Variational procedure for time-dependent processes

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A simple variational Lagrangian is proposed for the time development of an arbitrary density matrix, employing the ''factorization'' of the density. Only the ''kinetic energy'' appears in the Lagrangian. The formalism applies to pure and mixed state cases, the Navier-Stokes equations of hydrodynamics, transport theory, etc. It recaptures the least dissipation function condition of Rayleigh-Onsager and in practical applications is flexible. The variational proposal is tested on a two-level system interacting that is subject, in one instance, to an interaction with a single oscillator and, in another, that evolves in a dissipative mode.

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I. INTRODUCTION

Several basic aspects of stochastic dynamics remain controversial. (A critical ''state of art'' update in Ref. [1] shows this.) This situation contrasts with most physical theories, where the problems that arise are in the application of consensually accepted principles. It can perhaps be argued that the lack of an agreed-upon variational formulation of stochastic processes is at the root of the problem. As a remedy of this situation, this paper suggests a variational functional which is to be minimized and whose minimum is the true density matrix.

To be sure, in the past several principles of extrema have been proposed; these include Gauss's least constraint principle [2,3], the ''least dissipation function'' [4–6], minimum entropy production rate for steady states [7,8] (see also Ref. [9] for its violation), minimal energy generation rate [10], minimal scattering integral [11–13], least velocity error during pathway [3], the Yasue action for stochastic mechanics [14], a formulation involving a potential [15], and again, recently, maximum entropy production [16]. To these may be added several variational methods applicable to classical (i.e., not quantal) systems, such as those appropriate for general nonlinear problems [17], the ''governing principle for dissipative processes'' [18,19] and a generalized Hamiltonian principle [20]. Reviews of these and of other methods can be found in Refs. [21–24].

The present proposal for a variational procedure is based on the following new elements: (a) the factorization (to be discussed later in this paper) of the density matrix as introduced by Reznik [25] and utilized recently by Gheorghiu-Svirschevski [16], (b) a conventional Lagrangian similar to that used in mechanics to obtain the motion of a point particle subject to an external force, but in which the scalar potential is either absent or ignorable, (c) a vector potential that can be singular, without this having disastrous observational consequences, and (d) an appropriate use of minimization procedure, with origins going back to Gibbs [2]. The method covers a broad range of behaviors (''deterministic'' and stochastic, quantal and classical, electronic transport, discrete and continuous, Markovian and otherwise) and places in a new perspective certain aspects in currently employed theories of stochastic dynamics. Apart from these favorable (and *a priori* unexpected) features, a number of problems remain, which will have to be resolved by future efforts.

A pure state subcase (as opposed to a density matrix that can describe a statistical mixture of states) is the subject of a numerically worked out example in Sec. IV A and is equivalent to the (linear) time-dependent Schrödinger equation. For this several variational formulations are known in the literature [26–31]. The inter-relation between these was investigated in Ref. [30], where they were shown to be frequently equivalent. For the pure state case our density matrix variational method reduces to the McLachlan formalism [27], in which the variation of the function is carried out with respect to the time derivative only (while the function is kept fixed). Moreover, we give a justification of this procedure for stochastic processes.

Another application of the variational formalism, in Sec. IV B, includes a nonlinear, dissipative (non-Hermitian) mechanism and exemplifies quantum jumps.

II. THE VARIATION

The ''factorized'' form of the time-dependent density matrix ρ [16,25] reads in terms of the column and row vectors $\boldsymbol{\gamma}$ and (its Hermitian conjugate) $\boldsymbol{\gamma}^\dagger$,

$$\rho = \boldsymbol{\gamma}\boldsymbol{\gamma}^\dagger. \tag{1}$$

The above condensed notation is not trivially simple, so we give in Appendix A a ''tutorial'' on the notation. We now propose an action $S(T)$ (expressed in arbitrary units) this being the integral over time t with an arbitrary time end point T of a Lagrangian $\mathcal{L}$. It is the form of the Lagrangian that we seek: we propose that it has the ''quadratic'' form, as follows:

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$$S(T)=\int_0^T dt\mathcal{L}(t)=\int_0^T dt\mathrm{Tr}(i\dot{\gamma}^\dagger-\mathbf{A}^\dagger)\cdot(-i\dot{\gamma}-\mathbf{A}). \quad (2)$$

The variational equations based on the above action (these are the equations of motions for γ and $\gamma^\dagger$) are given below in Eq. (7) . Tr is the trace over all ''components'' of γ explained in Appendix A. Dots above symbols represent differentiation with time.

The variational equations are obtained in the usual way by varying the action $S(T)$ with respect to all components of the two vectors γ and $\gamma^\dagger$. Thus

$$\delta_\gamma S(T)=\int_0^T dt\,\delta\gamma(t)\left[\frac{\delta\mathcal{L}(t)}{\delta\gamma}-\frac{\partial}{\partial t}\frac{\delta\mathcal{L}(t)}{\delta\dot{\gamma}}\right]+\frac{\delta\mathcal{L}(T)}{\delta\dot{\gamma}}\delta\gamma(T) \quad (3)$$

and a similar expression for $\delta_{\gamma^\dagger}S(T)$. The last term, outside the integral, is the boundary term. It is assumed that the vectors γ are fixed initially at $t=0$, but not at any time later. (This will be discussed shortly.) If the boundary term can be made to vanish, so will be the whole variation, since in the absence of a scalar potential, the variations $\delta\mathcal{L}(t)/\delta\gamma$ and $\delta\mathcal{L}(t)/\delta\gamma^\dagger$ vanish. This follows, since for the postulated form of the Lagrangian these variations contain as a factor one or the other of the expressions $\delta\mathcal{L}(t)/\delta\dot{\gamma}$ and $\delta\mathcal{L}(t)/\delta\dot{\gamma}^\dagger$ and these vanish due to the boundary variation. (In fact, $\delta\mathcal{L}(t)/\delta\gamma$ and $\delta\mathcal{L}(t)/\delta\gamma^\dagger$ also vanish due to the presence of the same factors, but these variations do not form a sufficiently general basis for the variation procedure, since the vector potential $\mathbf{A}$ may not be a function of *all* components of the γ vector.)

