

## Noise in strong laser-atom interactions: Frequency fluctuations and nonexponential correlations

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We extend our study of the effects of jump-type noise on laser-atom interactions to frequency-telegraph noise. Such noise can be used as a model of collisional effects, in which the atomic transition frequency randomly jumps, or as a model of finite laser bandwidth effects, in which the laser frequency exhibits random jumps. We show that these two types of frequency noise can be distinguished in light-scattering spectra. We also discuss examples which demonstrate both temporal and spectral motional narrowing, nonexponential correlations, and non-Lorentzian spectra. Its exact solubility in finite terms makes the frequency-telegraph noise model an attractive alternative to the white-noise Ornstein-Uhlenbeck frequency noise model which has been previously applied to laser-atom interactions.

### I. INTRODUCTION

The radiative interactions of atoms and molecules are affected by processes, such as collisions of various kinds, which cannot be followed in microscopic detail. There is a long tradition in which these processes are treated theoretically by statistical methods.<sup>1</sup> This subject has been reopened in recent years<sup>2</sup> because the strength of laser-induced radiative interactions can invalidate earlier results based on harmonic-oscillator (Lorentz model) pictures of atomic electrons.<sup>1</sup>

A central assumption in most stochastic theories of collisional line shape is that the subject atom or molecule experiences a brief, perhaps even instantaneous, shift in its internal eigenenergies and transition frequencies due to a collision. It is also possible that the laser frequency undergoes fluctuations at the same time. Such fluctuations would contribute to a finite bandwidth of the laser light. As theoretical treatments have become more detailed, there has arisen experimental interest in testing these theories by taking careful account of laser noise<sup>3</sup> and by deliberately injecting controlled laser frequency fluctuations<sup>4</sup> into multiphoton absorption experiments.

In this paper we present a theory of strong laser-atom interactions, using a simple model of frequency fluctuations that has some common features and some advantages over previous models. Its features include analytic simplicity and a flexible form for the basic free correlation function, which may be nonexponential. Its principal advantage, which follows directly from the model's simplicity, is that the fundamental atomic or molecular response functions can be evaluated exactly, in finite terms. Even the most complicated dipole correlation function or scattered light spectrum requires, at worst, the

inversion of a finite matrix. This is in strong contrast to the situation encountered with the previously most-used model, in which the instantaneous frequency is assumed to be an Ornstein-Uhlenbeck (OU) Gaussian stochastic process with an exponential autocorrelation. In the OU model formal expressions can be obtained for response functions, but only in terms of infinite hierarchies of one kind or another.<sup>5</sup>

It is important to mention a few technical features of our model. It is based on the random telegraph.<sup>6</sup> It is not a Gaussian model but rather "pre-Gaussian" and has a Gaussian limit. We have discussed other pre-Gaussian models.<sup>7</sup>

Our pre-Gaussian model shares with the OU model the important non-Lorentzian property. As Georges and Lambropoulos have emphasized,<sup>8</sup> non-Lorentzian models can be essential for accurate prediction of some phenomena.<sup>9</sup> Also, it is well to keep in mind when comparing various models that to date none has been shown to have deep connections with the microscopic dynamics of frequency fluctuations, and that is true of our model as well.

In Sec. II, after an elementary introduction to the random telegraph model of laser frequency fluctuations, we derive the laser power spectrum and its non-Lorentzian band shape. For such a simple model the atomic response to such fluctuations can be easily established. In Sec. III we illustrate the atomic response of a two-level atom coupled to an external laser field. We discuss the time evolution of the two-level population and the non-Lorentzian band shape effects in far-wing ionization, for systems subjected to laser frequency-telegraph noise. In Sec. IV we consider a non-Lorentzian band shape in the case of the fluorescence spectrum of a strongly driven two-level atom. In Sec. V we discuss the fluorescence spectrum of a

strongly driven atom with frequency-interrupting collisional noise. Finally, some concluding remarks are given in Sec. VI.

## II. ELEMENTARY TREATMENT OF STOCHASTIC FIELD

Bivalued instantaneous frequency-telegraph noise consists of random jumps between two possible frequency values (states)  $\alpha$  and  $-\alpha$ . We assume that these states are distributed with probability:  $g(\alpha) = \frac{1}{2}\delta_{\alpha,a} + \frac{1}{2}\delta_{\alpha,-a}$ . The instantaneous frequency at time  $t$  can be written in the following form:  $\mu(t) = \alpha(-1)^{n(t,0)}$ , where  $n(t,0)$  is the number of times the telegraph changes its state between 0 and  $t$ . The random telegraph is a Markov process, i.e.,  $n(t,0) = n(t,t_1) + n(t_1,0)$  for  $t \geq t_1 \geq 0$ , with a Poisson distribution of  $n$ . The mean of  $n$  is proportional to the time interval  $t$  through the mean dwell time  $T$  of the telegraph:  $\langle n(t,0) \rangle_{\text{av}} = t/T$ . From these definitions<sup>10</sup> we calculate in a straightforward way that  $\langle \mu(t) \rangle = 0$ , and

$$\begin{aligned} \langle \mu(t+\tau)\mu(t) \rangle &= a^2 \langle (-1)^{n(t+\tau,0)} (-1)^{n(t,0)} \rangle \\ &= a^2 \langle (-1)^{n(t+\tau,t)} \rangle \\ &= a^2 \exp \left[ -\frac{2}{T} |\tau| \right]. \end{aligned} \quad (2.1)$$

This formula indicates that  $T/2$  has the clear physical interpretation as the coherence time of the frequency fluctuations. From the Markov property we conclude that  $\mu(t)$ , defined as a random telegraph, is not a Gaussian stochastic process though it is entirely defined by its two-point correlation function (2.1). For example, we have the following recurrence relation:

$$\langle \mu(t_1) \cdots \mu(t_n) \rangle = \langle \mu(t_1)\mu(t_2) \rangle \langle \mu(t_3) \cdots \mu(t_n) \rangle \quad \text{if } t_1 > t_2 > t_3 > \cdots > t_n. \quad (2.2)$$

This formula shows a decorrelation pattern that is clearly different from the well-known Gaussian decorrelation of moments higher than second.

