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Power spectrum of light scattered by a strongly driven Morse oscillator

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We evaluate the power spectrum of light scattered by a weakly nonlinear Morse oscillator driven by a strong single-mode classical electromagnetic field, taking radiation damping into account as the only relaxation process. The spectrum is calculated in the small-noise limit under a near-resonant condition (the resonance is considered in terms of an effective detuning parameter). We show that the single-peak structure that appears in the weak-field limit splits into a symmetric doublet centering around the driving field frequency in the presence of an intense field when the Rabi frequency far exceeds the natural damping rate.

I. INTRODUCTION

It is well known that the steady-state fluoresence spectrum[1] of an ensemble of two-level atoms irradiated by a strong near-resonant cw laser field is a symmetric spectrum about the laser frequency consisting of three spectral components. The interest in this problem was mainly initiated by Mollow[2] who, using a classical description of the light field, first gave a complete theoretical calculation of the spectrum that was subsequently observed experimentally by several independent groups.[3–5] From the theoretical point of view, full quantum treatments[6] have also confirmed this result. The theory of resonance fluorescence has been further extended to a number of other model systems, such as many two-level atomic systems,[7] three-level atoms,[8] the driven Dicke model,[9] etc., under a variety of different conditions, and also to account for the modification of the spectral characteristics in Raman scattering,[10] in optical double resonance,[11] in four-wave mixing,[12] and in other quantum optical effects in intense laser fields.

The purpose of this paper is to evaluate the power spectrum of the radiation scattered by a Morse oscillator driven by a strong incident field, which is assumed to oscillate harmonically near the resonance frequency. The oscillator is assumed to be weakly nonlinear and attains an equilibrium with the driving field through the effect of radiation damping only. We omit all other relaxation processes and also neglect the wave-mixing effects from the present analysis. We show that the single-peak structure in the presence of a weak driving field splits up into a doublet in the limit of a strong driving field (Rabi frequency Ω_0 much greater than the natural relaxation rate 2γ) as a result of an appreciable contribution of the inelastic scattering. The power spectrum of the scattered field in this limit consists of two peaks at the displaced frequencies $\omega\pm\Omega_0$, with their widths proportional to $K\Omega_0\gamma$, where $K+\frac{1}{2}$ denotes the number of bound states and is a measure of nonlinearity of the oscillator. The doublet structure of the power spectrum is reminiscent[13] of the vacuum field Rabi oscillations in a two-level atom in a cavity with a finite quality factor and loss rate, conveniently described in terms of dressed states in the current literature.

The basis of the present analysis is the model that consists of a Morse oscillator described[14,15] in terms of the generators of su(2) Lie algebra, interacting with a classical electric field. The effect of radiation damping is incorporated through the usual linear coupling of the oscillator to the quantized electromagnetic-field modes treated as a bosonic heat bath. Starting from the master equation for the reduced density operator of the weakly nonlinear Morse oscillator valid under Born-Markov and rotating-wave approximation, we formulate the appropriate Fokker-Planck equation with the help of P representation using spin coherent states for the generators pertaining to su(2). The corresponding Ito-Langevin stochastic differential equations under a small-noise linearization scheme can then be employed to evaluate analytically the relevant correlation function whose Fourier transform, apart from some simple factors, represents the power spectrum of the scattered light.

The rest of the paper is organized as follows: The model, the master equation, and the associated Fokker-Planck equation are presented in Sec. II. The Ito-Langevin equations and their mean-field solutions are given in Sec. III. To obtain nonstationary solutions in the limit of large driving field and small nonlinearity, the small-noise linearization has been carried out on the stochastic differential equations, which then reduce to the description of a multivariate Ornstein-Uhlenbeck process. The power spectrum is presented in Sec. IV. The paper is concluded in Sec. V.

II. THE MODEL AND THE BASIC EQUATIONS

The Hamiltonian of a Morse oscillator driven by a classical radiation field and coupled to heat bath is given by

$$H=H_M+H_{MF}+H_B+H_{MB}\ , \tag{2.1}$$

where

$$H_M=\hbar f(S_0+S_-S_+)\ , \tag{2.2a}$$

$$H_{MF}=\hbar\Omega(t)(S_+ + S_-) , \quad (2.2b)$$

$$H_B=\hbar\sum_i \omega_i b_i^\dagger b_i , \quad (2.2c)$$

and

$$H_{MB}=\hbar\sum_i (\chi_i b_i S_+ + \chi_i^* b_i^\dagger S_-) \quad (2.2d)$$

represent the Morse oscillator (H_M), the classical driving field term (H_{MF}), the harmonic oscillator heat bath (H_B), and its linear interaction[16] with the system (H_{MB}), i.e., the Morse oscillator, respectively. S_+, S_-, and S_0 are the generators of the su(2) Lie algebra[14] which act on the basis $|K,V\rangle$, where $K+\frac{1}{2}$ and V denote the number of bound states and the quantum level number of the oscillator, as follows:

