

Pre-Gaussian model of laser noise: Application to resonant two-photon ionization

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We present a theoretical treatment of resonant two-photon ionization in the presence of a noisy laser. The atom is modeled by a two-level system, while the statistical fluctuations of the field are described within the framework of the pre-Gaussian model. We show that previous results obtained by using Ornstein-Uhlenbeck statistics are recovered in the appropriate limit in the case of frequency fluctuations. We also give new results in the case of phase and amplitude noise.

I. INTRODUCTION

It is by now recognized that nearly all kinds of laser-atom interactions can be strongly affected by laser noise. There is, first of all, a practical reason to this fact, since many experiments require very high powers which can only be obtained in pulsed operation, and thus at the expense of poorly stabilized laser beams. Furthermore, strong fields imply a highly nonlinear coupling between matter and radiation, thus enabling the statistical fluctuations to enter the process in a nonperturbative way.

This is particularly true in the case of resonant interactions. For such processes, moreover, it should be noted^{1,2} that the new time scale introduced in the problem by the laser bandwidth can often be of the same order of magnitude as other typical rates, corresponding, for instance, to radiative decay, Rabi oscillations, or detuning. It is then obvious that the statistical properties of the laser beam can in no way be neglected in this case.

One of the most commonly used approaches for dealing with this problem is that of a two-level atom excited by a field whose statistical properties are described within the framework of the phase-diffusion model, the chaotic field model, or the Gaussian-amplitude model.³ This approach has been widely used, for example, in the study of ac Stark splitting⁴ and resonance fluorescence.^{3,5} More recently Eberly,⁶ Yeh and Eberly,^{1,2} and Jackson and Swain⁷ have considered an extension of this approach, in order to treat resonant multiphoton ionization (MPI). Their work emphasized, in particular, the importance of using statistical models, taking into account the possible non-Lorentzian features of the laser band shape.

On the other hand, an alternative approach to the use of Gaussian statistics has been proposed recently.⁸⁻¹² It is based on the random-telegraph model,⁸ and is called pre-Gaussian in the literature.¹⁰ It has been shown to be much easier to handle than the previous models and to reproduce, in the appropriate limit, the results of Gaussian statistics. It has already been applied to the study of resonance fluorescence⁸⁻¹¹ and nonresonant MPI.¹³

In this paper, our aim is to extend the analysis to the case of resonant MPI. Our treatment will parallel closely the phase-diffusion analysis of Yeh and Eberly,² and the pre-Gaussian model will be shown to reproduce correctly Ornstein-Uhlenbeck (OU) statistics in the case of frequency fluctuations. Our approach will also be shown to hold in the case of amplitude and phase fluctuations, and to provide exact solutions, since all calculations can be reduced to linear algebra.

II. THE PRE-GAUSSIAN MODEL

The pre-Gaussian model that we are considering here consists in the superposition of n statistically independent dichotomic Markov processes, also called random-telegraph signals.⁸ Assuming that each signal can take the two values $\pm a$, the resulting process $x(t)$ will be defined on the set of $n+1$ values: $-na, -(n-2)a, \dots, (n-2)a$, and na . The initial probability distribution for $x(t)$ is easily found to be^{9,10}

$$g_n(\alpha) = \frac{n!}{\left[\frac{n+\alpha}{2}\right]! \left[\frac{n-\alpha}{2}\right]!} \left(\frac{1}{2}\right)^n, \quad (1)$$

where α labels any possible state of the system. It should be noted that, since that distribution has a binomial form, the requirement that the variance of the process described by the pre-Gaussian model be independent of the number n of signals implies that the quantity a be renormalized in the following way:

$$a \rightarrow \bar{a} = a/\sqrt{n}. \quad (2)$$

Since the process is Markovian, the conditioned probability density function associated with it, namely, $P[\alpha\bar{a}, t | \alpha_0\bar{a}, t_0]$, is shown to satisfy the following Chapman-Kolmogorov equation:^{9,10}

$$\frac{\partial P[\alpha \bar{a}, t | \alpha_0 \bar{a}, t_0]}{\partial t} = -\frac{n}{T} P[\alpha \bar{a}, t | \alpha_0 \bar{a}, t_0] + \frac{n}{T} \left[\frac{n+\alpha}{2n} P[(\alpha-2)\bar{a}, t | \alpha_0 \bar{a}, t_0] + \frac{n-\alpha}{2n} P[(\alpha+2)\bar{a}, t | \alpha_0 \bar{a}, t_0] \right], \quad (3)$$