At this point the role of the boundary term is well worth reflecting upon. It is not present in, e.g., deterministic mechanics, where the values of the variables are fixed at both end points. However, it is well known that the boundary term arises when the value of the variant quantity, i.e., γ, is undetermined at a boundary. This is (physically) the case when a random force operates on the system. Thus, we are not allowed to neglect the boundary term. It is now a further ''bonus'' in the formalism that the vanishing of the boundary-term variation does not interfere with the vanishing of the body-term variation (i.e., it is neither contradictory to it nor incomplete with respect to it), but is by virtue of our choice of the Lagrangian precisely identical with it.

In summary, we have the variational equations

$$\frac{\delta\mathcal{L}(t)}{\delta\dot{\gamma}}=0 \quad (4)$$

and their complex conjugates

$$\frac{\delta\mathcal{L}(t)}{\delta\dot{\gamma}^\dagger}=0 \quad (5)$$

and these make the action extremal (and an absolute minimum) also when the ''vector potential''[1] $\mathbf{A}$ is a function of γ [32]. [To see this, note the remark after Eq. (7) .] This has the immediate consequence that in the expansion of the Lagrangian, shown in Eq. (2), the following expression (dissipation function) needs to be minimized:

$$F_D=\dot{\gamma}^\dagger\dot{\gamma}-i[\dot{\gamma}^\dagger\mathbf{A}-\dot{\gamma}\mathbf{A}^\dagger]. \quad (6)$$

This follows, since the $\mathbf{A}\cdot\mathbf{A}^\dagger$ term is independent of time derivatives and is not varied. We further note that (by the form of the vector potential) i times the square bracket is a real quantity. The quantity in the above equation is essentially of the form of Onsager and Machlup's dissipation function (the negative of Eqs. (4)–(25) in Ref. [5]. One recalls that their dissipation function is also minimized only with respect to the time derivatives of the variables just as in the procedure proposed in Ref. [27] and in the present work). To bring our ''DF'' precisely to the form of the dissipation function, we need to go from our variables γ and $\gamma^\dagger$ by a constant linear transformation (not necessarily a unitary one) to the variables α of Ref. [5]. One will then get, instead of the diagonal form $\dot{\gamma}^\dagger\dot{\gamma}$, a nondiagonal form which defines the ''generalized resistance matrix'' R_{ij} of Refs. [3,6]. [Since the use of extensive variables is to be preferred to intensive ones (and γ and $\gamma^\dagger$ are of the latter type) the transformation should include the system size. Alternatively, the action integral may be premultiplied by a size-dependent scale factor.]

Thus, the result of the variation are a set of equations

$$\dot{\gamma}=i\mathbf{A},$$

$$\dot{\gamma}^\dagger=-i\mathbf{A}^\dagger. \quad (7)$$

When $\mathbf{A}$ and $\mathbf{A}^\dagger$ are functions of γ and $\gamma^\dagger$, their (nonzero) derivatives come in with either $\dot{\gamma}-i\mathbf{A}$ or $\dot{\gamma}^\dagger+i\mathbf{A}^\dagger$ as factors and these factors vanish due to the above equations.

[We can illustrate this in the case of two components, for which we write the Lagrangian in Eq. (1) as

$$\mathcal{L}(t)=f_1^\dagger f_1+f_2^\dagger f_2 \quad (8)$$

and $f_1=-i\dot{\gamma}_1-\mathbf{A}_1(\gamma_k,\gamma_k^\dagger)$, etc., with the γ dependence explicitly put in A's. Recall that Eq. (7) means that for the variational solution $f_1=f_1^\dagger=f_2=f_2^\dagger=0$ at all times and that with this solution the minimal action is zero.

Upon varying with respect to, e.g., γ_1, the resulting Lagrange-Euler equations are now in full detail

$$-(\partial_{\gamma_1}A_1^\dagger)f_1+i\dot{f}_1^\dagger-f_1^\dagger(\partial_{\gamma_1}A_1)-(\partial_{\gamma_1}A_2^\dagger)f_2-f_2^\dagger(\partial_{\gamma_1}A_2)=0. \quad (9)$$

[1]We have named our frequently used quantity $\mathbf{A}$ the ''vector potential,'' in analogy with the quantity that enters as a cross term with the time derivative (''the particle velocity'') in the Lagrangian of classical mechanics [32], or inside the square with the canonical momentum and in distinction from the scalar potential ϕ, which appears in (nonrelativistic) Lagrangians as a separate term.

Since all the terms contain one of the f factors or their time derivatives (which are necessarily also zero) the above equation is satisfied for the proposed variational solution. There may be other solutions, though. If for these not all f's are zero (and therefore differ from the proposed solution), than the action (which consists of positive terms) is positive and larger than that for the solution given in Eq. (7).]

These are the equations of motion of the (independent) vector variables and can be regarded as having the status of the Langevin equations or Hamilton's equations for the set of conjugate variables γ_a and $\gamma_a^\dagger$. The processes considered in this section comprise a purely Hamiltonian process A_H (namely, energy preserving, ''elastic'') as well as some other, dissipative mechanisms. Thus, a Markovian scattering process represented by the symbol M_{ba} (designating half the probability per unit time of a scattering event taking the system from state a to b) can be written as (A_{out},A_{in}) to separate scattering out of and into a given state. We also add a stochastic, random process arising from, e.g., an external source, as A_r. Some other type of processes will be considered below in Sec. V.

