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Atomic Vibrations in the Thermodynamic Limit

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We present, on a simple model of a one-dimensional crystal lattice, the consequences of the assumption that the phases in the action–angle representation are random. We prove that this assumption amounts to the introduction of a stochastic measure which can be interpreted as a Gaussian noise. The presence of noise gives rise to a new spectral representation of states of the lattice. It is shown that this new spectral representation of states can also be extended on an infinite lattice through a rigorously defined transition to the thermodynamic limit. The traditional spectral representation, as a superposition of independent modes, of such states as atomic displacements leads to meaningless expressions in the thermodynamic limit. One of the main results is that under the random phase assumption the interactions lead to the appearance of equilibrium states. We obtain an explicit spectral representation of such states. This specific model illustrates how probabilistic behavior of an infinite system can be derived from classical laws of dynamics.

1. INTRODUCTION

Small perturbations may influence a classical or quantum dynamical system in such a way that it will approach statistical equilibrium. This can happen when the system is "large," which means that it has many, in fact infinite, degrees of freedom. In such a case even a relatively small perturbation which mixes the many degrees of freedom may give rise to irreversible behavior. In consequence the traditional description of dynamics in terms of trajectories which obey the Newtonian laws becomes meaningless. What can be done instead is to replace the time evolution of points by the evolution of probability densities or transition probabilities. Accordingly, the classical evolution equation can be replaced by an appropriate equation characterizing

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the irreversible evolution of densities, like the master equation. Nevertheless the natural question arises of how and under what conditions one can derive the probabilistic description from deterministic laws of dynamics. Confining ourself to Hamiltonian systems, we may ask: How does it happen that a system of N interacting particles which is reversible for finite N becomes irreversible when $N = \infty$? Another question is: Is it possible to obtain spectral representations of equilibrium states in the case $N = \infty$ in a similar way as can be done for finite N and, if so, what is the relation between them?

A first step toward resolution of these problems was taken by Pauli [1], who showed how in a quantum mechanical Hamiltonian system, $H = H_0 + \lambda V$, the approach to equilibrium can be produced by a relatively small perturbation V. Assuming that the free Hamiltonian H_0 has continuous spectrum and that the phases of the quantum mechanical amplitudes with respect to the eigenstates of H_0 are random, he derived the master equation from the Schrödinger equation. Pauli's approach was modified by van Hove [2], who derived the master equation under a weaker condition and introduced states (operators) with diagonal singularity which give meaning to equilibrium states.

Another approach to the problem of incorporating irreversibility on the fundamental level in large systems was proposed by Petrosky and Prigogine [3]. They proposed a new unified formulation of dynamics and thermodynamics through extended spectral representations.

States with diagonal singularity admit spectral representations in terms of the same spectral variables as other states of the Hamiltonian system. Thus the two classes of states, so different from the point of view of their behavior, can be connected. The difference is that the states with diagonal singularity are meaningless in the conventional Hilbert space formulation. Nevertheless they are derived, exploiting Hilbert space techniques, from finite systems (spectra) through a specific transition to the thermodynamic limit.

There are some problems with such a transition. It is not clear how the convergence of ordinary states to a state with diagonal singularity can be defined. Moreover, the natural representations of states, such as displacements of a particle, derived for systems with discrete spectra may become meaningless in the limit (see Section 2). The transition from discrete to continuous spectrum is, on the other hand, necessary since only in the limit do the master equation and the equilibrium states containing singularities of δ-function type acquire meaning.

A way round the difficulties associated with this transition to the thermodynamic limit has been proposed in refs. 4 and 5. It has been shown there that constructing a natural extension of quantum theory, it is possible to give rigorous meaning to such states as the microcanonical equilibrium. Nevertheless the related problem of transition from reversible finite system to irreversible infinite one has not been touched.

Looking for the origin of these difficulties, let us notice that equilibrium states correspond to probabilistic distributions on an infinite system. Such distributions can be derived as limits of probability distributions on finite systems. Therefore, trying to resolve the problem of the transition from a deterministic finite system to an infinite probabilistic one it is necessary to recognize where the randomness enters the classical Newtonian laws. It turns out that the only necessary probabilistic assumption is that proposed by Pauli.