Now we can calculate the field correlation of the electric field

$$E(t) = E_0 \exp[-i\omega_L t - i\phi_L(t)], \quad (2.3a)$$

where the field's phase is described by a random frequency telegraph:

$$\phi_L(t) = \int_0^t ds \mu(s). \quad (2.3b)$$

That is, by comparison with the preceding paper,<sup>11</sup> here the phase itself is not a telegraph but rather a smoothed telegraph. The physical consequences are not trivial, as we will show. First, the correlation of  $E(t)$  can be obtained from the following trivial equations for  $f(\tau) = \exp[i \int_t^{t+\tau} ds \mu(s)]$ :

$$\frac{\partial}{\partial \tau} f(\tau) = i\mu(t+\tau)f(\tau), \quad (2.4a)$$

$$f(\tau) = 1 + i \int_0^\tau ds \mu(t+s)f(s). \quad (2.4b)$$

By combining these we can obtain

$$\frac{\partial}{\partial \tau} f(\tau) = i\mu(t+\tau) \left[ 1 + i \int_0^\tau ds \mu(t+s)f(s) \right]. \quad (2.4c)$$

Now we calculate the stochastic average  $\langle f(\tau) \rangle$ , decorrelating  $\mu(t+s)$  from the function  $f(s)$  in Eq. (2.4) due to property (2.2). This procedure was used by us in a similar problem in our preceding paper,<sup>11</sup> Sec. III. It is justified in the Appendix of this paper [see the discussion of Eq. (A13)]. Thus  $\langle f(\tau) \rangle$  satisfies the equation

$$\frac{\partial}{\partial \tau} \langle f(\tau) \rangle = -a^2 \int_0^\tau ds e^{-(2/T)|\tau-s|} \langle f(s) \rangle, \quad (2.4d)$$

where we have inserted the correlation (2.1) derived above. The solution of this simple integro-differential equation can be found very easily:

$$\begin{aligned} \langle f(\tau) \rangle &= \frac{1}{2} \left[ \frac{1}{T\lambda} + 1 \right] e^{-[(1/T)-\lambda]|\tau|} \\ &\quad - \frac{1}{2} \left[ \frac{1}{T\lambda} - 1 \right] e^{-[(1/T)+\lambda]|\tau|}, \end{aligned} \quad (2.5)$$

where

$$\lambda^2 = \frac{1}{T^2} - a^2. \quad (2.6)$$

The electric field  $E(t)$  thus has the following Fourier spectrum

$$\begin{aligned} S_F(\omega) &= 2 \operatorname{Re} \int_0^\infty d\tau e^{i\omega\tau} \langle E(t)E^*(t+\tau) \rangle \\ &= \frac{\frac{8\pi a^2}{T} |E_0|^2}{(\omega - \omega_L)^4 + \left[ \frac{4}{T^2} - 2a^2 \right] (\omega - \omega_L)^2 + a^4}. \end{aligned} \quad (2.7)$$

It is clear from these formulas that depending on the sign of  $1/T^2 - a^2$  we can have a singlet or a doublet for the electric field power spectrum. With the increase of the dwell time  $T$ , the central part of the power spectrum centered around  $\omega = \omega_L$  splits into two components located at frequencies  $\omega = \omega_L \pm [a^2 - (1/T^2)]^{1/2}$ . In either case, singlet as well as doublet, an important feature of the frequency-telegraph jump model is a non-Lorentzian spectrum for the laser light. This remark is independent of the structure near line center. In all cases the far wing of the power spectrum given by Eq. (2.7) falls off as  $1/\omega^4$ , i.e., faster than Lorentzian.

Figure 1 shows examples of the field spectrum for the parameter choice  $a = 0.2$ . In these frames the solid curve ( $F$ ) is the field spectrum, and dotted curves ( $L$ ) show Lorentzian profiles (all curves are normalized to unit peak value). For short switching times, such as  $T = 1$  [Fig. 1(c)], we observe a single peak, nearly Lorentzian. For larger values of  $T$  the field spectrum becomes indistinguishable from a Lorentzian for frequencies  $|\omega - \omega_L| < 2/T$ , but for very large detunings the profile goes as  $\omega^{-4}$ . For longer dwell times  $T$  the spectrum splits into two components; see Figs. 1(a) and 1(b). The frequency peaks occur at  $\omega \cong \omega_L \pm a$  and the wings fall as  $\omega^{-4}$ . Both the central portion and the far wings differ from Lorentzian shape.

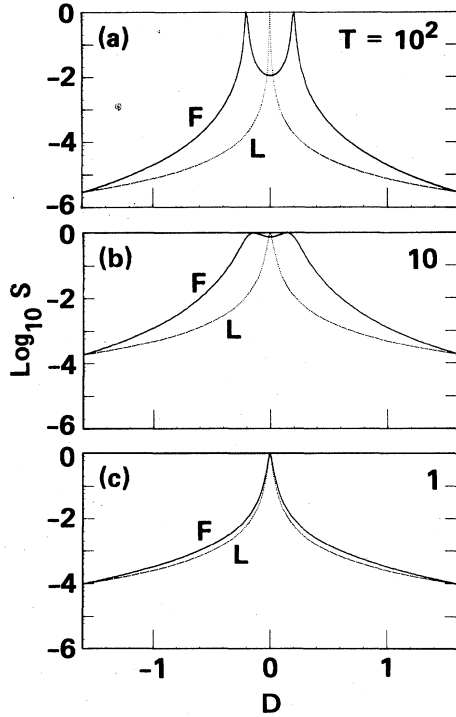


FIG. 1. Plot of  $\log_{10}S$  (solid curve  $F$ ) detuning  $D = \omega - \omega_L$  of the spectrometer frequency  $\omega$  from the mean laser frequency  $\omega_L$  for the spectrum  $S_F(D)$  of a laser field whose frequency undergoes bivalued random telegraph fluctuations of amplitude  $a = 0.2$  about the mean value  $\omega_L$ . (a) Mean interruption time  $T = 10^2$ , (b)  $T = 10$ , (c)  $T = 1$ . Dotted curve  $L$  shows Lorentzian profile, adjusted to agree with  $S_F(D)$  at edge of figure. All curves are normalized to unit peak value.

