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Polaron formation in a medium of damped excitations

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Abstract

Two schemes of the quantization of dynamical variables of dissipative systems are studied with an example of a two-level atom interacting with phonons scattered from an environment. Transformations of relevant Fock-space operators or rigged-Fock-space operators are proposed in order to diagonalize their equations of motion. It is shown that the damping of dispersionless phonons leads to the formation of a quasi-stationary state of the system—a polaron, which is found in both approaches. A time dependence of the polaron energy and the spectral lineshape of the two-level subsystem are evaluated. A transformation diagonalizing the equations of motion of the relevant classical problem (of a harmonic oscillator coupled to a field of damped waves) is found to be similar to the quantum case.

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1. Introduction

Rigged Hilbert spaces are the objects of recent studies on the possibility of reformulation of the quantum mechanics in order to include the instability of states [1]. Also, they are considered as a tool of the classical statistical mechanics [2] enabling one to extend the Koopman–von Neumann probabilistic formulation of the dynamics of large systems. The idea is based on the change of real eigenvalues—the energies into complex eigenvalues whose imaginary parts relate to the times of damping of particular (Gamov) modes of classical or quantum dynamical variables. One uses this formalism (e.g. [3]) for the description of resonances in the chaotic dynamics [4]. In order to express the extended quantum mechanics in the language of damped (quasi-) particles, rigged Fock spaces and Gamov algebras of operators [5] are constructed.

To this date, a number of studies on Gamov modes of different classical and quantum (linear or non-linear) models of physical systems are available (e.g. [5, 6]). The elementary processes in these systems lead to the decay of all particles involved in them, which makes it hard to interpret observations in time. The problem of the polaron formed in a system of

a two-level atom coupled to phonons (or to other excitations of a solid—magnons, etc) is the simplest one to study processes of the dressing of quantum states taking place without change in the atom subsystem. This dressing (the polaron formation) is not an instant process since the excited state of the atom (or an exciton) is created bare. One can determine characteristic times of the dephasing from measurements of the time dependence of an intensity of light emission from the atom or via observation how the energy of the emitted photons depends on time (e.g. [7]). I consider a mechanism of the polaron formation different from the dressing with phonons belonging to a frequency continuum that is accompanied by the emission from the atom of a small amount of the energy of the excited state via wave packets of phonons [8]. When the excitations (I will refer to them as phonons) are dispersionless (the phonon energy is independent of wavevector), they cannot form wave packets and the dressing process must be mediated by a dissipative environment. As we shall see, such a process can be described exactly using a Gamov algebra.

Different systems of a spatially confined particle interacting with optical (dispersionless) phonons can be considered for the observation of the effect. The present description is useful for the electron or exciton in a big-enough quantum dot since it effectively interacts (via the Frohlich coupling) with the longitudinal optical phonons of a very narrow band of frequencies. The width of this band decreases exponentially with the square of the quantum-dot diameter [9]. Then, the damping of phonons results in the acceleration of the initial decoherence compared to that which is due to their dispersion [10], however, the direct coupling of the carrier to the acoustical phonons leads to a complication of the polaronic state. Another candidate is a particle interacting with optical phonons confined in a nanostructure (a quantum dot, a nanocrystal, etc) since their energy spectrum is discrete.

The similarity of the probabilistic-classical and quantum formalisms is important for the development of the quantization methods [11]. However, the formulation of the classical mechanics in the framework of the rigged-Hilbert-space approach leads to non-Newtonian effects contradicting the standard description of the dissipation. For example, let us consider a damped harmonic oscillator with the free-oscillation frequency ω and the damping coefficient γ moving with the Hamiltonian and the relaxation function

$$\mathcal{H} = \tfrac{1}{2}(\pi^2 + \omega^2 x^2), \qquad \mathcal{R} = \gamma\pi^2, \tag{1}$$

where x, π denote the position and momentum satisfying the Hamilton equations

$$\dot{\pi} = -\frac{\delta\mathcal{H}}{\delta x} - \frac{\delta\mathcal{R}}{\delta(\delta\mathcal{H}/\delta\pi)} = -\omega^2 x - 2\gamma\pi, \qquad \dot{x} = \frac{\delta\mathcal{H}}{\delta\pi} = \pi. \tag{2}$$

The motion of an observable A is described with the equation

$$-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}A = LA \tag{3}$$

using the Liouville operator L determined via (2). Transforming canonically the momentum π into $p = \pi + \gamma x$, we write the Liouvillian with the complex variables

$$q^{\pm} = (\omega'/2)^{1/2}(x \mp \mathrm{i}p/\omega') \tag{4}$$

(here $\omega' = (\omega^2 - \gamma^2)^{1/2}$) as

$$L = \omega'\left(q^+\frac{\partial}{\partial q^+} - q^-\frac{\partial}{\partial q^-}\right) + \mathrm{i}\gamma\left(q^+\frac{\partial}{\partial q^+} + q^-\frac{\partial}{\partial q^-}\right). \tag{5}$$

Thus, the vectors of the decaying states (Gamov vectors) moving with the Liouvillian

$$L_{\mathrm{rHs}} = (\omega' - \mathrm{i}\gamma)\left(q^+\frac{\partial}{\partial q^+} - q^-\frac{\partial}{\partial q^-}\right) \tag{6}$$

are not functions of the phase-space variables of the damped harmonic oscillator. On the other hand, as we shall see with an example of the polaron problem, results of the quantum mechanics (especially those found with the Green-function techniques) coincide with predictions of the Gamov-algebra methods. Thus, the correspondence between the equations of motion of dynamical variables of a classical dissipative system and of its quantum counterpart demands investigation.

The classical counterpart of the polaron problem—the harmonic oscillator nonlinearly interacting with a deformation field of damped (via interaction with an environment) modes can be formulated with two different approaches, similar to the damped harmonic oscillator described above. It provides different schemes of its quantization. The model is the simplest one describing macroscopic systems displaying photo-induced phase transformations [12] (for example, a crystal of molecules changing its state from ionic to neutral under a light impulse).

In this work the polaron problem including the damping of the deformation (phonon) modes is solved with the use of the Hilbert-space and rigged-Hilbert-space methods for the classical and quantum cases. The differences of results of both approaches are interpreted. The harmonic oscillator (the classical case) and the two-level system (the quantum case) coupled to a deformation field of damped (phonon) modes are considered assuming that the interaction Hamiltonian is linear in the deformation. The quantum description is developed from the well-known 'independent-boson model' [13] applying the idea of the quantum damped oscillator (a review of [14]). A time dependence of the energy of the polaron (resulting from the inclusion of the dissipation) and the spectral intensity of the two-level subsystem are found using canonical transformations of the operators of the corresponding Fock spaces. These are modified versions of the transformation introduced by Lee, Low and Pines [15, 16]. Similar transformation of the phase-space variables of the classical model is found to diagonalize their equations of motion. I refer to some facts known from observations of relevant physical systems which can be predicted with the model.