where T denotes the mean switching time of the signals. The above equation involves finite differences with respect to the parameter $\alpha \bar{a}$. In the limit $n \rightarrow \infty$, this parameter becomes a continuous one and Eq. (3) reduces to a partial differential equation. In fact, it can be shown that the new quantity defined as

$$P(y, t; y_0, t_0) \equiv \lim_{n \rightarrow \infty} g_n(\alpha) P[\alpha \bar{a}, t | \alpha_0 \bar{a}, t_0] \quad (4)$$

satisfies the Fokker-Planck equation

$$\begin{aligned} \frac{\partial}{\partial t} P(y, t; y_0, t_0) &= \frac{\gamma^2 W}{2} \frac{\partial^2}{\partial y^2} P(y, t; y_0, t_0) \\ &+ \gamma \frac{\partial}{\partial y} [y P(y, t; y_0, t_0)], \end{aligned} \quad (5)$$

with

$$\gamma = \frac{2}{T}, \quad W = a^2 T. \quad (6)$$

$$\langle e^{i\phi(t) - i\phi(t')} \rangle_n = \left[\frac{1}{2} \left[\frac{1}{T\lambda} + 1 \right] \exp \left[- \left[\frac{1}{T} - \lambda \right] |t - t'| \right] - \frac{1}{2} \left[\frac{1}{T\lambda} - 1 \right] \exp \left[- \left[\frac{1}{T} + \lambda \right] |t - t'| \right] \right]^n, \quad (9)$$

where

$$\phi(t) \equiv \int^t x(s) ds, \quad \lambda^2 \equiv \frac{1}{T^2} - \bar{a}^2. \quad (10)$$

In the limit $n \rightarrow \infty$, Eq. (9) is easily found to reduce to the form¹⁰

$$\begin{aligned} \langle e^{i\phi(t) - i\phi(t')} \rangle_\infty \\ = \exp \left[-\frac{W}{2} \{ |t - t'| + [\exp(-\gamma |t - t'|) - 1] \} \right], \end{aligned} \quad (11)$$

which is well known from OU statistics.² As shown in Ref. 10, the laser power spectra associated with those three kinds of correlation functions are non-Lorentzian.

III. GENERAL THEORY

The simplest model for dealing with resonant MPI is the so-called "extended two-level model" proposed by Yeh and Eberly.² It contains two bound states $|1\rangle$ and $|2\rangle$ and a continuum $|c\rangle$. The bound-free dipole-matrix elements are assumed to depend only weakly on the energy, so that only equations of motion for the density-matrix elements ρ_{ij} associated with the bound states are required. They can be written, in the rotating-wave approximation,

$$\dot{\rho}_{12} = -(\beta + i\Delta)\rho_{12} - \frac{i}{2}\Omega(2\rho_{22} - n), \quad (12a)$$

This shows that, in the limit $n \rightarrow \infty$, one recovers from the pre-Gaussian model the well-known OU one. It corresponds to the fact that, in that limit, the pre-Gaussian model describes a random walk on the real line.

When considering a laser beam whose parameters fluctuate according to the pre-Gaussian model, three kinds of correlation functions have to be known.¹⁰ The first one, which is associated with amplitude fluctuations, is given by

$$\langle x(t)x(t') \rangle_n = a^2 e^{-2|t-t'|/T}, \quad (7)$$

where the index n indicates that the pre-Gaussian process involves n telegraph signals. Expression (7) is seen to be independent of n . The second one, which is associated with phase fluctuations, reads

$$\langle e^{ix(t) - ix(t')} \rangle_n = (\cos^2 \bar{a} + \sin^2 \bar{a} e^{-2|t-t'|/T})^n. \quad (8)$$

Finally, in the case of frequency fluctuations, we have

$$\dot{\rho}_{21} = -(\beta - i\Delta)\rho_{21} + \frac{i}{2}\Omega^*(2\rho_{22} - n), \quad (12b)$$

$$\dot{\rho}_{22} = -(A + R_{2c})\rho_{22} + \frac{i}{2}\Omega\rho_{21} - \frac{i}{2}\Omega^*\rho_{12}, \quad (12c)$$

$$\dot{n} = -R_{2c}\rho_{22}, \quad n \equiv \rho_{11} + \rho_{22}. \quad (12d)$$

