Rate processes and decay of metastable states inside an anharmonic well driven by a random zero-point field

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A procedure previously developed by us for the evaluation of the diffusion coefficient and drift velocity of a charged Newtonian particle in a zero-point field is applied here to a sextic potential-one-dimensional system, for which an exact solution to the related Hamilton-Jacobi-Yasue-Riccati equation is available. This potential energy function represents either a stiff single well or a triple well. For this model, the averaged diffusion coefficient has been evaluated by assuming that the particle under study is confined for a long time inside one anharmonic well, that is, in the limit of infinite relaxation time, and the results are compared with quantum-mechanical predictions. A double-well model potential is subsequently examined.

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I. INTRODUCTION

A new method of solving the equations of motion for a classical Newtonian particle (or even more general systems) interacting with a random field has been developed recently by us. This procedure provides a partitioning of the velocity of the particle into a Eulerian component, depending on position only, and a Lagrangian component, depending on initial conditions and time [1,2] whose most remarkable property is that the various components may assume, if properly defined, a clear physical significance, representing the drift component and diffusive component of velocity, respectively (see [3] for a review). Therefore they allow, upon averaging under suitable boundary conditions, to obtain a representation of the diffusion process of probability density in configuration space[1] [4].

This point of view has proved to be of some degree of usefulness in application to solve problems in stochastic electrodynamics. Actually, not only the harmonic oscillator problem has been solved exactly so as to obtain straightforwardly the Schrödinger equation for the ground state and fluctuations thereof but also the general problem has been solved nonrelativistically in the frame of the *frozen-trajectory approximation* (FTA).[2] It has been proved that the parameters of the diffusion equation, under general conditions of averaging, remain bounded and meaningful if the interaction time τ_c of the particle and random field approaches zero, which means that the speed of all the relevant variables is small toward the velocity of light. Therefore the results obtained here are independent of any cutting-off frequency.

According to a relativistic calculation made in Ref. [5], the main contributions to the high velocities come from the higher frequencies, which yield a contribution to the mean square velocity proportional to the frequency. These authors inferred that these higher frequencies cannot produce any relevant effect upon particle motion except for a pronounced damping upon the run-away effect. A similar result has been obtained by us by considering the singular solution proposed by Battezzati [1,6] to a nonrelativistic equation of motion for the electron introduced by Caldirola, which yields the nonrelativistic Lorentz-Dirac equation exempt from the run-away solutions [2], because it is satisfied simultaneously with a second-order equation, the Braffort-Marshall equation (see Eqs. (1.11a) and (1.11b) of [1]), which does not satisfy causality in very short-time intervals but retains validity for time scales larger than τ_c [7].

II. APPROPRIATE SOLUTION TO EQUATIONS OF MOTION TO DESCRIBE DIFFUSION

The basic formulas which are used below for specific calculations over concrete model systems are presented here following Ref. [8].

A classical three-dimensional Newtonian system separable in each coordinate q and conjugated canonical momentum Π, with mass m, is considered, whose Hamiltonian is

$$H(\Pi,q,t) = \frac{\Pi^2}{2m} + U(q) - qk(t). \tag{1}$$

$U(q)$ being the potential energy function and $k(t)$ being the random driving force, Gaussian with zero mean, whose autocorrelation function is denoted by

$$\Xi(t-s) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \exp\{i\omega(t-s)\}\breve{\Xi}(\omega)d\omega = \langle k(t)k(s)\rangle, \tag{2}$$

where the brackets denote as usual stochastic averages, while t and s are time coordinates. It is considered here $k(t)$ with the following spectrum:

$$\breve{\Xi}(\omega) = m\hbar\tau_c|\omega^3| \tag{3}$$

with $\hbar$ being the Planck constant and $\tau_c = \frac{2e^2}{3mc^3} = \frac{2}{3}(\frac{1}{137})^3$ in a.u., e being the electron charge and c being the velocity of

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[1]Each individual component of velocity may be complex valued, although the total velocity could be made real, by solving with the appropriate boundary conditions.

[2]This is the first term of an expansion of the diffusion coefficient, where the response function is expanded in inverse powers of the mass.

light. It is convenient to introduce also the following spectrum with the corresponding stochastic force $\lambda(t)$:

$$\breve{\Xi}_\lambda(\omega) = \frac{\hbar\tau_c|\omega^3|}{m(1+\tau_c^2\omega^2)}. \tag{4}$$

The spectrum of the random driving force determines the resistance experienced by the system, which leads to the nonrelativistic Lorentz-Dirac equation [5,7,9], whose regular solutions satisfy simultaneously the Braffort-Marshall equation which results in a second-order differential equation in the position variable $q(t)$ with driving force $\lambda(t)$,

$$\ddot{q} + \beta(q)\dot{q} + \frac{1}{m}U'(q) = \lambda(t) \tag{5}$$

with the corresponding Hamilton-Jacobi-Yasue equation for the action $\Sigma(q,t)$ [3,10].

$$\frac{1}{2m}\Pi(q,t)^2 + U(q) - mq\lambda(t) + \int \beta(q)\Pi(q,t)dq = -\frac{\partial\Sigma}{\partial t}. \tag{6}$$

As was stated in Sec. I, the two components of linear momentum of the particle are added together so as to yield the total momentum

$$\Pi(q,t) = p(q) + \tilde{p}(t,q), \tag{7}$$

where $p(q)$ is any solution of the Hamilton-Jacobi-Yasue-Riccati equation [8]. Here,

$$\frac{1}{2m}p(q)^2 + U(q) + D_0[p'(q) + m\beta(q)] + \int \beta(q)p(q)dq = E, \tag{8}$$

where E is a constant, while $\tilde{p}(t,q(t))$ is solved in terms of the generalized random force $\lambda(t)$ whose spectrum is given by Eq. (4) through the response function $G(t,s)$,

$$\begin{aligned} G(t,s) &= \exp\left\{-\int_s^t \beta(q)dq - \frac{1}{m}\int_s^t p'(q(\alpha))d\alpha\right\} \\ &\cong \exp\left\{-\bar{\beta}(t-s) - \frac{1}{m}\int_s^t p'(q(\alpha))d\alpha\right\}, \end{aligned} \tag{9}$$

where the primes denote derivatives over coordinate q and

$$\bar{\beta} = \frac{1}{m}\tau_c\langle U''(q)\rangle \tag{10}$$

is the average of the frictional coefficient which appears in Eqs. (5) and (8) [8]. Thus

$$\tilde{p}(t,q(t)) = \int_{-\infty}^{t} dsG(t,s)[m\lambda(s) + D_0p''(q(s)) + mD_0\beta'(q(s))] + O(\tau_c^2). \tag{11}$$

The above equations provide the splitting of the momentum $\Pi(q,t)$ which is appropriate in case that the averaged diffusion coefficient D_0 is known. They have been obtained by retaining terms up to $O(\tau_c)$ only [8].

