(\omega)$ and their sensitivity to the fluctuation characteristics of the driving field. All rates in our work are normalized to units of γ_1. We also assume that $\gamma_2=\gamma_1$, though it is fairly straightforward to include unequal values for these radiative widths. For all of the results presented here, we take $\Delta_1=20$ and $\Delta_2=30$.

We begin with the results for a field that follows the PDM, since these results are most relevant to fluctuation spectroscopy with diode lasers. The relevant noise parameters in this model ar the strength of the noise, b, and the bandwidth of the frequency fluctuations, β. In Fig. 2(a) is the spectrum $P(\omega)$ when $\beta=100$ and $b=1$, and since $\beta\gg b$, the field line shape is a Lorentzian. The driving field is weak and has a Rabi frequency of $\Omega=\sqrt{0.2}$. The spectrum clearly reveals two peaks at 20 and 30, which correspond to the detunings of the field from $|1\rangle$ and $|2\rangle$, and agrees well with the analytic results of Ref. [11]. Note that in this spectrum, the crossover response corresponding to the $|1\rangle\leftrightarrow|2\rangle$ transition is not visible. Since the basis for this spectroscopy lies in utilizing the spectral density in the wings of the laser power spectrum, we next look at the effect of reducing the spectral density in these wings. This can be done easily in our model by reducing β to 10, while keeping $b=1$. This still corresponds to a field spectrum that is close to Lorentzian, but with less spectral density in the wings than the field in Fig. 2(a). The results of using a field with a sharper cutoff frequency are shown in Fig. 2(b), which still reveal the presence of the two peaks, however diminished in size. This reduction in the signal is a direct consequence of exciting the transitions with smaller laser power [while the numbers on the vertical axis are in arbitrary units, one can compare the relative size of the numbers in Figs. 2(a) and 2(b) to get an estimate of the relative strengths of the resonances].

As previously stated, the utility of the Monte Carlo lies in its ability to accommodate a wide range of field strengths and bandwidths. While the work of Ref. [11] was restricted to weak driving fields, since it was based on a linear-response theory, here we can study the effect of strong fields that saturate the two transitions. In Fig. 2(c), Ω is increased to $\sqrt{20}$, while keeping $\beta=10$ and $b=1$. The two peaks at detunings of 20 and 30 are still prominent, as is the crossover resonance by its absence. A careful look at the spectrum indicates that the two peaks are now shifted to higher frequencies. This shift in the position of the peaks can be explained on the basis of the Stark effect. The expected shift in frequency is given by the ratio $2\Omega^2/\Delta$, where Δ is the detunings of the relevant transition. Thus, for $\Omega=\sqrt{20}$ and $\Delta_1=20$, we expect the shift in frequency to be 2, which agrees well with our results. In the presence of strong fields, fluctuation spectroscopy is thus not only useful in extracting atomic level structure information, but also in determining the extent of the Stark splitting.

While the results presented so far could be derived analytically [11], following techniques developed for multiplicative stochastic process, we now consider parameters where analytical solutions are either very difficult or impossible. One example is the Brownian motion phase-diffusion process which describes fields with frequency fluctuations on a time scale comparable to atom relaxation rates. In Fig. 2(d) is the spectrum when $\Omega=\sqrt{0.2}$ and we choose $\beta=1$ and $b=10$. The noise variance, which is the product of $b\beta$, is thus the same as for the results in Fig. 2(b). However, the physical situation is quite different, since the field spectrum now is a Gaussian (since $b\gg\beta$), with a FWHM related to the product of $b\beta$. A Gaussian spectrum has a sharp cutoff and so we expect the signal to diminish significantly, which is precisely the effect shown in Fig. 2(d). One still finds the two allowed resonances, but they are much weaker than in Fig. 2(b). The result of Figs. 2(a), 2(b), and 2(d) drive home the importance of having long tails to the driving field spec-