### III. ELEMENTARY TREATMENT OF ATOMIC RESPONSE

As the first simple example of elementary applications of the two-state random telegraph model of frequency fluctuations, we consider the direct calculations of the atomic population in the familiar two-level picture for the atom. In the Heisenberg picture and in the rotating wave approximation, the atomic equations of motion take the following form:<sup>12</sup>

$$\dot{\hat{\sigma}}_{12} = -(A/2 + i\Delta)\hat{\sigma}_{12} + \frac{i}{2}\Omega(t)(\hat{\sigma}_{22} - \hat{\sigma}_{11}), \quad (3.1a)$$

$$\dot{\hat{\sigma}}_{21} = -(A/2 - i\Delta)\hat{\sigma}_{21} - \frac{i}{2}\Omega^*(t)(\hat{\sigma}_{22} - \hat{\sigma}_{11}), \quad (3.1b)$$

$$\dot{\hat{\sigma}}_{11} = A\hat{\sigma}_{22} + \frac{i}{2}\Omega(t)\hat{\sigma}_{21} - \frac{i}{2}\Omega^*(t)\hat{\sigma}_{12}, \quad (3.1c)$$

$$\dot{\hat{\sigma}}_{22} = -A\hat{\sigma}_{22} - \frac{i}{2}\Omega(t)\hat{\sigma}_{21} + \frac{i}{2}\Omega^*(t)\hat{\sigma}_{12}, \quad (3.1d)$$

where we denote laser-atom detuning and radiative decay rate by  $\Delta$  and  $A$  and the atomic operators are (in the frame rotating with frequency  $\omega_L$ ) as usual  $\hat{\sigma}_{ij} = |i\rangle\langle j|$ . In Eqs. (3.1) the instantaneous Rabi frequency is defined to be

$$\Omega(t) = \frac{2d_{21}}{\hbar} E_0 \exp[-i\phi_L(t)] = \Omega_0 e^{-i\phi_L(t)}, \quad (3.2)$$

where  $d_{21}$  is the two-level transition dipole matrix element and  $\phi_L(t)$  is determined by the random telegraph  $\mu(t)$  as in (2.3b).

We calculate the inversion of a two-level atom described by Eq. (3.1) in the limit of exact resonance. This problem has been approached with a different noise model in our paper immediately preceding this one.<sup>11</sup> We can eliminate the dipole variables exactly, and from Eq. (3.1) we obtain the following integro-differential equation for  $w = \sigma_{22} - \sigma_{11}$ :

$$\dot{\hat{w}} = - \int_0^t ds e^{-(A/2)(t-s)} \times \text{Re}[\Omega^*(t)\Omega(s)]\hat{w}(s) - A(\hat{w} + 1). \quad (3.3)$$

The inversion and Rabi frequency decorrelate in this case, due to relation (2.2). Thus the stochastic expectation value of the inversion ( $w$  without circumflex) satisfies the equation

$$\dot{w} = -\Omega_0^2 \int_0^t ds [\langle f(t-s) \rangle e^{-(A/2)(t-s)} w(s)] - A(w + 1), \quad (3.4)$$

where  $\langle f(\tau) \rangle$  denotes the correlation function (2.5) derived above. It is easy to show that Eq. (3.4) is equivalent to a third-order differential equation with constant coefficients:

$$\begin{aligned} \ddot{w} + \left[ 2A + \frac{2}{T} \right] \dot{w} + \left[ \Omega_0^2 + a^2 + A \left[ \frac{2}{T} + \frac{5A}{4} \right] \right] w \\ + \left[ \frac{2\Omega_0^2}{T} + \frac{A\Omega_0^2}{2} + A^2 \left[ \frac{1}{T} + \frac{A}{4} \right] + Aa^2 \right] w \\ + \left[ A^2 \left[ \frac{1}{T} + \frac{A}{4} \right] + Aa^2 \right] = 0. \end{aligned} \quad (3.5)$$

We are mostly interested here in the effect of the frequency jump on the atomic response in the strong-field case ( $\Omega_0 > A$ ). We also take  $1/T \gg A$ . In that case Eq. (3.5) reduces to

$$\ddot{w} + \frac{2}{T}\dot{w} + (\Omega_0^2 + a^2)w + \frac{2\Omega_0^2}{T}w = 0. \quad (3.6)$$

Note that this differs from the corresponding equation derived in Sec. III of the preceding paper. We will discuss elsewhere the relation of these equations to similar third-order equations derived and studied by Burshtein.<sup>6</sup>

Equation (3.5) can be solved exactly, of course, in terms of the roots of a cubic polynomial. Just as in the preceding paper, it is easier to consider the two interesting limits of very slow and very fast switching of the frequency telegraph. For  $T \rightarrow \infty$  we obtain from Eq. (3.6) the following solution:

$$w(t) = \frac{a^2 + \Omega_0^2 \cos \sqrt{\Omega_0^2 + a^2} t}{a^2 + \Omega_0^2} w(0). \quad (3.7)$$

Note that this is a familiar expression in a different con-

text. This formula is equivalent to the *noise-free* but off-resonant evolution of the atomic inversion, with the detuning given by the frequency jump size  $a$ . For infinite but very large values of  $T$  the atomic inversion performs damped oscillations with frequency and damping rate given by

$$\Omega = \sqrt{\Omega_0^2 + a^2} \quad \text{and} \quad \gamma = \frac{2a^2}{a^2 + \Omega_0^2} \frac{1}{T}. \quad (3.8)$$

On the other hand, if  $T$  is very short ( $1/T \ll a, \Omega_0$ ) the frequency and damping rate are easily found to be quite different:

$$\Omega = \Omega_0 \quad \text{and} \quad \gamma = \frac{a^2 T}{4}. \quad (3.9)$$

These conclusions should be compared with our discussion of phase interruption presented in the preceding paper, keeping in mind that the parameter labeled  $a$  is physically different.

