Spectrum of an Oscillator with Jumping Frequency and the Interference of Partial Susceptibilities

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We study an underdamped oscillator with random frequency jumps. We describe the oscillator spectrum in terms of coupled susceptibilities for different-frequency states. Depending on the parameters, the spectrum has a fine structure or displays a single asymmetric peak. For nanomechanical resonators with a fluctuating number of attached molecules, it is found in a simple analytical form. The results bear on dephasing in various types of systems with jumping frequency.

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Oscillators with varying frequency are studied in many contexts. The frequency change underlies high-resolution mass sensing with nanomechanical resonators, which is based on the change being proportional to the mass of a particle attached to the resonator [1–4]. It is also used in dynamic atomic force microscopy and in high-resolution magnetic force microscopy [5,6]. In quantum systems, the Fock states of a vibrational mode of a trapped electron were detected from the frequency change of a nonlinearly coupled mode [7]. Recently it was proposed to use such change for quantum measurements of mechanical shot noise in an optomechanical system [8].

The change of the oscillator frequency can be often thought of as a jump; it occurs over a much smaller time than the oscillator decay time or the typical interjump interval. This is the case, e.g., where the jumps result from attachment and detachment of molecules (or nanoparticles) to a resonator [1,9,10] or from transitions between well-separated energy levels in a system coupled to the oscillator [7,8,11]. For random jumps, the oscillator dynamics is determined by the interrelation between the characteristic frequency change in a jump Δ, the jump rate W, and the oscillator decay rate Γ. Of utmost interest for classical and quantum measurements is the range where W, Γ, Δ are small compared to the oscillator eigenfrequency ω_0 in the absence of jumps.

In this Letter we consider the susceptibility of an oscillator with respect to resonant driving. The susceptibility is advantageous as it can be directly measured in the experiment and often gives the oscillator power spectrum. We develop an approach that allows finding it for an arbitrary interrelation between the relevant parameters, including Γ, Δ, and W. The sensitivity of the spectra makes it possible to use them for determining the characteristics of the frequency jumps.

Random frequency jumps are noise [9,10]. They lead to spectral broadening. This is a major dephasing mechanism [12], and the analysis below immediately extends to systems other than the oscillator. For oscillators, the spectral broadening due to continuous or quasicontinuous frequency fluctuations has been discussed in the literature, see [11,13–15] and references therein.

The spectral broadening should be qualitatively different depending on the ratio Δ/W. For small and frequent jumps, where $\Delta/W \ll 1$, one can think of the jumps as causing diffusion of the oscillator phase. The resulting spectral broadening should be of the order of the phase diffusion coefficient $\sim\Delta^2/W$.

The case of an arbitrary Δ/W is more complicated. It was studied in the celebrated Anderson paper [16] in the context of spectral broadening of two-level systems and a formal solution in the matrix form was obtained. Here we show that the problem can be formulated in terms of partial spectra and the corresponding partial susceptibilities. Such formulation is not limited to systems with detailed balance. It provides a new insight into the problem of dephasing and allows one to consider, in particular, an arbitrarily large number of states with different frequencies N. For nanoresonators with attaching-detaching molecules we obtain an explicit expression for the susceptibility for an arbitrary Δ/W. Our formulation reveals a connection between dephasing and the paradox of the quantum harmonic oscillator [17–20].

One might expect that the oscillator susceptibility $\chi(\omega)$ is a sum of independent partial susceptibilities $\chi(N;\omega)$ in different-frequency states N. They would be proportional to the state populations $P(N)$, and Im $\chi(N;\omega)$ would be a Lorentzian centered at the state frequency $\omega_0 + \Delta_N$ ($|\Delta_N - \Delta_{N\pm1}| \sim \Delta$). However, this is the case only in the limit of large Δ/W.

The inapplicability of the picture of independent partial susceptibilities can be understood by noticing that, in order to resolve frequencies separated by Δ, one should measure the system for time $\gtrsim 1/\Delta$. Since the frequency changes over time $\lesssim W^{-1}$, for $W \gtrsim \Delta$ the frequencies $\omega_0 + \Delta_N$ cannot be resolved. As we show, the susceptibility can still be formally described as a sum of partial susceptibilities, but the latter are no longer independent. Rather, the partial

susceptibilities are coupled, their shape is strongly changed compared to the $\Delta/W \gg 1$ limit, and the overall susceptibility can be described as a result of their interference.