For the first two processes we have

$$A_H = -H\gamma,$$

$$A_H^\dagger = -\gamma^\dagger H^\dagger = -\gamma^\dagger H,$$

$$(A_{out})_a = i\sum_b M_{ba}\gamma_a,$$

$$(A_{out}^\dagger)_a = -i\gamma_a^\dagger \sum_b M_{ba},$$

$$(A_{in})_a = -i\frac{1}{N}\left(\sum_b M_{ab}\gamma_b\gamma_b^\dagger\right)(\gamma_a^\dagger)^{-1},$$

$$(A_{in}^\dagger)_a = i(\gamma_a)^{-1}\frac{1}{N}\left(\sum_b M_{ab}\gamma_b\gamma_b^\dagger\right). \tag{10}$$

When we add to these the random force, we obtain in addition

$$(A_r)_a = -if_a\gamma_a, \quad (A_r^\dagger)_a = i\gamma_a f_a. \tag{11}$$

f_a represent the components of the random time-dependent force with zero mean and a finite self-correlation. (N is the number of states in the ensemble, see Appendix A.)

The vector potential $\mathbf{A_{in}}$ is singular. However, singularities in vector potentials are well known (as, e.g., in those for solenoidal or monopole fields). To cancel these singularities we shall follow the procedure of Reznik [25] and Gheorghiu-Svirschevski [16], who multiply $\boldsymbol{\gamma}^\dagger$ into $\dot{\gamma}$, $\boldsymbol{\gamma}$ into $\dot{\gamma}^\dagger$, add, and obtain the (master) equations for the density matrix.

After substituting the quantities from Eqs. (7), (10), and (11) we obtain by this procedure for a diagonal element (say) aa of the density matrix

$$\dot{\rho}_{aa} = (\dot{\gamma}\boldsymbol{\gamma}^\dagger + \boldsymbol{\gamma}\dot{\gamma}^\dagger)_{aa}$$

$$= i([\rho,H])_{aa} - 2\rho_{aa}\sum_b M_{ba} + 2\sum_b M_{ab}\rho_{bb} + 2\rho_{aa}f_a. \tag{12}$$

When one writes out the equation, similar to Eq. (12) , for the time derivatives of the off-diagonal matrix elements $\rho_{ab}(a\neq b)$, one finds singularities in them, due to the above mentioned singularities in the vector potentials. For a macroscopic system these singularities cancel, when one takes into account the phase decoherence between different states of macroscopic bodies. (The subject of microscopic to macroscopic transitions does not belong here. It was studied by various methods and has summaries in, e.g., Refs. [33,34].)

III. POTENTIAL FLUID DYNAMICS

An interesting application of the preceding complex factor-density formalism is for the well known potential flow (namely, fluid dynamics without vorticity) as presented in many hydrodynamic text books, e.g., Ref. [35]. If a flow satisfies the condition of zero vorticity, i.e., the velocity field $\vec{v}$ is such that $\vec{\nabla}\times\vec{v}=\vec{0}$, then there exists a function ϕ satisfying $\vec{v}=\vec{\nabla}\phi$.

In this section we answer the question, what form of the vector potential **A** appearing in Eq. (2) will ensure that upon variation of the action containing these vector potentials we shall obtain precisely the well known equations of potential flow hydrodynamics. These equations are

$$\frac{\partial\rho}{\partial t} + \vec{\nabla}\cdot(\rho\vec{\nabla}\phi) = 0, \tag{13}$$

$$\frac{\partial\phi}{\partial t} = -\frac{1}{2}(\vec{\nabla}\phi)^2 - h - \Phi - \nu\nabla^2\phi. \tag{14}$$

In these equations the physical meaning of the quantities is that ρ is the mass density, h is the specific enthalpy, ν is the viscosity coefficient, and Φ is some function representing the potential of an external force acting on the fluid. The first of these equations is the continuity equation, while the second is a modified Bernoulli's equation which takes into account some viscous effects. (A full viscous flow is of course not a potential flow and contains vorticity.) ρ and ϕ play the roles of the squared amplitude and of the phase angle, respectively. Both are real quantities.

The final results for the desired vector potentials and their complex conjugates are shown, below, in Eqs. (23) and (24). To obtain them, we first express the variational variables γ and $\gamma^\dagger$ that we have used so far in terms of the physical variables (ρ,ϕ). The variation will now be carried out with respect to the latter variables. The transformation is

$$\gamma = \sqrt{\rho}e^{i\phi}, \tag{15}$$

$$\gamma^\dagger = \sqrt{\rho}e^{-i\phi}. \tag{16}$$

Though all variables are now functions of the positions, and are thus continuous variables, we shall label them, as before, by the subscripts a, etc. The following relations (with no summations over repeated symbols) arise simply from the inverse transformation:

$$\dot{\phi}_a=(\dot{\gamma}_a\gamma^{\dagger}{}_a-\gamma_a\dot{\gamma}_a^{\dagger})/2i\rho_{aa}, \tag{17}$$

$$\vec{\nabla}\phi_a=(\vec{\nabla}\gamma_a\gamma^{\dagger}{}_a-\gamma_a\vec{\nabla}\gamma^{\dagger}{}_a)/2i\rho_{aa}, \tag{18}$$

$$\nabla^2\phi_a=(\nabla^2\gamma_a\gamma^{\dagger}{}_a-\gamma_a\nabla^2\gamma^{\dagger}{}_a)/2i\rho_{aa}, \\ -[(\vec{\nabla}\gamma_a\gamma^{\dagger}{}_a)^2-(\gamma_a\vec{\nabla}\gamma^{\dagger}{}_a)^2]/2i\rho_{aa}^2, \tag{19}$$

$$\vec{\nabla}\rho_{aa}=(\vec{\nabla}\gamma_a\gamma^{\dagger}{}_a+\gamma_a\vec{\nabla}\gamma^{\dagger}{}_a). \tag{20}$$