In those equations Δ denotes the detuning from the intermediate resonance between levels 1 and 2, and Ω is the Rabi frequency (at resonance) which is defined by

$$\Omega = 2\mathbf{D}_{21} \cdot \mathcal{E} / \hbar. \quad (13)$$

The electric field is given, in the dipole approximation, by

$$\mathbf{E}(t) = \mathcal{E} e^{-i(\omega t + \varphi)} + \text{c.c.}, \quad (14)$$

so that $\mathcal{E}$ is the amplitude of the field and φ its phase (these two quantities being likely to fluctuate) and $\mathbf{D}_{21}$ is the dipole-matrix element between levels 1 and 2. Moreover, A is Einstein's coefficient for spontaneous emission and the quantity R_{2c} accounts for the direct relaxation from level 2 into the continuum. It is defined as

$$R_{2c} = 2\pi(D_{2c}^2 / \hbar) |\mathcal{E}|^2. \quad (15)$$

We have also introduced in Eqs. (12) the quantity

$$\beta \equiv \frac{1}{2}(A + R_{2c}). \quad (16)$$

In what follows, three kinds of laser fluctuations will be considered. In the case of amplitude fluctuations, we have to perform in Eqs. (12) the replacement

$$\mathcal{E} \rightarrow \mathcal{E}_0[1+x(t)] , \quad (17)$$

where $\mathcal{E}_0$ denotes the mean value of the electric field strength. For phase fluctuations it becomes (in the rotating-wave approximation)

$$\mathcal{E} \rightarrow \mathcal{E}_0 e^{-ix(t)} , \quad (18)$$

while for frequency fluctuations the replacement $\mathcal{E} \rightarrow \mathcal{E}_0 e^{-i\phi(t)}$, combined with the change of variables

$$\rho'_{12} = e^{i\phi(t)} \rho_{12} , \quad (19a)$$

$$\rho'_{21} = e^{-i\phi(t)} \rho_{21} , \quad (19b)$$

$$\rho'_{22} = \rho_{22} , \quad (19c)$$

$$n' = n , \quad (19d)$$

is easily shown to result in the substitution

$$\Delta \rightarrow \Delta - \dot{\phi}(t) = \Delta - x(t) , \quad (20)$$

where we have used Eq. (10).

From now on, the equations satisfied by the density-matrix elements $\rho_{ij,n}$ (or $\rho'_{ij,n'}$) will be written in the symbolic form

$$\dot{\mathbf{V}}(t) = \mathbf{M}[x(t)]\mathbf{V}(t) , \quad (21)$$

where the vector $\mathbf{V}(t)$ is defined as

$$\mathbf{V}(t) \equiv (\rho_{12}(t), \rho_{21}(t), \rho_{22}(t), n(t))^T , \quad (22)$$

and the $\mathbf{M}$ matrix depends on the kind of fluctuations considered.

The quantity we are interested in is the total ionization rate R_I . It is defined by the requirement that¹

$$\int_0^\infty [\langle n(t) \rangle - e^{-R_I t}] dt = 0 , \quad (23)$$

which is the same as

$$R_I = 1 / \int_0^\infty \langle n(t) \rangle dt , \quad (24)$$

where $\langle \rangle$ denotes an average with respect to the field fluctuations. Note that, in the case of a purely coherent field (no fluctuations), the resolution of Eqs. (12) yields the “coherent” ionization rate¹

$$R_I^{\text{coh}} = R_{2c} \frac{R_{12}(\Delta)}{2R_{12}(\Delta) + A + R_{2c}} , \quad (25a)$$

where

$$R_{12}(\Delta) \equiv \frac{1}{4} |\Omega|^2 \frac{2\beta}{\Delta^2 + \beta^2} . \quad (25b)$$