We will consider an oscillator driven by a resonant field $F\exp(-i\omega t)$ + c.c., $|\omega - \omega_0| \ll \omega_0$, and weakly coupled to a thermal bath, with the decay rate being essentially the same for all relevant frequencies $\omega_0 + \Delta_N$, ω cf. Ref. [20]. Frequency jumps will be considered as imposed externally. This is a good approximation in many cases, molecule attachment-detachment and nonlinear coupling to an off-resonance mode being examples. Between the jumps and in the absence of the driving and coupling to the bath the oscillator is described by Hamiltonian $H_0 = \hbar(\omega_0 + \Delta_N)a^\dagger a$ which almost stepwise varies in time; here $a^\dagger$ and a can be defined as raising and lowering operators of an oscillator with frequency ω, if one disregards energy corrections $\propto (\omega_0 + \Delta_N - \omega)^2/\omega$.

The driving field does not cause transitions between different-N states. The oscillator response to the field is thus determined by the diagonal with respect to N matrix elements $\rho(N)$ of the density operator $\hat\rho$ [$\rho(N)$ are operators with respect to the oscillator Fock states]. Changing to the rotating frame with the canonical transformation $U(t) = \exp(-i\omega a^\dagger a t)$ and using the rotating wave approximation, we obtain the master equation

$$\dot\rho(N) = i(\delta\omega - \Delta_N)[a^\dagger a, \rho(N)] + i[F'a^\dagger + F'^* a, \rho(N)] - \hat\Gamma\rho(N) + \hat W\rho(N),$$
$$\delta\omega = \omega - \omega_0, \tag{1}$$

where $F' = F/(2M\hbar\omega)^{1/2}$ (M is the oscillator mass). The operator $\hat\Gamma$ describes oscillator decay, $\hat\Gamma\rho = \Gamma(\bar n + 1)\times(a^\dagger a\rho - 2a\rho a^\dagger + \rho a^\dagger a) + \Gamma\bar n(aa^\dagger\rho - 2a^\dagger\rho a + \rho aa^\dagger)$, where $\bar n = [\exp(\hbar\omega_0/k_BT) - 1]^{-1}$ is the Planck number.

The operator $\hat W$ describes transitions between different-frequency states N,

$$\hat W\rho(N) = \sum_r [W(N - r, r)\rho(N - r) - W(N, r)\rho(N)], \tag{2}$$

where r enumerates the number of states over which the transition is made. This model refers, in particular, to molecule attachment-detachment where molecules attach to a narrow region on the nanoresonator. The resonator frequency is then determined by the total number of attached molecules N. If molecules do not interact with each other, they attach or detach one by one,

$$W(N, 1) = WN_0, \qquad W(N, -1) = WN,$$
$$W(N, r) = 0 \quad \text{for} \quad |r| > 1; \qquad \Delta_N = -N\Delta. \tag{3}$$

Here, N_0 is determined by the externally controlled molecule flux; for molecules of mass $m_{\rm mol} \ll M$, $\Delta \propto m_{\rm mol}\omega_0/M$. The oscillator velocity jump from a mass change is small, it does not cause phase accumulation in time and can be disregarded.

The linear response of the oscillator to the driving is characterized by the susceptibility $\mathcal{X}(\omega)$ which relates the mean oscillator coordinate to the driving force, $\langle q(t)\rangle = \mathcal{X}(\omega)F\exp(-i\omega t)$ + c.c. For ω close to ω_0 we have $\mathcal{X}(\omega) = (\hbar/2M\omega)^{1/2}\langle a\rangle/F$, where the expectation value of operator a is given by the stationary solution of Eq. (1). Setting $\dot\rho(N) = 0$ in Eq. (1), multiplying this equation by a and taking trace over the oscillator Fock states for a given N (denoted by Tr_0 below), we obtain $\mathcal{X}(\omega) = (2M\omega)^{-1}\chi(\omega)$, where

$$\chi(\omega) = \sum_N \chi(N;\omega),$$
$$[\Gamma - i(\delta\omega - \Delta_N)]\chi(N;\omega) - \hat W\chi(N;\omega) = iP(N). \tag{4}$$

Here, $\chi(N;\omega) = \mathrm{Tr}_0 a\rho(N)/F'$, whereas $P(N) = \mathrm{Tr}_0\rho(N)$ is the stationary probability to find the oscillator in an Nth state. From Eq. (1), $P(N)$ is given by equation

$$\hat W P(N) = 0, \qquad \sum_N P(N) = 1. \tag{5}$$

Equation (4) describes the scaled susceptibility $\chi(\omega)$ as a sum of complex partial susceptibilities $\chi(N;\omega)$ for each eigenfrequency state N. These susceptibilities are given by a set of linear equations. They are coupled to each other, and since they are complex and the phase relations are important, one can say that they interfere, with $\chi(\omega)$ determined by the result of this interference.