Using these, we first rewrite Eq. (13) as

$$\dot{\rho}_{aa}=(\dot{\gamma}_a\gamma^{\dagger}{}_a+\gamma_a\dot{\gamma}_a^{\dagger}) \\ =-\rho_{aa}\nabla^2\phi_a-\vec{\nabla}\rho_{aa}.\vec{\nabla}\phi_a\equiv R_a(\gamma_a,\gamma^{\dagger}{}_a). \tag{21}$$

R_a being a well-defined, real function of the variational variables and of their first and second spatial derivatives (but independent of the time derivatives). Likewise, one obtains the rate equation for the phase, Eq. (14) , as

$$\dot{\phi}_a=-(\dot{\gamma}_a\gamma^{\dagger}{}_a-\gamma_a\dot{\gamma}_a^{\dagger})/2i\rho_{aa} \\ =-\frac{1}{2}(\vec{\nabla}\phi)^2-h-\Phi-\nu\nabla^2\phi\equiv N_a(\gamma_a,\gamma^{\dagger}{}_a) \tag{22}$$

in which N_a has properties similar to R_a [36].

We can solve for the two quantities, $\dot{\gamma}_a\gamma^{\dagger}{}_a$ and $\gamma_a\dot{\gamma}_a^{\dagger}$ from the preceding two equations, and then divide by $\gamma^{\dagger}{}_a$ and γ_a, respectively, to obtain the time derivatives. However, by Eq. (2) the time derivatives are just the vector potentials. Thus we finally obtain

$$\dot{\gamma}_a=(R_a+2i\rho_{aa}N_a)\frac{1}{2\gamma^{\dagger}{}_a}=iA_a \tag{23}$$

and the complex conjugates

$$\dot{\gamma}_a^{\dagger}=\frac{1}{2\gamma_a}(R_a-2i\rho_{aa}N_a)=-iA_a^{\dagger}. \tag{24}$$

We have thus found the vector potentials which have to be inserted in the action, so as to yield variationally the hydrodynamic equations, Eqs. (13) and (14). It is evident that the complex representation is a natural way to obtain variationally equations of motion for two such dissimilar quantities as amplitude and velocity. The physical extrema are certainly global minima (although the functional may have additional minima). Detailed applications will be undertaken in the future including the problem of a general viscous flow.

IV. APPLICATIONS

A. A periodically varying Hamiltonian

We shall now apply the proposed variational procedure to yield, in one case exactly and in another case approximately, the solution for a (Hermitian) Hamiltonian that has a periodic variation. The cases chosen are such that analytical solutions are known exactly ([37–39]), so that we can compare to them the variational solutions to be obtained here. Specifically, we consider the time development of a doublet subject to a Schrödinger equation whose Hamiltonian in a doublet representation is

$$H(t)=(G/2)\begin{pmatrix}-\cos(\omega t) & \sin(\omega t)\\ \sin(\omega t) & \cos(\omega t)\end{pmatrix}. \tag{25}$$

Here ω is the angular frequency of an external disturbance. The eigenvalues of Eq. (25) are $G/2$ and $-G/2$. If $G>0$, then in the ground state the amplitude in the upper component $\binom{1}{0}$ in Eq. (25) is

$$C_g^u=\cos(Kt)\cos(\omega t/2)+(\omega/2K)\sin(Kt)\sin(\omega t/2) \\ +i(G/2K)\sin(Kt)\cos(\omega t/2), \tag{26}$$

with

$$K=0.5\sqrt{G^2+\omega^2}. \tag{27}$$

The amplitude C_g^l of lower component $\binom{0}{1}$ in the ground state has a similar form, which we shall not bother to write out. For the variational procedure we postulate a superposition of complex circular functions

$$C_g^u=\sum_{m=-M,M+1}A_m^u\exp^{i(m-1/2)\omega t}, \tag{28}$$

and similarly for C_g^l. The complex coefficients A_m^u and A_m^l are determined by minimization of the variational action, Eq. (2), subject to the normalization condition that the sum of the absolute squares of the coefficients is unity. m takes integral values between the limits and we have taken for our trial functions $M=2$, that is, six terms in each component. The half integer in the exponent is suggested by the acquisition of a Berry phase after a full period $2\pi/\omega$. For the same reason we have taken the range of the integration in Eq. (2) to be twice the period. So as to create realistic conditions for the implementation of the variational procedure, we have chosen a *finite* range for the time variable, although, as can be seen from the formulas in Eqs. (26) and (27), for a general value of G the solution is not time periodic. We illustrate the procedure for two cases.

1. A periodic case

This comes about when G is such that the Rabi frequency K in Eq. (27) and ω are commensurate. We have chosen $G=\sqrt{15}\omega$, so that $K=2\omega$.

Minimizing the action with respect to the coefficients subject to the normalizing conditions turns out to be equivalent to diagonalizing a 12×12 matrix, whose elements are the

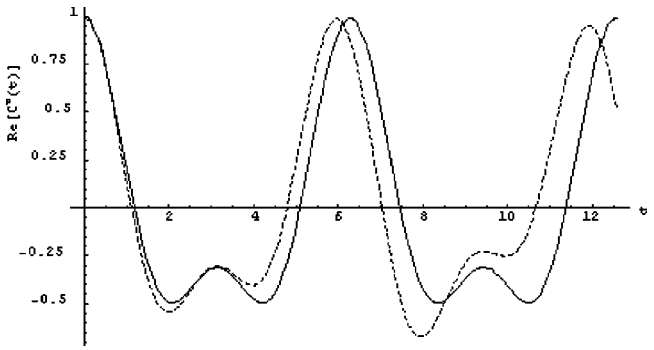

FIG. 1. Time dependence of the real part of the amplitude in the upper electronic component state. Parameter values $G=3$, $\omega=1$. Full line—variationally obtained state. Broken line—the exact, algebraic solution.

action integral computed with the circular function shown in Eq. (28) . There are 12×12 matrix elements, rather than just 6×6, since the upper and lower states are coupled by the off-diagonal terms in Eq. (25) .