The important point for calculating statistical averages as in (24) is now as follows. Combining the Chapman-Kolmogorov equation (3) for the probability density function and the dynamical equation (21) for $\mathbf{V}(t)$, a *master equation* can be derived for the so-called marginal averages $\mathbf{V}_\alpha(t)$.^{9,10,14} It reads (in components)

$$\begin{aligned} \dot{V}_{\alpha,i}(t) = & \left[[M(\alpha \tilde{a})]_i \delta_\alpha^\beta - \frac{n}{T} \delta_\alpha^\beta \delta_i^j + \frac{(n+\alpha)}{2T} \delta_{\alpha-2}^\beta \delta_i^j \right. \\ & \left. + \frac{(n-\alpha)}{2T} \delta_{\alpha+2}^\beta \delta_i^j \right] V_{\beta,j}(t) , \end{aligned} \quad (26)$$

the indices α, β taking the values $-n, -n+2, \dots, n-2$, and n , and the indices i, j taking the values 1, 2, 3, and 4. This is an equation in the product space of atomic and stochastic variables. It will be written for convenience in the symbolic form

$$\dot{V}_{\alpha,i}(t) = Y_{\alpha,i}^{\beta,j} V_{\beta,j}(t) . \quad (27)$$

Finally, the average values of the density-matrix elements, namely, $\langle \mathbf{V}(t) \rangle$, can be obtained from the solutions of (27) by the relation

$$\langle \mathbf{V}(t) \rangle = \sum_\alpha g_n(\alpha) \mathbf{V}_\alpha(t) . \quad (28)$$

Let $\tilde{V}_{\alpha,i}(z)$ be the Laplace transform of $V_{\alpha,i}(t)$. From Eq. (27) we have

$$\tilde{V}_{\alpha,i}(z) = [(z - Y)^{-1}]_{\alpha,i}^{\beta,j} V_{\beta,j}(0) . \quad (29)$$

Since the quantity we want to obtain is simply $\int_0^\infty \langle n(t) \rangle dt = \tilde{V}_4(0)$, we can use (28) and (29) to find

$$\int_0^\infty \langle n(t) \rangle dt = - \sum_\alpha g_n(\alpha) (Y^{-1})_{\alpha,4}^{\beta,j} V_{\beta,j}(0) . \quad (30)$$

Assuming the system to be initially in its ground state, the initial conditions are

$$V_{\beta,j}(0) = \delta_{j,4} . \quad (31)$$

Putting the results (24), (30), and (31) together we finally have

$$R_I = \left[- \sum_{\alpha,\beta} g_n(\alpha) (Y^{-1})_{\alpha,4}^{\beta,j} \right]^{-1} . \quad (32)$$

It is now apparent that, within the framework of the pre-Gaussian model, an exact solution can be found for the ionization rate R_I , since only the inversion of a finite matrix is required. Moreover, it can readily be performed numerically.

IV. RESULTS AND DISCUSSION

We begin by discussing the effect of pre-Gaussian *frequency* fluctuations, since this case can be compared directly with the OU results of Ref. 2. We shall make use of the quantity called by Yeh and Eberly the “differential incoherence of the ionization,” namely,

$$\delta R(\Delta) = \frac{R_I^{\text{PG}}(\gamma, W; \Delta) - R_I^{\text{coh}}(\Delta)}{R_I^{\text{coh}}(\Delta)} , \quad (33)$$

where $R_I^{\text{PG}}(\gamma, W; \Delta)$ is the ionization rate obtained from the relation (32), with the field parameters γ and W defined as in (6), and $R_I^{\text{coh}}(\Delta)$ is the ionization rate corresponding to a purely coherent field and is given by (25). In Fig. 1, $\delta R(\Delta)$ is given as a function of the detuning Δ , for various values of the parameter γ and in the case of a pre-Gaussian noise driven by eight telegraph signals. The results are in perfect agreement with those of Ref. 2, obtained with OU statistics.¹⁵ As could be expected from the form of the spectra corresponding to the correlation functions (9) and (11) (see Refs. 2 and 10), the parameter γ is seen to act as a cutoff for large values of Δ . However, when γ becomes infinite, the cutoff is no longer