III. EVALUATION OF DIFFUSION COEFFICIENT

The diffusion coefficient in the FTA approximation [3,8] of a classical charged particle in a zero-point field (ZPF) is obtained from the following expression, in which diffusion is described by an operator acting on the two-time transition probability density of particles[3]:

$$\begin{aligned} &\hat{D}(t,t_0)_q^{(\mathrm{FTA})}\langle\delta(q(t)-q)\rangle \\ &= \int_{-\infty}^{t} d\tau\left\langle \exp\left\{-\int_\tau^t \beta(q)dq\right\}\int_{-\infty}^{\tau} d\sigma\delta(q(t)-q)G(\tau,\sigma)\right. \\ &\quad \left.\times\int_{-\infty}^{t} dsG(\tau,s)\right\rangle\Xi_\lambda(s-\sigma). \end{aligned} \tag{12}$$

Integrating over dq and taking the mean over time of the frictional coefficient yield the averaged diffusion coefficient D_0 resulting in the more simple expression,

$$\begin{aligned} D(t,t_0)_0^{(\mathrm{FTA})} &= \int_{t_0}^{t} d\tau\exp\{-\bar{\beta}(t-\tau)\} \\ &\quad \times\left\langle\int_{-\infty}^{\tau} d\sigma G(\tau,\sigma)\int_{-\infty}^{t} dsG(\tau,s)\right\rangle\Xi_\lambda(s-\sigma). \end{aligned} \tag{13}$$

This has been evaluated in [8] in terms of the double Fourier transform $\breve{\Phi}(\omega,\varpi)$ of the function

$$\begin{aligned} \Phi(\tau-\sigma,\tau-s) &= \exp\{-\bar{\beta}(\tau-\sigma)h(\tau-\sigma) - \bar{\beta}(\tau-s)h(\tau-s)\} \\ &\quad \times\left\langle\exp\left\{-\frac{1}{m}\int_\sigma^\tau p'(q(\alpha))d\alpha - \frac{1}{m}\int_s^\tau p'(q(\alpha))d\alpha\right\}\right\rangle, \end{aligned} \tag{14}$$

where $h(t)$ is Heaviside function of the variable t, defined as

$$h(t) = 0 \quad \text{if } t < 0, \qquad h(t) = 1 \quad \text{if } t > 0. \tag{15}$$

In the limit $t - t_0 \to +\infty$ above relation (13) may be transformed into

[3] Markovian evolution of transition probability density is assumed in order to cancel the memory term [4].

$$D_0^{(\mathrm{FTA})} = \frac{-i\hbar\tau_c}{4\pi^2 m\bar{\beta}} \int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\varpi \frac{\breve{\Phi}(\omega,\varpi)|\varpi^3|}{(1+\tau_c^2\varpi^2)(\omega+\varpi-i\varepsilon)} \tag{16}$$

with ε infinitesimal >0. Expanding this expression in power series of $\bar{\beta}$ and denoting by $\breve{\Phi}(\omega,\varpi)_0$ the limiting value of the function $\breve{\Phi}(\omega,\varpi)$ as $\bar{\beta}\to 0$, the following expression of the averaged diffusion coefficient in a stationary state results [9]:

$$D_0^{(\mathrm{FTA})} = \frac{i\hbar}{2m}\left[1 - \frac{m}{\pi^2\langle U''(q)\rangle}\int_{-\infty}^{+\infty} d\omega \int_0^{+\infty} d\varpi \frac{\breve{\Phi}(\omega,\varpi)_0\varpi^3}{\omega+\varpi-i\varepsilon} + \frac{1}{2\pi^3}\int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\varpi \breve{\Phi}(\omega,\varpi)_0 \tau_c\omega \ln|\tau_c\omega|\right]. \tag{17}$$

Equation (17) has been obtained from Eq. (16) by expanding $\breve{\Phi}(\omega,\varpi)$ and retaining only the singular terms which are $O(\bar{\beta}\ln\tau_c)$. The sign of the real part of the diffusion coefficient depends on the signs of $\langle U''(q)\rangle$ and $\breve{\Phi}(\omega,\varpi)_0$, which are expected to assume prevalently positive values in stationary states, because of Eq. (10) and

$$\int_{-\infty}^{+\infty} d\omega \int_{-\infty}^{+\infty} d\varpi \breve{\Phi}(\omega,\varpi) = 4\pi^2. \tag{18}$$

IV. EVALUATION OF SINGLE-WELL RESPONSE FUNCTION

Since main scope here is the evaluation of the averaged diffusion coefficient, according to Eqs. (12) and (13), for a particle confined for an infinite time inside a single anharmonic well, the main task will be the calculation of response function (14) under these assumptions. To this end averages are computed by the method of cumulants, according to the method proposed in Kubo's [11] stochastic theory of line shape: Eq. (14) is rewritten in the form

$$\begin{aligned}\breve{\Phi}(\omega,\varpi) = &\int_{-\infty}^{+\infty} \exp\{i\varpi(\tau-s)\}d(\tau-s)\exp\{i\omega(\tau-\sigma)\}d(\tau-\sigma)\\ &\times\int_{-\infty}^{+\infty}\exp\{i\varpi(\tau-s)\}d(\tau-s)\\ &\times\exp\Big\{-\bar{\beta}(\tau-\sigma)h(\tau-\sigma)-\bar{\beta}(\tau-s)h(\tau-s)\\ &-\frac{1}{m}\langle p'(q)\rangle(2\tau-\sigma-s)\Big\}\\ &\times\left\langle \exp\left\{i\int_\sigma^\tau \omega_1(\alpha)d\alpha + i\int_s^\tau \omega_1(\alpha)d\alpha\right\}\right\rangle\end{aligned} \tag{19}$$

with

$$i\omega_1(\alpha) = -\frac{1}{m}[p'(q(\alpha)) - \langle p'(q)\rangle]. \tag{20}$$