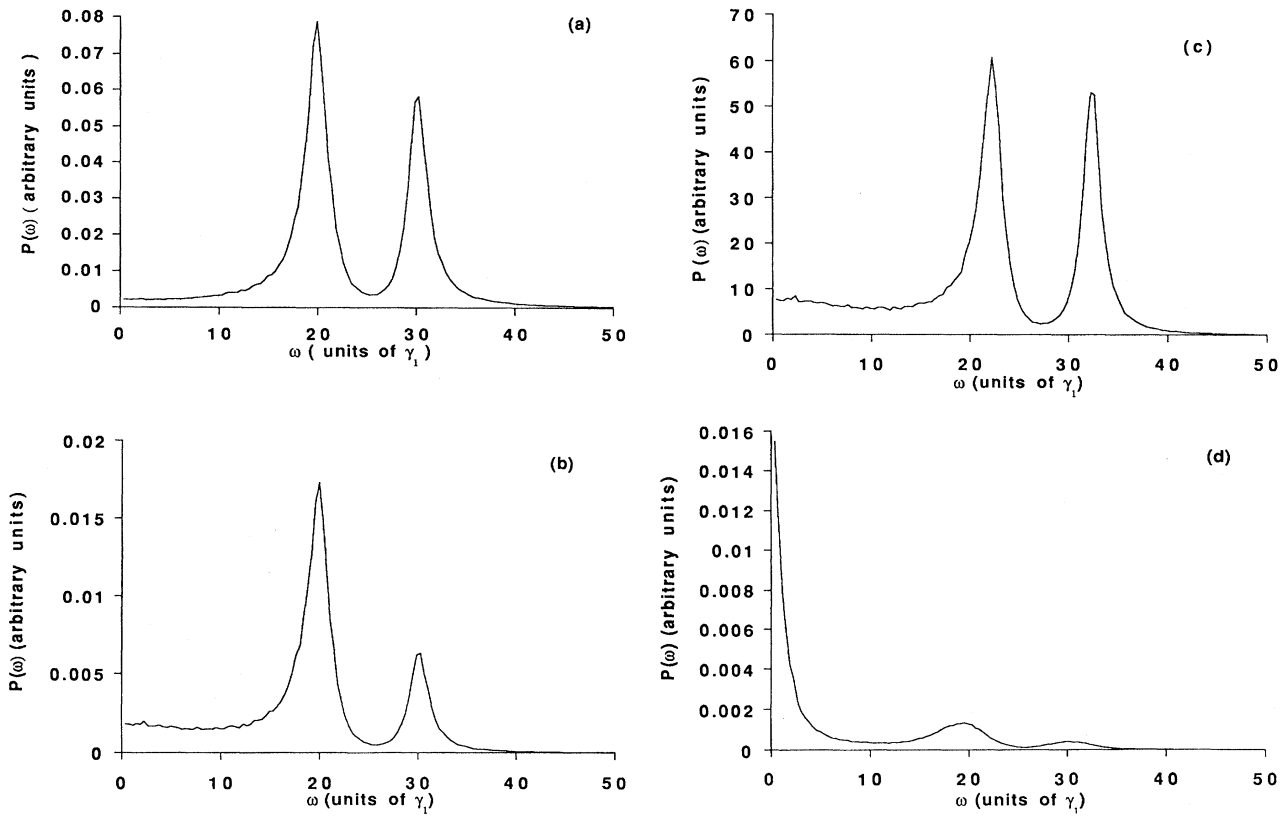

FIG. 2. Homodyne spectrum for the phase-diffusion model, for $\Delta_1=20$ and $\Delta_2=30$, and (a) $\beta=100$, $b=1$, and $\Omega=\sqrt{0.2}$, (b) $\beta=10$, $b=1$, and $\Omega=\sqrt{0.2}$, (c) $\beta=10$, $b=1$, and $\Omega=\sqrt{20}$, (d) $\beta=1$, $b=10$, and $\Omega=\sqrt{0.2}$.

trum for fluctuation spectroscopy to be successful. While we have not shown similar results for strong fields, identical behavior is seen there too.