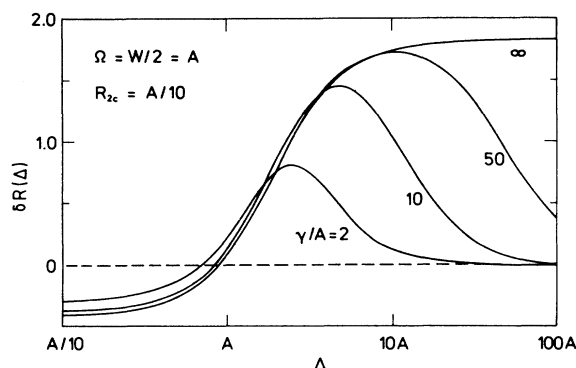


FIG. 1. The quantity $\delta R(\Delta)$ defined by Eq. (33), as a function of the detuning Δ , in the case of a pre-Gaussian frequency noise driven by eight telegraph signals. The values of the parameters are $\Omega = A$, $W = 2A$, $R_{2c} = A/10$, and $\gamma = 2A$, $10A$, $50A$, and ∞ , respectively.

present, and the case of Wiener-Levy (WL) statistics is recovered. For $\Delta > \Omega$, the correction due to the laser noise is always positive; this is because the spectral broadening of the field enhances the resonant transition between the levels 1 and 2. As in Ref. 2, we can also check that, in the limit of large detunings, all bandwidth-dependent effects behave like Δ^{-4} . This is in contrast to the case of a Lorentzian laser power spectrum (as in the WL case, for instance) where these effects are of order Δ^{-2} . This is readily checked by plotting the quantity $\Delta^2 \delta R(\Delta)$ as a function of Δ , as is done in Fig. 2. It is seen that for large values of Δ , the curves saturate. Once again, this figure perfectly agrees with Fig. 3 of Ref. 2.

It is interesting to note that this conclusion concerning the behavior of bandwidth effects for large detunings also

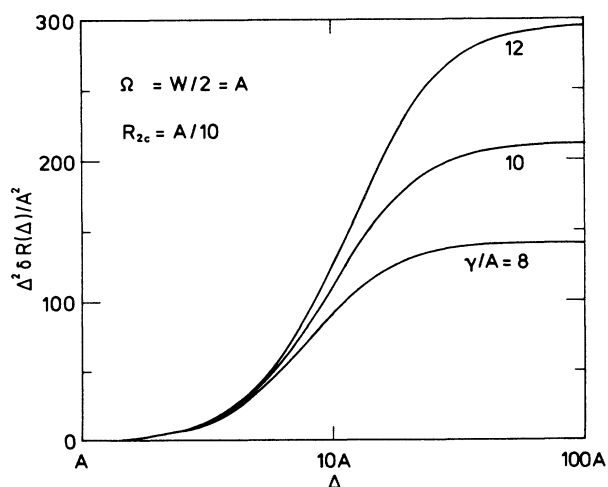


FIG. 2. The quantity $\Delta^2 \delta R(\Delta)/A^2$, as a function of the detuning Δ , in the case of a pre-Gaussian frequency noise driven by eight telegraph signals. The values of the parameters are $\Omega = A$, $W = 2A$, $R_{2c} = A/10$, and $\gamma = 8A$, $10A$, and $12A$, respectively.

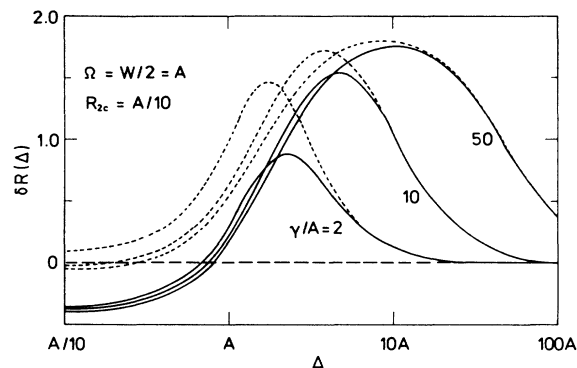


FIG. 3. Solid curves, same as in Fig. 1, but in the case of a pre-Gaussian frequency noise corresponding to a single random-telegraph signal. Dotted curves, corresponding results obtained by using the far-wing approximation of Eq. (36).