We find a pair of zero eigenvalues (actually eigenvalues of about 10^{-15}), whose meaning is the value of the action integral in the transformed representation; in other words, our variational solution is exact. Also, the numerical values of the variationally obtained coefficients A_g^u agree with those in the analytic solution in Eq. (26) . For comparison, the other eigensolutions have ''eigenvalues'' of orders 1–20.

2. A nonperiodic case

With the choice of, e.g., $G=3$ and $K=\sqrt{10}/2$, the analytic solution shown in Eq. (26) is not periodic and an exact solution cannot be achieved variationally while having a finite t range in the action integral. Moreover, a larger spread of the basic set in Eq. (28) is needed. Still, so as to estimate the efficacy of the variational procedure under nonoptimal conditions, we used the same t range and the same set size as in (a). The lowest eigenvalues (=the values of the action integral) are about 0.35, compared with others eigenvalues, which are again in the range of 1–20.

The results are also shown graphically, by comparing the variational solution (full lines) with the exact, analytic solution (broken lines) in Eq. (26) for real and imaginary parts in Figs. 1 and 2, respectively. The similarities are quite good for

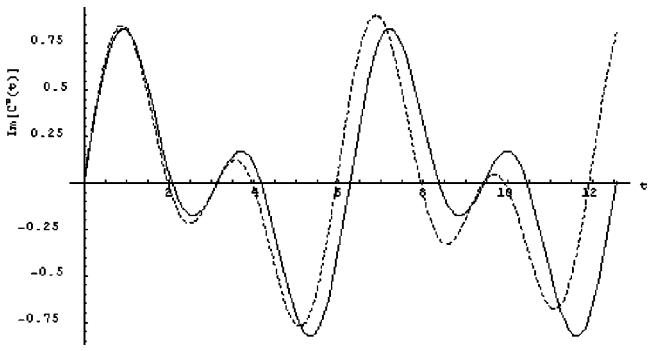

FIG. 2. Same as in the previous figure but for the imaginary part of the amplitudes.

the first half (which comprises the period of the Hamiltonian), but is worse in the second half and further deteriorates later, say in the time range $(4\pi/\omega,6\pi/\omega)$. On the other hand, had we taken the time range of the variational procedure [the upper limit of integration T in Eq. (2)] up to $6\pi/\omega$, we would have obviously got a somewhat different solution, which would have improved the approximation in the latter range and spoiled somewhat the agreement in the earlier range. In general, the approximate solution depends on the time range of the action. In our view, this endows a flexibility to the practical application of the method, in the sense that, depending on which time range is of interest, better approximation can be achieved for that range. Of course, when the ''approximate'' solution is identical with the true solution in some range, it will remain so, by analytical continuation, for all ranges.

In conclusion, one notes a successful application of the variational principle for a purely Hermitian case, whose solution, though available by algebra, is not trivial.

B. A nonlinear evolution

The continuous passage of an initially prepared pure state to transitions resembling quantum jumps was recently studied in Ref. [40], based on a form of the Liouville–von Neumann–Lindblad (LvNL) equation. The actual form used originated in a representation of fast level crossing in molecular systems involving two states [41]. It was noted in Ref. [40] that the factorization formalism, called the ''square root operator'' method of Ref. [25], represents an alternative way of showing how a dissipative term in the Hamiltonian can cause decoherence. To apply our variational formalism to this case, we first formulate the evolution equations in the factorization scheme and solve the resulting equations [this is done in Appendix A below] and, second, we obtain an approximation to the solution by minimizing the action with respect to some parameters appearing in the assumed γ's [this is carried out in in Appendix B].

1. Decoherence by the square root operator method

The vector potentials consisting of a Hamiltonian and a dissipative (non-Hermitian) part are now written for the two factors (γ_1,γ_2) of the density matrix as

$$\mathbf{A}_1=-\frac{1}{2}G\cos(\omega t)\gamma_1-J\gamma_2 - i\Gamma[\gamma_1-|\gamma_2|^2(1+\mu|\gamma_1|^2)/\gamma_1^\dagger,$$

$$\mathbf{A}_2=\frac{1}{2}G\cos(\omega t)\gamma_2-J\gamma_1-i\Gamma[\gamma_2-|\gamma_1|^2(1-\mu|\gamma_2|^2)/\gamma_2^\dagger. \tag{29}$$

One notices the similarity of these expressions with the corresponding formulas in Refs. [40,41] (where the interpretation of the terms is spelt out) and also the divisor γ on the extreme right, characteristic of the factorization formalism for dissipative processes, Eq. (10) . The trace of the density

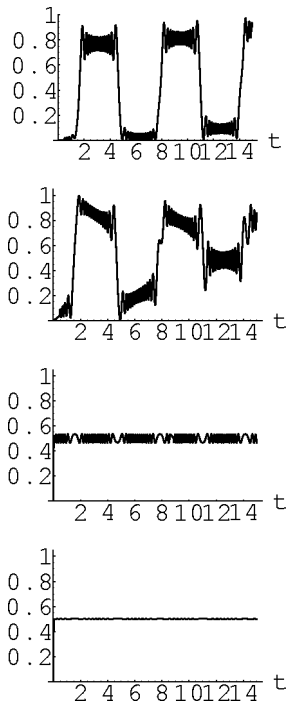

FIG. 3. Unitary evolution. The diagonal element of the density matrix ρ_{22} under a successively increasing dissipative parameter Γ. The parameters in Eq. (29) are $G=45$ (25 in the bottom drawing, chosen to facilitate the computation), $J=3$, $\omega=1$, μ (superlinearity coefficient)$=0$. Then, in top drawing—$\Gamma=0$, N (ensemble size)$=1$; in second drawing—$\Gamma=0.05$, $N=1$; in third drawing—$\Gamma=20$, $N=1$; in bottom drawing—$\Gamma=20$ (as in previous, but) $N=4$. The initial slope in the last two drawings is too steep to be visible and so are tiny fluctuations in the horizontal part of the bottom drawing. A nonzero value of the superlinear parameter μ, of the order of 1, hardly changes the curves.