There follows from the above equations that the main difficulty of the present problem will be the computation of the cumulant averages included in the exponential of the function $i\omega_1(\alpha)$. In a first approximation, these averages may be computed making use of the free oscillator correlation functions of the coordinate, imbedded in a zero-point field, which would suffice to yield the exact cumulant averages up to second order in the perturbation. Higher-order perturbative effects on the correlation functions could in principle be accounted for, but they would not modify the averages in short times because the small perturbations modify the correlation functions only in the frequency range

$$0 \le |\omega| \le |\omega_1| \ll \omega_0 \tag{21}$$

or in the time domain

$$|\alpha-\alpha'| > \frac{1}{|\omega_1|}. \tag{22}$$

From these basic arguments there follows [11]

$$\begin{aligned}&\left\langle \exp\left\{i\int_\sigma^\tau \omega_1(\alpha)d\alpha + i\int_s^\tau \omega_1(\alpha)d\alpha\right\}\right\rangle\\ &= \exp\left\{-\frac{1}{2}\Delta^2\left[\int_\sigma^\tau d\alpha\int_\sigma^\tau d\gamma\phi(\alpha-\gamma)\right.\right.\\ &\left.\left.+2\int_\sigma^\tau d\alpha\int_s^\tau d\gamma\phi(\alpha-\gamma)+\int_s^\tau d\alpha\int_s^\tau d\gamma\phi(\alpha-\gamma)\right]\right\},\end{aligned} \tag{23}$$

where

$$\begin{cases}\phi(\alpha-\gamma) = \dfrac{\langle\omega_1(\alpha)\omega_1(\gamma)\rangle - \langle\omega_1(\alpha)\rangle\langle\omega_1(\gamma)\rangle}{\Delta^2},\\ \Delta^2 = \langle\omega_1^2\rangle - \langle\omega_1\rangle^2.\end{cases} \tag{24}$$

The correlation function of the coordinate $\langle q(\alpha)q(\gamma)\rangle$ of the unperturbed harmonic oscillator in a ZPF can be computed straightforwardly, making use of the definitions of velocity components given in Sec. II with $p(q)$ equal to the single-well limit of Eqs. (31) and (41) below, which means $b\to 0, a\to 0$. Assuming "natural boundary conditions" which were defined, for instance, in Ref. [12],[4] there results that for $\sigma < t_0 < t$ [3,13],

$$\frac{\delta q(t)}{\delta\lambda(\sigma)} = \exp\left\{-\frac{1}{2}\bar{\beta}(t-\sigma)\right\} \times \frac{\exp\{i\Omega(t-\sigma)\} - \exp\{-i\Omega(t+\sigma-2t_0)\}}{2i\Omega} \tag{25}$$

with $\Omega > 0$ being the unperturbed frequency of oscillation, while for $t_0 < \sigma < t$,

[4]The coordinate is fixed at time $t=t_0$, while the velocity is in equilibrium with the random field. For a more detailed definition of boundary conditions, see Ref. [12].

$$\frac{\delta q(t)}{\delta\lambda(\sigma)} = \exp\left\{-\frac{1}{2}\bar{\beta}(t-\sigma)\right\} \times \frac{\exp\{i\Omega(t-\sigma)\} - \exp\{-i\Omega(t-\sigma)\}}{2i\Omega}. \quad (26)$$

Then, the coordinate correlation function may be computed by using Novikov' theorem in its most simple form [14],

$$\langle q(\alpha)q(\gamma)\rangle = \int_{-\infty}^{\alpha} ds \int_{-\infty}^{\gamma} d\sigma \frac{\delta q(\alpha)}{\delta\lambda(s)} \frac{\delta q(\gamma)}{\delta\lambda(\sigma)} \Xi_\lambda(s-\sigma) + \langle q(\alpha)\rangle\langle q(\gamma)\rangle, \quad (27)$$

therefore, by omitting transients and going to the limit $\tau_c \to 0$,

$$\begin{aligned}\langle q(\alpha)q(\gamma)\rangle &= \frac{\hbar\tau_c}{4m\bar{\beta}\Omega(1+\tau_c^2\Omega^2)}\exp\left\{-\frac{1}{2}\bar{\beta}|\alpha-\gamma|\right\}\left[\left(\Omega+\frac{1}{2}i\bar{\beta}\right)^2\exp\{i\Omega|\alpha-\gamma|\} - \left(\Omega-\frac{1}{2}i\bar{\beta}\right)^2\exp\{-i\Omega|\alpha-\gamma|\}\right] \\ &\quad + \frac{\hbar\tau_c}{2\pi i m\bar{\beta}\Omega}\int_{-\infty}^{0} d\eta \frac{\eta^3}{1-\tau_c^2\eta^2}\left[\frac{\exp\{\eta|\alpha-\gamma|\}}{\left(\Omega+\frac{i}{2}\bar{\beta}\right)^2+\eta^2} - \frac{\exp\{\eta|\alpha-\gamma|\}}{\left(\Omega-\frac{i}{2}\bar{\beta}\right)^2+\eta^2}\right] \\ &\cong \frac{\hbar\tau_c\Omega}{2m\bar{\beta}}\exp\left\{-\frac{1}{2}\bar{\beta}|\alpha-\gamma|\right\}\cos\{\Omega|\alpha-\gamma|\} \\ &= \frac{\hbar}{2m\Omega}\exp\left\{-\frac{1}{2}\bar{\beta}|\alpha-\gamma|\right\}\cos\{\Omega|\alpha-\gamma|\}. \end{aligned} \quad (28)$$