In Fig. 2 we show an example of random frequency-telegraph influence on atomic populations  $\langle \hat{\sigma}_{11} \rangle$  and  $\langle \hat{\sigma}_{22} \rangle$ , taking  $a=1.25$  and three different switching rates  $T^{-1}$  (all parameters in this paper are expressed in units of the Rabi frequency  $\Omega_0$ , and we set  $\Omega_0=1$ ). The change of

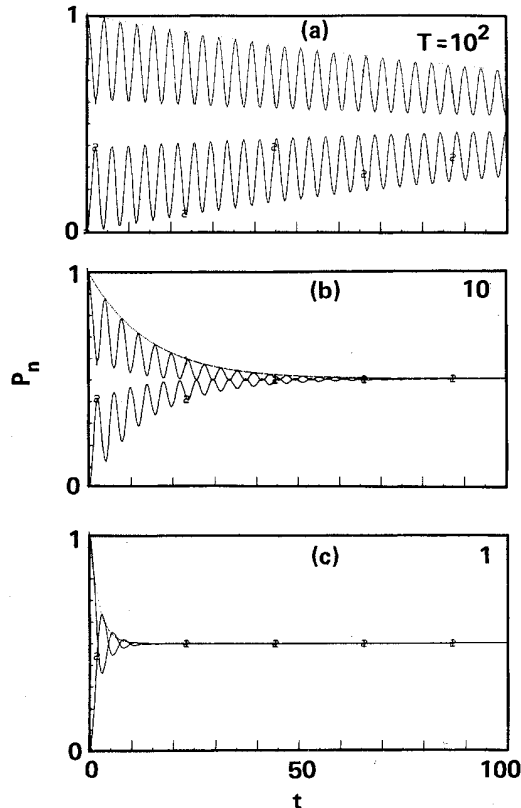


FIG. 2. Populations  $P_n(t) = \langle \hat{\sigma}_{nn}(t) \rangle$  vs  $t$  for two-level atom-excited frequency noisy laser of Fig. 1. Frames show results for  $a=1.25$  and for different mean interruption times  $T$ : (a)  $T=10^2$ , (b)  $T=10$ , (c)  $T=1$ . Dotted curve is envelope with effective relaxation rate. Mean Rabi frequency is set to unity,  $\Omega_0=1$ .

the time evolution of the atomic populations with the increase of the jumping time  $T$  can be clearly seen in these figures.

As our second elementary example to illustrate the effect of non-Lorentzian behavior of the frequency, we calculate the ionization rate of a two-level atom, including the effect of an intermediate resonance. We define the direct rate of relaxation from the excited level into the continuum to be  $R_{2c}$ .<sup>13</sup>

Only the far-wing ionization rate is interesting here, so we take  $\Delta \gg \Omega_0$ , where  $\Delta$  and  $\Omega_0$  are the detuning and Rabi frequency of the bound-bound excitation. In this case the total ionization rate is given by the following convolution integral:<sup>14</sup>

$$R_I = \frac{R_{2c}}{4\beta} \text{Re} \int_0^\infty ds e^{-(\beta+i\Delta)s} C_F(s), \quad (3.10)$$

where  $\beta = \frac{1}{2}(A + R_{2c})$  and  $C_F(\tau)$  is the correlation function of the driving electric field amplitudes. As in (3.1),  $A$  is the Einstein coefficient for spontaneous emission. Simple calculation leads to the following expression:

$$R_I = \frac{R_{2c}\Omega_0^2}{4\beta} \frac{\left[ \beta + \frac{2}{T} \right] \left[ \beta^2 + \frac{2\beta}{T} + a^2 \right] + \Delta^2 \beta}{\left[ \Delta^2 - \beta^2 - \frac{2\beta}{T} - a^2 \right]^2 + 4\Delta^2 \left[ \beta + \frac{1}{T} \right]^2}. \quad (3.11)$$

If  $\Delta$  is larger than any of the other parameters involved in the ionization rate ( $\Delta > \beta, 1/T$ ), i.e., if only very far-wing behavior of  $R_I$  is investigated, we obtain from Eq. (3.11) the simplified expression

$$R_I \simeq R_{2c} \frac{\Omega_0^2}{4\Delta^2} \left[ 1 + \frac{\left[ \beta^2 + \frac{2}{T}\beta + a^2 \right] \left[ 1 + \frac{2}{T\beta} \right]}{\Delta^2} \right]. \quad (3.12)$$

This formula indicates that the bandwidth-dependent effects occur first in the term proportional to  $1/\omega^4$ , as in the OU case.<sup>13</sup>

#### IV. LIGHT SCATTERING WITH FREQUENCY FLUCTUATION

In this section we investigate the light scattering spectrum<sup>15</sup> of a nondegenerate two-level atom resonantly excited by an external laser whose frequency fluctuates according to (2.1) or (2.3). In the steady-state limit the stationary spectrum is proportional to

$$S(\omega) = 2 \text{Re} \int_0^\infty d\tau e^{-(\gamma_s + i\omega)\tau} C(\tau), \quad (4.1)$$

where  $C(\tau)$  is the stationary dipole correlation function,

$$C(\tau) = \lim_{t \rightarrow \infty} \langle \hat{\sigma}_{21}(t+\tau) \hat{\sigma}_{12}(t) \rangle, \quad (4.2)$$

and  $\gamma_s$  is the bandwidth of the spectrometer being used to analyze the scattered light.<sup>16</sup>

It is clear that the spectrum is the Laplace transform of  $C(\tau)$ . An equation for  $C(\tau)$  can be obtained as follows.