We apply this idea to a simple model of a one-dimensional crystal lattice. We assume that the phases in action–angle representation of a crystal lattice are random and show that this assumption leads to a rigorous formulation of the thermodynamic limit and representations of states of the system in this limit. The stochasticity introduced by the random phases becomes a Gaussian noise in the thermodynamic limit. States, such as displacements of particles, become now stochastic processes represented by stochastic integrals. In addition, there appear diagonal singularities of states, corresponding to interactions. We also give a rigorous meaning to the statement that in the thermodynamic limit the Kronecker delta becomes Dirac's delta.

The assumption of stochasticity which we impose has been suggested by Petrosky and Prigogine [6]. It is similar to Pauli's random phase condition as modified by van Hove. Randomness is therefore introduced on the fundamental level. In a free system randomness of phases does not give any significant irreversible effects, although this assumption allows one to represent such states as atomic displacements in the thermodynamic limit. Moreover, the original representation can be recovered by taking the average. However, small but persistent interactions between these stochastic variables cause the creation of new states— states with diagonal singularities. In consequence our approach also allows us to derive extended spectral representations [3–5].

The additional advantage of such a description is that performing the transition $N \to \infty$, we need not switch from trajectories to densities. We can stay on the level of trajectories, although now a trajectory becomes a realization of a stochastic process. This makes the transition from reversibility to irreversibility more transparent. We can also observe where we have to depart from the traditional Hilbert space description. The states which correspond to a free system can be regarded as elements of a Hilbert space even in the new stochastic representation. Only the interactions create a new class of states.

2. RANDOM PHASES AND REPRESENTATION OF STATES

We consider a one-dimensional crystal lattice with an infinite number of atoms of mass $m = 1$ free to move along the line. The infinite lattice will

be treated as the limit of linear chains of N atoms when $N \to \infty$. Let us denote by $u_n = u_n(t)$ the displacement of the nth atom from its equilibrium position. We assume that the distance between any two neighboring atoms in the equilibrium position is a. Under this assumption the potential energy U is of the form

$$U = \frac{1}{2} \sum_{n,n'} A_{n,n'} u_n u_{n'} + U_0 \tag{1}$$

where U_0 is the potential in equilibrium. Consequently the quation of motion is

$$\frac{d^2 u_n}{dt^2} = -\sum_{n'} A_{nn'} u_{n'} \tag{2}$$

where the coefficients $A_{nn'}$ depend only on $n - n'$, $A_{nn'} = A(n - n')$, and satisfy $\Sigma_{n'} A(n - n') = 0$. The requirements concerning the coefficients $A_{nn'}$ are valid for N atoms under the cyclic condition on the displacement $u_{n+N} = u_n$. Under these conditions the general solution of Eq. (2) is [7]

$$u_n = \sum_k q_k e^{ikna} \tag{3}$$

where k is a multiple of $2\pi/aN$, $-\pi/a < k \leq \pi/a$, and $q_k = q_k(t)$ are such that $q_{-k} = q_k^*$ and satisfy the equation

$$\frac{d^2 q_k}{dt^2} + \omega_k^2 q_k = 0$$

and ω_k is the frequency associated with k,

$$\omega_k^2 = \sum_l A(l) e^{ikla}$$

Following the traditional approach [8], we express the displacements u_n in the action–angle variables (J_k, α_k) as the real part of

$$\tilde{u}_n = \frac{1}{N^{1/2}} \sum_k e^{ikna} \left(\frac{J_k}{\omega_k} \right)^{1/2} e^{i\alpha_k} \tag{4}$$

(we omitted here the factor $\sqrt{2}$). We would like to discuss first the behavior of $u_n = \Re \tilde{u}_n$ in the thermodynamic limit when the number of atoms N and the volume L in which they are enclosed tends to infinity while the ratio N/L remains finite. We shall identify N with L. Therefore we are interested in the study of the limit of (4) as $N \to \infty$ in the case when the values J_k and ω_k are reasonably chosen, for example if the ratio J_k/ω_k is bounded on $(-\pi/a, \pi/a]$.

It is obvious that, unless $J_k \equiv 0$, the sum in (4) is of order N and the limit does not exist. This may lead to the conclusion that in the thermodynamic

limit the displacements are meaningless, or at least cannot be represented in terms of action–angle variables. The first possibility must, of course, be excluded since we know that nothing dramatic happens to displacements when the number N of atoms in the lattice increases. Still the second possibility makes the difference between the case of finite and infinite number of particles enormous. This may cast some doubt on the validity of other reasoning based on the thermodynamic limit arguments.