V. SYMMETRIC TRIPLE-WELL-MODEL POTENTIAL

It is considered the following example, in which the sextic potential function $W(q)$ is substituted for $U(q)$ into Eq. (1),

$$W(q) = \frac{1}{2}m\omega_0^2 q^2 - \frac{1}{3}mbwq^4 + \frac{1}{18}mb^2q^6, \quad (29)$$

where ω_0 and b are real parameters. If the frequency w verifies the following relation,

$$w = \sqrt{\omega_0^2 - \frac{1}{4}\bar{\beta}^2 + 4\tau_c bD_0 w + 2ibD_0}, \quad (30)$$

then Eq. (8) admits the following solution to $O(\tau_c bq^2)$ included (terms of higher order in the product of the three variables are excluded):

$$p(q) = -m\left[iw + \frac{1}{2}\beta(q)\right]q + \frac{i}{3}mbq^3. \quad (31)$$

$W(q)$ with $b>0$ represents a symmetric triple well with minima at $q=0$, $q \cong \pm\sqrt{\frac{3\omega_0}{b}}$, and maxima at $q \cong \pm\sqrt{\frac{\omega_0}{b}}$, whose values are approximately 0, 0, and $\frac{2}{9}m\omega_0^2/b$, respectively. In the limit $b \to 0$ the depth and width of the wells increase indefinitely; therefore it is assumed that the expressions displayed in Sec. III for the evaluation of diffusion coefficient are applicable, and the quadratic approximation for the potential is a good first approximation. Since Eq. (12) involves integrals over infinite time, even the small varying part of the diffusion coefficient results to be sensibly a constant over all the region accessible to the particle because the final part of the trajectory would not contribute significantly to the mean values over time. Therefore it is obtained from Eqs. (20), (28), and (31) [14]

$$i\omega_1(\alpha) = -ib[q(\alpha)^2 - \langle q(\alpha)^2\rangle], \quad (32)$$

$$\begin{aligned}\langle\omega_1(\alpha)\omega_1(\gamma)\rangle_c &= b^2(\langle q(\alpha)^2q(\gamma)^2\rangle - \langle q(\alpha)^2\rangle\langle q(\gamma)^2\rangle) \\ &= 2b^2\langle q(\alpha)q(\gamma)\rangle^2 \\ &= \frac{b^2\hbar^2}{2m^2\omega_0^2}\phi_b(\alpha-\gamma) \\ &= \Delta_b^2\phi_b(\alpha-\gamma),\end{aligned} \quad (33)$$

$$\phi_b(\alpha-\gamma) = \exp\{-\bar{\beta}|\alpha-\gamma|\}\cos^2\{w(\alpha-\gamma)\}. \quad (34)$$

In order to obtain this correlation function the approximate linearity of the response [see Eq. (27)] has been used. Thus, in the limiting case that is considered, w is real and positive. Moreover, in the limit $\bar{\beta} \to 0$,

$$\begin{aligned}&\int_s^\tau d\alpha \int_\sigma^\tau d\gamma \phi_b(\alpha-\gamma) \\ &= \frac{1}{2}[\tau^2 - \tau(\sigma+s) + s\sigma] - \frac{1}{8w^2}[\cos 2w(\tau-\sigma) \\ &\quad + \cos 2w(\tau-s) - \cos 2w(\sigma-s) - 1].\end{aligned} \quad (35)$$

Using this integral and assuming the validity of Eq. (28) in the limit of small oscillations and $2\tau-\sigma-s < \Delta_b^{-1}$, the response function $\Phi_b(\tau-\sigma,\tau-s)_0$ is evaluated as

$$\Phi_b(\tau-\sigma,\tau-s)_0 \cong \exp\left\{i\left(w-\frac{1}{\sqrt{2}}\Delta_b\right)(2\tau-\sigma-s)-\frac{1}{4}\Delta_b^2(2\tau-\sigma-s)^2\right\}$$
$$\times\exp\left\{\frac{\Delta_b^2}{4w^2}\left[2\cos w(2\tau-\sigma-s)\cos w(\sigma-s)-\frac{1}{2}\cos 2w(\sigma-s)-\frac{3}{2}\right]\right\}$$
$$=\exp\left\{i\left(w-\frac{1}{\sqrt{2}}\Delta_b\right)(2\tau-\sigma-s)-\frac{1}{4}\Delta_b^2(2\tau-\sigma-s)^2\right\}$$
$$\times\left[1+\frac{\Delta_b^2}{4w^2}\left[2\cos w(2\tau-\sigma-s)\cos w(\sigma-s)-\frac{1}{2}\cos 2w(\sigma-s)-\frac{3}{2}\right]\right]+O(\Delta_b^4). \tag{36}$$

The large frequency behavior of this response function is evaluated from the very short-time expansion as

$$\breve{\Phi}_b(\omega,\varpi)_0 \approx 2\pi\delta(\omega-\varpi)\sqrt{\frac{2\pi}{\Delta_b^2}}\exp\left\{-\frac{\left[\frac{1}{2}(\omega+\varpi)+\frac{i}{m}\langle p'(q)\rangle\right]^2}{2\Delta_b^2}\right\} \tag{37}$$

