THERMORELAXATION RESONANCE IN THE QUANTUM THEORY OF BROWNIAN MOTION

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The Brownian motion of a quantum particle in a thermal reservoir possessing a finite correlation time τ_c is considered. Non-Markov Langevin equations for a stationary nonequilibrium state are obtained. At low temperatures T of the thermal reservoir, the correlation time $\tau_c=\hbar/2\pi T$ is fairly long. It is shown that allowance for the damping γ of the particle momentum over the correlation times τ_c: $\gamma\tau_c\simeq 1$, leads to an oscillating temperature dependence of the relaxation coefficient $\gamma(1/T)$ in the region of low temperatures of the thermal reservoir.

1. It is well known that in many cases the interaction of a dynamical system with a thermal reservoir (a macroscopic system possessing infinitely many degrees of freedom) cannot be described in the framework of the Markov approximation. Such situations are analyzed by various methods that take into account the finite correlation time τ_c of the thermal reservoir [1—7]. The present paper is devoted to the development of the Langevin approach to the theory of the Brownian motion of a quantum system that was proposed in [7]. This microscopic approach makes possible a unified treatment of the kinetics and fluctuations in nonlinear quantum systems in both the thermodynamically equilibrium state as well as in a strongly nonequilibrium state. The method is based on the assumption of Gaussian statistics of the unperturbed variables of the thermal reservoir. This makes it possible to eliminate the variables of the thermal reservoir from the equations of motion of the dynamical subsystem by means of the Bloch—De Dominicis statistical theorem [8] and the Furutsu—Novikov quantum theorem that follows from it [7]. Gaussian statistics will describe the thermal reservoir, the Hamiltonian F of which is quadratic in the creation—annihilation operators (as in the case of photon or phonon dissipative subsystems). Similar statistics is obtained for the random field produced by collisions in gases or plasmas with long-range interaction potential when the relaxation and fluctuations of the Brownian particle are due to the superposition of a large number of collisions with small loss of phase coherence. The method presented below is similar to the method of elimination of the Bose operators used in [1]. However, in contrast to the well-known studies [1—6], the choice of a definite statistics of the thermal reservoir makes it possible to obtain a Langevin equation and find the correlation functions of the fluctuation forces for a nonequilibrium quantum system in a strongly nonequilibrium state. Our approach is also valid for the case of non-Gaussian statistics of the thermal reservoir [9] under the additional assumption that the macroscopic subsystem (the thermal reservoir) is only weakly influenced by the Brownian particle.

In this paper, we analyze some aspects of the non-Markov relaxation of the quantum particle. We shall assume that the finiteness of the correlation time τ_c of the thermal reservoir has a purely quantum origin. This is possible when the susceptibility $\chi(\omega)$ of the dissipative subsystem is characterized by a sufficiently large width $1/\tau_0$, and the response function $\varphi(\tau)$ by a short time scale τ_0. However, the spectral density $S(\omega)$ of the fluctuations of the thermal reservoir can, by virtue of the fluctuation—dissipation theorem

$$S(\omega)=\hbar\chi''(\omega)\,\mathrm{cth}(\beta\omega/2) \tag{1}$$

have an appreciable frequency dispersion at low temperatures T of the thermal reservoir, $\beta=\hbar/T$. In the case of the collisional relaxation mechanism, when the interaction energy of the dynamical subsystem with the thermal reservoir, $Q(r, t)$, is equal to the sum of the energies of the interaction of the Brownian particle with the individual atoms of a buffer gas or plasma ions, we shall understand by τ_0 the duration of one collision. In the approximation of weak dispersion for the susceptibility, $\chi''(\omega)=\alpha\omega$, the correlation time of the unperturbed variables of the thermal reservoir is inversely proportional to its temperature: $\tau_c=\hbar/2\pi T$ [10]. Effects associated with non-Markov behavior of such type were considered, in particular, in [10—12].

In this paper, we show that allowance for the frequency dispersion of the spectral density $S(\omega)$ of the fluctuations of the thermal reservoir, Eq. (1), leads to a qualitative change of the temperature dependence of the relaxation coefficient of the momentum of the Brownian particle in the quantum region of temperatures. The dependence $\gamma(1/T)$ acquires an oscillating nature with extrema near the points of thermorelaxation resonance: $T_n=\hbar\gamma/2\pi n$, $n=1, 2, \ldots$. To calculate this effect, it is

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necessary to go beyond the framework of the Markov approximation and take into account the damping of the dynamical system over the correlation times of the thermal reservoir: $\gamma\tau_c \simeq 1$.

