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PHYSICAL REVIEW A VOLUME 2, NUMBER 3 SEPTEMBER 1970

Liouville Energy Eigenvalue Problem

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The mathematical dilemma of the direct determination of the reduced density matrix for an N-particle system in a stationary state is examined. An energy eigenvalue equation pertaining to the statistical Hamiltonian of a subsystem which determines the reduced density matrix is formulated. It is shown that the construction of this equation represents a classical, semi-quantum-mechanical attitude. The dilemma, in fact, arises from this attitude.

I. INTRODUCTION

The problem at issue in this paper may be termed the "Liouville energy eigenvalue problem." It is concerned with the direct determination of the reduced density matrix of a subsystem contained in a total system which is in a stationary state.

It is an elementary procedure to conceive and derive such an eigenvalue equation. However, the hazards involved in correctly setting up the equation are worth reflection. The attitude adapted in deriving and applying this equation differs notably from that used in the Schrödinger energy eigenvalue problem. The equation incorporates two entirely irreconcilable physical disciplines: It makes contact with quantum-theoretic ingredients on one hand, and on the other, it welds itself into classical, macroscopic concepts. Due to this semiquantum classical origin and the well-known inconsistency between quantum mechanics and classical mechanics, one can see that the above-mentioned difficulties are intrinsic.

We believe that the perplexity and the enormous number of mathematical artifacts generated in the search of an analytical solution to this problem are created because the problem is often stated without adequate exposition of its philosophical background. We do not share the view that the problem could be approached by analytical devices alone.

II. OPERATOR MEAN VALUE

It is easily shown that the mean value of a quantum-mechanical operator of a system of N identical particles,

$$\underline{\Omega}^{(N)} = \sum_{i=1}^{N} \underline{\Omega}(i) + \frac{1}{2!} \sum_{i,j}^{N}{}' \underline{\Omega}(i,j)$$

$$+\cdots \frac{1}{m!} \sum_{i_1 \cdots i_m}^{N\,\prime} \underline{\Omega}(i_1 \cdots i_m) , \tag{1}$$

can be written, purely formally, in the form

$$\langle \underline{\Omega}^{(N)} \rangle_\psi = (N/m)\,\mathrm{tr}(\underline{R}^{(m)} \Theta_i^{(m)}) , \tag{2}$$

where $m < N$ and ψ refers to an arbitrary normalized state. The prime notation on the summation sign indicates the exclusion of any duplication in the indices. In Eq. (2), $\underline{R}^{(m)}$ is defined as

$$\underline{R}^{(m)} = \sum_{i=1}^{m} \underline{\Omega}(i) + \frac{N-1}{m-1} \sum_{i,j}^{m\,\prime} \underline{\Omega}(i,j) + \cdots \frac{(N-1)\cdots(N-m+1)}{(m-1)!} \sum_{i_1, \cdots i_m}^{m\,\prime} \underline{\Omega}(i_1, \cdots i_m) , \tag{3}$$

$$\underline{\rho}^{(m)} = \mathrm{tr}^{(N-m)} \underline{\rho}^{(N)} , \tag{4}$$

where

$$\underline{\rho}^{(N)} = |\psi\rangle\langle\psi| . \tag{5}$$

The superscript on tr stands for the order with which the trace operation is carried out.

To derive this expression, one takes advantage of three properties: the linearity of the quantum-mechanical averaging process, the fact that ψ is symmetrized, and the fact that $\underline{\Omega}^{(N)}$ involves only a sum of operators which is symmetric in each order. In this formalism the quantity $(1/m) \times \mathrm{tr}(R^{(m)}\rho^{(m)})$ may be interpreted as the one-particle contribution to the mean value of $\Omega^{(N)}$ in the state ψ, and hence $\mathrm{tr}(\underline{R}^{(m)}\underline{\rho}^{(m)})$ as the m-particle contribution. In the case that $\Omega^{(N)}$ is of such a form that $m < N$, both $\underline{R}^{(m)}$ and $\underline{\rho}^{(m)}$ are reduced operators in the sense that the particles involved are part of the system. $\underline{\rho}^{(m)}$ is the reduced density operator associated with the m-particle subsystem.