$$S_+|K,V\rangle=[(2K-V)(V+1)]^{1/2}|K,V+1\rangle , \quad (2.3a)$$

$$S_-|K,V\rangle=[(2K-V+1)V]^{1/2}|K,V-1\rangle , \quad (2.3b)$$

$$S_0|K,V\rangle=(K-V)|K,V\rangle . \quad (2.3c)$$

For the representation of interest the states $|K,V\rangle$ diagonalize[14] H_M and N, the total excitation operator with the eigenvalues $-\hbar f[(K+\frac{1}{2})-(V+\frac{1}{2})]^2$ and $2K$, respectively, where the range parameter a and the dissociation energy D_0 of the Morse oscillator are related to f and K as

$$\hbar f=a^2\hbar^2/2\mu$$

and

$$K+\tfrac{1}{2}=-(2\mu D_0)^{1/2}/a\hbar , \quad (2.4)$$

and μ is the mass of the oscillator. K can be considered as a parameter of nonlinearity in the present theory. In the harmonic limit $K\rightarrow\infty$.

The Liouville-von Neumann equation of motion for the reduced density operator for the system under Born-Markov and rotating-wave approximation is given by[17]

$$\frac{d\rho}{dt}=-if[(S_0+S_-S_+),\rho]-i\Omega(t)[(S_++S_-),\rho] + 2\gamma(S_-\rho S_+ - 1/2\rho S_+S_- - 1/2S_+S_-\rho) , \quad (2.5)$$

where $\gamma=2\pi g(\omega)|\chi(\omega)|^2$ and $g(\omega)$ represents the mode density evaluated at ω, the frequency of the classical external field.

Here 2γ denotes the Einstein spontaneous-emission coefficient and $\Omega(t)$ is related to Rabi frequency Ω_0 through the following relation:

$$\Omega(t)=\Omega_0\cos(\omega t) . \quad (2.6)$$

The first term in Eq. (2.5) signifies the free motion of the Morse oscillator. The next term is due to an external classical field that drives or pumps the oscillator. The γ term represents the loss of energy from the oscillator to the bath. We neglect the diffusion of fluctuation in the heat bath into the oscillator mode, i.e. we intend work in the zero-temperature limit.

The above formulation of the master equation for the reduced density operator in which the system is described by su(2) operators, while the bath is described by boson operators, is commonly encountered in the Dicke model.[7,9] The damping term in the master equation has been used by Kilin[9] and others in their treatment of the Dicke model with interacting atoms where the system operators are nonlinear in su(2) operators. What is implicit in these (and also in the present) treatments is the assumption that the irreversible terms in the master equation remain unaffected by the nonlinearity of the free oscillator. Haake *et al.*,[18] however, have shown that for such a procedure to be adequate we require, in addition to the weak coupling of the system and heat bath, (i) weakness of the nonlinearity, i.e., K is not very small; (ii) the Rabi frequency $\Omega_0\gg2\gamma$, the natural relaxation rate; and (iii) the number of thermal quanta is effectively zero (note that we are working here in the zero-temperature limit). Secondly, the master equation (2.5) assumes that the driving and the damping act independently. This commonly used[2,7,9] assumption of independence, however, is true so long as the driving field is not strong enough to modify the unperturbed levels, significantly setting an upper limit to Ω_0.

The assumption of weakness of the nonlinearity of the free oscillator has two additional implications. Since we are considering a situation where the Morse oscillator is strongly driven, such as $\Omega_0\gg2\gamma$, an oscillator with a high degree of nonlinearity may dissociate or a chaotic dynamics may set in as a precursor to the process of dissociation.[19] Secondly, this assumption allows us, as we shall see in Sec. III, to use the method of small-noise expansion[20] in the stochastic differential equation in the limit of large driving field. Since noise is often small, this method has been widely used. The linearized stochastic differential equation then describes an Ornstein-Uhlenbeck process which is solvable analytically.