while the double Fourier transform of the expanded form of Eq. (36) is

$$\breve{\Phi}_b(\omega,\varpi)_0=\exp\left\{-\frac{3\Delta_b^2}{8w^2}\right\}2\pi\delta(\omega-\varpi)\frac{2\sqrt{\pi}}{|\Delta_b|}\exp\left\{-\frac{\left(\varpi+w-\frac{1}{\sqrt{2}}\Delta_b\right)^2}{\Delta_b^2}\right\}$$
$$+\frac{|\Delta_b|}{4w^2}\left[2\pi\delta(\varpi-\omega+2w)\sqrt{\pi}\exp\left\{-\frac{\left(\varpi+3w-\frac{1}{\sqrt{2}}\Delta_b\right)^2}{\Delta_b^2}\right\}+2\pi\delta(\varpi-\omega+2w)\sqrt{\pi}\ \exp\left\{-\frac{\left(\varpi-w-\frac{1}{\sqrt{2}}\Delta_b\right)^2}{\Delta_b^2}\right\}\right.$$
$$+2\pi\delta(\varpi-\omega-2w)\sqrt{\pi}\exp\left\{-\frac{\left(\varpi+w-\frac{1}{\sqrt{2}}\Delta_b\right)^2}{\Delta_b^2}\right\}+2\pi\delta(\varpi-\omega+2w)\sqrt{\pi}\exp\left\{-\frac{\left(\varpi+w-\frac{1}{\sqrt{2}}\Delta_b\right)^2}{\Delta_b^2}\right\}$$
$$\left.-\pi\delta(\varpi-\omega+4w)\sqrt{\pi}\exp\left\{-\frac{\left(\varpi+3w-\frac{1}{\sqrt{2}}\Delta_b\right)^2}{\Delta_b^2}\right\}-\pi\delta(\varpi-\omega-4w)\sqrt{\pi}\exp\left\{-\frac{\left(\varpi-w-\frac{1}{\sqrt{2}}\Delta_b\right)^2}{\Delta_b^2}\right\}\right]. \tag{37'}$$

As a consequence of the expansion of the exponential function in Eq. (36), this distribution has narrower width $(\Delta_b^2/2)^{1/2}$ instead of $|\Delta_b|$ which appears in Eq. (37).

Evaluating the averaged diffusion coefficient it is made use of Eq. (17). The double Fourier transform operation on $\Phi_b(\tau-\sigma,\tau-s)_0$ with respect to variable $\zeta=\sigma-s$ whose argument is $\frac{1}{2}(\varpi-\omega)$ and variable $\eta=2\tau-\sigma-s$, which yields the argument $\frac{1}{2}(\omega+\varpi)$ in $\Phi(\omega,\varpi)$, is written down explicitly. The integral over variable ζ is evaluated first, followed by integration over $d\omega$. Then the remaining integration over $d\varpi$ is interchanged with that over $d\eta$ because this allows us to take account easily, in the limit of vanishing $|\Delta_b/\omega_0|$, of the pole contributions. There follows, by retaining only the dominant pole terms in the ϖ integration in this limit,

$$D_0^{(\mathrm{FTA})} \cong \frac{i\hbar}{2m} - \frac{i\hbar\Delta_b^2}{8\pi\langle W''(q)\rangle w^2}\int_{-\infty}^{+\infty} d\eta \int_0^{+\infty} d\varpi\varpi^3 \exp\left\{-\frac{1}{4}\Delta_b^2\eta^2\right\}\left[\frac{\exp\left\{i\left(\varpi - w - \frac{1}{\sqrt{2}}\Delta_b\right)\eta\right\}}{2(\varpi - w) - i\varepsilon} - \frac{\exp\left\{i\left(\varpi - w - \frac{1}{\sqrt{2}}\Delta_b\right)\eta\right\}}{4(\varpi - 2w) - i\varepsilon}\right] + \frac{i\hbar}{\pi^2 m}\int_{-\infty}^{+\infty} d\varpi\,\tau_c\varpi\ \ln|\tau_c\varpi|\frac{\sqrt{\pi}}{|\Delta_b|}\exp\left\{-\frac{\left(\varpi + w - \frac{1}{\sqrt{2}}\Delta_b\right)^2}{\Delta_b^2}\right\}$$
$$\cong \frac{i\hbar}{2m} - \frac{i\hbar\Delta_b^2}{8\pi\langle W''(q)\rangle w^2}\int_{-\infty}^{+\infty} d\eta\int_{-\infty}^{+\infty} d\varpi\ \exp\left\{-\frac{1}{4}\Delta_b^2\eta^2 + i\left(\varpi - w - \frac{1}{\sqrt{2}}\Delta_b\right)\eta\right\} \times\left[\frac{1}{4}\varpi^2 - \frac{1}{2}w^2 + \frac{w^3}{2(\varpi - w - i\varepsilon)} - \frac{2w^3}{\varpi - 2w - i\varepsilon}\right] + \frac{i\hbar}{\pi m}\int_{-\infty}^{+\infty} d\varpi\,\tau_c\varpi\ \ln|\tau_c\varpi|\,\delta\left(\varpi + w - \frac{1}{\sqrt{2}}\Delta_b\right)$$
$$\cong \frac{i\hbar}{2m}\left(1 - 0.25\times\frac{\Delta_b}{\omega_0}\right) + \sqrt{\frac{\pi}{e}}\frac{\hbar|\Delta_b|}{8m\omega_0} - \frac{i\hbar}{\pi m}\tau_c\omega_0\ \ln\ \tau_c\omega_0 + O\left(\frac{\Delta_b^2}{\omega_0^2}\right). \tag{38}$$

Notice that only the pole $\varpi = w$contributes significantly to the real part of diffusion coefficient. The result has been evaluated approximately, but it shows clearly that in the stage of a single occupied well the process has no time-reversal symmetry.

This equation is also applicable if $b<0$, in which case potential $W(q)$ is a stiff single-well potential. It has been shown that, besides a small change in the dominant imaginary diffusion coefficient, the main effect of nonlinearity of the potential is the appearance of a *positive* real part of D_0, which is $O(\Delta_b|D_0|/\omega_0)$. Accordingly the oscillation frequencies of fluctuations over the ground state are shifted, in first order, to

$$\frac{E_n - E_0}{2mD_0} = -ni\omega_0 + n(n+1)b\frac{D_0}{\omega_0}. \tag{39}$$

Consequently, with $b<0$ density fluctuations over the stable equilibrium state are moderately damped, while in the opposite case (triple well), they are unstable and growing up in amplitude. They cannot grow indefinitely because of flow across the saddle point. It is pointed out that although the system might appear to absorb energy from the surroundings, this is only true at the primitive stage in which the system evolves near the bottom of the potential well since the small real part of D_0 is likely to be strongly modified as soon as the particle approaches or overflows the saddle point, thus approaching equilibrium in the adjacent well. Actually, at complete equilibrium time inversion symmetry should be restored.