In the second section, the quantum system is analysed using Green-function and Gamov-algebra methods while the solution of the classical problem including the dissipation is presented in the third section. In the fourth section, the quantization of the dissipative equations of motion is studied with relevance to the atom-damped-phonon problem. Finally, applicability of different formalisms is considered in conclusions.

2. Formation and decay of the polaron in a quantum system

The Hamiltonian of a quantum two-level system interacting with phonons which are damped due to their coupling to the environment consists of the terms representing the free Hamiltonians of the atom (an exciton), the phonons, and the environment and of two terms of the interaction, $\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{ph}} + \mathcal{H}_{\text{ex-ph}} + \mathcal{H}_{\text{env}} + \mathcal{H}_{\text{ph-env}}$. The atom–phonon part takes the form

$$\mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{ph}} + \mathcal{H}_{\text{ex-ph}} = \omega_{\text{ex}} a^{\dagger} a + \sum_{\mathbf{k}} \Omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + \frac{1}{N^{1/2}} \sum_{\mathbf{k}} F_{\mathbf{k}} a^{\dagger} a \left(b_{\mathbf{k}} + b_{-\mathbf{k}}^{\dagger} \right). \tag{7}$$

Here, $a^{(\dagger)}$, $b_{\mathbf{k}}^{(\dagger)}$ denote the annihilation (creation) operators of the exciton and of the phonon, respectively, N denotes the number of lattice sites. The effective interaction formfactors $F_{\mathbf{k}}$ differ from those of the corresponding translation–invariant interactions relevant to bulk systems (e.g. Frohlich interaction [13]). They can be obtained from the bulk formfactors by changing the Kronecker delta expressing the conservation of the momentum in an elementary process of the particle collision into the integral $\int \psi(\mathbf{x})^* \, \mathrm{e}^{\mathrm{i}\mathbf{k}\mathbf{x}} \psi(\mathbf{x}) \, \mathrm{d}^3\mathbf{x}$, where $\psi(\mathbf{x})$ denotes the wavefunction of the excited state of the atom (or the exciton).

The solution of the problem of the electron (exciton) interacting with phonons using Green functions exists and can be adapted to our system. Following [17], the causal Green function of the exciton $G(t) = -\mathrm{i}\langle 0|\mathcal{T}\{a(t)a^{\dagger}\}|0\rangle$ is determined via the mass operator

$$\Sigma(\omega) = \frac{\mathrm{i}}{2\pi N}\sum_{\mathbf{k}}\int_{-\infty}^{\infty}\mathrm{d}\omega'\, F_{\mathbf{k}}^2\Gamma_{\mathbf{k}}(\omega+\omega',\omega')G(\omega+\omega')D_{\mathbf{k}}(\omega'), \tag{8}$$

where the phonon Green function is defined as $D_{\mathbf{k}}(\omega) = -\mathrm{i}\langle 0|\mathcal{T}\left\{b_{\mathbf{k}}(t)+b_{-\mathbf{k}}^{\dagger}(t),\, b_{-\mathbf{k}}+b_{\mathbf{k}}^{\dagger}\right\}|0\rangle$, and $\Gamma_{\mathbf{k}}(\omega,\omega')$ denotes the Fourier transform of the vertex function

$$\Gamma_{\mathbf{k}}(t,t') = -\frac{N^{1/2}}{F_{\mathbf{k}}}\frac{\delta G^{-1}(t)}{\delta\left\{\left[b_{\mathbf{k}}+b_{-\mathbf{k}}^{\dagger}\right](t')\right\}}. \tag{9}$$

The phonon function can be decomposed into a sum of advanced and retarded one-particle functions $D_{\mathbf{k}}(\omega) = G_{\mathbf{k}}^{(R)}(\omega) + G_{\mathbf{k}}^{(A)}(\omega) = -\mathrm{i}\theta(t)\langle 0|b_{\mathbf{k}}(t)b_{\mathbf{k}}^{\dagger}|0\rangle - \mathrm{i}\theta(-t)\langle 0|b_{-\mathbf{k}}b_{-\mathbf{k}}^{\dagger}(t)|0\rangle$. Neglecting temperature corrections, they take the form

$$G_{\mathbf{k}}^{(R)}(\omega) = \frac{1}{\omega-\Omega_{\mathbf{k}}'+\mathrm{i}\gamma_{\mathbf{k}}}, \qquad G_{\mathbf{k}}^{(A)}(\omega) = -\frac{1}{\omega+\Omega_{\mathbf{k}}'-\mathrm{i}\gamma_{\mathbf{k}}}. \tag{10}$$

Their complex frequency $\Omega_{\mathbf{k}}' - \mathrm{i}\gamma_{\mathbf{k}}$ can be determined via the lowest order in $\mathcal{H}_{\mathrm{ph-env}}$ contribution to the polarization operator $\Pi(\omega,\mathbf{k})$ as $\Omega_{\mathbf{k}}' - \mathrm{i}\gamma_{\mathbf{k}} = \Omega_{\mathbf{k}} + \Pi(\Omega_{\mathbf{k}}+\mathrm{i}0^{+},\mathbf{k})$. In the calculations of $\Sigma(\omega)$, the vertex function is changed into a constant $Z \approx 1$ playing the role of the renormalization factor of the exciton Green function. This approximation discussed in detail in [18] corresponds to neglecting reducible diagrams of the perturbation expansion of $G(\omega)$ in the atom-phonon interaction [19] and leads to

$$\Sigma(\omega) = \frac{1}{N}\sum_{\mathbf{k}}F_{\mathbf{k}}^2\frac{Z^{-1}}{\omega-\Omega_{\mathbf{k}}'-\omega_{\mathrm{ex}}-\Sigma(\omega-\Omega_{\mathbf{k}}')+\mathrm{i}\gamma_{\mathbf{k}}}. \tag{11}$$

For simplicity of the form of the correlation function $A(\omega) = -2\,\mathrm{Im}\,G(\omega+\mathrm{i}0^{+})$, we assume that the phonons are dispersionless and that their lifetime is independent of the wavevector ($\Omega_{\mathbf{k}} = \Omega$, $\gamma_{\mathbf{k}} = \Gamma$). In particular, this assumption is valid for optical phonons when only the phonons belonging to a small part of the Brillouin zone effectively interact with the atom. Calculations of the coefficients of damping of the optical phonons due to the anharmonic decay into phonons of different modes (which is believed to be the most effective mechanism of the scattering) performed for different polar materials show that the wavevector-dependent contributions to them are small compared with their parts independent of wavevector (e.g. [20]). For example, this condition is satisfied in a system of the long-wavelength longitudinal-optical phonons interacting with the atom via the Frohlich coupling. Usually, they are damped due to the spontaneous decay into a pair of longitudinal-acoustical phonons (the so-called Klemens process) or into a pair of a big-energy phonon and a transverse-acoustical phonon (the Valee-Bogani or Ridley processes) [21].

