Thermal Schrödinger equation

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A form of the Schrödinger equation is given in which the time variable is replaced by a complex timetemperature variable and the interaction with the heat bath is represented by a stochastic interaction operator with Gaussian white noise. It is shown that at large times the ensemble averaged solution yields the equilibrium spatial probability density of quantum statistical mechanics. As equilibrium is approached, we also show how the master equation for a canonical ensemble can be derived from this form of the Schrödinger equation.

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I. INTRODUCTION

The incorporation of temperature into the equations of quantum theory is an important theme running through the history of the field. One of the first contributions to this problem was made by Slater [1], who introduced the function

$$S(\mathbf{r},\boldsymbol{\beta}) = \sum_{n} |\phi_{n}(\mathbf{r})|^{2} \exp(-\beta E_{n})$$
(1)

now known as the Slater sum, where E_n are the energy eigenvalues of the Hamiltonian, ϕ_n are its eigenfunctions, and the reciprocal temperature β is the inverse of the product of Boltzmann's constant times the absolute temperature. The equilibrium probability density $P(\mathbf{r},\beta)$ is calculated by integrating the Slater sum over all space to obtain the normalization factor and dividing the Slater sum by the normalization factor. Slater showed that although the eigenfunctions of the excited states had numerous oscillations and varied rapidly with position, the equilibrium probability density was a relatively slowly varying function of position. Instead of solving the time independent Schrödinger equation for the eigenvalues and eigenfunctions, one could obtain the equilibrium probability density by solving the Bloch equation [2]

$$\frac{\partial B}{\partial \beta}(\mathbf{r},\mathbf{r}',\beta) + HB = 0 \tag{2}$$

and then setting $S(\mathbf{r},\beta) = B(\mathbf{r},\mathbf{r},\beta)$ to get the Slater sum.

The properties of a nonrelativistic isolated quantum system are given by the Schrödinger equation. If the system interacts with another system then a time-dependent interaction operator V can be added to the Hamiltonian operator H of the isolated systems

$$i\hbar \frac{\partial|\psi\rangle}{\partial t} = H|\psi\rangle + V|\psi\rangle$$
 (3)

Quantum equations involving a complex time variable have been considered previously in various contexts [3-5]. In this paper we will consider the Schrödinger equation (3) with a stochastic interaction operator and a complex time variable. By using results obtained for stochastic differential equations we will show that ensemble averaged solutions of the Schrödinger equation give equilibrium spatial probability densities at large time. It will also be shown how this thermal quantum equation can be used to derive a form of the master equation.

The first quantum mechanical derivation of the master equation was given by Pauli [6], who considered a microcanonical ensemble. The derivation was carried out by applying time-dependent perturbation theory to the Schrödinger equation and using the random phase approximation at all times during the development of the system in time. A significant advance was made by Van Hove [7,8], who showed that the master equation could be derived by assuming random phases at some initial time if additional assumptions were made regarding the perturbation. Another derivation of the master equation was given by Nakajima [9], who used Liouville and projection operator methods. It has also been shown that the master equation can be derived by using a secondquantized Hamiltonian in the occupation number representation [10-12]. Swenson [13] presented a simplified derivation in which he showed that the master equation is obtained if a perturbation expansion is made, a diagonal singularity condition is assumed and the limit of infinite volume is taken. Using the density matrix with weak perturbation theory, Zwanzig [14] derived the Pauli master equation. Among his assumptions were that the density matrix is initially diagonal, the system has many degrees of freedom, time is relatively large, and the matrix elements are smooth functions of energy. By considering the time development of the density matrix, Montroll [15] derived an equation that is very close to the Pauli master equation and showed that it reduced to the Pauli master equation under certain conditions such as that the number of degrees of freedom of the assembly is very large, the volume of the assembly is very large, the density is constant, and the coupling constant is very small. Subsequently Montroll [16] derived an exact equation for the diagonal elements of the density matrix which depends only on the fact that the off diagonal elements are postulated as zero at the initial time. Although he did not give a derivation of the Pauli master equation he discussed some problems that would occur in the derivation. Van Kampen [17] has given a quantum

i

 $|\rho_{rr}|$

(12)

mechanical derivation of the master equation which closely parallels a classical derivation and holds that the introduction of an ensemble is neither necessary nor sufficient. Relationships between some of these derivations have been clarified by Zwanzig [18], who proved the equivalence of several versions of the master equation.

