

# Quantum Wave Packet Dynamics: Langevin Equations for Hamiltonian Systems imbedded into a Heat Bath

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**Abstract.** In the last decade the relation between classical and quantum statistics was carefully investigated. Here we study this problem by making use of a wave packet dynamics. The theory is applied to oscillators and atomic electrons embedded into an infinite bath of oscillators.

## 1. INTRODUCTION

The generalization of classical to quantum statistics is an old but still not yet completely solved problem of physics [1, 2, 3, 4, 5]. A problem of special interest is the formulation of the second law in the quantum regime [1, 6, 7, 8]. Here we will study the behavior of atomic electrons embedded in several statistical environments in the semiclassical regime. We will use for our investigations the method of wave packet dynamics which was successfully applied to particle physics and plasma physics problems [9, 10, 11, 12, 13, 14]. Starting from the quantum dynamical variational principle one derives classical equations of motion for the parameters of a given trial wavefunction, which are Hamiltonian-like under certain conditions. Because of the resulting Hamiltonian equations, this approach is found to provide some insights into the problem of fluctuations in the quantum regime.

After a brief review of the method, we first discuss the statistical properties of a harmonic quantum oscillator within the wavepacket approach. These results are used to investigate the wavepacket dynamics of a harmonic quantum Brownian particle embedded in an oscillator bath. Based on this model we study the ionization behavior of an hydrogen like ion subject to strong fluctuations.

## 2. WAVE PACKET DYNAMICS AND THE VARIATIONAL PRINCIPLE

In the following we briefly review the method we will use through out the paper, which is based on the quantum dynamical variational principle. As proposed in [11] Gaussian wavepackets will be chosen as an approximate wavefunction. The equations of motion

are derived from the variational formulation of the Schrödinger equation

$$\delta \int_{t_1}^{t_2} \langle \psi | i\hbar \partial_t - \hat{H} | \psi \rangle dt = 0. \quad (1)$$

For special applications the exact wavefunction is approximated by a trial wavefunction  $\varphi(\mathbf{x}, t; \mathbf{q})$ , which should be adapted to the problem under consideration. The shape of the trial wavefunction is characterized by a set of  $N$  parameters  $\mathbf{q} = (q_1, q_2, \dots, q_N)$ , whose equations of motion derived from Eq. 1 are given by [11, 12]

$$\partial_t \mathbf{q} = \{A\}^{-1} \cdot \nabla_{\mathbf{q}} H, \quad A_{\mu\nu} = \frac{\partial L}{\partial \dot{q}_\nu \partial q_\mu} - \frac{\partial L}{\partial q_\nu \partial \dot{q}_\mu}, \quad (2)$$

where  $H$  is the expectation value of the Hamiltonian operator. For certain choices of the trial wavefunctions the matrix  $A$  is of canonical type and Eq. 2 turns into Hamiltonian equations, where the function  $H$  plays the role of an effective Hamiltonian. One class of wavefunctions that fulfill this condition are Gaussian wavepackets of the form

$$\varphi(\mathbf{x}; \mathbf{r}, \mathbf{p}, \beta, p_\beta) = \left( \frac{d}{2\pi\beta} \right)^{d/4} \exp \left( - \left( \frac{d}{4\beta} - ip_\beta \right) (\mathbf{x} - \mathbf{r})^2 + ip(\mathbf{x} - \mathbf{r}) \right). \quad (3)$$

Here the variables  $\mathbf{r}$ ,  $\mathbf{p}$ ,  $\beta$ , and  $p_\beta$  characterize the shape of the wavefunction according to

- $\mathbf{r}$  - average position of the particle,
- $\mathbf{p}$  - average momentum,
- $\beta$  - width of the wavepacket,
- $p_\beta$  - time variation of the width.