Taking the vertex function constant, the plot of the correlation function $A(\omega)$ (for a weak coupling case [13], $\mathrm{Re}\,\Sigma(\omega_{\mathrm{pol}}) < \Omega$) consists of two peaks—one of them is centred at the frequency of the polaron $\omega_{\mathrm{pol}} = \omega_{\mathrm{ex}} + \mathrm{Re}\,\Sigma(\omega_{\mathrm{pol}})$ and the second one is centred at the frequency of the bound state of the polaron and one phonon $\omega_{\mathrm{pol}} + \Omega'$. The half width of the one-phonon peak equal to Γ corresponds to the inversion of the initial decoherence time (the time of the polaron formation). One can reproduce the spectral intensity of the atom excitation solving the equations of motion of its annihilation (creation) operators of the relevant Gamov algebra a_{out} $\left(a_{\mathrm{in}}^{\dagger}\right)$. They create the pair of Gamov vectors of the atom subsystem $|e\rangle_{\mathrm{in}}, {}_{\mathrm{out}}\langle e|$ such that $\lim_{t\to-\infty}|e\rangle_{\mathrm{in}} = 0$, $\lim_{t\to\infty}({}_{\mathrm{out}}\langle e|) = 0$, and ${}_{\mathrm{out}}\langle e|e\rangle_{\mathrm{in}} = 1$ acting on the state vector of the unexcited atom following $|e\rangle_{\mathrm{in}} = a_{\mathrm{in}}^{\dagger}|g\rangle$, ${}_{\mathrm{out}}\langle e| = \langle g|a_{\mathrm{out}}$. The exact correlation function

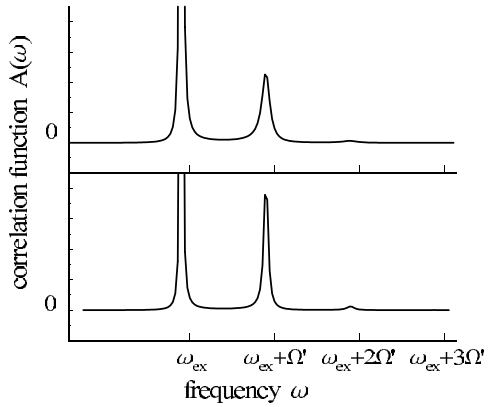

Figure 1. The correlation function $A(\omega)$ for parameters: $\mathrm{Re}\,\Delta/\Omega' = 0.1$ and $\mathrm{Re}\,\Delta/\Gamma = 2$ (top curve), $\mathrm{Re}\,\Delta/\Gamma = 4$ (bottom curve).

$A(\omega)$ found from the retarded Green function $G(t) = -\mathrm{i}\theta(t)\langle 0|\big[a_{\mathrm{out}}(t), a_{\mathrm{in}}^{\dagger}\big]|0\rangle$ is plotted in figure 1. It differs from the approximate correlation function by maxima corresponding to many-phonon bound states at the high-frequency region. Let us study the exact solution.

Modifying the Hamiltonian (7) by changing the creation and annihilation operators of the Fock algebra into the operators of the Gamov algebra, one removes the terms $\mathcal{H}_{\mathrm{env}}$, $\mathcal{H}_{\mathrm{ph\text{-}env}}$ from the Hamiltonian introducing complex frequencies of the non-interacting unstable particles. The non-Hermitean Hamiltonian takes the form

$$\mathcal{H} = z_{\mathrm{ex}} a_{\mathrm{in}}^{\dagger} a_{\mathrm{out}} + \sum_{\mathbf{k}} Z_{\mathbf{k}} b_{\mathbf{k}\mathrm{in}}^{\dagger} b_{\mathbf{k}\mathrm{out}} + \frac{1}{N^{1/2}} \sum_{\mathbf{k}} F_{\mathbf{k}} a_{\mathrm{in}}^{\dagger} a_{\mathrm{out}} \big(b_{\mathbf{k}\mathrm{out}} + b_{-\mathbf{k}\mathrm{in}}^{\dagger}\big), \tag{12}$$

where $z_{\mathrm{ex}} = \omega_{\mathrm{ex}} - \mathrm{i}0^{+}$, $Z_{\mathbf{k}} = \Omega_{\mathbf{k}}' - \mathrm{i}\gamma_{\mathbf{k}}$, and the creation and annihilation operators $a_{\mathrm{out}}, a_{\mathrm{in}}^{\dagger}$ commute to unity similar as $b_{\mathbf{k}\mathrm{out}}$ and $b_{\mathbf{k}\mathrm{in}}^{\dagger}$. The model is solvable using the non-unitary transformation $\Lambda = \mathrm{e}^{s}$ of the Gamov vectors, where

$$s = \frac{1}{N^{1/2}} \sum_{\mathbf{k}} \frac{F_{\mathbf{k}}}{Z_{\mathbf{k}}} a_{\mathrm{in}}^{\dagger} a_{\mathrm{out}} \big(b_{\mathbf{k}\mathrm{out}} - b_{-\mathbf{k}\mathrm{in}}^{\dagger}\big), \tag{13}$$

conserving the averages over the Gamov states. The inverse transformation takes the form $\Lambda^{*} = \mathrm{e}^{-s}$, where the operation $(\cdot)^{*} \equiv [(\cdot)^{\dagger}]'$ is defined as the conjunction of the Hermitean coupling—denoted $(\cdot)^{\dagger}$ and of the complex conjugate of the particle frequencies—denoted $(\cdot)'$. One finds new creation (annihilation) operators diagonalizing the Hamiltonian

$$\mathcal{H} = z_{\mathrm{pol}} \tilde{a}_{\mathrm{in}}^{\dagger} \tilde{a}_{\mathrm{out}} + \sum_{\mathbf{k}} Z_{\mathbf{k}} \tilde{b}_{\mathbf{k}\mathrm{in}}^{\dagger} \tilde{b}_{\mathbf{k}\mathrm{out}}, \tag{14}$$

where for any operator O; $\tilde{O} \equiv \Lambda O \Lambda^{*}$. Here $z_{\mathrm{pol}} = z_{\mathrm{ex}} - N^{-1} \sum_{\mathbf{k}} F_{\mathbf{k}}^{2} / Z_{\mathbf{k}} = z_{\mathrm{ex}} - \Delta$. For constant phonon frequencies $Z_{\mathbf{k}} = Z_0$, following the standard calculations developed for the 'independent-boson model' [13], we use the relations

$$\tilde{a}_{\mathrm{out}} = a_{\mathrm{out}}\, \mathrm{e}^{-\eta}, \qquad \tilde{a}_{\mathrm{in}}^{\dagger} = \mathrm{e}^{\eta} a_{\mathrm{in}}^{\dagger}, \qquad \tilde{a}_{\mathrm{out}} \tilde{a}_{\mathrm{in}}^{\dagger} = a_{\mathrm{out}} a_{\mathrm{in}}^{\dagger} \tag{15}$$

where $\eta = N^{-1/2} \sum_{\mathbf{k}} F_{\mathbf{k}} / Z_{\mathbf{k}} \big(b_{\mathbf{k}\mathrm{out}} - b_{-\mathbf{k}\mathrm{in}}^{\dagger}\big)$, and $a_{\mathrm{out}} a_{\mathrm{in}}^{\dagger} a_{\mathrm{out}} = a_{\mathrm{out}}$, $a_{\mathrm{in}}^{\dagger} a_{\mathrm{out}} a_{\mathrm{in}}^{\dagger} = a_{\mathrm{in}}^{\dagger}$, to evaluate the one-particle Green function of the atom-excitation

$$G(t) = -\mathrm{i}\theta(t) \exp\{-\mathrm{i} z_{\mathrm{pol}} t - \Delta / Z_0 (1 - \mathrm{e}^{-\mathrm{i} Z_0 t})\}. \tag{16}$$