2. We obtain a non-Markov Langevin equation for a quantum particle with mass m and charge e, interacting with a thermal reservoir in the presence of a constant external field E. The Hamiltonian of the system consisting of the particle and the thermal reservoir has the form

$$H=\frac{p^2}{2m}-\sum_k Q_k(t)\,e^{ikr(t)}-eEr(t)+F. \tag{2}$$

Here, r and $p(t)$ are the coordinate and momentum operators of the particle, F is the Hamiltonian of the thermal reservoir, $Q_k(t)$ are the harmonics of the energy of the interaction of the particle with the dissipative environment:

$$Q(r,t)=\sum_k Q_k(t)e^{ikr(t)}.$$

In the case of the phonon relaxation mechanism, we can understand by $Q(r, t)$ the Fröhlich potential or the deformation potential.

From (2) we obtain the Heisenberg equations of motion for the coordinate $r_j(t)$ ($j=1, 2, 3$):

$$\ddot{r}_j(t)=\frac{1}{m}\sum_k ik_j Q_k^{\mathrm{H}}(t)e^{ikr(t)}+\frac{e}{m}E_j. \tag{3}$$

This equation contains $Q_k^{\mathrm{H}}(t)$, the total Heisenberg operator of the thermal reservoir, which includes allowance for the change of the reservoir under the influence of the Brownian particle. For Gaussian statistics of the unperturbed variables $Q_k(t)$ of the thermal reservoir, the expansion of $Q_k^{\mathrm{H}}(t)$ with respect to the operators e^{ikrt} of the dynamical subsystem conjugate to the variable $Q_k(t)$ of the thermal reservoir has the form [7,9]

$$Q_k^{\mathrm{H}}(t)=Q_k(t)+\int dt_1\,\varphi_k(t-t_1)\,e^{-ikr(t_1)}. \tag{4}$$

Here,

$$\varphi_k(t-t_1)=\langle i[Q_k(t),\ Q_{-k}(t_1)]_-\rangle\eta(t-t_1)$$

is the response function (retarded Green's function) of the free thermal reservoir; $\eta(\tau)=1$ for $\tau>0$, $\eta(\tau)=0$ for $\tau<0$, $\hbar=1$. In the relation (4), we have omitted the nonlinear terms proportional to the response functions of higher orders:

$$\varphi_{k,k_1,k_2}(t;\,t_1,\,t_2)=\frac{i^2}{2!}P_{12}[[Q_k(t),\,Q_{k_1}(t_1)]_-,\,Q_{k_2}(t_2)]_-\eta(t-t_1)\eta(t_1-t_2),$$

where P_{12} is the operator of the sum of cyclic permutations of the indices. For Gaussian statistics of the variables $Q_k(t)$, the operator $[Q_k(t), Q_{k_1}(t)]_-$ will be a c number by virtue of the Bloch—De Dominicis theorem [8], and formula (4) gives the exact expression for the total operator of the dissipative subsystem. As is shown in [9], the additional terms in the expansion of $Q_k^{\mathrm{H}}(t)$ in the variables of the Brownian particle $e^{ikr(t)}$ that are associated with the non-Gaussian condition of the free operators $Q_k(t)$ are proportional to higher powers of the particle—thermal-reservoir coupling constant. If this last is small, the non-Gaussian state of the free thermal reservoir need not be taken into account.

Substituting (after preliminary symmetrization) the expression (4) in the equation of motion (3), we arrive at exact stochastic equations for the operators of the coordinates of the Brownian particle:

$$\ddot{r}_j(t)=\frac{1}{m}\sum_k ik_j\frac{1}{2}[Q_k(t),\,e^{ikr(t)}]_+ +\frac{1}{m}\sum_k ik_j\int dt_1\varphi_k(t-t_1)\frac{1}{2}[e^{ikr(t)}\cdot e^{-ikr(t_1)}]_- +\frac{e}{m}E_j. \tag{5}$$