agrees with the analysis made in Ref. 11. Indeed, the far-wing ionization rate was obtained there as

$$R_I = \frac{R_{2c}}{4\beta} \operatorname{Re} \int_0^\infty ds e^{-(\beta + i\Delta)s} C_F(s), \quad (34)$$

where¹⁶

$$C_F(s) \equiv \langle \Omega^*(t+s) \Omega(t) \rangle. \quad (35)$$

Using Eqs. (9), (13), and (14) it is found from Eqs. (34) and (35) that, in the case of a single random-telegraph signal ($n = 1$),¹¹

$$R_I = \frac{R_{2c} \Omega_0^2}{4\beta} \frac{(\beta + 2/T)(\beta^2 + 2\beta/T + a^2) + \Delta^2 \beta}{(\Delta^2 - \beta^2 - 2\beta/T - a^2)^2 + 4\Delta^2(\beta + 1/T)^2}. \quad (36)$$

This expression falls off like Δ^{-4} in the limit $\Delta \rightarrow \infty$. Furthermore, the values of $\delta R(\Delta)$ obtained from this formula are seen to be in good agreement with the exact values corresponding to the case $n = 1$ (see Fig. 3) provid-

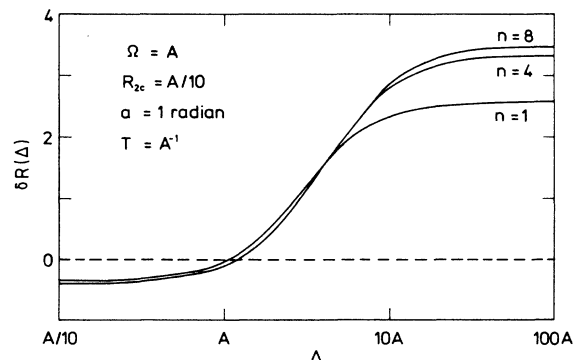


FIG. 4. The quantity $\delta R(\Delta)$ as a function of Δ in the case of a pre-Gaussian phase noise driven by one, four, and eight telegraph signals, respectively. The parameters are $\Omega = A$, $R_{2c} = A/10$, $T = A^{-1}$, and $a = 1$ radian.

TABLE I. Maximal value of $\delta R(\Delta)$ in the case of frequency fluctuations for various values of γ and n . Other parameters are as in Fig. 1.

$\gamma \backslash n$	1	2	4	8	10
2	0.895 13	0.843 57	0.819 40	0.808 27	0.806 12
5	1.314 58	1.247 66	1.218 68	1.205 14	1.202 51
10	1.534 44	1.480 75	1.456 59	1.445 09	1.442 83
50	1.775 46	1.739 17	1.731 25	1.727 35	1.726 57
∞	1.818 17	1.818 18	1.818 18	1.818 18	1.818 18

ed Δ is not too small, the agreement becoming better as Δ increases. We also see that the results of the pre-Gaussian model converge very rapidly when the number n of signals is increased. This is exhibited in Table I, where the maxima of $\delta R(\Delta)$, as shown in Figs. 1 or 3, for instance, are given for various values of γ and n . Finally, it should also be stressed that in the case $\gamma \rightarrow \infty$, the well-known WL result²

$$R_I^{\text{WL}} = \frac{\frac{\Omega^2}{4} \left[\frac{A+W+R_{2c}}{A+R_{2c}} \right]}{\Delta^2 + \left[\frac{A+W+R_{2c}}{2} \right]^2 + \frac{\Omega^2}{2} \left[\frac{A+W+R_{2c}}{A+R_{2c}} \right]} R_{2c} \quad (37)$$

must be recovered. Indeed, we find that the results from our pre-Gaussian calculation agree with this analytical result to within 10^{-7} in the case of eight telegraph signals, while even in the case of a single telegraph signal the relative error was always found to be less than 10^{-5} .

Let us now turn to the case of phase fluctuations. Once again we display in Fig. 4 the behavior of $\delta R(\Delta)$ as a function of Δ for various values of n . The correction due to phase noise is still seen to be negative for $\Delta \lesssim \Omega$. However, the cutoff appearing for large values of Δ in the case of frequency fluctuations is no longer observed here. This is easily understood from the form of the laser power spectrum corresponding to pre-Gaussian phase fluctuations.¹⁰ Although non-Lorentzian, it still