First we define an operator vector  $\hat{V}(t, \tau)$  with the following four components:

$$\hat{V}_1(t, \tau) = \hat{\sigma}_{21}(t + \tau) \hat{\sigma}_{12}(t), \quad (4.3a)$$

$$\hat{V}_2(t, \tau) = \hat{\sigma}_{12}(t + \tau) \hat{\sigma}_{12}(t) \exp[2i\phi_L(t + \tau)], \quad (4.3b)$$

$$\hat{V}_3(t, \tau) = \hat{\sigma}_{11}(t + \tau) \hat{\sigma}_{12}(t) \exp[i\phi_L(t + \tau)], \quad (4.3c)$$

$$\hat{V}_4(t, \tau) = \hat{\sigma}_{22}(t, \tau) \hat{\sigma}_{12}(t) \exp[i\phi_L(t + \tau)]. \quad (4.3d)$$

With the aid of Eqs. (3.1) and (3.2), a matrix equation for  $\hat{V}$  can be found. The average of its first component (in the limit  $t \rightarrow \infty$ ) is just the desired  $C(\tau)$ . The equation for  $\hat{V}(t, \tau)$  is

$$\frac{d}{d\tau} \hat{V}(t, \tau) = -iM_L[\mu(t + \tau)] \hat{V}(t, \tau), \quad (4.4)$$

where  $M$  is given by the stochastic matrix

$$M_L[\mu(t + \tau)] = \begin{pmatrix} -\frac{iA}{2} - \Delta & 0 & -\frac{\Omega_0}{2} & \frac{\Omega_0}{2} \\ 0 & -\frac{iA}{2} + \Delta - 2\mu(t + \tau) & \frac{\Omega_0}{2} & -\frac{\Omega_0}{2} \\ -\frac{\Omega_0}{2} & \frac{\Omega_0}{2} & -\mu(t + \tau) & iA \\ \frac{\Omega_0}{2} & -\frac{\Omega_0}{2} & 0 & -\mu(t + \tau) - iA \end{pmatrix}. \quad (4.5)$$

Because  $\hat{\sigma}_{ij}(t) \hat{\sigma}_{jk}(t) = \hat{\sigma}_{ik}(t)$ , the required initial condition takes the form

$$\hat{V}(\infty, 0) = \lim_{t \rightarrow \infty} (\hat{\sigma}_{22}(t), 0, \hat{\sigma}_{12}(t) \exp[i\phi_L(t)], 0). \quad (4.6)$$

The Appendix is devoted to the full solution of dynamical equations that can be cast in the form given by Eq. (4.4). We shall not give here the explicit analytical solution which can be found in the Appendix, but instead we

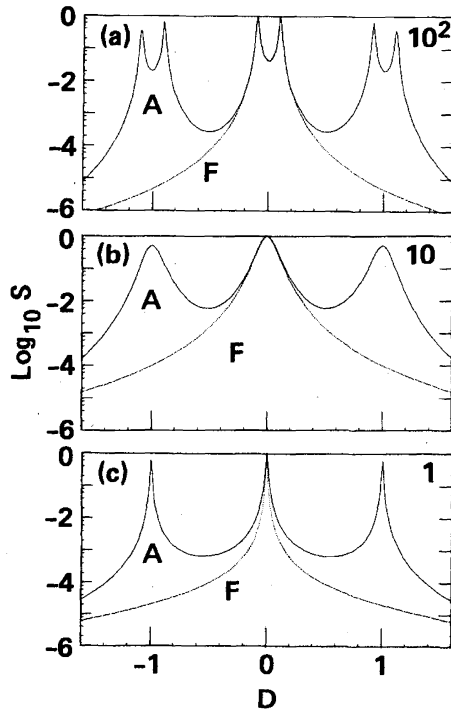


FIG. 3. Plots of  $\log_{10} S$  vs detuning  $D$  for emission spectrum of two-level atom (solid curve  $A$ ) and laser field (dotted curve  $F$ ) for laser frequency fluctuations,  $a=0.1$ . As in Fig. 2,  $\Omega_0=1$ , and as in Figs. 1 and 2 frames show results with different mean interruption times: (a)  $T=10^2$ , (b)  $T=10$ , (c)  $T=1$ . Other parameters: radiative decay rate  $A=10^{-7}$ , spectrometer width  $\gamma_S=10^{-8}$ .

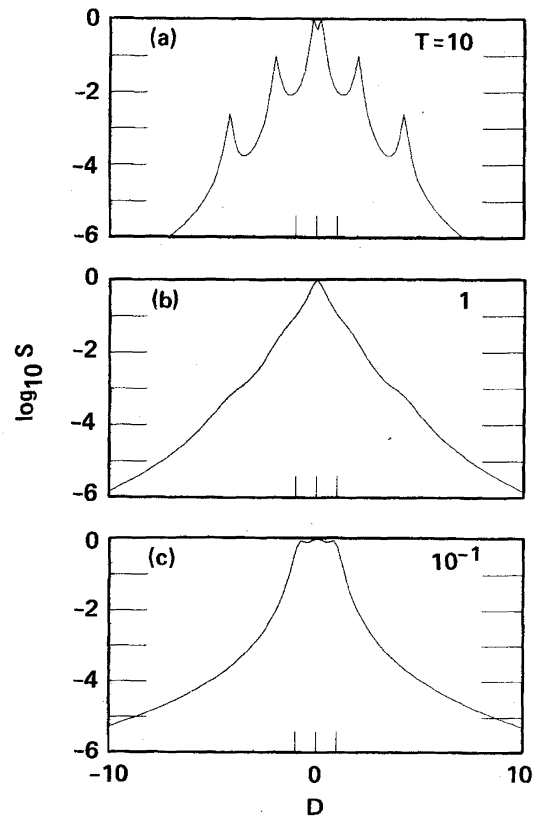


FIG. 4. Plots of  $\log_{10} S$  as in Fig. 2, but with a frequency jump  $a=2.0$  and different mean interruption times: (a)  $T=10$ , (b)  $T=1$ , (c)  $T=10^{-1}$ . Other parameters are the same as in Fig. 2.

show curves of the light scattering spectrum calculated numerically directly from Eq. (A.7). Figure 3 shows the scattered light spectrum of a two-level atom (solid curves) and the spectrum of the driving field (dotted curves) for excitation on resonance by a frequency noisy laser with  $a=0.1$  and three values of  $T$ . For  $T=1$  the ac Stark peaks have line shapes very close to Lorentzian profiles except in the very far wing. For larger values of  $T$  the non-Lorentzian profile of the scattered light spectrum fluorescence is clear (see also Fig. 2). For  $T=10^2$  and 10 we have  $\lambda^2 < 0$  [see Eq. (2.6)] and the two components  $\exp(\pm i|\lambda|)$  of the laser power spectrum given by Eq. (2.7) split the resonance lines of the fluorescence light.