It was noted by Petrosky and Prigogine [6] that in order to obtain a finite limit one should assume that angle variables α_k must behave as "stochastic variables." We shall give a rigorous meaning to a modification of this idea and show that the limit of (4) can be represented as a stochastic integral.

The assumption of stochasticity of α_k is similar to the Pauli's assumption of random phases at all times [1], discussed also by van Hove [2]. He argued that although the assumption of random phases of the quantum mechanical amplitudes is necessary to derive Pauli's master equation, the original Pauli random phase assumption can only hold at all times if the system is in equilibrium. He proposed instead the assumption of random phases only for the initial state of the system.

We face here a similar situation. We may assume that α_k are random variables. However, since $\alpha_k = \omega_k t + \delta_k$, as follows from the equations of motion, we may assume that only the phases $\delta_k = \alpha_k(0)$ are random and consider instead of (4) the sums

$$\tilde{u}_n = \frac{1}{N^{1/2}} \sum_k e^{i(kna+\omega_k t)} \left(\frac{J_k}{\omega_k} \right)^{1/2} e^{i\delta_k} \tag{5}$$

The possibility of the assumption that α_k are random variables will be discussed later.

Actually, the problem of representation of states in the thermodynamic limit concerns not only displacements. Any complex state $s_n = s_n(t)$ of a one-dimensional lattice which satisfies the cyclic condition $s_n = s_{n+N}$ can always be represented as a superposition of independent modes. We can therefore use the same arguments as in the case of displacements. In particular the stochastic representation of such states can be written as

$$s_n = \frac{1}{N^{1/2}} \sum_k e^{i(kna+\omega_k t)} c_k e^{i\delta_k} \tag{5'}$$

The problem under consideration can be reduced, for a fixed time t, as follows. Consider only even numbers N, since in such a case we have, for each N, the division of the interval $(-\pi/a, \pi/a]$ on N disjoint subintervals Δ_k of equal length, and denote by f_N the step function which equals

$e^{i(kna+\omega_k t)}(J_k/\omega_k)^{1/2}$ [respectively $e^{i(kna+\omega_k t)}c_k$, if we consider a general state (5′)] on the interval Δ_k. We ask about the convergence, as $N \to \infty$, of the sums

$$\frac{1}{\sqrt{N}} \sum_k f_N(k) e^{i\theta_k} \tag{6}$$

where θ_k (θ_k depend also on N) are independent, identically distributed random variables. If *a priori* there is no preference concerning phases, we may assume that the random variables θ_k are uniformly distributed in the interval $[-\pi,\pi]$. Putting

$$M_N(\Delta_k) = \frac{1}{\sqrt{N}} e^{i\theta_k}$$

we define a *stochastic measure* on the (finite) σ-algebra generated by intervals Δ_k. Recall that the term stochastic measure means that to each measurable set is assigned a random variable. By the assumption of independence of θ_k each measure M_N is independently scattered on the σ-algebra generated by the intervals Δ_k, i.e., for any two sets A_1, A_2 such that $A_1 \cap A_2 = \emptyset$ the random variables $M_N(A_1)$ and $M_N(A_2)$ are independent. In this terminology the expression (6) can be interpreted as the stochastic integral

$$\int_{-\pi/a}^{\pi/a} f_N(k) M_N(dk) \tag{7}$$

Thus our original problem of the existence of the limit (6) has been reduced to the problem of the existence of the limit (7) and, of course, the meaning of the convergence itself. We shall show that this limit is also of the the form of a stochastic integral with respect to some stochastic measure M defined on the σ-algebra of Borel subsets of $(-\pi/a, \pi/a]$.

In order to determine the measure M, which must also be some limit of M_N, let us consider an interval Δ from the dyadic division of the interval $(-\pi/a, \pi/a]$, i.e., for some $N = 2^m$. Thus $M_N(\Delta)$ is correctly defined for each $N = 2^{m+l}$, $l = 0, 1, 2, \ldots$, and we have

$$M_N(\Delta) = M_N(\Delta_1) + \ldots + M_N(\Delta_{2^l})$$

where $\Delta_1, \ldots, \Delta_{2^l}$ is the dyadic division of Δ on 2^l parts.