$(\rho=\gamma\gamma^{\dagger})$ stays constant during the motion and this property is maintained unchanged also by the superlinear terms which enter with the coefficient μ.

It may be noted that the equations of motions for $\gamma_1(t)$ and $\gamma_2(t)$ and of their conjugates, given in Eq. (7) , lead to the master equations for the matrix elements of the density operator. For the diagonal elements these are of the LvNL form (when $\mu=0$), but not for the nondiagonal ones. This property has already been noticed in Ref. [25].

We next solve two equations for γ's, subject to the pure state initial conditions $\gamma_1=1$, $\gamma_2=0$ at $t=0$, then form from the solutions the diagonal matrix elements of the density matrix and finally show the results in Fig. 3. The quantity changed between the upper three drawings is the strength Γ of the dissipative term. As this increases, a transition takes place from the slow to the fast decoherence regime. We note the remarkable similarity of the results obtained here by the factorization method to those in Fig. 1 of the above papers, except that for strong dissipation their drawings show little oscillations, unlike our third drawing from above. In this, drawn for $\Gamma/\omega=20$, after a very steep initial slope (not visible in the figure), both diagonal density matrix elements oscillate about the asymptotic value of $\frac{1}{2}$.

The majority of calculations whose results are shown in this section are carried out for a density matrix referring to an ''assembly'' consisting of one system. This means that in Appendix A, one has $N=1$ and the summation over α in Eq. (A2) is trivial. We have also worked out the density matrices when there is a nontrivial summation, namely, when initial conditions on the two factors are $\gamma_1(0)=e^{i\pi/2},e^{2i\pi/2},e^{3i\pi/2},e^{4i\pi/2}$ and $\gamma_2(0)=0$ (so that $N=4$), instead of having only $\gamma_1(0)=1$ (and $N=1$), as before. The resultant density tends now to an almost perfectly straight line. This is similar to the graphs shown in both Ref. [41] and in Ref. [40] for strong dissipation and elucidates the meaning of system averaging in the density matrix. (In more complex systems, one would require an averaging in the density matrix over a much larger number of states, such that $N\rightarrow\infty$.) We have also worked out the $N=4$ case for the upper three graphs in Fig. 3. For these graphs, there was hardly any perceptible change from those shown.

For the detailed interpretation of these results, which is not the primary subject of this work, we again refer to Refs. [40,41], and references therein. Also, we do not delve here into the details or extensions of the results, but turn to the variational treatment of time development to be got from minimizing the action.

2. Solving variationally

We note that the strongly oscillatory factor in the solution arises from the driving term $(G/2)\cos(\omega t)$ and that this term was already present in the Hamiltonian case considered in the preceding section, in Eq. (26). (We have, however, eliminated there the fast oscillating factor by subtracting from the Hamiltonian the so-called dynamic phase.) So in this section we shall put $G=0$, which also makes the numerical aspect of the variation considerably simpler. We then set up pair of suitable trial $\gamma(t)$'s, containing parameters to be varied.

In contrast to the preceding section (which was a linear problem and in which a large number of Fourier coefficients were varied), in the present problem only one variational parameter v is introduced. However, to make progress, we must consider critical regions of the time domain, namely, $t=0$ and $t=\infty$. At the former, it is easy to see that in order that the singularity due to the zero divisor $\gamma^{\dagger}{}_2(0)$ in the vector potential be matched by the time derivative of $\gamma_2(t)$ at $t=0$, this function must take there the form of

$$\lim_{t\rightarrow 0}\gamma_2(t)\rightarrow\sqrt{4e^{i\beta}\Gamma t}+O(t), \tag{30}$$

with the constant phase angle β arbitrary. Similarly, it can be shown that, asymptotically for large t, the same solution must have the form

$$\lim_{t\rightarrow\infty}\gamma_2(t)\rightarrow e^{-iJt}\sqrt{[1+ve^{-2(\Gamma-iJ)t}+o(e^{-2\Gamma t})]/2} \tag{31}$$

or some other form equivalent to this. A constant phase factor was ignored here. The parameter v cannot be found from

the equations of motion. We seek to obtain the variationally best v, such that the condition at $t=0$ is also satisfied. After some elementary algebra one finds that in terms of the variational parameter, the density factor γ_2 can have the form

$$\gamma_2(t)=e^{-iJt}\sqrt{\left[1+ve^{-2(\Gamma-iJ)t}-(1+v)\exp-\{(v+2)\Gamma-ivJ\}\frac{2t}{1+v}\right]}/2. \tag{32}$$

The previously introduced phase β was varied independently and found to be small. So we put $\beta=0$. The other density factor $\gamma_1(t)$ was so constructed that the exponentials inside the square root had the opposite signs to those in γ_2 and normalizing factors were added so that $|\gamma_1|^2+|\gamma_2|^2=1$ at all times.