These derivations are concerned with the master equation for a microcanonical ensemble, in which the probabilities that the eigenstates are occupied become equal at equilibrium. Here we will show how the master equation can be obtained for a canonical ensemble, in which the probabilities that the eigenstates are occupied approach the Boltzmann factor at equilibrium. For an arbitrary initial distribution it will be shown that the thermal Schrödinger equation gives the equilibrium distribution for a canonical ensemble at large times. In addition we show how the master equation for a canonical ensemble can be obtained from the thermal Schrödinger equation for systems approaching equilibrium. In this approach the Boltzmann factor is not assumed but emerges as a result of the derivation. A relationship between the rate constant for the approach to equilibrium and the square of the magnitude of the fluctuating potential, which can be regarded as a form of the fluctuation-dissipation theorem, will also be derived for such systems.

II. THERMAL QUANTUM EQUATION

Let us replace the time in the partial derivative of the Schrödinger equation (3) by the complex variable

$$\zeta = t - i\hbar\alpha/2 , \qquad (4)$$

$$i\hbar\frac{\partial|\psi\rangle}{\partial\zeta} = H|\psi\rangle + V_S|\psi\rangle , \qquad (5)$$

where α is a reciprocal temperature variable and specify that the stochastic interaction operator V_S is zero except when $\alpha = 0$. We are given the wave function of the quantum system at time t = 0 and want to calculate the wave function at some later time as the quantum system interacts with a heat bath at reciprocal temperature β . To do this, (A) we integrate (5) from $\alpha = \beta$ to $\alpha = 0$ keeping time constant at t = 0, then (B) we integrate (5) from t = 0to t keeping α constant at $\alpha = 0$, and finally (C) we integrate (5) from $\alpha = 0$ to $\alpha = \beta$ keeping time constant at t.

The state $|\psi\rangle$ is expanded in terms of the eigenfunctions of the Hamiltonian H

$$|\psi(t,\alpha)\rangle = \sum_{n} a_{n}(t,\alpha) |\phi_{n}\rangle , \qquad (6)$$

where

$$H|\phi_n\rangle = E_n|\phi_n\rangle . \tag{7}$$

The initial state is specified by the constants c_n

 $a_n(0,\beta)=c_n$.

The result of the first integration from $\alpha = \beta$ to $\alpha = 0$ at constant time t = 0 is

$$|\psi(0,0)\rangle = \sum_{n} a_{n}(0,0) |\phi_{n}\rangle ,$$
$$a_{n}(0,0) = c_{n} \exp(\beta E_{n}/2) .$$

When $\alpha = 0$, interaction between the system and the surrounding heat bath occurs, and it is convenient to introduce the density matrix

$$\rho_{nm}(t) = a_n(t,0)a_m^*(t,0), \quad \alpha = 0$$

We consider a stochastic interaction operator of the form

$$V_{S} = \eta(t)h(-\alpha)v_{rs}[|\phi_{r}\rangle\langle\phi_{s}| + |\phi_{s}\rangle\langle\phi_{r}|], \qquad (8)$$

where $\eta(t)$ is Gaussian white noise, h(x) is the Heaviside function with h(x)=0 for x < 0 and h(x)=1 for $x \ge 0$, r=r(t) and s=s(t) are very rapidly changing random integers designating states of the system with $r \ne s$, and v_{rs} are coefficients specifying the strength of the interaction. At any instant of time, r and s have particular values and designate a particular pair of states. The time interval during which r and s are constant is exceedingly small. For the very short time interval during which the random integers r and s are constant, the equations of change for elements of the density matrix are