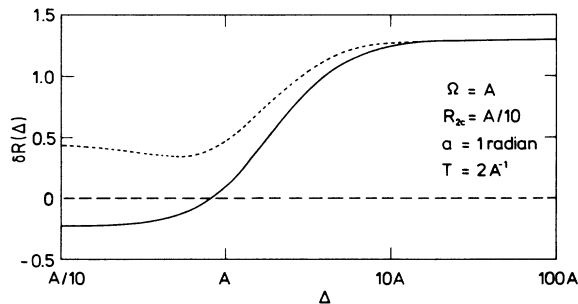


FIG. 5. Solid curve, same as in Fig. 4 but for $T=2A^{-1}$ and in the case of a single random-telegraph signal. Dotted curve, corresponding far-wing approximation obtained by using Eq. (38).

falls off like Δ^{-2} in the wings. Note that a formula analogous to (36) can readily be derived in the case of a single random-telegraph signal, with the help of Eqs. (8), (13), (14), and (35). One finds, in agreement with Wodkiewicz and Eberly,¹⁷ that

$$R_I = \frac{R_{2c} \Omega_0^2}{4\beta} \left[\cos^2 a \frac{\beta}{\beta^2 + \Delta^2} + \sin^2 a \frac{\beta + \frac{2}{T}}{\left[\beta + \frac{2}{T} \right]^2 + \Delta^2} \right]. \quad (38)$$

Figure 5 shows that this result again converges rapidly towards the exact one as Δ is increased.

The case of amplitude fluctuations is slightly more involved. This is because amplitude noise not only causes Ω to fluctuate, but also R_{2c} . If, for a moment, we choose to neglect this last fact, a formula analogous to (38) can again be derived with the help of Eqs. (7), (13), (14), (34), and (35). It reads (for any n)

$$R_I = \frac{R_{2c} \Omega_0^2}{4\beta} \left[\frac{\beta}{\beta^2 + \Delta^2} + \tilde{a}^2 \frac{\beta + \frac{2}{T}}{\left[\beta + \frac{2}{T} \right]^2 + \Delta^2} \right]. \quad (39)$$

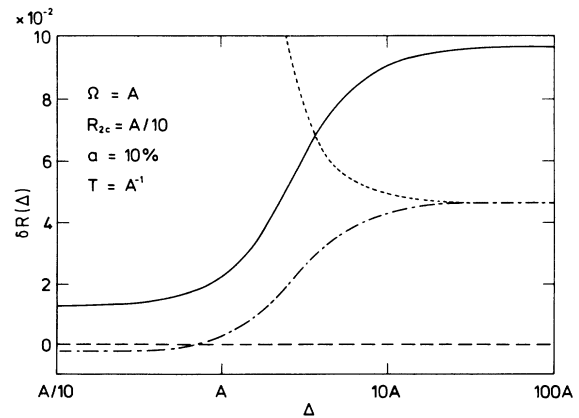


FIG. 6. The quantity $\delta R(\Delta)$ as a function of Δ in the case of pre-Gaussian amplitude noise. The parameters are $\Omega=A$, $R_{2c}=A/10$, $T=A^{-1}$, and $a=1\%$. Solid curve, exact result. Dashed-dotted curve, result obtained by neglecting the fluctuations of R_{2c} . Dotted curve, far-wing approximation of Eq. (39).

Comparison with the "exact" result (in which the fluctuations of R_{2c} are neglected, however) is given in Fig. 6. The behavior is seen to be qualitatively similar to the one obtained for phase fluctuations: $\delta R(\Delta)$ is negative for small values of Δ , positive for $\Delta > \Omega$, and reaches a constant value for $\Delta \gg \Omega$. Including the possibility for R_{2c} to fluctuate modifies this behavior; as shown also in Fig. 6, the negative part of $\delta R(\Delta)$ is now suppressed. Note that these results are stable with respect to n . This is a consequence of the invariance of the correlation function (7) with respect to the number of signals involved in the process.

V. CONCLUSION

In this work, we have extended the use of the pre-Gaussian model of laser noise to the study of two-

photon resonant ionization. We have shown that the results obtained with the help of OU statistics can be easily recovered in this way. Moreover, even a single telegraph signal was found, in the case of frequency fluctuations, to provide surprisingly accurate results. We also showed that interesting results could be obtained in the same way for amplitude or phase fluctuations. These results indicate that the pre-Gaussian model is a very convenient one for analyzing the effects of laser noise on resonant photoionization processes.

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