One feature that has been elusive so far is the crossover resonance, corresponding to the $|1\rangle\leftrightarrow|2\rangle$ transition, which is not reflected in the power spectrum. However, for this spectroscopy to be an effective tool, it is desirable to find a way to highlight this transition in the spectrum. To do this, we calculate the fourth-order contribution to the signal, which is the contribution one would measure in direct detection, instead of homodyne detection. In Fig. 3(a), $\beta=100$, $b=1$, and $\Omega=\sqrt{0.2}$. Now we find clear evidence of the crossover peak at a frequency of 10. The physical origin of this peak in the spectrum can be understood in terms of a mixing process between components at frequencies of Δ_1 and Δ_2 in the laser spectrum. The appearance of the crossover resonance in the signal spectrum [Fig. 3(a)] is due to an interference effect between these components, giving rise to a signal at $\Delta_2-\Delta_1$. For the atom and field configurations chosen in this work, the peak arising from the mixing process can be identified with the frequency differences between the upper two atomic levels. This association stems from the fact that we have a single field that is approximately tuned to the two transitions, thereby creating the detunings Δ_1 and Δ_2, and $\Delta_2-\Delta_1$ in our work (see Fig. 1) is precisely the crossover frequency. On increasing the field strength, as in Fig. 3(b), to $\Omega=\sqrt{20}$, we still find a strong signal at the crossover resonance frequency. Since the two allowed resonances are Stark shifted by different amounts, depending on the values of Δ_1 and Δ_2, we expect the crossover peak also to be shifted from its original position. While we have checked that this is true in general, for the result shown here, the difference between Stark shifts for the two transitions is too small for the effect to be visible on the crossover peak. We find this peak to be very sensitive to the bandwidth of the frequency fluctuations. On reducing β from 100 to 10, and all other parameters being the same as in Fig. 3(a), we find that the crossover peak disappears, a shown in Fig. 3(c), but the allowed resonances are still prominent. There is, of course, a reduction in the overall signal level also, due to the sharper cutoff of the laser spectrum. In the case of strong fields also, a similar effect is found (results not shown). The results show that a large value of β is necessary to highlight the crossover resonance in the spectrum, and since diode-laser radiation is accompanied by long tails (which means large β), they are ideal candidates for use in noise spectroscopy. To further illustrate the importance of β to the resolution of the crossover peak, in Fig. 3(d) we consider the case when $\beta=100$ and $b=10$. This spectrum, for strong fields, shows a dramatic increase in the signal level corresponding to the crossover resonance, while almost completely eliminating the allowed resonances. The noise is so large here ($b\beta=1000$) that the two allowed resonances are lost. However, since the crossover resonance is found only in the fourth-order contribution, which does not directly include the field noise unit, it is much less susceptible to being lost. It is important to men-