If one defines the scalar product of an operator $\underline{\Theta}_i$ with respect to $\underline{\Theta}_j$

$$\mathrm{tr}(\underline{\Theta}_i \underline{\Theta}_j^\dagger), \tag{6}$$

the relation between $\underline{\Theta}_i$ and $\underline{\Theta}_j$

$$\mathrm{tr}(\underline{\Theta}_i \underline{\Theta}_j^\dagger) = \delta_{ij} \tag{7}$$

may be called an orthogonality condition.[1] By the so-called method of expansion into orthogonal operators,[2] one may write Eq. (2) as

$$\langle \underline{\Omega}^{(N)} \rangle_\psi = (N/m) \sum_i R_i^{(m)} \rho_i^{(m)} = \sum_i \Omega_i , \tag{8}$$

where

$$R_i^{(i)} = \mathrm{tr}(R^{(m)} \underline{\Theta}_i^{(m)\dagger}), \tag{9}$$

$$\rho_i^{(m)} = \mathrm{tr}(\underline{\rho}^{(m)} \underline{\Theta}_i^{(m)\dagger}), \tag{10}$$

and $\{\underline{\Theta}_i^{(m)}\}$ is an arbitrary complete set of linearly independent m-particle operators. The set may be non-Hermitian, but one is allowed to make it Hermitian by suitable transformation. $\langle \Omega^N \rangle_\psi$ now is expressed as the scalar product of $R^{(m)}$ and $\underline{\rho}^{(m)}$ in a vector space spanned by $\{\underline{\Theta}_i^{(m)}\}$, $R_i^{(m)}$ and $\underline{\rho}_i^{(m)}$ being the respective components in this vector space. If the $\underline{\Theta}_i^{(m)}$ are Hermitian, we see that $\underline{\rho}^{(m)}$ is representable over a real field. Further, if each of the $\underline{\Theta}_i^{(m)}$ corresponds to a physical observable, the representation is in terms of physical data, i.e., the mean values of $\underline{\Theta}_i^{(m)}$. The characterization of state and observable in this vector space formalism has been called the Liouville representation of quantum mechanics.[3] Since the complete set of $\{\rho_i^{(m)}\}$ are experimentally determinable, one can then talk about the determination of $\underline{\rho}^{(m)}$, $m < N$, by means of Eq. (10) without reference to the state of the N-particle system.

It is interesting to note that the information in $\underline{\rho}^{(m)}$ is extracted from $\underline{\rho}^{(N)}$. The passage from the description of the N-particle $\underline{\rho}^{(N)}$ to the m-particle $\underline{\rho}^{(m)}$ usually does not stand in a unique and reciprocal relation. In terms of the matrix elements of $\underline{\rho}^{(N)}$ one has in the formalism of a composite system the form

$$\rho_{ij}^{(m)} = \sum_\alpha \rho_{i\alpha, j\alpha}^{(N)} ,$$

where the products $i\alpha$ and $j\alpha$ denote the eigenvalues which label some tensor product representation of $\underline{\rho}^{(N)}$. While describing a pure state, $\underline{\rho}^{(N)}$ conveys to us the best obtainable, or the maximal, information about a system. In principle, it satisfies the Schrödinger equation, and the probabilities of finding given events in such a state are regulated by the uncertainty principle. The term "irreducible probability" is therefore used by Margenau in this context.[4] On the other hand, unless the subsystem is truly isolated and symmetrization of the total system is not effected, $\underline{\rho}^{(m)}$ is a mixed state (of von Neuman,[5] of Landau,[6] and of Dirac[7]). It contains an imprecise or incomplete knowledge of a system. The probabilities involved therein have no direct reference to quantum-mechanical laws and cannot be derived from them. Since they are mixed up with classical origins, they can be manipulated at will by physical means, and therefore, are said to be "reducible."[4] Owing to this reason, the treatment of physical phenomena in terms of reduced density matrix exhibits the nature of macroscopic method in classical

physics. Quantum precision is diluted or replaced by classical ignorance. The calculation of the individual Ω_i in formula (8) in the case $m < N$, evidently does not show an ordinary way of computing a quantum-mechanical expectation value.