VI. DOUBLE-WELL MODEL POTENTIAL

Similar calculations were carried out for a soluble model of a (quasi)symmetric double-well potential, without detecting, to the present order of approximation, any deviation from the quantum-mechanical value of diffusion coefficient.

The following potential energy function is introduced, with real parameters ω_0 and a

$$V(q) = -imaD_0q + \frac{1}{2}m\omega_0^2q^2 - \frac{1}{2}mauq^3 + \frac{1}{8}ma^2q^4, \tag{40}$$

then Eq. (8) is satisfied to $O(\tau_c aq^2)$ included by

$$\frac{1}{m}p(q) = -\left[iu + \frac{1}{2}\beta(q)\right]q + \frac{i}{2}aq^2 \tag{41}$$

with

$$u = \sqrt{\omega_0^2 - \frac{1}{4}\bar{\beta}^2 - \frac{3}{2}\tau_c a^2 D_0}. \tag{42}$$

Potential $V(q)$ is a (quasi)symmetric double well, which in the limit $\hbar\to 0$, $\bar{\beta}\to 0$ has maxima and minima in the points $q=0$, $q\cong\frac{\omega_0}{a}$, $q\cong\frac{2\omega_0}{a}$, being thoroughly symmetric. Since the main scope here is the evaluation of the function $\Phi_a(\tau-\sigma,\tau-s)$ for short-time intervals, which yields the Fourier transform $\breve{\Phi}_a(\omega,\varpi)$ for large values of the argument, by using Kubo's stochastic theory of line shape [11] through Eqs. (19), (20), and (41) it is obtained

$$i\omega_1(\alpha) = -ia[q(\alpha) - \langle q(\alpha)\rangle]. \tag{43}$$

In the single-well harmonic approximation for the response function leading to Eq. (28), the variable $q(\alpha)$ is a Gaussian random variable, consequently, the cumulant expansion of $\ln\Phi_a(\tau-\sigma,\tau-s)$ can be stopped after the second cumulant average, yielding, in the range of values

$$|\bar{\beta}(\tau-\sigma)|,|\bar{\beta}(\tau-s)|,|\bar{\beta}(\sigma-s)| \ll 1 \tag{44}$$

the following result:

$$\int_\sigma^\tau d\alpha \int_s^\tau d\gamma \phi_a(\alpha-\gamma) = \frac{2\bar{\beta}}{\frac{1}{4}\bar{\beta}^2+u^2}\{h(\tau-\sigma)h(\tau-s)[|\tau-\sigma|h(\sigma-s)+|\tau-s|h(s-\sigma)] + h(\sigma-\tau)h(s-\tau)[|\tau-\sigma|h(s-\sigma)+|\tau-s|h(\sigma-s)]\} + \frac{\exp\left\{-\left(\frac{1}{2}\bar{\beta}+iu\right)|\tau-\sigma|\right\}+\exp\left\{-\left(\frac{1}{2}\bar{\beta}+iu\right)|\tau-s|\right\}-\exp\left\{-\left(\frac{1}{2}\bar{\beta}+iu\right)|\sigma-s|\right\}-1}{\left(\frac{1}{2}\bar{\beta}+iu\right)^2} + \text{c.c.} \tag{45}$$

There results that the very short-time behavior for $u|\tau-\sigma|,u|\tau-s|,u|\sigma-s|\ll 1$ is a Gaussian:

$$\left\langle \exp\left\{-\frac{1}{m}\int_\sigma^\tau p'(q(\alpha))d\alpha - \frac{1}{m}\int_s^\tau p'(q(\alpha))d\alpha\right\}\right\rangle \cong \exp\left\{-\frac{1}{m}\langle p'(q)\rangle(2\tau-\sigma-s)-\Delta_a^2(2\tau-\sigma-s)^2\right\}, \tag{46}$$

where

$$\Delta_a^2 = \frac{\hbar\tau_c u a^2}{2m\bar{\beta}} \cong \frac{\hbar a^2}{2m\omega_0} \tag{47}$$

Fourier transforming function (46) it is obtained

$$\breve{\Phi}_a(\omega,\varpi) \approx 2\pi\delta(\omega-\varpi)\sqrt{\frac{\pi}{\Delta_a^2}}\exp\left\{-\frac{\left[\frac{1}{2}(\omega+\varpi)+\frac{i}{m}\langle p'(q)\rangle\right]^2}{4\Delta_a^2}\right\}. \tag{48}$$

Therefore the effect of finite nonzero a (which means the effect of a side well which would disappear as $a\to 0$) is a Gaussian broadening of the spectrum around the main frequency of harmonic oscillations. For longer time intervals, which however satisfy inequality (44), going to the limit $\bar{\beta}\to 0$ it is obtained from Eq. (45)

$$\begin{aligned}\Phi_a(\tau-\sigma,\tau-s)_0 &= \exp\left\{-\frac{1}{m}\langle p'(q)\rangle(2\tau-\sigma-s)\right\}\exp\left\{-\frac{\Delta_a^2}{\omega_0^2}\left[6+2\cos\omega_0(\sigma-s)-8\cos\frac{1}{2}\omega_0(\sigma-s)\cos\frac{1}{2}\omega_0(2\tau-\sigma-s)\right]\right\}\\ &= \exp\left\{-\frac{1}{m}\langle p'(q)\rangle(2\tau-\sigma-s)\right\}\exp\left\{-\frac{2\Delta_a^2}{\omega_0^2}[3+\cos\omega_0(\tau-\sigma)\cos\omega_0(\tau-s)+\sin\omega_0(\tau-\sigma)\sin\omega_0(\tau-s)\right.\\ &\quad\left.-2\cos\omega_0(\tau-\sigma)-2\cos\omega_0(\tau-s)]\right\}.\end{aligned} \tag{49}$$