Thus one has to solve a set of Hamiltonian equations for  $2(d+1)$  parameters, where  $d$  is the dimension of the system. In this case the function  $H$ , which plays now the role of an effective Hamiltonian, is given by

$$H = \frac{p^2}{2m} + \frac{\hbar^2 d^2}{8\beta m} + \frac{2\hbar^2}{m} \beta p_\beta^2 + \langle \varphi | \hat{V}(\mathbf{x}) | \varphi \rangle. \quad (4)$$

Here the first three terms correspond to the kinetic energy of the particle.

Let us now discuss two special choices of the external potential, describing a one dimensional harmonic oscillator and an atomic electron in an external homogenous field.

### 3. THE HARMONIC OSCILLATOR

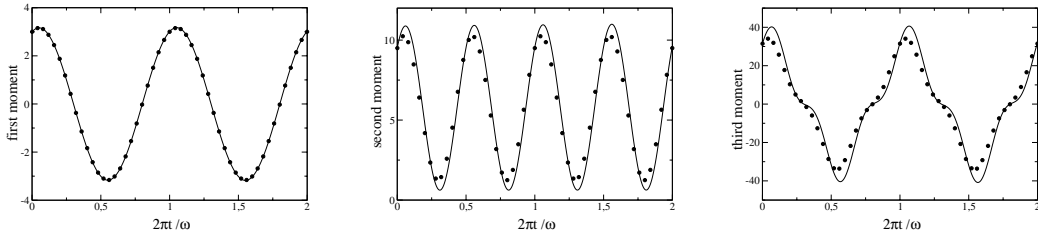
#### 3.1. Dynamics and Statistical Thermodynamics of Gaussian Wavepackets

Since the Schrödinger equation of an harmonic oscillator is separable for each of the degrees of freedom, it is sufficient to consider the case  $d = 1$ . In this case the equations

of motion for the parameters, derived from Eqs. 2 and 4, are given by

$$\begin{aligned} \dot{x} &= \frac{p}{m}, & \dot{p} &= -m\omega^2 r, \\ \dot{\beta} &= \frac{4\hbar^2}{m}\beta p_\beta, & \dot{p}_\beta &= \frac{\hbar^2}{m} \left( \frac{1}{8\beta^2} - 2p_\beta^2 \right) - \frac{1}{2}m\omega^2, \end{aligned} \quad (5)$$

with  $\omega$  being the frequency of the oscillator. In contrast to the potential presented in the following section the dynamics of the width  $\beta$  does not couple to the dynamics of the wavepacket center. The width shows an oscillating behavior, except for the fix point  $r = p = p_\beta = 0$  und  $\beta = \frac{\hbar}{2m\omega}$ , which corresponds to the exact quantum mechanical groundstate energy  $H = \frac{1}{2}\hbar\omega$ . Beside the groundstate the system Eq. 5 does not account for stable states, since the excited eigenstates can not be described with the trail function Eq.3. However, the dynamics of a wavefunction, which is of Gaussian type initially, is well described by Eq. 5, because such a function stays Gaussian under the action of the corresponding quantum mechanical time evolution operator. In Fig. 1 we compare the time evolution first three moments of the probability density calculated from the Schrödinger equation and from Eqs. 3 and 5.



**FIGURE 1.** Time evolution of the first three moments of the probability density of an initially Gaussian wavepacket moving in an harmonic oscillator. The solid lines show the dynamics calculated from Eqs. 3 and 5, whereas the points are obtained from a numerical solution of the corresponding Schrödinger equation..