$$i\hbar \frac{d\rho_{nm}}{dt} = (E_n - E_m)\rho_{nm}, \quad m, n \neq r, s \quad , \tag{9}$$

$$\pi \frac{d\rho_{rm}}{dt} = (E_r - E_m)\rho_{rm} + \eta(t)v_{rs}\rho_{sm}, \quad m \neq r,s \quad , \qquad (10)$$

$$i\hbar \frac{d\rho_{sm}}{dt} = (E_s - E_m)\rho_{sm} + \eta(t)v_{rs}\rho_{rm}, \quad m \neq r,s \quad (11)$$

$$i\hbar \frac{d}{dt} \begin{vmatrix} \rho_{rs} \\ \rho_{sr} \\ \rho_{ss} \end{vmatrix} = \begin{vmatrix} 0 & -\eta(t)v_{rs} & \eta(t)v_{rs} & 0 \\ -\eta(t)v_{rs} & E_r - E_s & 0 & \eta(t)v_{rs} \\ \eta(t)v_{rs} & 0 & E_s - E_r & -\eta(t)v_{rs} \\ 0 & \eta(t)v_{rs} & -\eta(t)v_{rs} & 0 \end{vmatrix} \begin{vmatrix} \rho_{rr} \\ \rho_{rs} \\ \rho_{sr} \\ \rho_{ss} \end{vmatrix}.$$

These equations have the form

$$\frac{d\rho}{dt} = J\rho + \eta(t)K\rho , \qquad (13)$$

where ρ is a vector whose elements are functions of t; in this case these elements are given by the elements of the column vector of (12). We want to obtain the stochastic average of these elements, which is an average over an ensemble of systems each of which is subjected to Gaussian white noise $\eta(t)$ with zero mean and unit spectral density. Here J and K are matrices whose elements do not depend on time. For this case the stochastically averaged form of (13) is given by [19,20]

$$\frac{d\bar{\rho}}{dt} = J\bar{\rho} + \frac{1}{2}K^2\bar{\rho} . \qquad (14)$$

Solving (14) is equivalent to solving (13) for each member of the ensemble and then averaging over all members of the ensemble. Applying (14) to (10), (11), and (12) we obtain

$$\frac{d\bar{\rho}_{rm}}{dt} = \frac{E_r - E_m}{i\hbar} \bar{\rho}_{rm} - \frac{v_{rs}^2}{2\hbar^2} \bar{\rho}_{rm}, \quad m \neq r, s \quad , \tag{15}$$

$$\frac{d\bar{\rho}_{sm}}{dt} = \frac{E_s - E_m}{i\hbar} \bar{\rho}_{sm} - \frac{v_{rs}^2}{2\hbar^2} \bar{\rho}_{sm}, \quad m \neq r, s \quad , \tag{16}$$

$$\frac{d\bar{\rho}_{rr}}{dt} = \frac{v_{rs}^2}{\hbar^2} (-\bar{\rho}_{rr} + \bar{\rho}_{ss}) , \qquad (17)$$

$$\frac{d\bar{\rho}_{ss}}{dt} = \frac{v_{rs}^2}{\hbar^2} (-\bar{\rho}_{ss} + \bar{\rho}_{rr}) , \qquad (18)$$

$$\frac{d\bar{\rho}_{rs}}{dt} = \frac{E_r - E_s}{i\hbar}\bar{\rho}_{rs} + \frac{v_{rs}^2}{\hbar^2}(\bar{\rho}_{sr} - \bar{\rho}_{rs}) , \qquad (19)$$

$$\frac{d\bar{\rho}_{sr}}{dt} = \frac{E_s - E_r}{i\hbar}\bar{\rho}_{sr} + \frac{v_{rs}^2}{\hbar^2}(\bar{\rho}_{rs} - \bar{\rho}_{sr}) . \qquad (20)$$