In the absence of driving, the power spectrum of the oscillator near its peak for $\omega \approx \omega_0$ is $Q(\omega) = \pi^{-1}\,\mathrm{Re}\int_0^\infty dt\exp(i\omega t)\tilde Q(t)$ with $\tilde Q(t) = \langle a(t)a^\dagger(0)\rangle$. For the considered model $Q(\omega) = (\bar n + 1)\pi^{-1}\,\mathrm{Im}\chi(\omega)$. Correlator $\tilde Q(t)$ can be found from Eq. (1) with $F' = \delta\omega = 0$ using the quantum regression theorem. The formal solution is $\tilde Q(t) = \tilde Q_0(t)\tilde Q_W(t)$, where $\tilde Q_0(t)$ is the correlator in the absence of frequency jumps, $\tilde Q_0(t) = (\bar n + 1)\times\exp(-i\omega_0 t - \Gamma t)$, whereas $\tilde Q_W(t) = \sum_{N,N'}\{\exp[(-i\hat\Delta + \hat W)t]\}_{N'N}P(N)$ describes the effect of frequency jumps; matrix $\hat\Delta$ is diagonal with respect to N, $(\hat\Delta)_{NN'} = \Delta_N\delta_{NN'}$.

The factor $\tilde Q_W(t)$ coincides with the Anderson result for the power spectrum of a two-level system [16]. It can be evaluated by diagonalizing the matrix $-i\hat\Delta + \hat W$ [16]. However, direct diagonalization becomes complicated for large characteristic N. The solution of Eq. (4), on the other hand, is facilitated by the rapid fall-off of $\chi(N;\omega)$ for sufficiently large N.

A simple explicit solution of Eq. (4) for large characteristic N can be found if $W(N, r)$ and Δ_N smoothly vary with N and in Eq. (2) $|r| \ll \bar N$, where $\bar N \gg 1$ is the position of the maximum of $P(N)$. This limit is of interest for nanoresonators if the number of attached molecules is large, on average. Considering N in Eq. (4) as a continuous variable and expanding $W(N - r, r)$, $\chi(N - r;\omega)$ to second order in r, one obtains

$$[\Gamma - i(\delta\omega - \bar\Delta - \Delta'\delta N)]\chi - \nu\partial_N(\delta N\chi) - D\partial_N^2\chi = i(\nu/2\pi D)^{1/2}e^{-\nu(\delta N)^2/2D},$$
$$\delta N = N - \bar N. \tag{6}$$

Here, $\bar{N}$ is given by condition $\sum_r rW(\bar{N}, r) = 0$,

$$\nu = -\partial_N \sum_r rW(N, r), \qquad D = \frac{1}{2}\sum_r r^2 W(\bar{N}, r),$$

$\bar{\Delta} = \Delta_{\bar{N}}$, and $\Delta' = \partial_N \Delta_N$; all derivatives are calculated for $N = \bar{N}$, and we assume $\bar{N}^2 \gg D/\nu \gg 1$, $|\Delta'|/\nu$.

Equation (6) can be solved using the method below,

$$\chi(\omega) = i\int_0^\infty dt \exp[-\Gamma t + i(\delta\omega - \bar{\Delta})t] \times \exp[-(D\Delta'^2/\nu^3)(\nu t - 1 + e^{-\nu t})]. \quad (7)$$

Because of the frequency jumps, the absorption cross section Im $\chi(\omega)$ becomes substantially non-Lorentzian, but it remains symmetric in the large-$\bar{N}$ limit.

Another limiting case of interest is where the oscillator frequencies in different states N are strongly different, $|\Delta_N - \Delta_{N'}| \gg \Gamma, W$ for $N \neq N'$. Here, the partial spectra $\chi(N; \omega)$ are almost independent from each other. Near resonance, $\omega \approx \omega_0 + \Delta_N$, from Eq. (4)

$$\chi(N; \omega) \approx iP(N)[\gamma_N - i(\delta\omega - \Delta_N)]^{-1}, \quad (8)$$

where $\gamma_N = \Gamma + \sum_r W(N, r)$. From Eq. (8), Im $\chi(N; \omega)$ has a Lorentzian peak at frequency $\omega_0 + \Delta_N$ [16]. The area of the peak is determined by the state population $P(N)$. The half-width γ_N depends on the oscillator decay rate Γ and the total probability to switch from state N to other states. This is a familiar result for systems with strongly different transition frequencies [17].