After we have shown that the dynamics of a Gaussian wavepacket is well described by the present approach, let us now investigate the statistical and thermodynamical properties of the harmonic oscillator on the basis on this wavepacket treatment. Since the time evolution of the parameters is described by Hamiltonian equations one may interpret these parameters as classical phase space variables and formally introduce the density of states in parameter phase space according to

$$\rho(\Gamma) = Z^{-1} \exp(-H(\Gamma)/k_B\tau), \quad (6)$$

where the corresponding phase space partition function is given by

$$Z = \int d\Gamma e^{-H/k_B\tau}. \quad (7)$$

Here  $\tau$  is an effective temperature which characterizes the dispersion in parameter space. As we will see, in the classical high temperature limit the parameter space dispersion

and the thermodynamical temperature approach each other (up to a constant factor). However, the constant  $\tau$  can not be interpreted as the absolute temperature even in the classical limit [16] and one has to take care about the meaning of this quantity. A relation between the parameter space dispersion  $\tau$  and the absolute temperature  $T$  has to be determined from known thermodynamical quantities

$$\frac{1}{T} = \frac{\partial S}{\partial U}, \quad S = -k_B \sum_n p_n \ln p_n. \quad (8)$$

Here  $U = 2k_B\tau + \frac{1}{2}\hbar\omega$  has been derived from Eq. 7 and  $S$  is the quantum mechanical entropy, calculated from the average occupation number of the  $n$ 'th eigenstate of the oscillator

$$p_n = \left\langle \left| \langle \psi_n(x) | \varphi(x; r, p, \beta, p_\beta) \rangle \right|^2 \right\rangle = \int \rho(\Gamma) |\langle \psi_n(x) | \varphi(x; \Gamma) \rangle|^2 d\Gamma. \quad (9)$$

Evaluation of the integrals and substitution into Eq. 8 leads to the relation

$$k_B T = \frac{\hbar\omega}{\ln\left(1 + \frac{\hbar\omega}{2k_B\tau}\right)}. \quad (10)$$

which connects the parameter space dispersion with the physical temperature  $T$ . Let us introduce a "parameter-space temperature"

$$k_B \tilde{T} = 2k_B\tau + \frac{1}{2}\hbar\omega = \frac{1}{2} \coth\left(\frac{\hbar\omega}{2k_B T}\right), \quad (11)$$

so that in the classical high temperature limit both temperatures are identical. Note that only one of these temperatures can have an thermodynamical meaning, e.g. if one of the quantities  $\tilde{T}$  and  $T$  is fixed the other one depends on the eigenfrequency of the oscillator.

The temperature dependence of the average energy obtained from Eq. 7 together with Eq. 10

$$U = \frac{\hbar\omega}{2} \coth\left(\frac{\hbar\omega}{2k_B T}\right). \quad (12)$$

yields the known result for a quantum harmonic oscillator.

### 3.2. Derivation of the Langevin equation

After the preliminary study of the statistical thermodynamics of 1-d oscillators we will introduce now the quantenmechanical Langevin-Gleichung for a Brownian oscillator, on the basis of the wavepacket approach. Further we will investigate the fluctuation dissipation theorem. Here we discuss the interpretation of the two temperatures  $\tilde{T}$  and  $T$ , e.g. the relation between the basic frequency of the oscillator central  $\omega_0$  and the running frequencies of bath modes, having in mind the problems discussed earlier by Klimontovich [1].

The Hamiltonian operator for a Brownian particle embedded in bath of harmonic oscillator may be written as [4, 5]

$$\begin{aligned}\hat{H}_{tot} &= \hat{H} + \hat{H}_B + \hat{H}_I \\ &= \hat{H} + \sum_k \frac{\hat{p}_k^2}{2m_k} + \frac{1}{2} m_k \omega_k^2 \hat{q}_k^2 + \sum_k c_k \hat{q}_k \hat{x} + \frac{c_k^2}{2m_k \omega_k^2} \hat{x}^2.\end{aligned}\quad (13)$$