Let us consider the real part of $M_N(\Delta)$,

$$\Re M_N(\Delta) = 2^{-(m+l)/2} \cos\theta_1 + \ldots + 2^{-(m+l)/2} \cos\theta_{2^l}$$

and put

$$\xi_{lj} = 2^{-(m+l)/2} \cos\theta_j, \quad j = 1, \ldots, 2^l$$

For each l the random variables ξ_{lj} are independent and identically distributed with the mean value

$$E\xi_{lj} = 2^{-(m+l)/2} \frac{1}{2\pi} \int_{-\pi}^{\pi} \cos x \, dx = 0$$

and the variance

$$\text{Var } \xi_{lj} = \sigma_{lj}^2 = 2^{-m-l} \frac{1}{2\pi} \int_{-\pi}^{\pi} \cos^2 x \, dx = 2^{-m-l-1}$$

Therefore $\max_{1 \le j \le 2^l} \sigma_{lj} \to 0$ as $l \to \infty$, and $\Sigma_{j=1}^{2^l} \sigma_{lj}^2 = 2^{-m-1}$. In consequence the array

$$\{\xi_{lj}\}_{\substack{l=1,2,\ldots \\ j=1,\ldots,2^l}}$$

satisfies the assumptions of the central limit theorem [9], which says that there exists a limit of the distributions of $\xi_{l1} + \ldots + \xi_{l2^n}$ as $l \to \infty$. Since the random variables $\cos \theta_j$ are also bounded, it is easy to check that the array

$$\{\xi_{lj}\}_{\substack{l=1,2,\ldots \\ j=1,\ldots,2^l}}$$

satisfies the normal convergence criterion [9, Sec. 22]. Consequently the distributions of $\Re M_N(\Delta)$ converge to the normal distribution $\mathcal{N}(0, |\Delta|)$ with the mean zero and the variance $|\Delta|$. We obtain the same result for the imaginary part of $M_N(\Delta)$.

Using the same arguments, we can show, under reasonable assumptions on the values J_k/ω_k, the convergence of (6) in the sense of distributions as $N \to \infty$.

Let us note that the limit of the distributions of the real and complex parts of $M_N(\Delta)$ will be either Gaussian or zero independently of the choice of random phases θ_k, provided the Gaussian central limit theorem is satisfied. We can choose θ_j being arbitrary independent with respect to j identically distributed random variables such that $Ee^{i\theta_j} = 0$. The best choice, therefore, is to consider the Gaussian stochastic measure from the beginning, as it gives the same type of distributions of $M_N(\Delta)$ for both finite N as well as in the limit $N \to \infty$, and it will allow us to obtain a much stronger, mean square convergence, which in turn will allow "interactions" to be tackled and described by multiple stochastic integrals.

Let us therefore assume that the "noise" added in the action–angle variables representation (9) of u_n corresponds to values of an independently scattered Gaussian stochastic measure M on the Brillouin zone, i.e., the real and the complex parts of $M_N(\Delta_k) = M(\Delta_k)$ are independent Gaussian random variables with mean 0 and variance $|\Delta_k|/2$ for each Δ_k belonging to the Nth division of $(-\pi/a, \pi/a]$.

Both the real and complex parts of M can be determined by a Gaussian process $\{X_k\}_{k \in (-\pi/a, \pi/a]}$ with independent increments, $EX_k = 0$, and the correla-

tion function E $X_k X_{k'}$, $= \min\{k, k'\}/2$. We set for the real (complex) part of $M((k, k'])$ the value $X_{k'} - X_k$, and then extend M to a countably additive measure on all Borel subsets of $(-\pi/a, \pi/a]$ [10].

We would like to stress here that the stochastic process X_k is not parametrized by time t, but by the wave numbers k. In order to avoid such confusion we prefer to speak about a stochastic measure instead of a stochastic process.

It follows from the above considerations that the random phase assumption amounts to the assumption that $\tilde{u}_n = \tilde{u}_n^N$ is of the form

$$\tilde{u}_n = \sum_k e^{i(kna+\omega_k t)} \left(\frac{J_k}{\omega_k} \right)^{1/2} M(\Delta_k) \tag{8}$$