In the opposite limit of small frequency change, $|\Delta_N|/\Gamma \to 0$, all partial spectra have the same shape,

$$\chi(N; \omega) = iP(N)(\Gamma - i\delta\omega)^{-1} \qquad (|\Delta_N| \ll \Gamma). \quad (9)$$

The jump rate does not affect the solution in the limit $|\Delta_N|/\Gamma \to 0$. The spectrum as a whole is Lorentzian centered at frequency ω_0. This is closely related to the paradox of the quantum harmonic oscillator, which occurs in the absence of frequency jumps [17–19]. The oscillator susceptibility can be presented as a superposition of partial susceptibilities corresponding to transitions between neighboring energy levels [20]. These susceptibilities are coupled by dissipation-induced interlevel transitions described by the operator $\hat{\Gamma}\rho$ in Eq. (1). The role of this coupling with respect to the oscillator Fock states is similar to that of the transitions described by $\hat{W}\rho$. For $\Delta_N = W = 0$, as a result of the interference, all partial susceptibilities have the same shape [20]. If the interference were disregarded as in Eq. (8), the spectrum would have a completely different shape.

In the limit $W \gg |\Delta_N|, \Gamma$ the term $\hat{W}\chi(N; \omega)$ is the leading order term in Eq. (4) for $\chi(N; \omega)$, and to first order in W^{-1},

$$\chi(\omega) = i[\gamma - i(\delta\omega - \bar{\Delta})]^{-1},$$

$$\bar{\Delta} = \sum \Delta_N P(N), \quad (10)$$

$$\gamma = \Gamma + \sum P(M)\Delta_N \Delta_M x_\alpha(N)\tilde{x}_\alpha(M)(-\lambda_\alpha)^{-1},$$

where $\mathbf{x}_\alpha$, $\tilde{\mathbf{x}}_\alpha$ and λ_α are the right and left eigenvectors and nonzero eigenvalues of matrix $\hat{W}$, $\mathbf{x}_\alpha \cdot \tilde{\mathbf{x}}_\beta = \delta_{\alpha\beta}$ (the stability of the oscillator stationary state implies Re $\lambda_\alpha < 0$). From Eq. (10), Im $\chi(\omega)$ is again a Lorentzian peak, but now centered at the average frequency $\omega_0 + \bar{\Delta}$ and with half-width γ that exceeds Γ by $\sim\Delta^2/W$ cf. [16].

One of the most interesting and important for applications models of frequency jumps is the model of molecule attachment-detachment Eq. (3). To find $\chi(\omega)$ it is convenient to write $\chi(N, \omega)$ as a Fourier transform,

$$\chi(N; \omega) = \int_0^\infty dt e^{i\delta\omega t}\tilde{\chi}(N; t), \qquad \tilde{\chi}(N; 0) = iP(N).$$

Then Eq. (4) becomes a set of homogeneous differential-difference equations for functions $\tilde{\chi}(N; t)$. It can be solved using the discrete Laplace transform method, i.e., changing to $x(z; t) = \sum_N z^N \tilde{\chi}(N; t)$. This leads to a first-order linear partial differential equation for $x(z; t)$. Its solution immediately gives $\tilde{\chi}(t) = \sum_N \tilde{\chi}(N; t)$,

$$\tilde{\chi}(t) = ie^{-\Gamma t + iWN_0\xi t}\exp[N_0\xi^2(1 - e^{-(W - i\Delta)t})],$$
$$\chi(\omega) = \int_0^\infty dt e^{i\delta\omega t}\tilde{\chi}(t); \qquad \xi = \Delta/(W - i\Delta). \quad (11)$$

Expression (11) for the susceptibility has the form of an integral of an elementary function. It goes over into Eq. (7) for $N_0 \gg 1$, $|\Delta|/W$. The shape of the susceptibility depends on the interrelation between the frequency change per jump Δ, the jump rate W, and N_0. It is convenient to analyze it by rewriting Eq. (11) as

$$\chi(\omega) = \sum_{k=0}^\infty \phi_k(\omega),$$
$$\phi_k(\omega) = e^{N_0\xi^2}(-N_0\xi^2)^k/k! \times [\Gamma - i(\delta\omega + WN_0\xi) + k(W - i\Delta)]^{-1}. \quad (12)$$

Formally, Eq. (12) for $\chi(\omega)$ looks like a sum of partial spectra. However, functions $\phi_k(\omega)$ differ from the partial susceptibilities $\chi(k; \omega)$ introduced earlier. They are close only in the limit of large frequency jumps, $|\Delta| \gg W$, as seen by comparing Eqs. (8) and (12). In this limit Im $\chi(\omega)$ is a set of equally spaced Lorentzian lines with half-widths $\Gamma + (k + N_0)W$. From Eq. (12) it is easy to find corrections to the line shape $\sim W/\Delta$.