Here  $\hat{H}$ ,  $\hat{H}_B$  and  $\hat{H}_I$  denote the Hamilton operator of the Brownian particle, of the bath and the interaction between particles and bath. The variables  $\hat{q}_k$  and  $\hat{p}_k$  describe the dynamics of the modes of the bath and  $\hat{x}$  is the coordinate operator of the Brownian particle. As we have shown above, the dynamics of harmonic oscillators is well described by the Gaussian wavepacket approach. Assuming that all the masses are different  $m_k \neq m_l$  ( $\forall k \neq l$ ) no symmetrization of the wavefunctions is required and we may use a simple product representation of Gaussian wavepackets. This way we obtain the effective Hamiltonian

$$\begin{aligned}H_{tot} &= H + \sum_k \frac{p_k^2}{2m_k} + \frac{\hbar^2}{m_k} \left( \frac{1}{8\beta_k} + 2\beta_k p_{\beta_k} \right) \\ &\quad + \sum_k \frac{1}{2} m_k \omega_k^2 \left[ \left( x_k + \frac{c_k}{m_k \omega_k^2} x \right)^2 + \beta_k + \left( \frac{c_k}{m_k \omega_k^2} \right)^2 \beta \right],\end{aligned}\quad (14)$$

where the parameters  $x_k$ ,  $p_k$ ,  $\beta_k$  and  $p_{\beta_k}$  describe the mean position, the mean momentum and the width of the Gaussian wave packets of the bath modes. Using the effective Hamiltonian Eq. 14 one derives the following classical Langevin equation for the parameters [17]

$$\begin{aligned}\dot{x} &= p, \\ \dot{p} &= -\frac{\partial H}{\partial x} - \int_0^t \gamma(t-s) \dot{x}(s) ds + \xi(t),\end{aligned}\quad (15)$$

where

$$\begin{aligned}\xi(t) &= \sum_k c_k \left[ \left( x_k(0) + \frac{c_k}{m_k \omega_k^2} x(0) \right) \cos \omega_k t + \frac{p_k(0)}{m_k \omega_k} \sin \omega_k t \right], \\ \gamma(t) &= \sum_k \frac{c_k^2}{m_k \omega_k^2} \cos \omega_k t\end{aligned}\quad (16)$$

is a gaussian noise term and the friction kernel due to the interaction with the heat bath. The time correlation function  $\langle \xi(0) \xi(t) \rangle$ , e.g. the time correlation function of the expectation values of the fluctuation operators, has no physical relevance, because for the calculation of average values of observables, such as the energy of the Brownian particle, one needs to know the expectation value of the correlation function  $\frac{1}{2} \langle [\hat{\xi}(0) \hat{\xi}(t)]_+ \rangle$ ,

where  $[\hat{a}, \hat{b}]_+ = \hat{a}\hat{b} + \hat{b}\hat{a}$  denote the anticommutator of two operators. By using the explicit form of the Drude-Ullersma spectrum [18, 8] for the coupling to the bath modes and performing the necessary integrations we arrive at the following expression

$$\begin{aligned} \gamma(t) &= \gamma \cdot \delta(t) \\ \langle \xi(0) \xi(t) \rangle &= \frac{2\gamma\Delta}{\pi} \sum_k \left[ 2k_B\tau + \frac{\hbar}{2}\omega_k \right. \\ &\quad \left. + \frac{2\gamma\Delta}{\pi} \left( \frac{k_B\tau}{m\omega^2 + \frac{4\gamma\Delta}{\pi m_k \omega_k^2}} + \frac{\hbar}{\sqrt{4m^2\omega^2 + \frac{16\gamma\Delta}{\pi m_k \omega_k^2}}} \right) \right] \cos \omega_k t \\ &= \frac{2\gamma}{\pi} \int \left( 2k_B\tau + \frac{1}{2}\hbar\omega \right) \cos \omega t d\omega = \frac{2\gamma}{\pi} \int 2k_B\tilde{T} \cos \omega t d\omega, \quad (17) \end{aligned}$$

where  $\tilde{T}$  is related to the temperature  $T$  according to Eq. 11. The question is now, how to interpret these two temperatures. In the last section we have called  $\tilde{T}$  the parameter space temperature and  $T$  the physical temperature defined through the quantum mechanical von Neumann entropy. Therefore, one should demand thermal equilibrium with respect to the temperature  $T$ . This leads to a frequency dependent  $\tilde{T}$  and thus we arrive at the known Nyquist form of the fluctuation-dissipation theorem (FDT)

$$\langle \xi \xi \rangle_\omega = \gamma \hbar \omega \coth \left( \frac{\hbar \omega}{2k_B T} \right). \quad (18)$$