[for the rest of this section we confine ourselves to the displacement $\tilde{u}_n$ of the form (5), since the general case (5′) can be treated in the same way]. Instead of (8) we write simply

$$\tilde{u}_n = \sum_k f_N(k) M(\Delta_k) \tag{9}$$

It follows from the theory of stochastic integrals (see, for example, ref. 11) that there exists a mean-square limit of (9) as $N \to \infty$ if and only if the sequence $\{f_N\}$ converges in $L^2_{(-\pi/a, \pi/a]}$, i.e., if there exists a function f on $(-\pi/a, \pi/a]$ such that

$$\lim_N \int_{-\pi/a}^{\pi/a} \left| f_N(k) - f(k) \right|^2 dk = 0$$

In such a case f is stochastically integrable and we put

$$\int f(k) M(dk) = \text{l.i.m.}_N \sum_k f_N(k) M(\Delta_k)$$

where l.i.m.$_N$ denotes the limit, as $N \to \infty$, in the norm $\|\cdot\| = (E|\cdot|^2)^{1/2}$. Moreover we have

$$\mathrm{E} \left| \int f(k) M(dk) \right|^2 = \int \left| f(k) \right|^2 dk$$

In order to apply these facts to our specific model, recall that numbers k form, as $N \to \infty$, a dense subset of $(-\pi/a, \pi/a]$. It is enough to assume that, for example, the ratios J_k/ω_k can be extended to a continuous and integrable function $J(k)/\omega(k)$. In such a case we obtain the following formula for the mean-square limit of $\tilde{u}_n = \tilde{u}_n^N$ and its averages:

$$\text{l.i.m.}_N \, \tilde{u}_n = \int e^{i(kna+\omega(k)t)} \left(\frac{J(k)}{\omega(k)} \right)^{1/2} M(dk) \tag{10}$$

and

$$\lim_N \langle |\tilde{u}_n|^2 \rangle = \lim_N \mathrm{E}|\tilde{u}_n|^2 = \int \frac{J(k)}{\omega(k)}\, dk = \lim_N \frac{1}{N} \sum_k \frac{J_k}{\omega_k} \tag{11}$$

Equation (10) together with (11) gives a rigorous meaning to the conversion formula of vector-wave sums to integrals [see ref. 6, (2.10)] in the mean-square averages.

Assuming random phases at the initial time and, consequently, that $\tilde{u}_n$ is of the form (8), we see that the time evolution $t \to \tilde{u}_n(t)$ becomes a stationary stochastic process with the correlation function

$$\langle \tilde{u}_n(t+s)\tilde{u}_n(t) \rangle = \int e^{i\omega(k)s} \frac{J(k)}{\omega(k)}\, dk$$

Another consequence of the random phase assumption is that we can give a rigorous meaning to the statement that in the limit $N \to \infty$ the "weighted" Kronecker delta $N\delta^{\mathrm{kr}}_{kk'}$ becomes the Dirac delta $\delta(k - k')$. To see this, let us note that for each N, there is one-to-one correspondence between the wave numbers k and the intervals Δ_k. Moreover, the Kronecker delta can be expressed as

$$\delta^{\mathrm{kr}}_{kk'} = \mathrm{E}\, M(\Delta_k)\overline{M(\Delta_k)}$$

Indeed, for $k \neq k'$, $M(\Delta_k)$ and $M(\Delta_k')$ are independent random variables with mean zero, thus $N\delta^{\mathrm{kr}}_{kk'} = 0$. For $k = k'$, $\delta^{\mathrm{kr}}_{kk'} = E|M(\Delta_k)|^2 = |\Delta_k| = 1/N$. Therefore

$$N\delta^{\mathrm{kr}}_{kk'} = \begin{cases} 0 & \text{for} \quad k \neq k' \\ 1 & \text{for} \quad k = k' \end{cases}$$

To check the convergence $N\delta^{\mathrm{kr}}_{kk'} \to \delta(k - k')$, let us fix $k \in (-\pi/a, \pi/a]$ and extend $\delta^{\mathrm{kr}}_{kk'}$ on all k' putting $N\delta^{\mathrm{kr}}_{kk'} = 1$ if $k' \in \Delta_k$ and 0 otherwise, where Δ_k is now the interval which contains k. Thus if φ is a continuous function, then

$$\int \varphi(k')N\delta^{\mathrm{kr}}_{kk'}\, dk' = \frac{1}{|\Delta_k|}\int_{\Delta_k} \varphi(k')\, dk' \to \varphi(k) = \int \varphi(k')\delta(k - k')\, dk'$$

as $N \to \infty$.