In Fig. 4 we show curves of the same power spectrum, but with a much larger jump parameter of the frequency,  $a=2$ . These three curves clearly show that the Rabi splitting is affected by  $a$  according to Eq. (3.8). For small values of  $T$  the resonance peaks merge and the Stark splitting is given by  $\Omega_0$  only. Note that for  $T=10^{-1}$  we observe the motional narrowing of the spectral profiles discussed for phase fluctuations in the preceding paper. For  $T=10$  the pattern is more complicated. The peak positions can be predicted by a lengthy analysis of the matrix  $M_L$  to lie at positive and negative values given by  $a$ ,  $2a$ , and  $\Omega_0^2/2a$ , in good agreement with the figure.

#### V. LIGHT SCATTERING WITH ENERGY-SHIFTING COLLISIONS

In this section we investigate the scattered light spectrum of a nondegenerate two-level atom resonantly driven by strong monochromatic laser light in the presence of energy-shifting collisions. The incoherences due to collisions can be described by fluctuations of the atomic detuning.<sup>1</sup> The simplest model of such phase-interrupting collisions assumes that the atomic detuning  $\Delta$  in the atomic equations (3.1) should be replaced by

$$\Delta(t) = \omega_{21}(t) - \omega_L = \omega_{21} + \mu(t) - \omega_L = \Delta + \mu(t), \quad (5.1)$$

where  $\mu(t)$  is the instantaneous deviation of the atomic energy due to collision. This instantaneous frequency is a random telegraph signal described by Eqs. (2.1)–(2.6), and

$$M_C[\mu(t+\tau)] = \begin{pmatrix} -\frac{iA}{2} - \Delta - \mu(t+\tau) & 0 & -\frac{\Omega_0}{2} & \frac{\Omega_0}{2} \\ 0 & -\frac{iA}{2} + \Delta + \mu(t+\tau) & \frac{\Omega_0}{2} & -\frac{\Omega_0}{2} \\ -\frac{\Omega_0}{2} & \frac{\Omega_0}{2} & 0 & iA \\ \frac{\Omega_0}{2} & -\frac{\Omega_0}{2} & 0 & -iA \end{pmatrix}. \quad (5.3)$$

The required initial condition is

$$\hat{V}(\infty, 0) = \lim_{t \rightarrow \infty} [\hat{\sigma}_{22}(t), 0, \hat{\sigma}_{12}(t), 0]^T. \quad (5.4)$$

Note that the collisional interaction matrix  $M_C$  is different from the laser fluctuation interaction matrix given

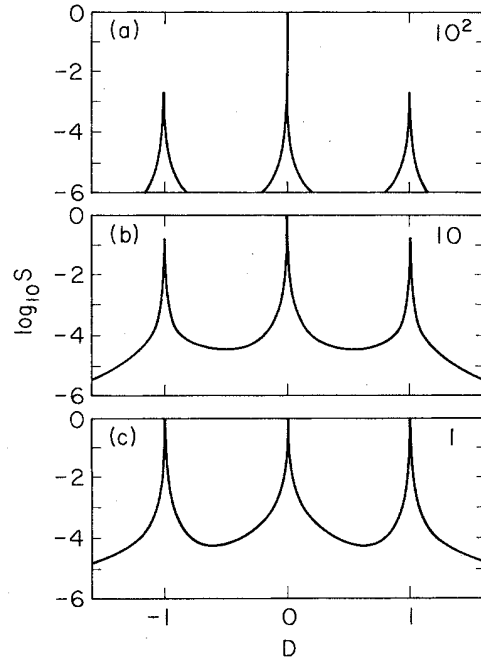


FIG. 5. Plots of  $\log_{10} S$  vs detuning  $D$  for emission spectrum of a two-level atom for collisional frequency fluctuations,  $a=0.1$ . As in Figs. 2 and 4,  $\Omega_0=1$  and consecutive frames show results with different mean interruption times: (a)  $T=10^2$ , (b)  $T=10$ , (c)  $T=1$ .

the two parameters  $a$  and  $T^{-1}$  can be interpreted, respectively, as the strength and frequency of collisions.

The correlation function  $C(\tau)$ , for the calculation of the fluorescence spectrum [see Eq. (4.1)], can be calculated from the same operator vector  $\hat{V}(t, \tau)$  given in Eqs. (4.3). In this case the field is assumed monochromatic so  $\phi_L(t) = \text{const}$ . For simplicity we take  $\phi_L(t) = 0$ . The equation for  $\hat{V}(t, \tau)$  follows from Eqs. (3.1) and (5.1):

$$\frac{d}{d\tau} \hat{V}(t, \tau) = -iM_C[\mu(t+\tau)] \hat{V}(t, \tau), \quad (5.2)$$

where the matrix  $M_C$  is given by

by Eq. (4.5).