In fact, this form of the FDT contradicts the second law of thermodynamics, since the average energy calculated with the help of Eq. 18 depends on the kinetic coefficient  $\gamma$ . This was pointed out in [6] and investigated later [7, 8].

The assumption of different parameter space temperatures for each oscillator is justified if there is no interaction between them, which is the case if the influence of the Brownian particle on the bath modes can be neglected. But if there is a small nonlinear interaction between the bath oscillators or if the indirect coupling between the bath modes due to the term  $c_k x_k x/2$  appearing in Eq. 14 has to be taken into account, the assumption of different parameter space temperatures breaks down. In that case it is not clear how to treat the two temperatures  $\tilde{T}$  and  $T$ .

Let us point out here a connection between earlier work of Klimontovich [6] related to this problem. We may demand that the equilibrium must be defined in parameter space due to the interaction term in Eq. 14. If the Brownian particle is also in equilibrium with the bath we can relate the parameter space temperature with the system temperatures via the properties of the central oscillator and arrive at the FDT

$$\langle \xi \xi \rangle_\omega = \gamma \hbar \omega_0 \coth \left( \frac{\hbar \omega_0}{2k_B T} \right), \quad (19)$$

which was postulated by Klimontovich and shown to explain some results for the Lamb shift known from quantum electrodynamics [6]. Obviously, this form of the FDT does

not contradict the second law of thermodynamics. In the small coupling limit Eq. 18 goes over into Eq. 19 [6, 8].

This way, we may state here that in dependence on the context, the present wavepacket approach leads either to the Nyquist form or to the Klimontovich form of the FDT. However, the debate for the right treatment of this problem is not settled down yet.

## 4. DYNAMICS OF ATOMIC ELECTRONS

### 4.1. Atom subject to a strong periodic field

We start by studying an atom in vacuum under the influence of an external periodic field. According to the formalism described in section 2 the effective Hamiltonian for an atomic electron subject to a linearly polarized laser pulse is given by (atomic units are used, if not stated otherwise)

$$H = \frac{p^2}{2} + \frac{9}{8\beta} + 2\beta p_\beta^2 - \frac{1}{r} \operatorname{erf} \left( \sqrt{\frac{3}{2\beta}} r \right) + \mathcal{E}(t) \mathbf{E} r \cos(\omega t), \quad (20)$$

where  $\mathcal{E}(t) < 1$  describes the temporary shape of the pulse. The fourth term in Eq. 20 corresponds to an effective Coulomb potential seen by the electronic wavepacket. Due to the absence of the Coulomb singularity the effective Hamiltonian provides one stable stationary point in the case of an static external field ( $\omega = 0$ ,  $\mathcal{E}(t) = 1$ )

$$\begin{aligned} \mathbf{r}_0 \cdot \mathbf{E} &= -\alpha \cdot E + O(E^2), \quad \mathbf{r}_0 \times \mathbf{E} = 0, \quad \mathbf{p}_0 = \mathbf{0} \\ \beta_0 &= \frac{27}{32Z^2} \pi + \frac{\pi^4}{2} \left( \frac{27}{32Z^2} \right)^3 E^2 + O(E^4), \quad p_{\beta_0} = 0. \end{aligned} \quad (21)$$

As can be seen in table 1, the groundstate energy and the polarizability of the model atom determined from Eqs. 20 and 21 shows reasonable agreement with the quantum mechanical values.