To eliminate the free thermal-reservoir variables $Q_k(t)$ from this equation, we use the quantum generalization [7] of the Furutsu—Novikov theorem. Assuming that the coordinate and momentum of the Brownian particle are functionals of the Gaussian random field $Q_k(t)$, we find by means of the Bloch—De Dominicis theorem [8]

$$\left\langle\frac{1}{2}[Q_k(t),\,e^{ikr(t)}]_+\right\rangle=\int dt_1 M_k(t-t_1)\left\langle\frac{\delta e^{ikr(t)}}{\delta f_{-k}(t)}\right\rangle\Bigg|_{f=0}, \tag{6}$$

where f_k is an auxiliary deterministic force additive with respect to the random potential $Q_k(t)$, and

$$M_k(t-t_1)=\langle{}^1\!/_2[Q_k(t),\ Q_{-k}(t_1)]_+\rangle$$

is the correlation function of the variables $Q_k(t)$. For the functional derivative in (2), we can obtain the relation

$$\frac{\delta e^{ikr(t)}}{\delta f_{-k}(t_1)}\bigg|_{f=0}=\frac{i}{\hbar}[e^{ikr(t)},e^{-ikr(t_1)}]_-\eta(t-t_1).$$

Using formula (6), we write the equation of motion (5) of the Brownian particle in the form of the Langevin equation

$$\ddot{r}_j(t)-\frac{1}{m}\sum_k ik_j\int dt_1\left\{\tilde{M}_k(t-t_1)i[e^{ikr(t)},e^{-ikr(t_1)}]_-+\varphi_k(t-t_1)\frac{1}{2}[e^{ikr(t)},e^{-ikr(t_1)}]_+\right\}=\frac{1}{m}\xi_j(t)+\frac{e}{m}E_j \tag{7}$$

with fluctuation source having zero mean value, $\langle\xi_k\rangle=0$,

$$\xi_j(t)=\sum_k ik_j\frac{1}{2}[Q_k(t),e^{ikr(t)}]_+-\sum_k\int dt_1\,ik_j\tilde{M}_k(t-t_1)i[e^{ikr(t)},e^{-ikr(t_1)}]_-,\quad \tilde{M}_k(\tau)=M_k(\tau)\eta(\tau). \tag{8}$$

The explicit expression (8) for the fluctuation force $\xi_j(t)$ enables us to calculate the statistical properties of the quantum Brownian particle in both thermodynamic equilibrium and in a strongly nonequilibrium state.

3. We use Eq. (5) to analyze the non-Markov effects due to the finiteness of the correlation time τ_c of the thermal reservoir. We note that τ_c determines the time scale of the correlation function $M_k(\tau)$. In the general case, the time scale of the response function $\varphi_k(\tau)$ is given by a different time τ_0. In many physics problems, the function $\varphi_k(\tau)$ can be approximated by the expression

$$\varphi_k(\tau)=\lambda_k\exp(-\tau/\tau_0)\eta(\tau).$$

Then for the susceptibility $\chi_k(\omega)$ of the thermal reservoir, the Fourier transform of the function $\varphi_k(\tau)$, we have $\chi_k(\omega)=\lambda_k\tau_0/(1-i\omega\tau_0)$. If the characteristic times of the dynamical subsystem and, in particular, the relaxation time $1/\gamma$ of the momentum of the Brownian particle are much longer than the time τ_0, then for $\chi_k''(\omega)$ the approximation of low frequency dispersion is valid: $\chi_k''(\omega)=\alpha_k\omega$, where $\alpha_k=\lambda_k\tau_0^2$. As follows from the fluctuation–dissipation theorem (1), the correlation time τ_c of the fluctuations of the thermal reservoir, which is proportional to the inverse width of the spectrum $S_k(\omega)$, can be very appreciable even at small times τ_0. It is possible that $\gamma\tau_c\simeq 1$, and the Markov approximation is violated in the quantum region $T<\hbar\gamma$ of temperatures of the thermal reservoir.

With allowance for what we have said above, we simplify Eq. (5). We restrict ourselves to the one-dimensional case, and we also consider a nonequilibrium stationary state, in which the particle momentum will consist of a component mV, where V is the constant (in time) drift velocity, and a component $q(t)$ that relaxes to a value zero: $p(t)=mV+q(t)$. To close Eq. (5) or the Langevin equation (7), we assume that at times $t-t_1\sim\tau_c$ the operator $q(t)$ satisfies the relaxation equation

$$\dot{q}(t)+\gamma q(t)=0$$

with as yet unknown damping γ. The evolution of $q(t)$ during an interval $t-t_1\sim\tau_c$ can then be approximately described by the relation

$$q(t)=\exp[-\gamma(t-t_1)]q(t_1). \tag{9}$$

Here, the influence of the fluctuation source on $q(t)$ is ignored. In what follows, the relaxation coefficient will be found self-consistently. The assumption concerning the evolution law of $q(t)$, which will be justified subsequently, makes it possible to calculate the equal-time commutators in the nonlinear equation (7) and reduce it to a simpler form. Indeed, with allowance for (9) the evolution of the coordinate of the Brownian particle is approximately described by the expression

$$x(t)=x(t_1)+V(t-t_1)+\frac{1}{m\gamma}[1-e^{-\gamma(t-t_1)}](p(t_1)-mV). \tag{10}$$