Finally, let us discuss briefly a possibility of the assumption that the angle coordinates α_k are random variables [6]. If we simply replace $\alpha_k = \alpha_k(t)$ by random variables $\theta_k = \theta_k(t)$, then the corresponding random measure $M = M_t$ also depends on t. However, our previous considerations concerning determination of the measure M were time independent. Therefore $M_t(\Delta)$ must have for each t a normal distribution with mean zero and variance $|\Delta|$. This implies that, for example, the distribution of

$$\tilde{u}_n = \tilde{u}_n(t) = \text{l.i.m.}_N \sum_k e^{ikna}\left(\frac{J_k}{\omega_k}\right)^{1/2} M(\Delta_k) = \int e^{ikna}\left(\frac{J_k}{\omega_k}\right)^{1/2} M_t(dk) \quad (12)$$

does not depend on t either. Note that the assumption $\alpha_k(t) = \theta_k(t)$ is equivalent to $\alpha_k(t) = \omega_k t + \theta_k(t)$, which means that phases are random at all times. We have therefore reached the same conclusion as quoted above [2], that the assumption of random phases at all times implies that the system is in equilibrium.

3. STOCHASTIC REPRESENTATION OF INTERACTIONS

Having presented the above idea of introducing random phases to the action–angle variables, we may now consider interactions $u_n u_{n'}$, $u_n u_{n'} u_{n''}$, ..., or, in general, any interactions between two or more particles in the thermodynamic limit, in particular, anharmonic lattices with the potential energy U of the form [7]

$$U - U_0 = \frac{1}{2}\sum_{n,n'} A_{n,n'} u_n u_{n'} + \frac{1}{6}\sum_{n,n',n''} B_{n,n',n''} u_n u_{n'} u_{n''} \quad (13)$$

[compare with (1)].

In the case of interacting particles there appears an additional phenomenon which is not present on the level of one particle and which can not be treated rigorously in the traditional "Hilbert space approach"—the appearance of "diagonal singularities" [2–6].

We would like now to present very briefly this, perhaps most important consequence of the introduction of random phases. We shall show the appearance of diagonal singularities in the thermodynamic limit on the case of two interacting particles. Although this case may seem to be oversimplified, it exhibits all the main features of interactions. Physically more interesting, but technically more complicated, interactions of many particles such as in anharmonic lattices can be treated in the same manner. More rigorous treatment of this problem will be presented elsewhere.

The basic tool which allows the study of interactions in the thermodynamic limit is the multiple stochastic integral, which has its origin in the pioneering work of Wiener on "polynomial chaos" [12]. Although there is a vast literature devoted to this subject, the multiple stochastic integral is usually defined for functions which equal zero on diagonals. Such an approach allows one to simplify the theory of the integrals, but is not adequate for our purposes. It turns out that diagonals of integrands play a crucial role in problems connected with irreversibility. For this reason we base our further considerations on refs. 13 and 14, which concern only double stochastic integrals, but in the most general form.

Let us therefore consider as an example the states S_N which can be expressed in terms of displacements of u_n as

$$S_N = \sum_{n,n} C_{n,n'} u_n u_{n'} \tag{14}$$

Introducing action–angle coordinates and assuming further that $\omega_{-k} = \omega_k$ and $J_{-k} = J_k$, we can transform (14) into

$$S_N = \frac{1}{N} \sum_{k,k',\varepsilon_1,\varepsilon_2} \left(\frac{J_k J_{k'}}{\omega_k \omega_{k'}} \right)^{1/2} V_{kk'} e^{i(\varepsilon_1 \alpha_k + \varepsilon_2 \alpha_{k'})} \tag{15}$$

where $V_{kk'} = \Sigma_{n,n'} C_{n,n'} e^{i(kn+k'n')a}$. The summation in (15) is over all k, k' and over all signs ε_1, $\varepsilon_2 = \pm$.

Similar considerations as those which led to formulation (9) lead us now to the expression

$$S_N = \sum_{k,k',\varepsilon_1,\varepsilon_2} g_N(k, k') M_{\varepsilon_1}(\Delta_k) M_{\varepsilon_2}(\Delta'_k) \tag{16}$$

where $M_+ = M$, $M_- = \overline{M}$, and g_N is a step function defined on the square $(-\pi/a, \pi/a] \times (-\pi/a, \pi/a]$, which equals $(J_k J_{k'}/\omega_k \omega_{k'})^{1/2} V_{kk'} e^{i(\varepsilon_1 \omega_k + \varepsilon_2 \omega_{k'})t}$ on $\Delta_k \times \Delta_{k'}$.