**TABLE 1.** Comparison of the groundstate energy and the polarizability of a hydrogen-like ion calculated quantum mechanically and from Eqs. 20 and 21.

	wavepacket model	quantum result	discrepancy
groundstate energy	-0.424 <i>a.u.</i>	-0.5 <i>a.u.</i>	15%
polarizability	3.2 <i>a.u.</i>	4.5 <i>a.u.</i>	29%

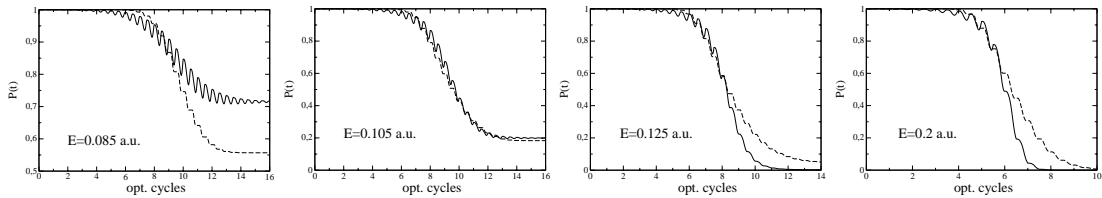
After having assure good static properties of our model atom, let us now discuss the dynamical behavior in a strong laser pulse, especially the ionization process. In dependence on the laser parameters  $E_0 = |\mathbf{E}_0|$  and  $\omega$  there are different channels for the electron to leave the core, multi-photon ionization [19], quasistatic tunneling [20] and barrier supression ionization (BSI) [21]. Obviously, neither tunneling nor photon

absorption can be described within the wavepacket model discussed above, since neither discrete transitions between certain states nor a splitting of the wavefunction is included in the used ansatz of a single Gaussian wavepacket. However, for sufficiently large field strength, the Coulomb barrier may be suppressed below the ionization threshold, so that the electron is able to escape over this barrier. In this barrier suppression ionization (BSI) regime, classical and semiclassical pictures have been found to work well. In [14] we have shown that the discussed model provides a reasonable extension to the case of three dimensional Coulomb potentials of the so called "wavepacket spreading model" which has been successfully applied to BSI in an one dimensional screened softcore potential [22].

If the field is strong enough so that nearly all excited states are destroyed the ionization probability can be well approximated by the groundstate population according to  $P_I(t) = 1 - P_G(t)$ , which is obtained from the overlap of the initial groundstate wavepacket (Eq. 21) and the time dependent wavefunction

$$P_G = \left( \frac{36\beta_0\beta}{D} \right) \exp \left( -\frac{3}{2D} \left[ (9B + p_\beta C) r^2 + 4\beta_0\beta B p^2 - C \mathbf{pr} \right] \right), \quad (22)$$

where  $B = \beta + \beta_0$ ,  $C = 16p_\beta\beta_0\beta^2$ ,  $D = 9B^2 + p_\beta\beta_0C$  and  $\beta_0$  is given by Eq. 21. In Fig. 2 we compare the calculated ionization probability as a function of time for a frequency of  $\omega = 0.1 \text{ a.u.}$ , a Gaussian pulse shape containing 20 laser cycles and different field amplitudes and compare this result with a semiempirical rate formula which has been obtained by a fit to numerical ab initio solutions of the full quantum problem [21]. For low field strength tunneling is the dominant process and thus the curve calculated from the wavepacket dynamics decreases slowly. For field strength larger than the BSI threshold the agreement rapidly becomes better since the tunneling does not contribute in that case. With increasing field strength both curves start to deviate at larger times, because the long distance behavior of the true groundstate wavefunction is not exactly described by the Gaussian ansatz.



**FIGURE 2.** Groundstate population as a function of time an external frequency of  $\omega = 0.1 \text{ a.u.}$ , a pulse length of  $N = 20$  and different field strength. The solid line shows the results of the wavepacket dynamics together with Eq. 22 and the dashed line shows the population calculated from the ionization rate proposed in [21].