The imaginary part of its Fourier transform

$$G(\omega) = \mathrm{e}^{-\Delta/Z_0} \sum_{l=0}^{\infty} \frac{(\Delta/Z_0)^l}{l!} \frac{1}{\omega - z_{\mathrm{pol}} - lZ_0} \tag{17}$$

determines the spectral intensity plotted in figure 1.

Let us note that the quantity $-\mathrm{Im}\,\Sigma(\omega_{\mathrm{pol}}) \approx -\mathrm{Im}\,z_{\mathrm{pol}} = 1/N \sum_{\mathbf{k}} F_{\mathbf{k}}^2 \gamma_{\mathbf{k}} \big/ \left(\Omega_{\mathbf{k}}'^2 + \gamma_{\mathbf{k}}^2\right) = -\mathrm{Im}\,z_{\mathrm{pol}}$ represents the damping coefficient of a particle dressed with phonons (the polaron). Here, according to [16], $F_{\mathbf{k}}^2 \big/ \left[N\left(\Omega_{\mathbf{k}}'^2 + \gamma_{\mathbf{k}}^2\right)\right]$ is equal to the probability that there is excited one phonon of the wavevector $\mathbf{k}$ when the system is in the polaronic state. Thus, it is predicted by both solutions that the non-zero probability per time unit of the decay of phonons contributing to the polar cloud surrounding the atom (equal to $2\gamma_{\mathbf{k}}$) results in a finite lifetime of the polaronic state. Since there are no processes changing the two-level subsystem, the damping (polaron decay) is connected to the instability of the cloud. Similar instability was suggested to be observed as slow dephasing of the Wannier exciton confined in a semiconductor quantum dot [22, 10].

3. Classical polaron formation

A macroscopic system of polarizable molecules coupled to the deformation field is considered as a model system displaying photo-induced structure changes. In our classical treatment of its dynamics, oscillatory variables x, π, and $X_{\mathbf{k}}$, $\Pi_{\mathbf{k}}$ represent the total polarization and the momentum of the molecule subsystem, and a deformation and momentum of a mode characterized with the wavevector $\mathbf{k}$, respectively. The non-interacting excited molecule is treated as an oscillator of the frequency ω and the dephasing of the complex variables $q^{\pm} = (\omega/2)^{1/2}(x \mp \mathrm{i}\pi/\omega)$ results from a time dependence of the amplitude of the local elementary-dipole oscillation. Thus, the dephasing of the variable $q^{\pm}$ is a consequence of a change of the system structure compared to its structure just after the photo-excitation. The Hamiltonian is of the form

$$\mathcal{H}\left(q^+, q^-, X_{\mathbf{k}_1}, X_{\mathbf{k}_2}, \ldots, \Pi_{\mathbf{k}_1}, \Pi_{\mathbf{k}_2,\ldots}\right) = \omega q^+ q^- + \frac{1}{2}\sum_{\mathbf{k}} \left(\Pi_{\mathbf{k}}^2 + \Omega_{\mathbf{k}}^2 X_{\mathbf{k}}^2\right) + \frac{1}{N^{1/2}} \sum_{\mathbf{k}} (2\Omega_{\mathbf{k}}')^{1/2} F_{\mathbf{k}} q^+ q^- X_{\mathbf{k}}. \tag{18}$$

Writing the Liouville operator with the complex variables $q^{\pm}$, $Q_{\mathbf{k}}^{\pm} \equiv (\Omega_{\mathbf{k}}'/2)^{1/2}(X_{\mathbf{k}} \mp \mathrm{i}P_{\mathbf{k}}/\Omega_{\mathbf{k}}')$, where $P_{\mathbf{k}} \equiv \Pi_{\mathbf{k}} + \gamma_{\mathbf{k}} X_{\mathbf{k}}$ denotes the canonically transformed momentum, we include the dissipation introducing the damping of the deformation modes. Following the considerations in introduction, the relaxation function $\mathcal{R}\left(\Pi_{\mathbf{k}_1}, \Pi_{\mathbf{k}_2,\ldots}\right) = \sum_{\mathbf{k}} \gamma_{\mathbf{k}} \Pi_{\mathbf{k}}^2$ leads to the Liouvillian

$$\begin{aligned} L = {} & \omega\left(q^+\frac{\partial}{\partial q^+} - q^-\frac{\partial}{\partial q^-}\right) + \sum_{\mathbf{k}} \Omega_{\mathbf{k}}'\left(Q_{\mathbf{k}}^+\frac{\partial}{\partial Q_{\mathbf{k}}^+} - Q_{\mathbf{k}}^-\frac{\partial}{\partial Q_{\mathbf{k}}^-}\right) + \mathrm{i}\sum_{\mathbf{k}} \gamma_{\mathbf{k}}\left(Q_{\mathbf{k}}^+\frac{\partial}{\partial Q_{\mathbf{k}}^+}\right. \\ & \left. + Q_{\mathbf{k}}^-\frac{\partial}{\partial Q_{\mathbf{k}}^-}\right) + \frac{1}{N^{1/2}}\sum_{\mathbf{k}} F_{\mathbf{k}}\left(q^+\frac{\partial}{\partial q^+} - q^-\frac{\partial}{\partial q^-}\right)\left(Q_{\mathbf{k}}^- + Q_{-\mathbf{k}}^+\right) \\ & - \frac{1}{N^{1/2}}\sum_{\mathbf{k}} F_{\mathbf{k}} q^+ q^-\left(\frac{\partial}{\partial Q_{\mathbf{k}}^-} - \frac{\partial}{\partial Q_{-\mathbf{k}}^+}\right) \end{aligned} \tag{19}$$

which will be diagonalized below (here $\Omega_{\mathbf{k}}' = (\Omega_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2)^{1/2}$). Using the fact that the product $J = q^+q^-$ is a non-negative constant of motion ($L(q^+q^-) = 0$) and that the kinetic energy of the deformation subsystem takes the simple form $T\left(\Pi_{\mathbf{k}_1}, \Pi_{\mathbf{k}_2}, \ldots\right) = 1/2 \sum_{\mathbf{k}} \Pi_{\mathbf{k}}^2$,

we find the deformation corresponding to the minimum of the potential energy $X'_{\mathbf{k}} = -(2\Omega'_{\mathbf{k}}/N)^{1/2} J F_{\mathbf{k}} / \Omega^2_{\mathbf{k}}$. The value $J = 0$ corresponds to the case when the molecular subsystem is neutral (unpolarized).