The last part of the integration path consists of integrating (5) from $\alpha = 0$ to $\alpha = \beta$, keeping time constant at t, which gives us

$$a_n(t,\beta) = \exp(-\beta E_n/2)a_n(t,0)$$
 (21)

It is convenient to introduce the stochastically averaged distribution matrix D, whose elements are

$$D_{nm} = \overline{a_n(t,\beta)a_m^*(t,\beta)} .$$
⁽²²⁾

The relation between the matrix elements is

$$\overline{\rho}_{nm}(t) = \exp[\beta(E_n + E_m)/2]D_{nm}(t) . \qquad (23)$$

III. KINETIC EQUATIONS

It will be shown that the matrix D becomes diagonal as time becomes very large. Let

$$G_{nm}(t) = |D_{nm}(t)|^2$$
(24)

be the absolute value squared of the matrix element $D_{nm}(t)$. We want to determine how $G_{nm}(t)$ changes with time. The time derivative of (24) is

$$\frac{dG_{nm}}{dt} = \frac{dD_{nm}^{*}}{dt} D_{nm} + D_{nm}^{*} \frac{dD_{nm}}{dt} .$$
 (25)

Substituting (15), (16), and (23) into (25) we obtain

$$\frac{dG_{rm}}{dt} = -\frac{v_{rm}^2}{\hbar^2}G_{rm}, \quad m \neq r,s \quad . \tag{26}$$

Since G_{rm} cannot be negative, the right-hand side of (26) shows that the absolute value of the matrix element D_{rm} must decrease until it reaches zero. Similarly, for the state s we have

$$\frac{dG_{sm}}{dt} = -\frac{v_{sm}^2}{\hbar^2}G_{sm}, \quad m \neq r, s \quad , \tag{27}$$

and the absolute value of the off-diagonal matrix elements

 D_{sm} must decrease with time. As for off-diagonal matrix elements D_{rs} and D_{sr} , the rate of change of their absolute values are obtained from (19), (20), (23), and (24), which give

$$\frac{dG_{rs}}{dt} = -\left[\frac{2v_{rs}}{\hbar}\mathrm{Im}(D_{rs})\right]^2,\qquad(28)$$

$$G_{sr} = G_{rs} \quad . \tag{29}$$

During the short time interval when the random numbers r and s are constant, the imaginary part of the matrix element D_{rs} might be zero, in which case (28) might indicate that the absolute value of this matrix element might not decrease but remain constant. However during the next time interval where the random numbers r and shave different values, the time development of this particular off-diagonal matrix element will now be determined by (26) or (27) and thus its absolute value must decrease further if it is not already zero. Therefore all off-diagonal elements of the matrix D tend to zero with increasing time.

Next let us consider the diagonal elements. During the short time interval when the stochastic potential (8) acts on states r and s, the diagonal matrix elements D_{rr} and D_{ss} can change, but the other diagonal matrix elements $D_{mm}, m \neq r, s$, do not change. From (17), (18), and (23) we obtain the equation for the rate of change of the matrix elements D_{rr} and D_{ss}

$$\frac{dD_{rr}}{dt} = \frac{v_{rs}^2}{\hbar^2} \left[-D_{rr} + \exp(\beta E_s - \beta E_r) D_{ss} \right], \qquad (30)$$

$$\frac{dD_{ss}}{dt} = \frac{v_{rs}^2}{\hbar^2} \left[-D_{ss} + \exp(\beta E_r - \beta E_s) D_{rr} \right] \,. \tag{31}$$

It is convenient to introduce the ratio

$$R = D_{rr} / D_{ss} . aga{32}$$

Combining (30), (31), and (32) yields the differential equation

$$\frac{dR}{dt} = \frac{v_{rs}^2}{\hbar^2} [1 + \exp(\beta E_r - \beta E_s)R] [\exp(\beta E_s - \beta E_r) - R] .$$
(33)

This equation shows that if $R < \exp(\beta E_s - \beta E_r)$, then dR/dt > 0 and the values of R will increase. If $R > \exp(\beta E_s - \beta E_r)$, then dR/dt < 0 and the value of R will decrease.