However, we may conclude that the dynamics of barrier suppression ionization caused by a time dependent and space independent external force is adequately described by the present wavepacket formalism.

## 4.2. Dissipative Dynamics - Ionization due a surrounding harmonic bath

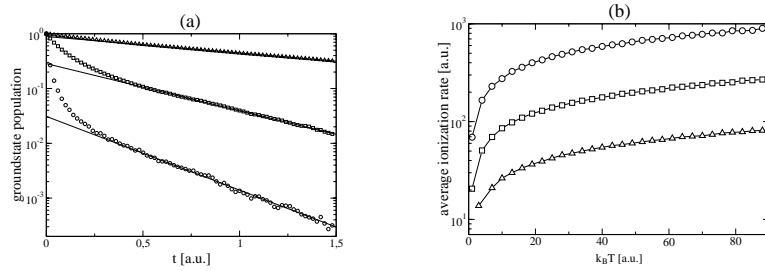
Since, the fluctuating force acting on an atomic electron due to a surrounding harmonic bath is of the type discussed in the previous section, we can expect a good description of the BSI-process in the framework of the wavepacket approach. According to section 3.2 the equations of motion for the wavepacket parameters read

$$\begin{aligned}\dot{\mathbf{r}} &= \mathbf{p}, & \dot{\mathbf{p}} &= \nabla_{\mathbf{r}} \left[ \frac{Z}{r} \operatorname{erf} \left( \sqrt{\frac{3}{2\beta}} r \right) \right] - \gamma \mathbf{p} + \xi(t), \\ \dot{\beta} &= 4p_{\beta} \beta, & \dot{p}_{\beta} &= \frac{9}{8\beta^2} - 2p_{\beta}^2 - Z \sqrt{\frac{3}{2\pi\beta^3}} \exp \left( -\frac{3r^2}{2\beta} \right).\end{aligned}\quad (23)$$

Since the BSI picture holds only in the case of strong external perturbations and a fast release of the electron wavepacket, we may restrict ourselves to the high temperature limit, where the spectrum of the fluctuations becomes

$$\langle \xi \xi \rangle_{\omega} = 2\gamma k_B T. \quad (24)$$

Analogous to the approach in the previous section we calculate the ionization probability as a function of time simple from the overlapp Eq. 22, where the parameters are now evolved by solving Eq. 23. The average ionization probability is obtained by averaging over an ensemble of 100000 trajectories. In Fig. 3a we show the resulting ionization probability for a fixed temperature and two different friction constants. As can be seen, after a short equilibration the system settles into an exponential decay region. From the slope of those curves we calculate the corresponding ionization rate, which is shown in Fig. 3b. Note that counterintuitively the ionization rate increases with a increasing damping constant.



**FIGURE 3.** Groundstate population as a function of time for  $k_B T = 20 a.u.$  (a) and fitted ionization rate (b) at three different damping constants:  $\gamma = 1 a.u.$  (circles),  $\gamma = 0.1 a.u.$  (squares) and  $\gamma = 0.01 a.u.$  (triangles).

## 5. CONCLUSIONS

In conclusion we have shown that the problem of quantum brownian motion in a harmonic oscillator can be treated in the framework of a wavepacket approach [9, 10, 11,

12, 13, 14]. The particle dynamics is described by classical statistics in parameter space of the used trial wavefunction. There appear two different temperatures  $\tilde{T}$  and  $T$  living in parameter space or in the Hilbert space of the wavefunction, respectively. In dependence on the temperature, chosen to define equilibrium, we arrive either at the Nyquist form or the Klimontovich form of the fluctuation-dissipation theorem.

As a second application of the wavepacket approach the semiclassical ionization behavior of a hydrogen like ion embedded into a heat bath has been studied. The calculated ionization rate is found to increase with both increasing temperature and damping constant.

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