The transformation of the dynamical-variable set $\{q^+, q^-, Q^+_{\mathbf{k}}, Q^-_{\mathbf{k}}\}$ into

$$\tilde{q}^{\pm} = q^{\pm} \exp\left\{ \pm \frac{1}{N^{1/2}} \sum_{\mathbf{k}} \frac{\Omega'_{\mathbf{k}} F_{\mathbf{k}}}{\Omega^2_{\mathbf{k}}} \left(Q^-_{\mathbf{k}} - Q^+_{-\mathbf{k}}\right) \pm \mathrm{i} \sum_{\mathbf{k}} \frac{\gamma_{\mathbf{k}} F_{\mathbf{k}}}{\Omega^2_{\mathbf{k}}} \left(Q^-_{\mathbf{k}} + Q^+_{-\mathbf{k}}\right) \right\},$$

$$\tilde{Q}_{\mathbf{k}}{}^{\pm} = Q^{\pm}_{\mathbf{k}} + \frac{1}{N^{1/2}} (\Omega'_{\mathbf{k}} \pm \mathrm{i}\gamma_{\mathbf{k}}) \frac{F_{\mathbf{k}}}{\Omega^2_{\mathbf{k}}} q^+ q^- \tag{20}$$

enables one to simplify the Liouvillian as

$$\begin{aligned} L = {} & \left(\omega - \frac{2}{N^{1/2}} \sum_{\mathbf{k}} \frac{\Omega'_{\mathbf{k}} F^2_{\mathbf{k}}}{\Omega^2_{\mathbf{k}}} \tilde{q}^+ \tilde{q}^- \right) \left(\tilde{q}^+ \frac{\partial}{\partial \tilde{q}^+} - \tilde{q}^- \frac{\partial}{\partial \tilde{q}^-} \right) + \sum_{\mathbf{k}} \Omega'_{\mathbf{k}} \left(\tilde{Q}^+_{\mathbf{k}} \frac{\partial}{\partial \tilde{Q}^+_{\mathbf{k}}} \right. \\ & \left. - \tilde{Q}^-_{\mathbf{k}} \frac{\partial}{\partial \tilde{Q}^-_{\mathbf{k}}} \right) + \mathrm{i} \sum_{\mathbf{k}} \gamma_{\mathbf{k}} \left(\tilde{Q}^+_{\mathbf{k}} \frac{\partial}{\partial \tilde{Q}^+_{\mathbf{k}}} + \tilde{Q}^-_{\mathbf{k}} \frac{\partial}{\partial \tilde{Q}^-_{\mathbf{k}}} \right) \\ & - \sum_{\mathbf{k}} \frac{2\mathrm{i}\gamma_{\mathbf{k}} F_{\mathbf{k}}}{\Omega^2_{\mathbf{k}}} \tilde{q}^+ \tilde{q}^- \left[(\Omega'_{\mathbf{k}} - \mathrm{i}\gamma_{\mathbf{k}}) \frac{\partial}{\partial \tilde{Q}^-_{-\mathbf{k}}} + (\Omega'_{\mathbf{k}} + \mathrm{i}\gamma_{\mathbf{k}}) \frac{\partial}{\partial \tilde{Q}^+_{\mathbf{k}}} \right]. \end{aligned} \tag{21}$$

Since $\tilde{q}^+ \tilde{q}^- = q^+ q^- = J$, one finds the equations of motion of the dynamical variables $\tilde{q}^{\pm}$, $\tilde{Q}^{\pm}_{\mathbf{k}}$ to be linear. The transformation inverse to (20) gives the time dependence of the deformation variable

$$X_{\mathbf{k}}(t) = \left(A_{\mathbf{k}} \mathrm{e}^{-\mathrm{i}\Omega'_{\mathbf{k}} t} + A^*_{\mathbf{k}} \mathrm{e}^{\mathrm{i}\Omega'_{\mathbf{k}} t} \right) \mathrm{e}^{-\gamma_{\mathbf{k}} t} - \frac{J F_{\mathbf{k}} (2\Omega'_{\mathbf{k}})^{1/2}}{N^{1/2} \Omega^2_{\mathbf{k}}}. \tag{22}$$

It tends to $X'_{\mathbf{k}}$ in the long-time limit in which the lattice stabilizes ($\dot{X}_{\mathbf{k}}(t \to \infty) = \Pi_{\mathbf{k}}(t \to \infty) = 0$).

The intensity of the light scattering from the molecular subsystem depends on the velocity of the transition of energy between it and the deformation subsystem. Let us calculate the energy of the molecular subsystem (the oscillator) as the function of time $\mathcal{H}_{\mathrm{m}}(t) = \omega J + N^{-1/2} \sum_{\mathbf{k}} (2\Omega'_{\mathbf{k}})^{1/2} F_{\mathbf{k}} J X_{\mathbf{k}}(t)$. We will denote its time derivative as $P(t)$. Assuming that only one phonon mode of the wavevector $\mathbf{k}_R$ is excited, we evaluate the differential intensity

$$[P(t) - P(0^-)]/P(0^-) = J_2/J_1 \left[\Omega'_{\mathbf{k}_R} / \gamma_{\mathbf{k}_R} \sin\left(\Omega'_{\mathbf{k}_R} t\right) + \cos\left(\Omega'_{\mathbf{k}_R} t\right) \right] \mathrm{e}^{-\gamma_{\mathbf{k}_R} t} - 1, \tag{23}$$

where J_1, J_2 denote the values of J before and after the photo-excitation at $t = 0$. The differential reflection spectra of some physical systems were found to display similar damped oscillations with the phonon frequency related to the wavelength of the scattered photons (e.g. organic charge-transfer crystal–tetrathiafalvalen-p-chloranil [23], collosal-magnetoresistance compound $Pr_{0.7}Ca_{0.3}MnO_3$ [24]).

4. Quantization of the dissipative equations of motion

In this section, the polaron formation is described via quantization of the equations of motion of the macroscopic variables of the third section. Thus, the description is performed with variables averaged over the degrees of freedom of the environment.