III. ENERGY APPROXIMATION BY REDUCED OPERATOR TECHNIQUE

The Hamiltonian operator of quantum-mechanical particle system has the form $\underline{H} = \underline{H}_0 + \underline{v}$, where $\underline{H}_0$ is a quadratic operator describing the free-particle system and $\underline{v}$ is the interaction operator. In dealing with the problem of approximating atomic or molecular energies, the Hamiltonian of the system is usually considered to involve only two-body interactions. Proceeding on this case, expressions (1)–(3) then take the following forms:

$$\underline{H} = \sum_{i=1}^{N} \underline{h}_0(i) + \tfrac{1}{2} \sum_{i,j}^{N}{}' \underline{v}(i,j) \ , \tag{11}$$

$$\langle \underline{H} \rangle_\psi = \tfrac{1}{2} N \operatorname{tr}(\underline{K}\underline{\rho}^{(2)}), \tag{12}$$

$$\underline{K} = \underline{h}_0^{(1)} + \underline{h}_0^{(2)} + (N-1)\underline{v}(1,2) \ . \tag{13}$$

Here, we have denoted the m-particle reduced operator corresponding to the N-particle Hamiltonian $\underline{H}$ by $\underline{K}$, which conventionally bears the name "reduced Hamiltonian." By changing the normalization covention for $\underline{\rho}^{(2)}$ one may write $\underline{K}$ in different forms. For convenience, let us choose a complete set of orthonormal two-particle operators $\{\underline{\Theta}_i^{(2)}\}$ and expand (12) into

$$\tfrac{1}{2} N \textstyle\sum_i k_i \rho_i^{(2)} = \tfrac{1}{2} N \sum_i \epsilon_i \ , \tag{14}$$

where

$$k_i = \operatorname{tr}(\underline{K}\underline{\Theta}_i^{(2)}) \ , \tag{15}$$

$$\rho_i^{(2)} = \operatorname{tr}(\underline{\rho}^{(2)}\underline{\Theta}_i^{(2)}) \ . \tag{16}$$

The physical meaning of ϵ_i is obvious: It represents the two-particle contribution to $\langle \underline{H} \rangle_\psi$ in the ith subspace. The problem then passes over the choosing of an optimal two-particle Liouville space in which the functional $\langle \underline{H} \rangle_\psi$ is to be minimized. Interest in applying the reduced density matrix technique to the ground-state energy eigenvalue problem for an atom or molecule has been stimulated by the original work of Löwdin[8] and Mayer.[9] The question now arises: Is there any fundamental rule which will enable us to characterize this optimal two-particle space? It is clear that at this point we are embarking on a statistical treatment. We do not know nor do we care to know the underlying dynamics.

IV. LIOUVILLE ENERGY EIGENVALUE EQUATION

An essential feature of statistical physics is that one employs theoretical constructs and physical quantities expressed in terms of a condensed set of more tractable parameters which serves to replace a large number of impractical dynamical variables. Since the great mass of raw data is organized into macroscopic terms, analysis and representation of this data is made in closer correspondence with laboratory results. We are thus led to a more phenomenological approach.

For convenience of presentation we shall express the time-independent Schröndinger equation in reduced operator language. We shall render explicit the connection and the philosophical difference between the approximation of the wave function of an N-particle system and that of the associated two-particle reduced density matrix.

To this end, let us apply the Rayleigh-Ritz procedure to establish a generic form for the approximate solutions to $\langle \underline{H} \rangle_\psi$ by the reduced density matrix method.

Consider the mean value of the N-particle Hamiltonian in a finite-dimensional direct-Hilbert subspace. Consider the case that the latter is formed from the subspace E_p and E_q of the two complementary subsystems consisting of 2 and $N-2$ particles, respectively. Take the dimensions of each E_p and E_q to be n and m. In the subspace $E_p \otimes E_q$, an arbitrary fictitious wave function ψ', not necessarily completely symmetrized, may be written in a biorthonormal development:

$$|\psi'\rangle = \sum_{i,j}^{n,m} c_{ij} |p_i q_i\rangle \ , \tag{17}$$

where C_{ij} is the N-particle amplitude matrix on the basis $|p_i q_i\rangle$, and $\{p_i\}$, $\{q_i\}$ are taken separately symmetrized. The two-particle reduced density matrix derived from ψ' related to particles 1, 2 on the basis $\{p_i\}$ then reads

$$\rho_{\alpha\beta}^{(2)} = \sum_{r=1}^{m} C_{\alpha r} C_{\beta r}^{\dagger}, \quad \alpha, \beta = 1, \ldots n \ . \tag{18}$$

Clearly, the inverse mapping of C_{ij} to $\rho_{\alpha\beta}^{(2)}$ is not unique. A family of C_{ij} differing within an arbitrary phase factor in each column vector of C_{ij} may yield the same $\rho_{\alpha\beta}^{(2)}$.