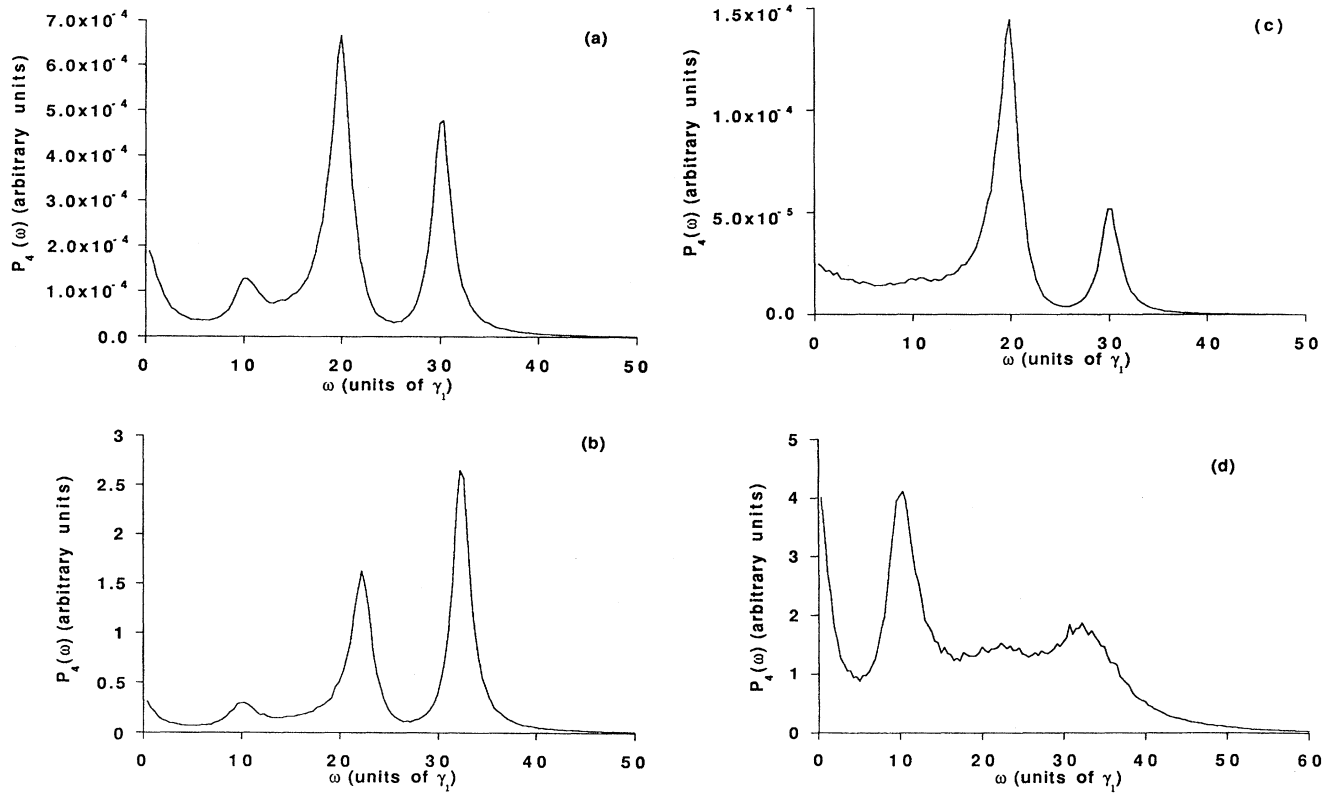

FIG. 3. Fourth-order contribution to the homodyne spectrum for the phase-diffusion model, with $\Delta_1=20$ and $\Delta_2=30$, and (a) $\beta=100$, $b=1$, and $\Omega=\sqrt{0.2}$, (b) $\beta=100$, $b=1$, and $\Omega=\sqrt{20}$, (c) $\beta=10$, $b=1$, and $\Omega=\sqrt{0.2}$, (d) $\beta=100$, $b=10$, and $\Omega=\sqrt{20}$.

tion here that the kind of spectroscopy discussed here works best when the field bandwidth is comparable to the atom relaxation rates (i.e., spectrally colored fields). Monochromatic fields do not have the frequency spread to drive all the atomic transitions, while broadband fields produce very weak resonances. This is the reason that the allowed transitions are almost nonexistent in Fig. 3(d), since even if one assumes that the parameters correspond to pure PDM, the field FWHM is 10 times the atomic natural width. Finally, we also mention that we have checked that $P_1(\omega)$, which gives the linear contribution to Eq. (2.11), produces only a dc contribution and is zero at all other frequencies (for PDM). This result can be shown analytically [11], and in fact is one of the major differences in atom response when exposed to fields obeying the PDM versus the CFM.