Once more the question of existence and representation of the state S_N in the limit $N \to \infty$ amounts to the existence of the limit of (16) and its representation. The answer is not, however, a straightforward generalization of the previous case. The existence of the L^2 limit $g(k, k')$ of $g_N(k, k')$ as $N \to \infty$ is a necessary but, in general, not sufficient condition for the existence of l.i.m.$_N$ S_N. In addition, the function $g_N(k, k')$ must be L^1-convergent on the diagonal $k = k'$ [13]. If this is the case, the double stochastic integral of $g(k, k')$ exists and

$$S_\infty = \sum_{\varepsilon_1 \varepsilon_2} \int \int g(k, k') M_{\varepsilon_1}(dk) M_{\varepsilon_2}(dk') = \underset{n}{\text{l.i.m.}}\, S_N \tag{17}$$

Therefore we can define the state S in the thermodynamic limit, putting $S = S_\infty$, if and only if the interactions $V_{kk'}$ can be extended to a function $V(k, k')$ defined on the square $(-\pi/a, \pi/a] \times (-\pi/a, \pi/a]$ which satisfies

$$\int \int \frac{J_k J_{k'}}{\omega_k \omega_{k'}} \left| V(k, k') \right|^2 dk\, dk' + \int \frac{J_k}{\omega_k} \left| V(k, k) \right| dk < \infty$$

Since our stochastic measure is Gaussian, we can derive from ref. 13 a more explicit representation of $S = S_\infty$. For example, that component of (17) which corresponds to $\varepsilon_1 = +$ and $\varepsilon_2 = -$ is of the form

$$\int \frac{J_k}{\omega_k} V(k, k)\, dk + \int\int \left(\frac{J_k J_{k'}}{\omega_k \omega_{k'}}\right)^{1/2} V_0(k, k') e^{i(\omega_k - \omega_{k'})t} M(dk)\overline{M}(dk') \quad (18)$$

where $V_0(k, k') = V(k, k')$ for $k \neq k'$, and $V_0(k, k) = 0$. The second component of (19) can be also regarded as a stochastic integral with respect to a stochastic measure which assumes 0 on the diagonal.

The above example indicates that it is possible to describe the class of all states involving interactions of two particles. Namely such a state can be identified with a stochastically integrable function $g(k, k')$ and represented as a stochastic integral. The time evolution of interactions is, however, more complicated than the states considered in the previous section. For example, the stochastic process corresponding to one component of $u_n u_{n'}$, which is represented in the thermodynamic limit by

$$\int\int e^{i[(kn+k'n')+(\omega_k - \omega_{k'})t]} \left(\frac{J(k)J(k')}{\omega(k)\omega(k')}\right)^{1/2} M(dk)\overline{M}(dk')$$

is not stationary.

4. INTERACTIONS AND DIAGONAL SINGULARITIES

We now connect the states introduced above, which are random variables, with states understood in the traditional way—as densities or operators. In order to avoid heavy notation let us consider a real Gaussian stochastic measure M and a state S associated with a real function g

$$S = \int\int g(k, k') M(dk) M(dk') \quad (19)$$

The state (19) is correctly defined if and only if g is square-integrable as a function of two variables and integrable on the diagonal. We can therefore associate with g an operator on $L^2_{[-\pi/2,\pi/2]}$ of the form

$$A = A^{\mathrm{d}} + A^{\mathrm{nd}} \quad (20)$$

The operator A^{d} is a multiplication operator

$$A^{\mathrm{d}}\varphi(k) = g(k, k)\varphi(k)$$

and A^{nd} is a kernel operator

$$A^{\mathrm{nd}}\varphi(k) = \int g(k, k')\varphi(k')\, dk'$$

Operators of the form (20) are called states, or operators, with *diagonal singularity* [2–5]. They belong to the dual of the Banach algebra of observ-

ables with diagonal singularity characterized in ref. 4. It follows from the above remarks that each state corresponding to interactions determines an operator with diagonal singularity.

The converse is also true if we restrict ourself to operators (20) where A^{d} is an operator of multiplication by an integrable function $a(k)$ and A^{nd} is a Hilbert Schmidt operator. In such a case we can define the corresponding state S through (19) taking as $g(k, k')$ the kernel of the operator A^{nd} modified in such a way that $g(k, k) = a(k)$.