The frequency fluctuations  $\mu(t)$  enter the interaction matrix  $M_C[\mu(t)]$  differently depending on whether their source is an atomic energy fluctuation or laser frequency fluctuation. It is interesting to note that these differences show up first in the equations of motion involving atomic

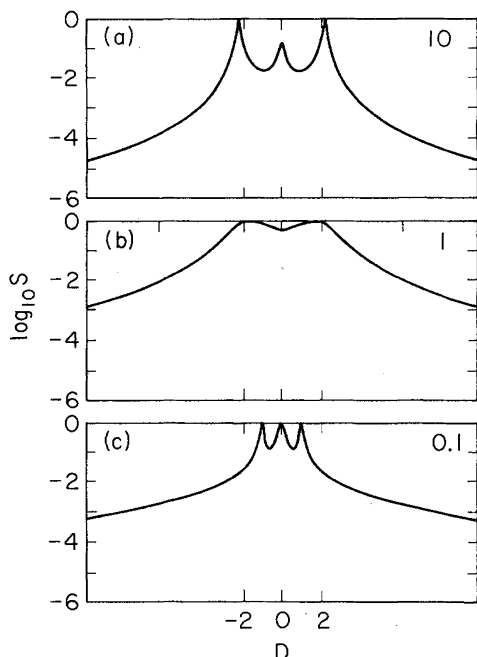


FIG. 6. Plots of  $\log_{10} S$  as in Fig. (5) but with a different collisional frequency jump  $a=2.0$  and different mean interruption times: (a)  $T=10$ , (b)  $T=1$ , (c)  $T=10^{-1}$ . Other parameters the same as in Fig. 5.

correlations. Single expectation values involving just dipole moments and atomic inversions do not show these differences. The solution for the light scattering spectrum is given in the Appendix.

Figure 5 shows the light scattering spectrum of a two-level atom for excitation on resonance with energy-shifting collisions with  $a=0.1$  and three values of  $T$ . The most important feature of these curves compared to laser frequency fluctuations is the disappearance of the additional splitting of the central and wing components. This can be seen from the following argument. For laser frequency fluctuations the Rabi amplitude in the proper limit has the form of  $\exp(\pm i|\lambda|)$  leading to a splitting of the spectral components. For collisional noise the Rabi frequency is independent of  $\mu(t)$  and only the atomic energy fluctuates. Figure 6 shows the same resonance fluorescence power spectrum but with a much larger jump parameter of the collisional noise  $a=2$ . We note that the peak splittings are just as should be expected from the two different effective Rabi frequencies already obtained in Eqs. (3.8) and (3.9) for short and long telegraph switching times.

## VI. SUMMARY

In this paper we have discussed characteristics of strong atom-field interactions in the presence of pre-Gaussian<sup>7</sup> frequency-telegraph noise. We have shown that when such a model is used for laser frequency fluctuations it leads to a nonexponential autocorrelation of the laser electric field and a non-Lorentzian laser power spectrum, one that falls off more rapidly in the wings. We have shown

that the telegraph model of laser frequency fluctuations leads to exactly soluble atomic equations of motion, i.e., the response functions of an atomic system exposed to such noise can be determined explicitly in finite terms. We have discussed the population dynamics as well as the light scattering spectrum and we have studied the effect of the non-Lorentzian bandshapes on far-wing absorption rates.

We have shown that a frequency telegraph can also be used to introduce collisional noise into the laser-atom interaction, and we have studied the atomic response to strong laser excitation under such conditions. Since only the difference of the laser and atomic frequencies (the detuning) enters in the rotating-wave-approximation atomic equations, it might be thought redundant to study collisional noise separately from laser noise. However, a comparison of the light scattering spectrum of the atom calculated in the two cases (laser frequency noise, collisional frequency noise) shows that the origin of the frequency noise makes a difference. This is perhaps surprising, but it has also been observed with other noise models and will be discussed elsewhere.<sup>17</sup>

These remarks point, of course, to the most obvious characteristic of our results. This is that the response of the atom to strong laser excitation not only exhibits interesting characteristics in the presence of noise, but that these characteristics can differ significantly depending on the model of the noise and even within a given noise model. The sharp spikes shown in our light scattering spectra in Figs. 3(a) and 4(a), as contrasted with those in Figs. 3(c) and 4(c), are clear evidence for this. The differences with spectra following from other noise models is just as great or greater. One conclusion is that, as experimental tests of various kinds become available, it will be valuable to have a variety of flexible theoretical models on hand.

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## APPENDIX

In our earlier publications<sup>7</sup> and in the preceding paper<sup>11</sup> we have given a more detailed description of telegraph signals as examples of a special class of Markov chains. We recall now the most important properties of such stochastic processes. It is well known that the dynamics of a Markov chain is completely described by the following Chapman-Kolmogorov-Smoluchowski (CKS) equation for the joint probability distribution  $p(at|\alpha_0 t_0)$  for the chain's possible states  $\alpha$  and  $\alpha_0$ :

$$\begin{aligned} \frac{\partial}{\partial t} p(\alpha, t | \alpha_0, t_0) = & -c(\alpha, t) p(\alpha, t | \alpha_0, t_0) \\ & + \sum_{\beta} c(\alpha, t) f(\alpha | \beta; t) p(\beta, t | \alpha_0, t_0), \end{aligned} \quad (\text{A1})$$

where  $c(\alpha, t)$  is the frequency that the telegraph in the state at time  $t$  will change its state in the interval  $t, t+dt$  and the function  $f(\alpha | \beta; t)$  can be interpreted as the conditional probability of making a change from  $\beta$  during the interval  $t, t+dt$ , given the transition ends in  $\alpha$ .

In this paper we specialize Eq. (A1) to a two-state ( $\alpha$  and  $-\alpha$ ) telegraph noise described in Sec. II. For the two-state telegraph noise we have

$$c(\alpha, t) = \frac{1}{T} \quad \text{and} \quad f(\alpha | \beta; t) = \delta_{\alpha, -\beta} \quad (\text{A2})$$

and accordingly the CKS equation (A1) takes the form

$$\begin{aligned} \frac{\partial}{\partial t} p(\alpha, t | \alpha_0, t_0) \\ = -\frac{1}{T} p(\alpha, t | \alpha_0, t_0) + \frac{1}{T} p(-\alpha, t | \alpha_0, t_0) \end{aligned} \quad (\text{A3})$$

with the initial condition  $p(\alpha, t_0 | \alpha_0, t_0) = \delta_{\alpha, \alpha_0}$ . This equation can be solved easily, leading for  $t \geq t_0$  to

$$\begin{aligned} p(\alpha, t | \alpha_0, t_0) = & \frac{1}{2} \delta_{\alpha, \alpha_0} (1 + e^{-2(t-t_0)/T}) \\ & + \frac{1}{2} \delta_{\alpha, -\alpha_0} (1 - e^{-2(t-t_0)/T}). \end{aligned} \quad (\text{A4})$$

With the help of this solution we calculate the correlation function (2.1) [with  $g(\alpha_0)$  defined in Sec. II]:

$$\begin{aligned} \langle \mu(t+\tau) \bar{\mu}(t) \rangle = & \sum_{\alpha, \alpha_0} \alpha \alpha_0 p(\alpha, t+\tau | \alpha_0, t_0) g(\alpha_0) \\ = & a^2 e^{-2|\tau|/T}, \end{aligned} \quad (\text{A5})$$

as predicted from the simple arguments used in Sec. II.