Minimizing the action integral for a set of parameter values for J and Γ yields optimized v's. Keeping $J=3$ fixed (as in Refs. [40,41] and selecting a set of dissipation parameter we obtain as follows:

$$J=3, \quad \Gamma=0.1, \quad v=-0.985,$$

$$J=3, \quad \Gamma=0.5, \quad v=-0.78,$$

$$J=3, \quad \Gamma=2, \quad v=-0.755.$$

For the middle case $\Gamma=0.5$ we compare in Fig. 4 the variational solution (thin continuous curve, with $v=-0.78$) and the solution from the equation of motion (thick continuous curve). In the asymptotic regime of large t, the behavior of the two curves is quite similar, though the amplitude of oscillation is clearly smaller in the variational solution than in the exact solution. The discrepancy appears more serious in that a different choice of the variational parameter, namely, $v=-0.98$ (also shown in the figure by broken lines), which has an action larger than the optimized one, comes nearer to the exact solution. However, we show in the inset that in the extremely short time region, the optimized solution is *qualitatively* better than the other choice. Because of the singular behavior of the short time region in the vector potential, this region dominates the value of the action. At the same time, yet another choice of the parameter $v=-0.05$ (shown in the figure by dotted lines) gives a definitely poorer resemblance to the correct curve.

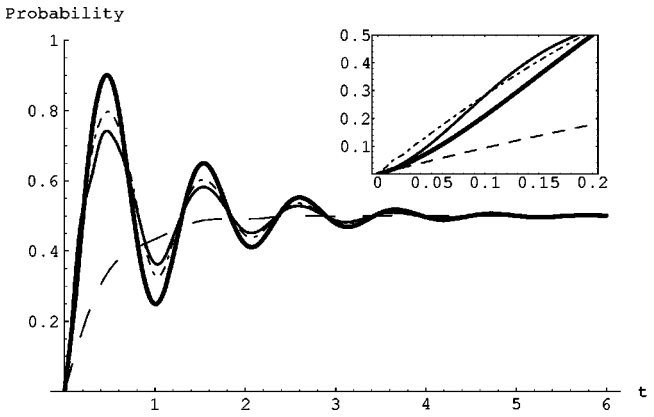

FIG. 4. Comparison of solutions. ρ_{22}, a diagonal component of the density matrix, for the following values of the parameters in Eq. (29): $G=0$, $J=3$, $\omega=1$, $\Gamma=0.5$, $\mu=0$. Solution of equation of motion—thick line. Variational solution in Eq. (32) with optimized parameter $v=-0.78$—thin line. Hypothesized solution as in Eq. (32) with $v=-0.98$—small broken lines. Hypothesized solution with $v=-0.05$—large broken lines. The inset shows the curves near $t=0$.

In conclusion, when it comes to describe subtle quantum mechanical ensemble properties, the factorization (or square root operator) method can be used either in its equation of motion form or variationally. In the present case, at least, the equation of motion approach was from a numerical viewpoint undoubtedly superior. So one may question the use of the variational method. However, on the one hand not all problems may be easily solvable. On the other hand, one should remember that the derivation of the equation of motion in Eq. (7) is itself based on a variational ansatz introduced in this paper.

V. FURTHER EXTENSIONS

To treat non-Markovian processes, the vector potentials have to be functions of the γ vectors at earlier times, but otherwise, no change in the formalism is needed.

Non-negativity of the entropy change follows from the master equations and properties of the scattering probabilities M_{ba} in Eq. (12) , as is shown in Ref. [42].

Transport processes can be treated simply. Thus, let us consider electronic conduction in a solid due to a spatial gradient in the potential (i.e., an electric field) or in the ambient temperature. The γ vectors are, normally, labeled by the reciprocal, k-vector index and are essentially small deviations from the square root of the equilibrium, Fermi-Dirac electronic distribution function. Following the standard treatment given in, e.g., Ref. [13], the time derivatives of the γ vectors (which are now real and identical to $\gamma^\dagger$) are proportional to the spatial gradients. The vector potentials represent the scattering integral. Then either equation of motion in Eq. (7) is simply the Boltzmann equation in an inhomogeneous form; namely, its left-hand side represents the source or the gradient and the right-hand side contains the desired distribution function under the integral over all wave vectors. The Lagrangian can be used to obtain the solution variationally. This variational formulation is, however, different from those given in Refs. [11,13]. (Of course, different variational procedures can lead to the same result.)

We have noted earlier that the postulated Lagrangian does not contain a potential. Adding a potential to the Lagrangian might apparently change the equations of motion. It seems to us, however, that under conditions prevailing in stochastic

processes, this will not happen. The reason for this can be stated in various forms and is rooted in the circumstance (already noted above) that in the presence of a random force one has no control over the value of the variables, only on its rate of change. (''Free terminal end point'' condition of Ref. [3], p. 9.) In Appendix B we give a formal proof for the following proposition. ''When the following conditions hold: (a) the potential is a non-negative quadratic form in all of its variables (γ's), (b) the vector potentials **A** are all real and positive, (c) the initial values of the γ vectors are suitably chosen, and (d) the variation is performed under conditions of fixed initial values of γ and $\dot{\gamma}$, then it follows that the action obtained from the variation of the velocities only, i.e., with the potential regarded as ignorable, is less than the action obtained from the variation of both the variables and their derivatives (namely, through the usual Lagrange equations, which are obtained under fixed initial and final boundary conditions).'' The result holds probably under a wider range of conditions, since in the proof we have not utilized the requirement imposed on the self-correlation of the random forces f_a by the ''second'' theorem of Mori [43]. (This requirement ensures, among other things, the time-shift invariance of the random process which is at the root of the Onsager-Machlup theory [5,6]. Needless to say, that the result obtained in Appendix A is not in conflict with the validity of the Euler-Lagrange equations, since these are obtained under conditions that the variables have fixed values at the final time.