Therefore, as time increases and the random numbers r and s of the stochastic potential (8) change, (26), (27), and (28) show that the off-diagonal elements approach zero

$$\lim_{t \to \infty} D_{mn}(t) = 0, \quad m \neq n$$
(34)

and (33) shows that the ratios of the diagonal elements approach the Boltzmann values

$$\lim_{t \to \infty} \frac{D_{mm}(t)}{D_{nn}(t)} = \frac{\exp(-\beta E_m)}{\exp(-\beta E_n)} .$$
(35)

During the short time when the random numbers r and

s of the stochastic potential (8) are constant, if the system is not in a steady state, there will be a change in the number of systems that are in states r and s but no change in the number of systems that are in other states. Some systems that are in state r before the interaction will be in state s after the interaction or vice versa. Among the systems that are in state r or state s, the fraction that are in state r is

$$f_r = \frac{D_{rr}}{D_{rr} + D_{ss}} \tag{36}$$

and the fraction that are in state s is

$$f_s = \frac{D_{ss}}{D_{rr} + D_{ss}} \ . \tag{37}$$

Combining (36) and (37) with (30) and (31) gives us the coupled differential equations

$$\frac{df_r}{dt} = \frac{v_{rs}^2}{\hbar^2} \left[-f_r + \exp(\beta E_s - \beta E_r) f_s - gf_r \right], \qquad (38)$$

$$\frac{df_s}{dt} = \frac{v_{rs}^2}{\hbar^2} \left[-f_s + \exp(\beta E_r - \beta E_s) f_r - g f_s \right], \qquad (39)$$

$$g = \exp(\beta E_s - \beta E_r)f_s + \exp(\beta E_r - \beta E_s)f_r - f_r - f_s .$$
(40)

At equilibrium, f_r and f_s have the values

$$f_r^{(eq)} = \frac{\exp(-\beta E_r)}{\exp(-\beta E_r) + \exp(-\beta E_r)} , \qquad (41)$$

$$f_s^{(eq)} = \frac{\exp(-\beta E_s)}{\exp(-\beta E_s) + \exp(-\beta E_s)} .$$
(42)

If the system is near equilibrium, then the values of f_r and f_s are close to $f_r^{(eq)}$ and $f_s^{(eq)}$: $|f_r(t)-f_r^{(eq)}| \ll 1$ and $|f_s(t)-f_s^{(eq)}| \ll 1$. For this case, (38) and (39) simplify to

$$\frac{df_r}{dt} = \frac{v_{rs}^2}{\hbar^2} \frac{2 \exp(-\beta E_s)}{\exp(-\beta E_r) + \exp(-\beta E_s)} \times [-f_r + \exp(\beta E_s - \beta E_r)f_s], \qquad (43)$$

$$\frac{df_s}{dt} = \frac{v_{rs}^2}{\hbar^2} \frac{2\exp(-\beta E_r)}{\exp(-\beta E_r) + \exp(-\beta E_s)}$$

$$\times [-f_s + \exp(\beta E_r - \beta E_s)f_r] . \tag{44}$$

Let p_j be the probability that the system is in state *j*. Since $f_r = p_r/(p_r + p_s)$ and only p_r and p_s change during the interaction, the sum p_r and p_s is constant and f_r is proportional to p_r . Thus (43) can be written in the form

$$\frac{dp_r^{(s)}}{dt} = -p_r k_{rs} + p_s k_{sr} , \qquad (45)$$

where

$$k_{rs} = \frac{2v_{rs}^2}{\hbar^2} \frac{\exp(-\beta E_s)}{\exp(-\beta E_r) + \exp(-\beta E_s)} , \qquad (46)$$

$$k_{sr} = \exp(\beta E_s - \beta E_r) k_{rs} \quad . \tag{47}$$

The superscript (s) in (45) indicates that the rate of change of p_r is a result of the interaction between states r and s.