As the next step we transform the master equation (2.5) to a c-number form by introducing the diagonal representation of the density matrix ρ,

$$\rho(t)=\int d^2\eta\, P(\eta,\eta^*,t)|\eta\rangle\langle\eta| , \quad (2.7)$$

where $|\eta\rangle$ denotes the spin coherent state[21] of Radcliff pertaining to su(2) operators as follows:

$$|\eta\rangle=\frac{1}{(1+|\eta|^2)^K}e^{\eta S_+}|K,0\rangle . \quad (2.8)$$

As usual, $|\eta|^2$ represents the average excitation of the Morse oscillator.

Using standard techniques,[17,20] we arrive at the following Fokker-Planck equation for $P(\eta,\eta^*,t)$, which is the c-number equivalent of the master equation (2.5):

$$\frac{dP}{dt}=-\left[\frac{\partial}{\partial\eta}\left[3if\eta-2ifK\eta\frac{1-|\eta|^2}{1+|\eta|^2}-2K\gamma\eta-i\Omega(t)+i\Omega(t)\eta^2\right]\right]P$$
$$+\left[\frac{\partial^2}{\partial\eta^2}[(\gamma+if)\eta^2]P\right]+\tfrac{1}{2}\left[\frac{\partial^2}{\partial\eta\,\partial\eta^*}(\gamma|\eta|^4)P\right]+\text{c.c.} \tag{2.9}$$

This c-number equation for a damped, driven Morse oscillator forms the basis of our further analysis. If we neglect the diffusion terms, i.e., all the second derivative terms, it reduces to a first-order equation. For deterministic systems such as those considered here, the density evolution is necessarily a first-order differential equation. Since from the quantum-mechanical point of view the motion is stochastic around the classical "mean motion," the complete equation (2.9) describes the quantum dynamics of joint probability density.

One important aspect of the two-dimensional Fokker-Planck equation (2.9) has to be clarified. Equation (2.9) does not have a positive-definite diffusion matrix. However, by taking η and η^* as independent complex variables, we can interpret Eq. (2.9) as a Fokker-Planck equation with a positive-definite diffusion matrix in a four-dimensional space. This implies that we are using a positive P representation,[22] which allows us to go over to the associated stochastic differential equations which we discuss in Sec. III. It may not be out of place, however, to mention that serious doubt has been cast on this method, which, fortunately, does not affect the present work because it uses the positive P representation only near a fixed point.[23]

III. THE ITO-LANGEVIN EQUATIONS AND THE MEAN-FIELD SOLUTIONS

Having obtained the Fokker-Planck equation (2.9), one can immediately write down the Langevin c-number equations of motion by inspection. We then have

$$\frac{d\eta}{dt}=3if\eta-2ifK\eta\frac{1-\eta\eta^*}{\eta\eta^*}-2K\gamma\eta+i\Omega(t)\eta^2$$
$$-i\Omega(t)+G_\eta(t)\ , \tag{3.1}$$

together with the independent equation for η^*. Here $G_\eta(t)$ and $G_{\eta^*}(t)$ are independent Langevin forces with zero reservoir average as follows:

$$\langle G_\eta(t)\rangle_R=0=\langle G_{\eta^*}(t)\rangle_R\ . \tag{3.2}$$

The moments of the Langevin force may be read off by inspection from Eq. (2.9):

$$\langle G_i(t)G_j(s)\rangle_R=2D_{ij}\delta(t-s)\ , \tag{3.3}$$

where D_{ij} are the diffusion coefficients; for example,

$$\langle G_\eta(t)G_{\eta^*}(s)\rangle_R=\gamma\eta^{*2}\eta^2\delta(t-s)\ ,$$
$$\langle G_\eta(t)G_\eta(s)\rangle_R=(\gamma+if)\eta^2\delta(t-s)\ . \tag{3.4}$$

To dispense with the undesirable fast time dependence of $\Omega(t)$ from Eq. (3.1), we invoke a slowly varying envelope approximation, whereby following Slusher *et al.*,[24] we assume the complex amplitude η oscillates at the driving frequency ω such that

$$\eta=\hat\eta(t)e^{i\omega t}\ , \tag{3.5a}$$

$$\eta^*=\hat\eta^*(t)e^{-i\omega t}\ . \tag{3.5b}$$

The c-number stochastic differential equations for independent slowly varying amplitudes $\hat\eta$ and $\hat\eta^*$ are obtained by substituting Eqs. (3.5) and (2.6) in Eq. (3.1) and equating the terms of the same frequency. We also point out that in the process we neglect the higher-order frequency mixing effects. The resulting equations are

$$\dot{\hat\eta}=-i\left[\omega-f\left[3-2K\frac{1-\hat\eta\hat\eta^*}{1+\hat\eta\hat\eta^*}\right]\right]\hat\eta-2K\gamma\hat\eta$$
$$+\frac{i\Omega_0}{2}\hat\eta^2-\frac{i\Omega_0}{2}+G_{\hat\eta}(t)\ , \tag{3.6}$$

together with the equation for $\hat\eta^*$.