In the limit $a\to 0$ the above expression can be expanded

$$\begin{aligned}\Phi_a(\tau-\sigma,\tau-s)_0 = \exp\left\{-\frac{1}{m}\langle p'(q)\rangle(2\tau-\sigma-s)\right\}\left\{1-\frac{2\Delta_a^2}{\omega_0^2}[3+\cos\omega_0(\tau-\sigma)\cos\omega_0(\tau-s)\right.\\ \left.+\sin\omega_0(\tau-\sigma)\sin\omega_0(\tau-s)-2\cos\omega_0(\tau-\sigma)-2\cos\omega_0(\tau-s)]+O\left(\frac{\Delta_a^4}{\omega_0^4}\right)\right\},\end{aligned} \tag{50}$$

so that the Fourier transform is easily obtained to the same order of approximation,

$$\begin{aligned}\breve{\Phi}_a(\omega,\varpi)_0 \cong{}& 4\pi^2\left[1-6\frac{\Delta_a^2}{\omega_0^2}\right]\delta\left(\omega+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi+\frac{i}{m}\langle p'(q)\rangle\right)-4\pi^2\frac{\Delta_a^2}{\omega_0^2}\left[\delta\left(\omega+\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi-\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\right.\\ &+\delta\left(\omega-\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi+\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)-2\delta\left(\omega+\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi+\frac{i}{m}\langle p'(q)\rangle\right)\\ &-2\delta\left(\omega-\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi+\frac{i}{m}\langle p'(q)\rangle\right)-2\delta\left(\omega+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi+\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\\ &\left.-2\delta\left(\omega+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi-\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\right].\end{aligned} \tag{51}$$

Then from Eq. (17) the single-well average diffusion coefficient in the frozen-trajectory approximation follows

$$D_0^{(\mathrm{FTA})} = \frac{i\hbar}{2m} + \frac{i\hbar}{2\pi^2 m\omega_0^2}\int_{-\infty}^{+\infty} d\omega \int_0^{+\infty} d\varpi \frac{\varpi^3}{(1+\tau_c^2\varpi^2)(\omega+\varpi)}$$
$$\times 4\pi^2 \frac{\Delta_a^2}{\omega_0^2}\left[\delta\left(\omega+\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi-\omega_0+\frac{i}{m}\langle p'(q)\rangle\right) - 2\delta\left(\omega+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi-\omega_0+\frac{i}{m}\langle p'(q)\rangle\right)\right]$$
$$+ \frac{i\hbar\tau_c}{4\pi^3 m}4\pi^2\int_{-\infty}^{+\infty} d\omega\int_{-\infty}^{+\infty} d\varpi\,\delta\left(\omega+\frac{i}{m}\langle p'(q)\rangle\right)\delta\left(\varpi+\frac{i}{m}\langle p'(q)\rangle\right)\frac{\omega\ln\tau_c|\omega|}{1+\tau_c^2\omega^2} + \mathrm{h.o.t.}$$
$$\cong \frac{i\hbar}{2m} - \frac{2i\hbar}{m\omega_0^2}\left(\frac{\Delta_a}{\omega_0}\right)^2 \frac{\left(\omega_0 - \frac{i}{m}\langle p'(q)\rangle\right)^3}{\frac{2i}{m}\langle p'(q)\rangle} - \frac{4i\hbar}{m\omega_0^2}\left(\frac{\Delta_a}{\omega_0}\right)^2 \frac{\left(\omega_0 - \frac{i}{m}\langle p'(q)\rangle\right)^3}{\omega_0 - \frac{2i}{m}\langle p'(q)\rangle} + \frac{\hbar\tau_c}{\pi m^2}\langle p'(q)\rangle \ln\tau_c\left|\frac{i}{m}\langle p'(q)\rangle\right|$$
$$= \frac{i\hbar}{2m} + \frac{\hbar\tau_c}{\pi m^2}\langle p'(q)\rangle\ln\tau_c\left|\frac{i}{m}\langle p'(q)\rangle\right| + o(a^5), \qquad (52)$$

since $\frac{i}{m}\langle p'(q)\rangle = \omega_0 + o(a)$.

VII. CONCLUSIONS

The diffusion coefficient for two particular anharmonic oscillators in a ZPF, whose static potential functions are given by Eqs. (29) and (40), has been evaluated up to second order in the anharmonicity parameter in the limit $\bar{\beta}\to 0$. Potential (40) is a slightly asymmetric one by a term which is first order in the anharmonicity parameter times $\hbar$, but is well behaved at infinity since it diverges as a^2q^4. Potential (29) is symmetric and diverges at infinity as b^2q^6. However, it behaves differently at intermediate values of coordinate, according to the sign of b. Consequently, the postulated stationary distribution is found to be stable or unstable with respect to spontaneous fluctuations for $b<0$ or $b>0$, respectively. By writing explicitly the real and imaginary parts of the diffusion coefficient, the quasistationary transient probability distributions inside the main well are obtainable in the form of Eqs. (A1) and (A2). Among these solutions, which are exact for $\bar{\beta}\to 0$, only that one in Eq. (A2) with $b<0$ is acceptable over the whole real axis of the coordinate q. In fact $P_a(q)$ is not bounded on one side, depending on the sign of a, while $P_b(q)$ with $b>0$ is unbounded on each side. This behavior is understandable since the diffusion coefficient which has been calculated here is actually an average value over the whole distribution of probability density in the well which is centered on the origin of coordinates, thus Eqs. (A1) and (A2) cannot represent correctly the distribution in full detail with regard to position far from the origin. The main term of the diffusion coefficient in the FTA approximation for the response is however a purely imaginary constant independent of the potential, as it results from Eq. (17), the small deviations from this constant value being the object of study in this work.

Notice that the assumed dependence of the potential energy on D_0 is not essential in the subsequent calculations leading to Eq. (38) to the required level of accuracy. Similar developments could be made by substituting ω_0 for w in both Eqs. (29) and (31), which would correspond to solving Eq. (8) by the Brillouin-Kramers-Wentzel method. Then, it would be possible to investigate on the effects of higher-order terms of the expansion of the action in powers of D_0.