The commutator of the operators $x(t)$ and $x(t_1)$,

$$[x(t),x(t_1)]_-=-(i\hbar/m\gamma)[1-e^{-\gamma(t-t_1)}]$$

is a c number. Then from the Baker–Hausdorff formula

$$e^{-ikx(t_1)}e^{ikx(t)}=e^{ik[x(t)-x(t_1)]}e^{\frac{k^2}{2}[x(t_1),x(t)]_-}$$

we find for the commutator and anticommutator of the functions $e^{ikx(t)}$ and $e^{-ikx(t_1)}$

$$i[e^{ikx(t)},e^{-ikx(t_1)}]_-=2\sin\left[\frac{k^2}{2m\gamma}(1-e^{-\gamma(t-t_1)})\right]e^{ik[x(t)-x(t_1)]},$$

$$\tfrac{1}{2}[e^{ikx(t)}, e^{-ikx(t_1)}]_+ = \cos\left[\frac{k^2}{2m\gamma}(1-e^{-\gamma(t-t_1)})\right] e^{ik[x(t)-x(t_1)]}.$$

With allowance for what was said above, we obtain from (7) a simpler Langevin equation for the momentum $p=m\dot{x}(t)$ of the Brownian particle:

$$\dot{p}(t) - \sum_k ik \int dt_1 R_k(t-t_1) e^{ikV(t-t_1)} \exp\left\{\frac{ik}{m\gamma}[1-e^{-\gamma(t-t_1)}]q(t_1)\right\} = \xi(t) + eE. \tag{11}$$

where

$$R_k(\tau) = 2\tilde{M}_k(\tau)\sin\left[\frac{k^2}{2m\gamma}(1-e^{-\gamma\tau})\right] + \varphi_k(\tau)\cos\left[\frac{k^2}{2m\gamma}(1-e^{-\gamma\tau})\right].$$

We assume that the relaxing component $q(t)$ is sufficiently small to ensure fulfillment of the inequality $|kq/m\gamma| \ll 1$ and linearization of Eq. (11) with respect to q. Separating from the right-hand side of the transformed equation (11) the component that is constant in time, and equating it to the external force eE, we obtain a nonlinear relation for the drift velocity V of the particle in the stationary nonequilibrium state:

$$\sum_k k \int d\tau\, R_k(\tau) \sin(kV\tau) = eE. \tag{12}$$

The remaining part gives an equation for $q(t)$:

$$\dot{q}(t) + \int dt_1\, \Lambda(t-t_1) q(t_1) = \xi(t), \tag{13}$$

with kernel

$$\Lambda(\tau) = \sum_k (k^2/m\gamma) R_k(\tau)(1-e^{-\gamma\tau})\cos(kV\tau). \tag{14}$$

We analyze the interaction of a fairly heavy Brownian particle with long-wavelength fluctuations of the thermal reservoir. Then $\hbar k_0^2/m\gamma \ll 1$, where k_0 is the maximum wave vector of the thermal reservoir. From the relations (13) and (14) we obtain the dispersion relation

$$-i\omega + \Lambda(\omega) = 0,$$

a solution of which we shall seek in the form $\omega = -i\gamma$. As a result, we arrive at a self-consistent equation for γ:

$$\gamma = \sum_k \frac{k^2}{2m\gamma}\Big\{\chi_k(-i\gamma+kV) + \chi_k(-i\gamma-kV) - \chi_k(kV) - \chi_k(-kV)$$

$$+ \frac{k^2}{m\gamma}[S_k(-i\gamma+kV) + S_k(i\gamma+kV) - 2S_k(kV)]\Big\}. \tag{15}$$

Note that our search for a solution of the dispersion relation in the form $\omega = -i\gamma$ agrees with the assumption made earlier concerning the nature of the evolution of the relaxing component of the momentum.