VI. CONCLUSION

The variational action (or Lagrangian) proposed in Eq. (2) for dynamical processes has the advantages of being simple, general, and flexible. It differs from previously employed variational procedures by the factorization ansatz in Eq. (1), by the absence of a scalar potential term, and the presence of a variable final time upper limit. The relation of the postulated Lagrangian to some basic invariance property (such as ''frame indifference'' [44]) remains to be explored, account being taken of the fact that, for vector potentials that are not all equal, the formalism is non-Abelian (namely, the vector potentials cannot be transformed away by a single gauge factor) [45].

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APPENDIX A: A TUTORIAL ON THE FACTORIZED DENSITY MATRIX FORMALISM

Though the factorized density matrix, written in an abstract form as $\rho=\boldsymbol{\gamma}\cdot\boldsymbol{\gamma}^{\dagger}$, has been employed before in Refs. [16,25], we shall explain its formalism here, following Band's introductory texts to the von Neumann matrix method [46,47]. Let Ψ_{α} be a possible wave function describing the quantum state of the α'th system in the ensemble ($\alpha=1,2,\ldots,N$). It can be expanded in terms of a set of eigenstates u_n as

$$\Psi_{\alpha}=\sum_n \gamma_n^{\alpha}u_n. \tag{A1}$$

As derived in Ref. [46] and other texts, the density matrix arises from the ensemble average over all systems in the sense that its nm component is

$$\rho_{nm}=\frac{1}{N}\sum_{\alpha}\gamma_n^{\alpha}\gamma_m^{\alpha+}. \tag{A2}$$

γ^{α}'s are best viewed as row vectors, distinct for each α (or system) and the $\gamma^{\alpha+}$'s as column vectors. The γ and $\gamma^{\dagger}$ derivatives in the text (which implement the variation procedure) are with respect to γ_n^{α} and $\gamma_m^{\alpha+}$. The ensemble averaging, namely, the summing over α and the subsequent division by N, is not explicitly written out in the text, but is designated by inserting a dot between γ symbols, so that the previous matrix element is written as

$$\rho_{nm}=\gamma_n\cdot\gamma_m^{\dagger}. \tag{A3}$$

It is clear that γ's are not vector quantities, but the traces over the dotted products are proper scalars.

APPENDIX B: PROOF OF THE MINIMAL ACTION UNDER ONE-POINT BOUNDARY CONDITION

Assumptions. In the action, Eq. (2) , the vector potential A is now assumed to be positive (non-negative) and real. We shall further subtract from the action (see below) a potential term, in which the potential V depends on the variables only, not on their derivatives. This potential is supposed to be monotonic, nondecreasing, and positive in the relevant range of its variables.

We start the proof for a single time-dependent variable g which replaces the earlier complex variable γ through

$$\dot{g}=\dot{g}(t)=-i\dot{\gamma}. \tag{B1}$$

The reality of g for all times will be evident. The one-point (initial time) boundary conditions fix $g(0)$ and $\dot{g}(0)$, while $g(t)$ at later times develops according to its equation of motion. We write the action, including the potential, in the single variable g as

$$S(T)=\int_0^T dt[\{\dot{g}-A(t)\}^2-2V(g(t))]. \tag{B2}$$

The boundary conditions fix the value of $g(0)>0$ and of $\dot{g}(0)$. We next minimize the above action in two ways and subsequently compare the resulting actions. The first is the usual Lagrange equation way in the presence of a potential V and the quantities arising from this method will be denoted by the superscript V. The second method pretends that there is no potential and the corresponding quantities will take the superscript 0. It is the second method that was used in the text.

$$\ddot{g}^V(t)=\dot{A}(t)-V'(g(t)), \tag{B3}$$

where the prime represents the derivative with respect to the argument g,

$$\dot{g}^0(t)=A(t). \tag{B4}$$

The latter equation imposes the following initial condition for the velocities:

$$\dot{g}^0(0)=A(0)=\dot{g}^V(0). \tag{B5}$$

Integrating Eq. (B3) once, we obtain

$$\dot{g}^V(t)=A(t)-P(t), \quad P(t)\equiv\int_0^t V'(g(t))dt, \tag{B6}$$

where $P(t)$ is zero at $t=0$ and is for positive times non-negative, since it is, by Eq. (B3) , the time integral of a positive quantity. Subtracting $\dot{g}^0$ shown in Eq. (B4) from the last equation and integrating, it is clear that g^V never exceeds (algebraically) g^0. Calculating the actions obtained in the two methods and subtracting we find:

$$\begin{aligned} S^V(T)&=\int_0^T dt[\{\dot{g}^V-A(t)\}^2-2V(g^V(t))] \\ &=\int_0^T dt[P(t)^2-2V(g^V(t))], \\ S^0(T)&=\int_0^T dt[\{\dot{g}^0-A(t)\}^2-2V(g^0(t))] \\ &=\int_0^T dt[-2V(g^0(t))]\Rightarrow S^V(T)-S^0(T) \\ &=\int_0^T dt[P(t)^2+2\{V(g^0(t))-V(g^V(t))\}]. \end{aligned} \tag{B7}$$

In the integrand the squared term is necessarily positive (non-negative) and so is the term containing the difference of potentials since the 0 argument is larger than the V argument and the potential is monotonic by supposition. Though obtained under restricted conditions, the result shows clearly that the two-point boundary conditions are necessary requisites for the validity of the Lagrange-Euler equations of motion. Generalization to several (real) variables $g_1, g_2, \ldots, g_N$ is immediate, when the potential is a positive quadratic form in these variables, since this can be diagonalized (with positive eigenvalues) simultaneously with the kinetic energy term. However, the initial point variables need to be chosen carefully in this case.

Finally, we have not proven that the action using equations of motion of the text is minimal, but only that is lower than that obtained with the (for this case, inappropriate) use of the Lagrange equations. Furthermore, it is not evident that the solutions obtained in this appendix satisfy conditions required from density matrices or probabilities (e.g., normalizations).

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