The state of the subsystem of the atom and phonons (A) is described with a reduced density operator ($\rho_{A(B)} = \mathrm{Tr}_{B(A)}(\rho)$, B denotes the environment). The time dependence of a Heisenberg-picture observable $\hat{O}_A = \mathrm{Tr}_B[\hat{O}(1_A \otimes \rho_B)]$ of this subsystem is expected to

satisfy equations of motion similar to classical equations of motion (including dissipation) if there exists a corresponding classical observable. Some elements of the set of the quantized canonically conjugated variables of the system averaged over a basis of the environment states are not expected to satisfy the canonical commutation relations (see [25, 26]), namely

$$[x, \pi] = \mathrm{i}, \qquad [X_{\mathbf{k}}, \Pi_{\mathbf{k}}] = \mathrm{i}\,\mathrm{e}^{-2\gamma_{\mathbf{k}}t}. \tag{24}$$

After performing the canonical transformation

$$P_{\mathbf{k}} = \Pi_{\mathbf{k}} + \gamma_{\mathbf{k}} X_{\mathbf{k}} \tag{25}$$

of the deformational variables, the equations of motion of the exciton-annihilation (-creation) operator $a'^{(\dagger)} = (\omega_{\mathrm{ex}}/2)^{1/2}(x + (-)\mathrm{i}\pi/\omega_{\mathrm{ex}})$ and of the operators of the phonon field and momentum operators can be written as

$$\begin{aligned}
&\mathrm{i}\dot{a}' = \omega_{\mathrm{ex}} a' + \left(\Omega_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2\right)^{1/4} \left(\frac{2}{N}\right)^{1/2} \sum_{\mathbf{k}} F_{\mathbf{k}} a' X_{\mathbf{k}}, \\
&\dot{P}_{\mathbf{k}} = -\left(\Omega_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2\right) X_{\mathbf{k}} - \gamma_{\mathbf{k}} P_{\mathbf{k}} - \left(\Omega_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2\right)^{1/4} \left(\frac{2}{N}\right)^{1/2} F_{\mathbf{k}} a'^{\dagger} a', \\
&\dot{X}_{\mathbf{k}} = P_{\mathbf{k}} - \gamma_{\mathbf{k}} X_{\mathbf{k}}.
\end{aligned} \tag{26}$$

In order to diagonalize the first equation of (26), I propose to transform the reduced-Fock-space operators O_A of the set $\{a', a'^{\dagger}, X_{\mathbf{k}}, P_{\mathbf{k}}\}$ into new operators $\tilde{O}_A$ (denoted $\{\tilde{a}', \tilde{a}'^{\dagger}, \tilde{X}_{\mathbf{k}}, \tilde{P}_{\mathbf{k}}\}$) with the unitary operation

$$\begin{aligned}
&\tilde{O}_A = \mathrm{e}^{s} O_A \,\mathrm{e}^{-s}, \\
&s \equiv \mathrm{i} \frac{2^{1/2}}{\left(\Omega_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2\right)^{1/4}} \sum_{\mathbf{k}} \Phi'_{\mathbf{k}} \tilde{a}'^{\dagger} \tilde{a}' \tilde{P}_{\mathbf{k}} + 2^{1/2} \left(\Omega_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2\right)^{1/4} \sum_{\mathbf{k}} \Phi''_{\mathbf{k}} \tilde{a}'^{\dagger} \tilde{a}' \tilde{X}_{\mathbf{k}}.
\end{aligned} \tag{27}$$

It results in the transformation of the equations of motion, which is performed expanding the operators $\dot{\tilde{O}}_A$ and O_A as

$$\begin{aligned}
&\dot{\tilde{O}}_A = \dot{O}_A + [\dot{s}, O_A] + [s, \dot{O}_A] + \tfrac{1}{2}[s, [\dot{s}, O_A]] + \tfrac{1}{2}[\dot{s}, [s, O_A]] + \tfrac{1}{2}[s, [s, \dot{O}_A]] + O(s^3), \\
&O_A = \tilde{O}_A + [\tilde{O}_A, s] + \tfrac{1}{2}[s, [s, \tilde{O}_A]] + O(s^3).
\end{aligned} \tag{28}$$

The conditions of diagonalization of the equation of motion of $\tilde{a}'$ take the form

$$\left[\left(\Omega_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2\right)^{1/2} \mp \mathrm{i}\gamma_{\mathbf{k}}\right] \Phi'_{\mathbf{k}} \pm \left[\left(\Omega_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2\right)^{1/2} \mp \mathrm{i}\gamma_{\mathbf{k}}\right] \Phi''_{\mathbf{k}} = N^{-1/2} F_{\mathbf{k}} \tag{29}$$

for $F_{\mathbf{k}} = F_{-\mathbf{k}}$. They determine $\Phi'_{\mathbf{k}}$, $\Phi''_{\mathbf{k}}$ to be similar as relevant coefficients of the transformation $q^{\pm} \to \tilde{q}^{\pm}$ in (20) and, for constant phonon frequencies $\Omega_{\mathbf{k}} = \Omega$, $\gamma_{\mathbf{k}} = \Gamma$, they lead to the final equations of motion

$$\begin{aligned}
&\dot{\tilde{a}}' = -\mathrm{i}[\omega_{\mathrm{ex}} - \Delta + \Delta(\mathrm{e}^{-2\Gamma t} - 1)]\tilde{a}', \\
&\dot{\tilde{P}}_{\mathbf{k}} = -(\Omega^2 - \Gamma^2)\tilde{X}_{\mathbf{k}} - \Gamma \tilde{P}_{\mathbf{k}} - \left(\frac{2}{N}\right)^{1/2} (\Omega^2 - \Gamma^2)^{1/4} F_{\mathbf{k}} (1 - \mathrm{e}^{-2\Gamma t}) \tilde{a}'^{\dagger} \tilde{a}', \\
&\dot{\tilde{X}}_{\mathbf{k}} = \tilde{P}_{\mathbf{k}} - \Gamma \tilde{X}_{\mathbf{k}},
\end{aligned} \tag{30}$$

where $\Delta = N^{-1}(\Omega^2 - \Gamma^2)^{1/2}/\Omega^2 \sum_{\mathbf{k}} F_{\mathbf{k}}^2$. The solution of the first equation is the operator

$$\tilde{a}'(t) = \exp\left\{-\mathrm{i}\left[(\omega_{\mathrm{ex}} - 2\Delta)t - \frac{\Delta}{2\Gamma}(\mathrm{e}^{-2\Gamma t} - 1)\right]\right\} \tilde{a}'. \tag{31}$$