Of interest is also the limit $N_0|\xi|^2 \ll 1$ where either the frequency shift is small, $|\Delta| \ll W$, or the average number of attached molecules is small, $N_0 \ll 1$. Here, the leading order term in $\chi(\omega)$ is $\phi_0(\omega)$. It gives a Lorentzian peak of Im $\chi(\omega)$ centered at $\delta\omega = -(W^2/\Delta)N_0|\xi|^2$, with half-width $\gamma = \Gamma + WN_0|\xi|^2$ cf. Eq. (10). The frequency-jump-induced broadening can be comparable with Γ even for small $N_0|\xi|^2$ provided $\Gamma \ll W$. For large $W/|\Delta|$ we have $W|\xi|^2 \propto 1/W$, the jump-induced broadening becomes weaker with increasing W in agreement with the picture of motional narrowing. The position of the spectral

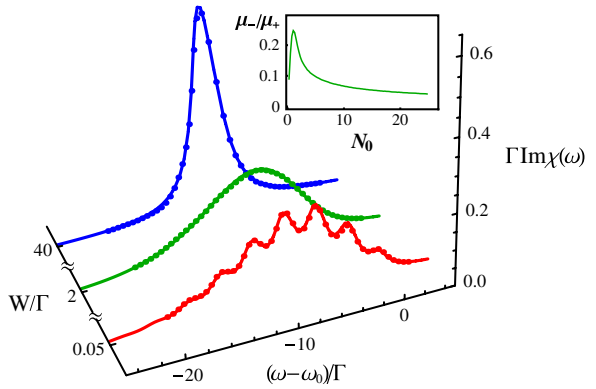

FIG. 1 (color online). The spectrum of the oscillator with attaching and detaching molecules (nanoparticles) for different scaled attachment rates W/Γ. The average number of attached molecules is $N_0 = 3$, and $\Delta/\Gamma = 3$. Data points show the results of numerical simulations obtained using the Gillespie algorithm [25]. Inset: the asymmetry parameter as function of N_0 for $W/\Gamma = 2$, $\Delta/\Gamma = 3$.

peak becomes $-N_0\Delta$, it is independent of W and linearly increases with N_0.

The evolution of the spectrum Im $\chi(\omega)$ with varying parameters is illustrated in Fig. 1. Already for moderate frequency jumps, $\Delta/\Gamma = 3$, the spectrum may display a well-pronounced fine structure. With increasing jump frequency this structure is smoothed out, but the spectrum is asymmetric, generally. The asymmetry can be characterized by the distances $\omega_\pm$ between the maximum and the half-maxima of Im $\chi(\omega)$. The ratio $\mu_-/\mu_+ = |\omega_- - \omega_+|/(\omega_- + \omega_+)$ displays a characteristic peak as function of N_0.

With the increase of W/Δ the spectrum becomes narrow. The position of the spectral peak depends on N_0. The sensitivity to N_0 can be used in experiments on nanoresonators, in particular, to measure the mass-dependent frequency shift Δ, since N_0 can be changed by varying the influx of molecules.

The above analysis can be extended to other types of systems with jumping frequency, to nonlinear response of such systems, and to situations where the jump rates are state dependent or the system causing the jumps has its own internal dynamics. An example is a qubit nonresonantly coupled to a cavity mode investigated in the experiment [21] and analyzed in [22–24]. Here, the qubit frequency depends on the mode state. For a driven mode, partial susceptibilities of the qubit will have off-diagonal components with respect to the Fock states of the mode N, and one should characterize the qubit by $\chi(N, N'; \omega)$. If the mode dynamics is Markovian in slow time, one will obtain a set of linear equations for $\chi(N, N'; \omega)$ which can be studied numerically.

The results of this Letter provide a general method of describing spectral broadening from discrete frequency jumps. They give a new insight into the problem of dephasing and reveal the connection between dephasing from frequency jumps and the spectra of multilevel systems. It is shown that, by studying coupled partial susceptibilities, one can follow the evolution of the spectrum from well-resolved fine structure, for comparatively large and rare jumps, to a broadened single peak, for small and frequent jumps or for a large number of states with different frequencies. The spectrum of a nanoresonator with attaching and detaching molecules or nanoparticles is found in the explicit form and its sensitivity to the parameters is analyzed.

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