Consider the maximal number of effective independent parameters in $\rho_{\alpha\beta}^{(2)}$. There are $2n^2$ parameters in these n^2 complex elements. Since $\rho_{\alpha\beta}^{(2)}$ is Hermitian, the hermiticity reduces this parameter number to n^2. The normalization reduces it to $n^2 - 1$. From the many possible choices of parameters one may take the real valued set $\{k_l\}$, $l = 1, \ldots, n^2$, with which to define a fictitious wave function ψ' such that

$$C_{ij}=\sum_{l=1}^{n^2} k_l \mid b_{ij}^{(l)}\rangle \quad , \tag{19}$$

where $b_{ij}^{(l)}$ are totally symmetrized and satisfy the orthonormality relations

$$\sum_{i,j}^{n,m} b_{ij}^{(l)} b_{ij}^{(l)\dagger} = \delta_{ll'} \quad . \tag{20}$$

The representation of $\mid \psi' \rangle$ and $\underline{\rho}^{(2)}$ are then of the following forms:

$$\mid \psi' \rangle = \sum_{l=1}^{n^2} \sum_{i=1}^{n} \sum_{j=1}^{m} k_l b_{ij}^{(l)} \mid p_i q_j \rangle = \sum_{l=1}^{n^2} k_l \mid \Phi_l \rangle \ , \tag{21}$$

$$\rho_{\alpha\beta}^{(2)} = \sum_{r=1}^{m} \left(\sum_{l=1}^{n^2} k_l b_{\alpha r}^{(l)} \right) \left(\sum_{l'=1}^{n^2} k_{l'} b_{\beta r}^{(l')\dagger} \right) \quad . \tag{22}$$

Subject to the constraint

$$\sum_{l=1}^{n^2} k_l^2 = 1 \ , \tag{23}$$

the minimization of $\langle \underline{H} \rangle_{\psi'}$ thus is the satisfaction of the variational equation

$$\delta \langle \underline{H} \rangle_{\psi'} + \lambda \delta \left(\sum_{l=1}^{n^2} \right) k_l^2 \quad = 0 \ , \tag{24}$$

where λ is a Lagrangian undetermined multiplier. By using Eqs. (21) and (23) and by performing some elementary algebraic manipulations, the following eigenvalue equation results immediately from (24):

$$\sum_{l=1}^{n^2} [\tfrac{1}{2} N \operatorname{tr}(\underline{K}\,\underline{T})^{(t,l)} + \lambda \delta_{t,l}] k_l = 0 \ , \tag{25}$$

where the operator $\underline{T}^{(t,l)}$ is defined by its matrix elements on $\{p_i\}$, $i = 1, \ldots, n$ as

$$T_{\alpha\beta}^{t,l} = \langle p_\alpha \mid \underline{T}^{(t,l)} \mid p_\beta \rangle = \sum_{r=1}^{m} \tfrac{1}{2} (b_{\alpha r}^{(l)} b_{\beta r}^{(t)\dagger} + b_{\alpha r}^{(t)} b_{\beta r}^{(l)\dagger})$$

$$\alpha, \beta = 1, \ldots, n, \quad t, l = 1, \ldots, n^2 \ . \tag{26}$$

$(\tfrac{1}{2}N) \operatorname{tr}(\underline{K}\,\underline{T})^{(t,l)}$ may be considered as the elements of an N-particle statistical Hamiltonian $\underline{H}_s$. That is $\underline{H}_s = \underline{P}\,\underline{H}\,\underline{P}^{-1}$ where

$$\underline{P} = \sum_{l=1}^{n^2} \mid \Phi_l \rangle \langle \Phi_l \mid$$

is the projection operator which projects out the subspace of $E_p \otimes E_q$ in which all parameters are statistically effective in $\underline{\rho}^{(2)}$. Likewise, one defines a two-particle statistical Hamiltonian $\underline{h}_s$ whose elements are $\operatorname{tr}(\underline{K}\,\underline{T})^{(t,l)}$, recognizing that $\operatorname{tr}(\underline{K}\,\underline{T})^{(t,l)}$ determines the components of $\underline{K}$ on the basis $\{\underline{T}^{(l,l)}\}$, $l = 1, \ldots, n^2$ in the Liouville representation. We now construct a new eigenvalue equation

$$\sum_{l=1}^{n^2} [\operatorname{tr}(\underline{K}\,\underline{T})^{(t,l)} + \lambda \delta_{t,l}] k_l = 0 \tag{27}$$

and call it the "Liouville energy eigenvalue equation." It provides a generic form for approximate solutions to the determination of $\underline{\rho}^{(2)}$. The components of $\underline{\rho}^{(2)}$ are given from the eigenvector in (27) as $\rho_l^{(2)} = \operatorname{tr}(\underline{\rho}\,\underline{T})^{(l,l)} = k_l^2$, corresponding to the eigenenergy

$$\sum_{l=1}^{n^2} k_l^2 \operatorname{tr}(\underline{K}\,\underline{T})^{(l,l)} \ .$$