The fluctuation-dissipation theorem has many interpretations. The fluctuation-dissipation theorem for a Brownian particle in a scattering medium establishes a relationship between the friction coefficient and the correlation function of the fluctuating forces acting on the particle [21]. The friction coefficient determines the rate at which a rapidly moving Brownian particle slows down and comes into thermal equilibrium with the surrounding medium. The fluctuation-dissipation theorem shows that the friction coefficient is proportional to the square of the magnitude of the fluctuating force. Similarly, (46) shows that the rate constant k_{rs} , which determines that rate at which the system approaches equilibrium, is proportional to the square of the magnitude of the fluctuating potential v_{rs} . According to this interpretation, (46) can be regarded as a form of the fluctuationdissipation theorem.

Next let us consider a set of N ensembles each of which has a different value of s and where each has the same value of r. The average change dp_r in p_r during the short time interval dt is

$$dp_{r} = \frac{1}{N} \sum_{s} dp_{r}^{(s)} .$$
(48)

Averaging (45) over the set of N ensembles and using (48) we obtain

$$\frac{dp_{r}}{dt} = -p_{r} \frac{1}{N} \sum_{s} k_{rs} + \frac{1}{N} \sum_{s} p_{s} k_{sr} .$$
(49)

Equation (49) supplemented by (47) is recognized as the master equation.

IV. EQUILIBRIUM PROBABILITY DENSITY

Next let us consider solutions of the thermal Schrödinger equation (5) in the limit where time approaches infinity. Equation (6) in the coordinate representation is

$$\psi(\mathbf{r},t,\beta) = \sum_{n} a_{n}(t,\beta)\phi_{n}(\mathbf{r}) .$$
(50)

It is convenient to introduce the function

$$F(\mathbf{r},t,\beta) = \overline{\psi(\mathbf{r},t,\beta)\psi^*(\mathbf{r},t,\beta)} .$$
 (51)

Combining (22), (50), and (51) yields

$$F(\mathbf{r},t,\beta) = \sum_{n} \sum_{m} D_{nm}(t,\beta)\phi_{n}(\mathbf{r})\phi_{m}^{*}(\mathbf{r}) .$$
(52)

According to (34), as time tends to infinity, the offdiagonal elements $D_{nm}(t)$ become zero. In this limit (52) simplifies to

$$F(\mathbf{r},t,\beta) = \sum_{n} D_{nn}(t) |\phi_n(\mathbf{r})|^2, \quad t \to \infty \quad .$$
 (53)

According to (35), as time tends to infinity the ratio of the diagonal elements approaches a ratio of Boltzmann factors. Combining (35) and (53) yields

$$\lim_{t \to \infty} \frac{F(\mathbf{r}, t, \beta)}{D_{11}(t)} = \exp(\beta E_1) \sum_n \exp(-\beta E_n) |\phi_n(r)|^2 .$$
(54)

The equilibrium probability density is given by

$$P(\mathbf{r},\beta) = \frac{\sum_{n} \exp(-\beta E_{n}) |\phi_{n}(\mathbf{r})|^{2}}{\sum_{n} \exp(-\beta E_{n})} .$$
(55)

Integrating (54) over space and noting that the wave functions are normalized, we obtain the result that in the limit of large time the function (51) divided by its spatial integral approaches the equilibrium probability density.

$$P(\mathbf{r},\beta) = \lim_{t \to \infty} \frac{F(\mathbf{r},t,\beta)}{\int F(\mathbf{r},t,\beta)d\mathbf{r}} .$$
 (56)

Thus, like the Bloch equation (2), the thermal Schrödinger equation (5) gives the equilibrium spatial probability density without explicitly introducing the Boltzmann factor $\exp(-\beta E_i)$.

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