It is convenient to define two quantities f' and δ as follows:

$$\delta=\omega-f'\ ,$$

where

$$f'=f\left[3-2K\frac{1-\hat\eta\hat\eta^*}{1+\hat\eta\hat\eta^*}\right]\ . \tag{3.7}$$

Equation (3.6) can then be rewritten as

$$\dot{\hat\eta}=-i\delta\hat\eta-2K\gamma\hat\eta+\frac{i\Omega_0}{2}\hat\eta^2-\frac{i\Omega_0}{2}+G_{\hat\eta}(t)\ . \tag{3.8}$$

If we neglect fluctuation, i.e., work within a mean-field limit, we may set $G_{\hat\eta}(t)=0$ to obtain a steady-state amplitude $\hat\eta_e$ from the following equation obtained by putting the left-hand side of Eq. (3.8) equal to zero:[25]

$$i\delta_e\hat\eta_e+2K\gamma\hat\eta_e-\frac{i\Omega_0}{2}\hat\eta_e^2+\frac{i\Omega_0}{2}=0\ , \tag{3.9}$$

where the subscript e refers to equilibrium values. The quantity δ_e then becomes $\delta_e=\omega-f_e'$, where

$$f_e'=f\left[3-2K\frac{1-|\hat\eta_e|^2}{1+|\hat\eta_e|^2}\right]\ .$$

The important point that has to be noted here is that although $\hat\eta$ and $\hat\eta^*$ are independent complex variables, the mean-field solutions $\hat\eta_e$ and $\hat\eta_e^*$ are complex conjugates since we have neglected the fluctuation in the steady state.

It is now easy to see that the quantity inside the large parentheses in the right-hand side of the last expression for f_e' modifies the oscillator frequency f through the nonlinear parameter K and the average excitation number $|\hat{\eta}_e|^2$. Assuming $(1-|\hat{\eta}_e|^2)/(1+|\hat{\eta}_e|^2)$ to be a slowly varying function of the average excitation number, one can interpret δ_e as an effective detuning parameter.

Under the near-resonant condition we may set the effective detuning parameter, $\delta_e \approx 0$, and obtain the steady-state deterministic solutions of $\hat{\eta}_e$ together with $\hat{\eta}_e^*$ from Eq. (3.9).

In the limit of the large driving field such that $\Omega_0 \gg 2\gamma$ and for the small nonlinearity of the Morse oscillator, a small-noise linearization process around the equilibrium solutions can be carried out[20] in the full equation (3.8) and the equation for η^*. We thus let $\hat{\eta}=\hat{\eta}_e+\xi$ and $\hat{\eta}^*=\hat{\eta}_e^*+\xi^*$, where ξ and ξ^* are the fluctuations around the steady state. The result is the Ito stochastic differential equation of the form

$$d\xi(t)=-\underline{A}\,\xi(t)dt+\underline{D}\,d\mathbf{W}(t)\ , \tag{3.10}$$

where $\underline{A}$ and $\underline{D}\,\underline{D}^T$ are the constant drift and diffusion matrices, $d\mathbf{W}(t)$ denotes the independent Wiener processes dW_1 and dW_2, and $\xi(t)$ represents a two-dimensional vector. The Ito stochastic differential equation (3.10) describes a multivariate Ornstein-Uhlenbeck process that is analytically solvable. In the Sec. IV we calculate the power spectrum from the knowledge of the drift and diffusion matrices.

IV. THE POWER SPECTRUM

We consider a near-resonant condition $\delta_e=0$. Then the power spectrum[20] centered at $\nu=\omega$ (the driving laser frequency) may be written as

$$\begin{aligned} S_{\xi\xi^*}(\nu-\omega) &= \int_{-}^{+}\exp[-i(\nu-\omega)]\langle\xi(t)\xi^*(0)\rangle dt \\ &= \{[\underline{A}+i(\nu-\omega)\underline{I}]^{-1} \\ &\quad\times \underline{D}\,\underline{D}^T[\underline{A}^T-i(\nu-\omega)\underline{I}]^{-1}\}_{\xi\xi^*}\ . \end{aligned} \tag{4.1}$$