In this paper we have dealt with the case in which the allowed number of independent parameters in a fully parametrized $\mid \psi \rangle$ is greater than $n^2 - 1$. Let this number be $g - 1$. We now give attention to the generic form for approximate solutions to the determination of

$$\mid \psi \rangle = \sum_{l=1}^{g} k_l \mid \Phi_l \rangle \quad ,$$

obtaining

$$\sum_{l=1}^{g} [\tfrac{1}{2} N \operatorname{tr}(\underline{K}\,\underline{T})^{(t,l)} + \lambda \delta_{t,l}] k_l = 0 \ . \tag{28}$$

As $g \to \infty$ with n and m, Eq. (28) constitutes the Schrödinger energy eigenvalue equation. The elements $\frac{1}{2} N \operatorname{tr}(\underline{K}\,\underline{T})^{(t,l)}$ pertain to the N-particle dynamical Hamiltonian.

It is instructive to examine the mapping from $\{k_l\}$, $l = 1, \ldots, g$, which is a solution to (28), to the solution of (27), $\{k_l\}$, $l = 1, \ldots, n^2$, $n^2 < g$. An attempt to ascertain $\underline{\rho}^{(2)}$ directly by (27) or by other means such as the variational technique always implies a statistical calculation performed without assurance of dynamical validity. The degree and types of dynamical ignorance are no longer discernable in $\underline{\rho}^{(2)}$. This includes all types of holonomic N-representability constraints.[10]

V. CONCLUDING REMARKS

The formulation and applications of the Liouville energy eigenvalue equation represent a classical semi-quantum-mechanical attitude. The dilemma in evaluating $\underline{\rho}^{(2)}$ may be depicted as our inability to tell in advance the characteristics of the operator $\underline{P}$, or the information about the optimal $\underline{T}^{(t,l)}$. This inability, in fact, is a direct result of the above-mentioned attitude. Quantum precision cannot be generated from classical ignorance. Nevertheless, the equation should have its complementary usage in situations where such precision is not required or one's good intuition could pragmatically make up for it.

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PHYSICAL REVIEW A VOLUME 2, NUMBER 3 SEPTEMBER 1970

Transition Radiation in a Periodically Stratified Plasma

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The solution to the problem of determining the transition radiation emitted when a charged particle moves uniformly in a periodically stratified cold, isotropic plasma is obtained. The electromagnetic field may be expressed in terms of an infinite number of normal modes. The conditions under which some of these modes are radiative are discussed, and expressions for the field components and the energy spectral density are obtained. It is found that radiation may be emitted for arbitrarily small particle velocities and that the strongest emission takes place in a frequency band just above the average plasma frequency. Numerical results are presented to illustrate the behavior of the spectra of the various radiative modes as the frequency and plasma parameters are varied.

I. INTRODUCTION

When a charged particle moves with constant speed through an inhomogeneous medium, there are two (macroscopic) mechanisms by which radiation may be emitted. Radiation of the Čerenkov[1,2] type is expected if the particle moves close to or through a region in which the phase velocity of light is less than the speed of the particle. For radiation of this type to occur, the speed of the particle must be greater than the smallest phase velocity encountered. Transition radiation,[3-5] on the other hand, may be expected to occur at any particle speed. As the charged particle moves uniformly in the continuously inhomogeneous medium, its images will not, in general, be in uniform motion, but will be accelerated. Transition radiation may be thought of as being emitted by these accelerated image charges. Since the nonuniform motion of the images will occur even if the moving charged particle is traveling slowly, there is no velocity threshold for transition radiation.

In a previous paper,[6] the emission of Čerenkov and transition radiation by a charged particle moving uniformly in a periodically stratified nondispersive dielectric medium was considered. It was found that the electromagnetic field excited by the passage of the charged particle may be expressed in terms of an infinite number of normal modes. Each of these normal modes was a modulated cylindrical wave, propagating in the direction of motion of the particle at a phase speed equal to the velocity of the particle. Some of these modes were also propagating in the outward direction, away from the track of the particle. Furthermore, the threshold velocity, cutoff frequency, and emission angles for each mode were also found. However, these results are not applicable to dispersive media.