The contribution of the parametric fluctuations of the thermal reservoir to the damping coefficient γ increases strongly as the points $\pm kV - i\gamma$ approach one of the poles $\omega_n = -2\pi i n T$ of the function $\coth(\hbar\omega/2T)$. We shall call this phenomenon thermorelaxation resonance. The absence of poles of the function $\tilde{S}_k(\omega)$, which is the Fourier transform of the causal correlation function $\tilde{M}_k(\tau) = M_k(\tau)\eta(\tau)$, in the upper half-plane of ω leads to a weak monotonic dependence of the drift velocity V on the temperature T. It follows from (12) that $V = \mu_0 E$, where the mobility $\mu_0 = e/m\gamma_0$, and γ_0 is the damping coefficient due to the reaction of the thermal reservoir to the influence of the particle:

$$\gamma_0 = \sum_k \alpha_k k^2/m.$$

Without particularizing further the scattering mechanism, we assume that $\alpha_k \sim 1/k^2$. Then in the most interesting case of low drift velocities $V \ll T/\hbar k_0$, we arrive, after some manipulations, at a nonlinear self-consistent equation for the normalized relaxation coefficient $G = \gamma/\gamma_0$ of the momentum of the Brownian particle:

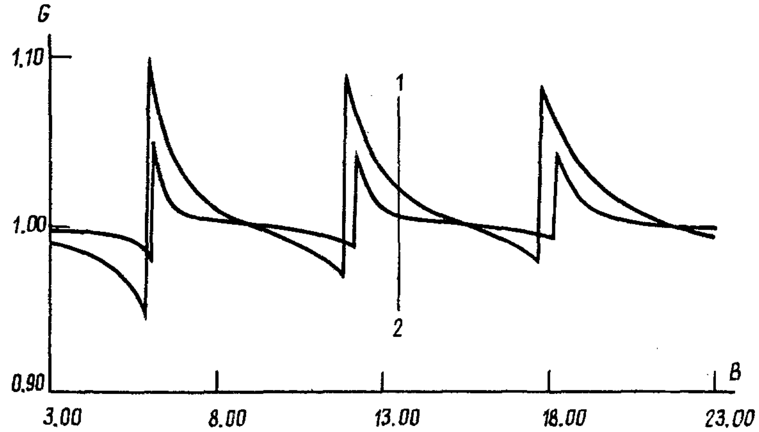

Fig 1

$$G=1+\frac{2s}{BGW}\left[\cos\left(\frac{BG}{2}\right)-\frac{2}{BG}\sin\left(\frac{BG}{2}\right)\right]\Phi(\theta), \tag{16}$$

where

$$\Phi(\theta)=\theta\left[1-\operatorname{arctg}\left(\frac{1}{\theta}\right)\right], \quad \theta=\frac{2}{BW}\sin\left(\frac{BG}{2}\right), \quad B=\frac{\hbar\gamma_0}{T}, \quad W=\frac{k_0V}{\gamma_0}, \quad s=\frac{\hbar k_0^2}{m\gamma_0}.$$

In the limits of both low ($T\to 0$, $kV>0$) and high ($T\gg\hbar kV$, $\hbar\gamma$) temperatures, the relaxation is mainly associated with the reaction of the thermal reservoir: $G\simeq 1$. Equation (16) is correct for $s\ll 1$, $BW\ll 1$. The numerical solution of this equation describing $G=\gamma/\gamma_0$ as a function of the reciprocal temperature $B=\hbar\gamma_0/T$ of the thermal reservoir for $W=10^{-5}$ and the two parameter values $s=0.05$ (curve 1) and $s=0.01$ (curve 2) is shown in Fig. 1. It is practically independent of the drift velocity in the region $10^{-6}\ll W\ll 10^{-2}$ and therefore encompasses the case of equilibrium relaxation. The oscillations begin at the temperature $T_1=\hbar\gamma/2\pi$ and extend into the region of lower temperatures with decreasing amplitude. Note the different nature of the oscillations of the function $G(B)$ directly at the points of thermorelaxation resonance $BG=2\pi n$ or $T_n=\hbar\gamma/2\pi n$, $n=1, 2, 3, \ldots$. With increasing parameter s, the contribution of the thermal fluctuations to the relaxation increases, but for $s\geq 0.1$ the oscillations of the function $G(B)$ become unstable, and therefore the value $s=0.05$ is optimal for observation of the effect.

For a heavy Brownian particle, for example, a proton with $m\simeq 10^{-24}$ g, interacting with a thermal reservoir possessing maximum wave vector $k_0\simeq 10^7$ cm^{-1} for damping coefficient $\gamma_0\simeq 2\cdot 10^{12}$ sec^{-1}, the parameter $s\simeq 0.05$ and the temperature of the first thermorelaxation resonance is $T_1\simeq 2$°K. The maximum amplitude of the oscillation of the momentum damping coefficient $\gamma(1/T)$ will then have the value $\Delta\gamma\simeq 0.15\gamma_0$, where γ_0 is the damping coefficient due to the reaction of the thermal reservoir to the influence of the particle.

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