The equivalence established above allows us to connect operators with diagonal singularity with probability densities commonly used in statistical mechanics. Notice first that an operator with diagonal singularity determines, via (19), the random variable S, which in turn determines the probability density. In order to find this density it is enough to find its characteristic function (Fourier transform).

Consider first a characteristic function of a noninteracting state which, like atomic displacements, can be represented in the form

$$\int f(k)M(dk) \tag{21}$$

As before, assume that f and M are real. Then its characteristic function is

$$\mathrm{E}e^{i\xi \int f(k)M(dk)} = e^{-1/2\xi^2 \int f^2(k)\, dk}$$

This means that the density function corresponding to state (21) is Gaussian with mean zero and variance $\int f^2(k)\, dk$. However, the characteristic function of state (19)

$$\phi(\xi) = \mathrm{E}e^{i\xi \int\int g(k,k')M(dk)M(dk')} \tag{22}$$

and consequently its density, is no longer Gaussian. To see this it is enough to put $g(k, k') = a(k)a(k')$, where $\int a^2(k)\, dk = 1$. Then

$$\int\int g(k, k')M(dk)M(dk') = \left[\int a(k)M(dk)\right]^2 \tag{23}$$

and the characteristic function of (23) is

$$\phi(\xi) = \frac{1}{(1 - 2i\xi)^{1/2}}$$

As a straightforward generalization of above case let us consider the function

$$g(k, k') = \sum_{n=1}^{\infty} \lambda_n a_n(k)a_n(k') \tag{24}$$

where $\int a_n(k)a_{n'}(k)\, dk = \delta_{nn'}$, and $\lambda_n \geq 0$, $\Sigma_n \lambda_n < \infty$. Then

$$\int\int g(k, k')M(dk)M(dk') = \sum_n \lambda_n \left[\int a_n(k)M(dk)\right]^2$$

Since $a_n(k)$ are pairwise orthogonal, so are their stochastic integrals,

$$X_n = \int a_n(k)M(dk)$$

Since X_n are Gaussian random variables, they are mutually independent. Consequently X_n^2 are also independent. Thus the characteristic function of the corresponding state (19) is of the form

$$\mathrm{E}e^{i\xi \int\int g(k,k')M(dk)M(dk')} = \prod_{n=1}^{\infty} \mathrm{E}e^{\lambda_n \xi X_n^2} = \prod_{n=1}^{\infty} \frac{1}{(1 - 2i\xi\lambda_n)^{1/2}} \tag{25}$$

Observe that the right-hand side of (25) is nothing but the Fredholm determinant associated with the kernel (24).

The above formula which relates the characteristic functions of some interacting states with Fredholm determinants can be applied directly to operators with diagonal singularity. For example, it is enough to assume that A^{nd} is a nonnegative tracial operator and take as λ_n its eigenvalues arranged in a nonincreasing order (including multiplicities). If $\int a(k)\, dk = \mathrm{Tr}(A^{\mathrm{nd}})$, where $a(k)$ is the multiplier of A^{d}, then the characteristic function of the corresponding state (19) is also of the form (25). To see this it is enough to apply Mercer's theorem [15].

Applying the above technique, we can also find distributions of $u_n(t)$ and $u_n u_{n'}(t)$. In order to do this it is enough to split (10) [resp. (18)] into real and complex parts and calculate the corresponding characteristic functions.

We shall not discuss in this article the Liouville space description of lattices such as presented in ref. 6, which pointed out the necessity to go beyond the traditional Hilbert space structure. It follows from (18) that even to describe admissible interaction functions V for two particles we cannot confine ourselves to the "natural" Hilbert space of square-integrable functions.

Generally, considering states which involve interactions of s distinguished atoms $n_1, \ldots, n_s$, we obtain expressions of the form

$$\int \ldots \int V(k_1, \ldots, k_s)M_1(dk_1) \ldots M_s(k_s) \tag{26}$$

where $V(k_1, \ldots, k_s)$ is a function of s variables, and $M_j = M$ or $\overline{M}$. The necessary and sufficient conditions of the existence of such integrals also involve integrability of V on diagonals.

The stochastic approach presented in this paper concerns systems where the noise is of short range and affects only closest neighbors. We may also

consider long-range noise, but in such a case the central limit theorem cannot be applied to the scaling $N^{-1/2}$. Another scaling, like $N^{-1/p}$, $0 < p < 2$, would lead to stable Lévy stochastic measures and processes.

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