The other equations of (30) give

$$\begin{aligned}\tilde{X}_{\mathbf{k}}(t) &= \frac{1}{2^{1/2}(\Omega^2-\Gamma^2)^{1/4}}\left(\mathrm{e}^{-\mathrm{i}(\Omega^2-\Gamma^2)^{1/2}t-\Gamma t}\tilde{b}_{\mathbf{k}}+\mathrm{e}^{\mathrm{i}(\Omega^2-\Gamma^2)^{1/2}t-\Gamma t}\tilde{b}^{+}_{-\mathbf{k}}\right)\\ &\quad -\frac{2^{1/2}(\Omega^2-\Gamma^2)^{1/4}}{N^{1/2}\Omega^2}F_{\mathbf{k}}(1-\mathrm{e}^{-2\Gamma t})\tilde{a}'^{\dagger}\tilde{a}',\\ \tilde{P}_{\mathbf{k}}(t) &= \mathrm{i}\frac{(\Omega^2-\Gamma^2)^{1/4}}{2^{1/2}}\left(\mathrm{e}^{\mathrm{i}(\Omega^2-\Gamma^2)^{1/2}t-\Gamma t}\tilde{b}^{+}_{-\mathbf{k}}-\mathrm{e}^{-\mathrm{i}(\Omega^2-\Gamma^2)^{1/2}t-\Gamma t}\tilde{b}_{\mathbf{k}}\right)\\ &\quad -\frac{2^{1/2}(\Omega^2-\Gamma^2)^{1/4}}{N^{1/2}\Omega^2}\Gamma F_{\mathbf{k}}(1+\mathrm{e}^{-2\Gamma t})\tilde{a}'^{\dagger}\tilde{a}'.\end{aligned} \tag{32}$$

The time of decrease of the energy of the quasiparticle $\tau_{\mathrm{f}} = 1/2\Gamma$ determined from (30) is also the time of the creation of a polaronic (deformation) cloud surrounding the excited atom, which is seen in the time dependence of $\tilde{X}_{\mathbf{k}}(t)$. Note that the momentum $\tilde{\Pi}_{\mathbf{k}}(t)$ satisfies the condition of stability of the polaronic cloud $\lim_{t\to\infty}\tilde{\Pi}_{\mathbf{k}}(t) = 0$.

The present formalism can also be applied in order to describe the two-level system interacting with overdamped excitations. Then, one would solve equations (26) with the condition $\Gamma > \Omega$ for which the field $X_{\mathbf{k}}$ cannot be canonically quantized. Thus, we would consider a mixed quantum-classical dynamics which was recently shown not to be relevant for non-dissipative (Hamilton) systems [27]. For $\Gamma \gg \Omega$, one would find the deformation field and momentum

$$\begin{aligned}X_{\mathbf{k}}(t) &= -\frac{2^{1/2}(\Omega^2-\Gamma^2)^{1/4}}{N^{1/2}\Omega^2}F_{\mathbf{k}}(1-\mathrm{e}^{-2\Gamma t})a'^{\dagger}a',\\ P_{\mathbf{k}}(t) &= -\frac{2^{1/2}(\Omega^2-\Gamma^2)^{1/4}}{N^{1/2}\Omega^2}\Gamma F_{\mathbf{k}}(1+\mathrm{e}^{-2\Gamma t})a'^{\dagger}a',\end{aligned} \tag{33}$$

and the time dependence of the polaron frequency of the form

$$\omega(t) = \omega_{\mathrm{ex}} + 2\Delta(\mathrm{e}^{-2\Gamma t}-1). \tag{34}$$

Similar time dependence of the luminescence frequency has been observed with the confined in a quantum dot excitons forming magnetic polarons in a paramagnetic medium [7, 28]. The polaron-formation time was found there to be equal to the spin relaxation time. However, the observation of the Frohlich-polaron formation is more difficult since optical-phonon relaxation is usually much faster than the spin relaxation.

In order to compare the present solution to that obtained with the Green-function method or using the Gamov algebra, we evaluate the function $A'(\omega)$ defined as the Fourier transform of the correlation function $A'(t) = \langle 0'|a'(t)a'^{\dagger}|0'\rangle$, where $|0'\rangle = |g\rangle \otimes |\mathrm{vac}\rangle$ denotes the conjunction of the vectors of the atom ground state $|g\rangle$ and of the phonon vacuum $|\mathrm{vac}\rangle$. The above function is defined as an average over the vacuum state of the subsystem while in the previous section we have calculated the relevant correlation function as an average over the vacuum of the whole system. In order to find

$$A'(\omega) = \int_{-\infty}^{\infty}\theta(t)\,\mathrm{e}^{\mathrm{i}(\omega+\mathrm{i}0^{+})t}\langle 0'|a'(t)a'^{\dagger}|0'\rangle\,\mathrm{d}t + \int_{-\infty}^{\infty}\theta(-t)\,\mathrm{e}^{\mathrm{i}(\omega-\mathrm{i}0^{+})t}\langle 0'|a'(t)a'^{\dagger}|0'\rangle\,\mathrm{d}t, \tag{35}$$

it is necessary to determine $\tilde{a}'(t)$ for a negative time. We use the finding of Bateman, who established that the description of the damped harmonic oscillator in the framework of the Hamilton formalism demands introduction of a dual dynamical variable [29]. It is a physical variable for $t < 0$ fulfilling the transformed via $\Gamma \to -\Gamma$ equations of motion of the damped harmonic oscillator. Thus, for $t < 0$, we arrive at

$$\tilde{a}'(t) = \exp\left\{-\mathrm{i}\left[(\omega_{\mathrm{ex}}-2\Delta)t + \frac{\Delta}{2\Gamma}(\mathrm{e}^{2\Gamma t}-1)\right]\right\}\tilde{a}'. \tag{36}$$

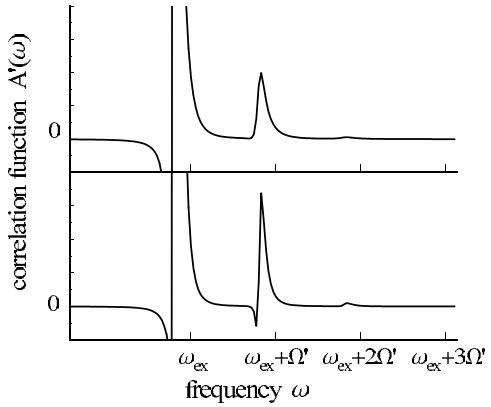

Figure 2. The correlation function $A'(\omega)$ for parameters: $\Delta'/\Omega = 0.1$ and $\Delta/\Gamma = 2$ (top curve), $\Delta/\Gamma = 4$ (bottom curve). Here $\Omega' \equiv (\Omega^2 - \Gamma^2)^{1/2}$.