The present calculations have the advantage of providing a highly accurate solution in the potential $W(q)$, the resulting potential energy function being real up to $O(b^2)$ included.

In case of potential $V(q)$ (double well) the variability of diffusion coefficient has been found to be irrelevant up to second order in the parameter of anharmonicity, while for potential $W(q)$ a small real part already appears in first order. Accordingly, the potential well results to be more stiff than quadratic for $b<0$, while two deep side wells appear for $b>0$, however small the perturbation parameter is. In the first circumstance fluctuations in the equilibrium state inside the central well are damped, while in the second instance they are unstable and growing up in amplitude, which clearly corresponds to jumping of the particle into the side wells. This effect is seemingly distinct from the following others which act in the same direction:

(i) Eq. (5) with Eq. (10) shows that damping is increased with curvature of the potential, while it becomes negative near the saddle point, where the potential is convex, thus favoring overflow [15]. However here it has been assumed a constant damping coefficient $\bar{\beta}$ and moreover it was taken the limit $\bar{\beta}\to 0$; therefore this effect is neglected.

(ii) The tunneling effect and symmetrization of the wave function is a quantum-mechanical effect which is also active here, being due to the imaginary dominant part of the diffusion coefficient. The quantum-mechanical approach to this problem [16–18] has been sketched in the Appendix for comparison.

The conclusions that follow are that, though the calculations reported here cannot display in full detail the spatial and temporal dependence of the diffusion parameters, however they show evidence without ambiguity that the main effect of the random ZPF acting on the system is an enhancement of the rates of decay and relaxation processes and consequently cannot be in conflict with thermodynamical equilibrium.

Similar conclusion about the action of a zero-point field affecting the behavior of a quantum system were reported in Ref. [9], though their approach was different apparently of the kind of point (i) of this discussion.

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APPENDIX: EXPANSION OF EXACT SOLUTIONS INTO POLARIZATION ORDERS

Since the potential function $V(q)$ is very close to a symmetric double well, with midpoint $q=u/a$, the present problem is suitable for a quantum-mechanical solution in terms of symmetry-adapted perturbation theory [19]. This is the one-dimensional analog of the expanded interaction between two attractive centers such as atoms or molecules in the limit of large internuclear separation, the small parameter playing the role of the inverse interatomic distance $1/R$. This aspect of the problem is also shared with the potential $W(q)$ with $b>0$, although in this case the perturbative center for the interaction is twofold.

The analytical solutions to the Schrödinger equation which are here computed exactly as $\tau_c \to 0$ are given by the square roots of $P_a(q)$ and $P_b(q)$, which are

$$P_a(q) \propto \exp\left\{ \frac{\mathrm{Im}(D_0) + i\mathrm{Re}(D_0)}{|D_0|^2} \left(-\frac{1}{2}uq^2 + \frac{1}{6}aq^3 \right) \right\}, \tag{A1}$$

$$P_b(q) \propto \exp\left\{ \frac{\mathrm{Im}(D_0) + i\mathrm{Re}(D_0)}{|D_0|^2} \left(-\frac{1}{2}wq^2 + \frac{1}{12}bq^4 \right) \right\}. \tag{A2}$$

By expanding these solutions in power series of the small parameter a or b and substituting into the Schrödinger equation, it is possible to isolate an infinite series of recurrent equations for the coefficients of the expansion, which are functions of the coordinate q verifying the required boundary conditions. Actually these functions are obtainable from the coefficients of a Rayleigh-Schrödinger perturbative power series in the expansion parameter. In quantum mechanics the leading terms of each of these series are, for potential $V(q)$,

$$\psi_0(q) \propto \exp\left\{ -\frac{m\omega_0 q^2}{2\hbar} \right\}, \tag{A3}$$

$$\psi_1(q) = \frac{maq^3}{\hbar}\psi_0(q), \tag{A4}$$

satisfying the first-order equation

$$\left(-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial q^2} + \frac{1}{2}m\omega_0^2 q^2 - \frac{1}{2}\hbar\omega_0 \right)\psi_1(q) = \frac{1}{2}(ma\omega_0 q^3 - a\hbar q)\psi_0(q) \tag{A5}$$

and for potential $W(q)$

$$\psi_0(q) \propto \exp\left\{ -\frac{m\omega_0 q^2}{2\hbar} \right\}, \tag{A6}$$

$$\psi_1(q) = \frac{1}{2}\left(\frac{b}{2\omega_0}q^2 + \frac{mb}{6\hbar}q^4 \right)\psi_0(q), \tag{A7}$$

$$\left(-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial q^2} + \frac{1}{2}m\omega_0^2 q^2 - \frac{1}{2}\hbar\omega_0 \right)\psi_1(q) = \left(\frac{1}{3}mb\omega_0 q^4 - \frac{b\hbar^2}{4m\omega_0} \right)\psi_0(q), \tag{A8}$$

where the first correction to the energy is

$$E_1 = -\frac{1}{3}mb\omega_0\langle q^4 \rangle = -\frac{b\hbar^2}{4m\omega_0}. \tag{A9}$$

The functions $\psi_i(q)$, $i=1,2,\ldots,\infty$ verify the following relation:

$$\psi_p(q) = \psi_0(q) + \sum_{i=1}^{\infty}\psi_i(q). \tag{A10}$$

where $\psi_p(q)$ is the polarization function. The functions $\psi_i(q)$ are the terms of a polarization expansion of the solution, satisfying the required boundary conditions for $q=\pm\infty$, and they can be used to evaluate long-range polarization and even exchange effects. The interesting fact here is the proof of the unusual feature that the sums of the series diverge at infinity [except for potential $W(q)$ with $b<0$], although each individual term converges to zero in that limit. The polarization function is in this example situated outside of the Hilbert space of normalizable functions. However, in a bound region of space near the origin, the exact solutions can be used to represent the distortion of the wave function due to the long-range perturbation.

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