The explicit calculations show that the expression (4.1) reduces to the following form:

$$S_{\xi\xi^*}=\frac{\gamma|\hat{\eta}_e|^2\{\Omega_0^2[2K(\gamma/\Omega_0)+\hat{\eta}_e^c]^2+\Omega_0^2\hat{\eta}_e^{r^2}+(\nu-\omega)^2\}}{\{(\nu-\omega)^2-\Omega_0^2[\hat{\eta}_e^{r^2}-4K(\gamma/\Omega_0)\hat{\eta}_e^c-\hat{\eta}_e^{c^2}-4K^2(\gamma/\Omega_0)^2]\}^2+\{4\Omega_0^2\hat{\eta}_e^r[2K(\gamma/\Omega_0)+\hat{\eta}_e^c]\}^2}\ , \tag{4.2}$$

where $\hat{\eta}_e^r$ and $\hat{\eta}_e^c$ are the real and imaginary parts of $\hat{\eta}_e$. In the limit of the strong driving field, i.e., $\Omega_0/2\gamma \gg 1$, the last expression can be further simplified if we see that in this limit the steady-state deterministic solution of Eq. (3.9), $\hat{\eta}_e$, is real such that $\hat{\eta}_e^2 \simeq 1$. We then have

$$S_{\xi\xi^*}=\frac{\gamma[\Omega_0^2+(\nu-\omega)^2]}{[(\nu-\omega)^2-\Omega^2]^2+(8K\gamma\Omega_0)^2}\ . \tag{4.3}$$

In the limit of very intense incident field, the spectrum is a superposition of Lorentzian functions at each of its maxima at $\nu=\omega+\Omega_0$ and $\nu=\omega-\Omega_0$. For a weak field we find, however, a single-peak structure at $\nu=\omega$. As an illustration, we have displayed these power spectra for a Morse oscillator with $K=20$ in Fig. 1 for different values of Ω_0. The doublet structure is reminiscent of the vacuum field Rabi oscillations of a single two-level atom in a cavity where a single quantum of energy is transferred back and forth between the atom and the cavity.[13] It is also interesting to note that, although the nonlinearity of the Morse oscillator has a profound effect on the widths, the magnitude of splitting in high field is weakly dependent on it. The appearance of Ω_0 in width is a typical power-broadening feature.

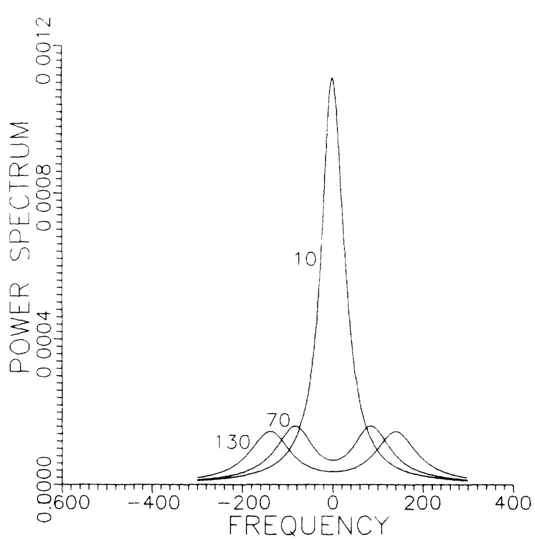

FIG. 1. The power spectra $\gamma S_{\xi\xi^*}(\nu-\omega)$ as a function of the frequency $(\nu-\omega)/\gamma$ for different values of Ω_0/γ and for $K=20$ (both scales arbitrary).

V. CONCLUSIONS

Although the problem of a damped driven Morse oscillator has received wide attention from various workers[19,26] over the last two decades in various contexts—particularly from the point of view of regular and chaotic dynamics of energy change during multiphoton excitation and dissociation—one aspect that still remains to be considered is the calculation of spectral features at the level of fundamental quantum optics. Our aim here is to address these points. Just as the model describing a two-level atom interacting with a single-mode field in the presence of radiation damping lies at the heart of quantum optics, the present model of a Morse oscillator interacting with a single-mode classical electromagnetic field in presence of a quantized heat bath may similarly serve as an analogous paradigm in molecular spectroscopy. The Morse oscillator has been realized in the present paper in terms of su(2) Lie algebra which discards the continuum [which can, however, be incorporated in terms of su(1,1)]. This is a convenient realization for the situations where one is not concerned with dissociation. It may, however, be mentioned in this context that the effect of the continuum in fluorescent spectra has been considered by Dalton[27] for the case of two-level atoms to treat internal atomic relaxation process. Leaving out coupling to the continuum may imply the neglect of this kind of relaxation process. Although the considerations on the relaxation processes are too idealistic, we still hope that the strong-field splitting described in the present paper can be realized experimentally in a suitably chosen diatomic molecule.

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