Though the CFM is not directly relevant to cw diode lasers, it is interesting nonetheless to study the differences in the homodyne signal when the driving field has amplitude fluctuations instead of frequency fluctuations. We find that the results are significantly different if one uses a chaotic field instead of a phase-diffusing field. Once again we separate the signal into linear, quadratic, and fourth-order contributions. For weak fields ($D\Gamma=0.2$), the quadratic term is shown in Fig. 4(a), with the two resonances at 20 and 30. The linear contribution, which only had a dc contribution for the PDM, now has the structure shown in Fig. 4(b). The resonances are still at 20 and 30, but they are now dispersive instead of Lorentzian. This dispersive contribution can be enhanced in experiments by increasing the atom density, or by increasing the laser beam spot size so that the field interactions with more atoms inside the vapor cell. Neither Fig. 4(a) nor 4(b) indicate the crossover resonance, which can be revealed only through the fourth-order contribution (just as for PDM). This is shown in Fig. 4(c) for the same parameter values as Figs. 4(a) and 4(b), and the crossover transition is clearly seen. We have also calculated the third-order contribution, but the results are not shown since that term does not contain any information not already contained in the quadratic term. The results for the strong fields indicate behavior very similar to that seen for PDM, where we find the Stark shifts of the atomic levels reflected in the spectra.

Finally, we note that even for the CFM, it is desirable to have a large field bandwidth to extract the crossover resonance in the fourth-order spectrum. This is clear from comparing Figs. 4(c) and 4(d), where Γ has been changed from 1 to 10. For the CFM, as define by us in Eq. (2.6), the field spectrum is always a Lorentzian with a FWHM given by 2Γ. Thus, in Fig. 4(c), the field bandwidth is comparable to the atomic width, while Fig. 4(d) corresponds to a broadband field. The crossover peak gets larger by orders of magnitude when Γ is increased, but the allowed resonances disappear for such large bandwidths.

IV. SUMMARY

In this paper we have presented a very general model to study the possibility of using diode-laser noise as a spectroscopic tool for extracting energy-level information on atoms