In the case of laser-atom interactions, the dynamical equations of motion, which have their origin in the basic equation (3.1), can be written in the following general form:

$$\frac{d\hat{V}}{dt} = -iM[x(t)]\hat{V}(t) \quad (\text{A6})$$

with a given matrix  $M$  which depends locally on the external arbitrary noise  $x(t)$ . For Markov chains we can write the following exact equation for the marginal average  $V_a(t)$  of the quantity  $V(t)$  with a random telegraph noise  $x(t)$  described by Eq. (A3):

$$\frac{d}{dt} V_a(t) = \left[ -iM(\alpha) - \frac{1}{T} \right] V_a(t) + \frac{1}{T} V_{-a}(t). \quad (\text{A7})$$

The stochastic expectation value of  $V$  is given by

$$\langle \hat{V}(t) \rangle = \sum_{\alpha} g(\alpha) V_{\alpha}(t) = \frac{1}{2} [V_a(t) + V_{-a}(t)]. \quad (\text{A8})$$

Equation (A7) is a special case of a master equation associated with the general CKS relation. Such master equations have been introduced into quantum optics first by Burshtein.<sup>6</sup> This master equation gives a full description of the stochastic expectation value of  $V$  and as such will play a fundamental role in all our applications. Due to the obvious symmetric (A8) and antisymmetric superpositions of the marginal averages forming the solutions of Eq. (A7) we can write the following closed system of equations involving only stochastic expectation values of  $V$ :

$$\frac{d}{dt} \begin{bmatrix} \langle \hat{V}(t) \rangle \\ V_A(t) \end{bmatrix} = \begin{bmatrix} -iM_S & -iM_A \\ iM_A & -M_S - \frac{2}{T} \end{bmatrix} \begin{bmatrix} \langle \hat{V}(t) \rangle \\ V_A(t) \end{bmatrix}, \quad (\text{A9})$$

where

$$V_A(t) = \frac{1}{2} [V_a(t) - V_{-a}(t)] \quad (\text{A10})$$

and

$$M_{S,A} = \frac{1}{2} [M(a) \pm M(-a)]. \quad (\text{A11})$$

For time-independent  $M$  matrices, a closed formula for the stochastic expectation value of the  $V$  operator can be obtained from Eq. (A9). From this equation we obtain the following exact integro-differential equation for  $V(t)$  [if  $V_A(0)=0$ ]:

$$\begin{aligned} \frac{d}{dt} \langle \hat{V}(t) \rangle = & -iM_S \langle \hat{V}(t) \rangle \\ & -M_A \int_0^t ds e^{[-iM_S - (2/T)](t-s)} M_A \langle \hat{V}(s) \rangle, \end{aligned} \quad (\text{A12})$$

which has the Laplace-transform solution

$$\langle \hat{V}(t) \rangle = \int \frac{dz}{2\pi i} \frac{e^{zt}}{z + iM_S + M_A \frac{1}{z + iM_S + \frac{2}{T}} M_A} \langle \hat{V}(0) \rangle. \quad (\text{A13})$$

This result can be generalized: The solution of any dynamical equation of motion that can be written in the form (A6) can be averaged over the telegraph noise  $x(t)$  exactly. Application<sup>18</sup> of Eq. (A13) with the definitions (4.3)–(4.6) or (5.1)–(5.4) leads to the stationary power spectrum of light scattering due to laser or collisional fluctuations, respectively. For  $A=0$  and  $\Delta=0$  the dynamical equations of motion for the atomic correlations, Eqs. (4.4) and (5.2), have a steady-state initial condition  $V_A(\infty)=0$ . Following Eq. (4.1) the light scattering spectrum is

$$S(\omega) = 2 \operatorname{Re} \langle \tilde{V}_1(z=i\omega + \gamma_S) \rangle, \quad (\text{A14})$$

where  $\langle \tilde{V}_1(z) \rangle$  is the Laplace transform of the atomic correlation function. For the case of  $A=0$  and no detuning we calculate from Eq. (A13) that these correlations have the following forms.



Laser frequency fluctuations,

$$\tilde{V}_1(z) = \frac{1}{2} \frac{(z+4b)(z+b-c) + \frac{\Omega_0^2}{2} d^2}{z(z+4b)(z+b-c) + \frac{\Omega_0^2 d^2 z}{2} + \frac{\Omega_0^2}{2} (z+4b)}. \quad (\text{A15})$$

Collisional fluctuations,

$$\tilde{V}_1(z) = \frac{1}{2} \frac{(z+b) + \frac{\Omega_0^2}{2}}{(z+b-c)[z(z+b+c) + \Omega_0^2]}. \quad (\text{A16})$$

In Eqs. (A15) and (A16) we have used the following nota-

tions:

$$\begin{aligned} b &= \frac{a^2 \left[ \left[ z + \frac{2}{T} \right]^2 + \frac{\Omega_0^2}{2} \right]}{\left[ z + \frac{2}{T} \right] \left[ \left[ z + \frac{2}{T} \right]^2 + \Omega_0^2 \right]}, \\ c &= \frac{a^2 \Omega_0^2}{2 \left[ z + \frac{2}{T} \right] \left[ \left[ z + \frac{2}{T} \right]^2 + \Omega_0^2 \right]}, \\ d &= 1 - \frac{2a^2}{\left[ \left[ z + \frac{2}{T} \right]^2 + \Omega_0^2 \right]}. \end{aligned} \quad (\text{A17})$$

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