Following standard calculations for the 'independent-boson model', we use the relations

$$\tilde{a}' = a'\,\mathrm{e}^{-\eta}, \qquad \tilde{a}'^{\dagger} = \mathrm{e}^{\eta}a'^{\dagger}, \qquad \tilde{a}'\tilde{a}'^{\dagger} = a'a'^{\dagger} \tag{37}$$

found with $a'a'^{\dagger}a' = a'$, $a'^{\dagger}a'a'^{\dagger} = a'^{\dagger}$. Here $\eta = \sum_{\mathbf{k}}\left[(-\Phi'_{\mathbf{k}} + \Phi''_{\mathbf{k}})\tilde{b}^{\dagger}_{-\mathbf{k}} + (\Phi'_{\mathbf{k}} + \Phi_{\mathbf{k}''})\tilde{b}_{\mathbf{k}}\right] - \mathrm{i}4\Gamma\Delta/\Omega^2$. Applying the operator-disentanglement procedure described in [13, 30], for $t > 0$, we have

$$\langle 0'|a'(t)a'^{\dagger}|0'\rangle = \mathrm{e}^{-\mathrm{i}\left[(\omega_{\mathrm{ex}}-2\Delta)t - \frac{\Delta}{2\Gamma}(\mathrm{e}^{-2\Gamma t}-1)\right]} \times \exp\left\{\frac{1}{2}\left(\Phi''^2_{\mathbf{k}} - \Phi'^2_{\mathbf{k}}\right)\left(1 + \mathrm{e}^{-2\Gamma t} - 2\,\mathrm{e}^{-\mathrm{i}(\Omega^2-\Gamma^2)^{1/2}t - \Gamma t}\right)\right\}. \tag{38}$$

The corresponding average for a negative time can be written when changing Γ into $-\Gamma$. Expanding the above average and performing its Fourier transformation one arrives at the correlation function of the form $A'(\omega) = 2\,\mathrm{Im}\{I(\omega)\}$, where

$$I(\omega) = \mathrm{e}^{-\frac{\Delta'}{2\Omega} - \mathrm{i}\frac{\Delta}{2\Gamma}} \sum_{l=0}^{\infty}\sum_{h=0}^{l} \frac{1}{h!(l-h)!}\left(\frac{\mathrm{i}\Delta}{2\Gamma} - \frac{\Delta'}{2\Omega}\right)^{l-h}\left(\frac{\Delta'}{\Omega}\right)^{h} \times \frac{1}{\omega - \omega_{\mathrm{ex}} + 2\Delta - h(\Omega^2-\Gamma^2)^{1/2} + \mathrm{i}(2l-h)\Gamma + \mathrm{i}0^+}, \tag{39}$$

and $\Delta' = \Delta\Omega/(\Omega^2 - \Gamma^2)^{1/2}$. The plot of $A'(\omega)$ (figure 2) consists of peaks positioned at the frequencies corresponding to the polaron and to the states of the polaron bound to one-or-more phonons as the plot of the spectral intensity $A(\omega)$ found in the second section. However, $A'(\omega)$ does not satisfy the condition of positivity of the spectral intensities of bosons and fermions at $\omega > 0$. It does not apply since $A'(t)$ is calculated with the operators averaged over the environment state. Thus, $A'(\omega)$ does not describe optical spectra, similar as the corresponding functions for systems described with Hamiltonians depending on time.

In [31], the spectra for such a system (an atom in a classical field periodic in time) were found calculating directly the optical transition probability per time unit. When we add the atom–photon-interaction term $\mathcal{H}_{\mathrm{int}} = V_\omega c^{\dagger}_{\omega}a_c + \mathrm{H.c.}$ to the Hamiltonian, (c_ω denotes the annihilation operator of the photon of the frequency ω, $a^{\dagger}_c$—the creation operator of the exciton interacting with the photon), it takes the form

$$\lim_{T\to\infty}\left|\left\langle c_\omega a^{\dagger}_c(T)\right\rangle\right|^2/T = \lim_{T\to\infty}|V_\omega|^2\left|\int_0^T \langle 0'|a'(t)a'^{\dagger}|0'\rangle\,\mathrm{e}^{\mathrm{i}\omega t}\,\mathrm{d}t\right|^2\Big/T. \tag{40}$$

The above method is not effective when $\int_0^T \langle 0'|a'(t)a'^{\dagger}|0'\rangle \, \mathrm{e}^{\mathrm{i}\omega t} \, \mathrm{d}t$ is not a periodic function of T as it is in our case. However, the total transition probability

$$|V_\omega|^2 \left| \int_0^\infty \langle 0'|a'(t)a'^{\dagger}|0'\rangle \, \mathrm{e}^{\mathrm{i}\omega t} \, \mathrm{d}t \right|^2 = |V_\omega|^2 |I(\omega)|^2 \tag{41}$$

can be estimated for all ω except one point $\omega = \omega_{\mathrm{ex}} - 2\Delta$, and the plot of $|I(\omega)|^2$ determines the spectral lineshape. Its characteristic topic is an asymmetry of the peaks not present in $A(\omega)$ of section 2.

5. Conclusions

We have solved the problem of the polaron in a medium of damped excitations with different approaches and referred to some experimental facts supporting their results. Let us note that the Gamov vectors of the system of the classical harmonic oscillator interacting with damped deformation waves can be determined, similarly as for the problem of the radiation damping in classical systems described by Petrosky *et al* [32]. Its Liouville operator received by the similar change of signs in the third term of (19) as described in the introduction is diagonalizable via the transformation of the variables $\{q^{\pm}, Q^{\pm}_{\mathbf{k}}\}$ into

$$\tilde{q}^{\pm} = q^{\pm} \exp\left\{ \pm \frac{1}{N^{1/2}} \sum_{\mathbf{k}} \frac{F_{\mathbf{k}}}{Z_{\mathbf{k}}} \left(Q^{-}_{\mathbf{k}} - Q^{+}_{-\mathbf{k}} \right) \right\}, \tag{42}$$

$$\tilde{Q}^{\pm}_{\mathbf{k}} = Q^{\pm}_{\mathbf{k}} + \frac{1}{N^{1/2}} \frac{F_{\mathbf{k}}}{Z_{\mathbf{k}}} q^{+} q^{-}. \tag{43}$$

Similar to the quantum case discussed in section 2, the irreversibility inherent in this model is not related to the reduction of the phase space. Here, the damping of the variable $\tilde{q}^{\pm}$ (a finite lifetime of the classical polaron) is analogous to the description of the Brownian motion while in the deterministic model of the third section the polaron mode is stable.

The preparation of the environment state before a measurement may be a key to the choice of the relevant scheme of quantization. As example observables, let us consider the correlators calculated in the second and fourth sections. The observation of $A(t)$ (the scalar product of the ket $a^{\dagger}|0\rangle$ and of the bra $\langle 0|a(t)$, where $|0\rangle$ denotes the absolute-vacuum state) demands preparation of the environment state while $A'(t)$ (the scalar product of the ket $a'^{\dagger}|0'\rangle$ and of the bra $\langle 0'|a'(t)$, where $|0'\rangle$ denotes the vacuum state of the atom–phonon subsystem, $a'(t) = \mathrm{Tr}_{\mathrm{B}}\{\rho_{\mathrm{B}} a(t)\}$) is determined with the environment being in a mixed state. In particular, since it is easier to prepare the environment of a single atom than the environment of an ensemble of atoms (because the excitation of all atoms cannot take place at exactly the same time), one can expect that different formalisms described here can be applied for the one-atom spectroscopy or for the spectroscopy of big systems.

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