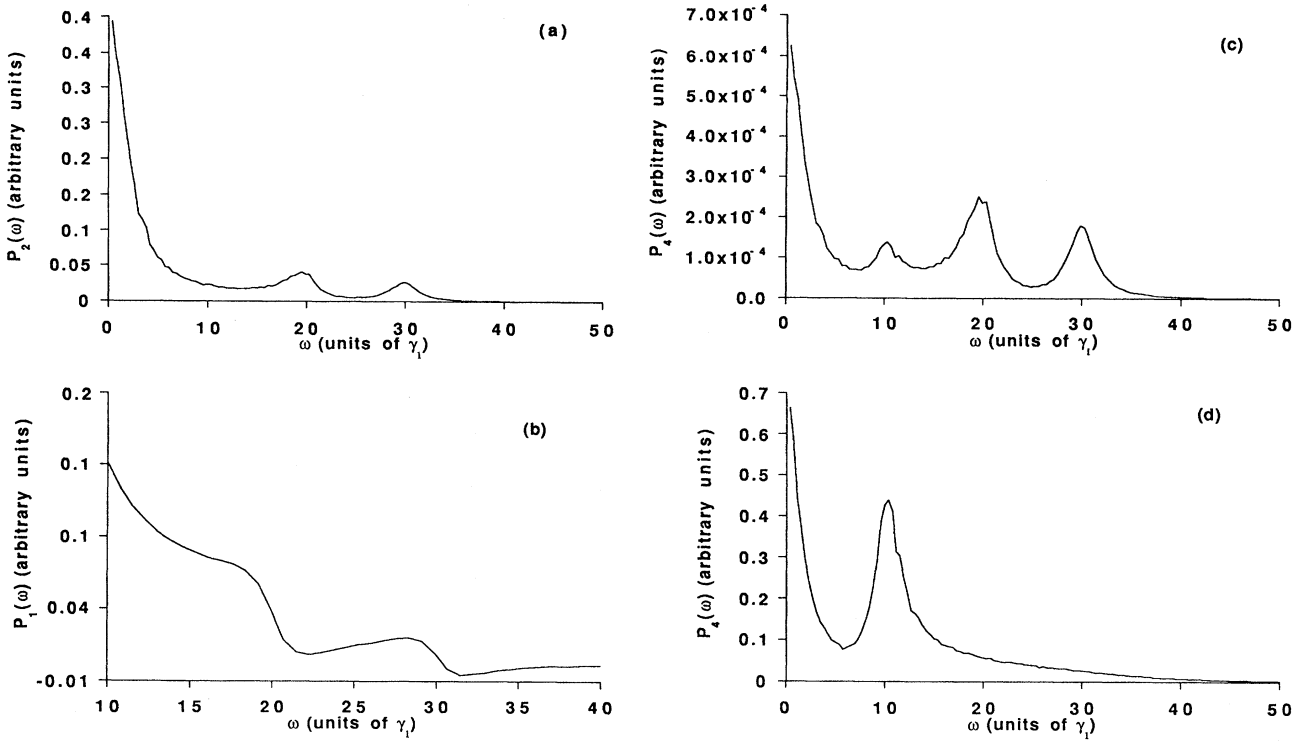

FIG. 4. Homodyne spectrum for chaotic fields, with $\Delta_1=20$ and $\Delta_2=30$, and (a) quadratic contribution with $\Gamma=1$, $D=0.2$, (b) linear contribution with $\Gamma=1$, $D=0.2$, (c) fourth-order contribution with $\Gamma=1$, $D=0.2$, (d) fourth-order contribution with $\Gamma=10$, $D=0.2$.

in a vapor cell. The scheme is based on homodyning the stochastic field transmitted through an atomic sample with the incident field, and spectrally analyzing the resulting photocurrent. The diode-laser radiation treated as a phase-diffusing field, and density-matrix equations for a three-level V system, interacting with a stochastic field, are solved using a Monte Carlo procedure. We present results for a wide range of field strengths and bandwidths, and also investigate the impact of using chaotic field instead of phase-diffusing fields. We find that it is most convenient to analyze the results by separating the signal into contributions that are linear, quadratic, and fourth-order in the radiated field. The linear and quadratic contributions can be experimentally extracted through a homodyne process, whereas the fourth-order contribution is the one that one would obtain through a direct detection of the signal.

We find that for the PDM, when a weak field is approximately tuned to the two allowed transitions of a V system, the linear contribution is nonexistent, and the quadratic contribution reveals allowed resonances. The strengths of the resonances are very sensitive to the bandwidth of the frequency fluctuations. In general, fluctuation spectroscopy works best for spectrally colored fields, i.e., when the field bandwidth is of the order of the atomic width. If the bandwidth is too large or too small compared to the atomic widths, the resonances are either too weak or nonexistent. A second desirable trait is that the field spectrum have long tails. We have verified this by considering the phase-diffusion model where the bandwidth of the frequency fluctuations is smaller than the strength of the fluctuations. This gives rise to a Gaussian field spectrum, which, because of its sharp cutoff, reduces the strength of the resonances significantly. When strong, saturating fields are used for spectroscopy, we find that the quadratic contribution again has the two allowed resonances in it, but they are now Stark shifted to higher frequencies. This indicates that the spectrum may be used as a measure of the Stark shift, if one already has information about the transition frequency.

The fourth-order contribution is found to reveal the crossover transition, in addition to the allowed transitions. This crossover peak does not appear in the quadratic contribution. The signal here is also very sensitive to the field spectrum, and, in particular, can be enhanced by increasing the field bandwidth. This leads to a rather paradoxical sitaution, where a field spectrum with FWHM much greater than the natural width strengthens the crossover transition in the spectrum, but also reduces or even eliminates the allowed transitions. Clearly, an intermediate bandwidth, i.e,. one comparable to the atomic widths, is best suited for extracting all the desired information.

Finally, we also report on the use of chaotic fields for spectroscopy. The most dramatic difference is in the linear contribution, which exhibits dispersive resonances instead of the Lorentzian resonances seen in the quadratic and fourth-order contributions. For strong fields we once again find evidence for the Stark shift, and the bandwidth dependence of the resonance is qualitatively similar to that seen for phase-diffusing fields.

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