Systematic nonperturbative approach for thermal averages in quantum many-body systems: The thermal-cluster-cumulant method

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We present in this paper a systematic nonperturbative cluster-cumulant method for deriving thermal averages of operators in quantum many-body systems. The method combines the advantages of the cumulant expansion scheme of thermodynamic perturbation theory, the approach of thermofield dynamics as a finite-temperature field theory, and the time-dependent coupled-cluster theory extended to "imaginary time." We have generalized the concepts of cumulants in a nonperturbative manner and have posited on the statistical operator an exponential-like ansatz containing connected, size-extensive operators in the exponent. These latter cumulantlike operators have been termed "cluster cumulants" by us. For a compact treatment, we have derived an alternative thermal field theory in which a time-ordered product is expanded in terms of "thermal normal products" of operators and thermal contractions—leading to a "thermal Wick's theorem." The thermal normal products are the finite-temperature analogs of the ordinary normal products and have zero thermal averages. Operators in these products commute (anticommute) under permutations for bosons (fermions). This thermal representation is shown to be unitarily related to the traditional thermofield dynamics formulation, but has the advantage of using only the physical variables. The imaginary-time evolution of the statistical operator is treated by our recently formulated time-dependent cluster-cumulant theory. The partition function is evaluated as an exponential of a connected quantity. As an illustrative example, we have computed the partition function of an anharmonic oscillator with equally weighted cubic and quartic perturbation for a wide range of coupling, extending to the strongly nonperturbative regime. We study the behavior of free energy in the low-temperature limit and verify numerically the validity of the Kohn-Luttinger theorem [Phys. Rev. **118**, 41 (1960)] for this system. We also show that our formalism is a natural nonperturbative analog of the thermodynamic perturbative theory by showing that a perturbative solution of the thermal-cluster-cumulant equations generates a variation of the Bloch–Balian–de Dominicis theory.

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I. INTRODUCTION

The problem of systematically computing equilibrium thermal averages of quantum-mechanical observables has always remained a formidable challenge ever since the inception of quantum statistical mechanics. For interacting many-particle systems, there are two interrelated levels of complexity: (i) discerning the hierarchy of a scheme for systematic inclusion of the subset of interactions of gradually diminishing importance and (ii) development of a compact method of performing the thermal average (i.e., the Boltzmann trace) of the set of terms generated in the process (i) above. Most of the earlier developments were based on a perturbative construction, using the apparatus of field theory at finite temperature [1–3]. Several independent formulations were put forward, for example, of computing the grand partition function of a system of bosons or fermions or both, starting with the "equation of motion" of the statistical operator $e^{-\beta(H-\mu N)}$ [2,3]. The relationship between them is far from trivial, and there is vast literature in this subject. We mention a few selected references, viz., of Bloch, Balian, and deDominicis [4–6], following the earlier leads by Lee and Yang [7] and Montroll and Ward [8]. Although widely used, the perturbation method has two obvious limitations: it behaves poorly in the strongly correlated regime and it offers no systematic method of selectively summing important classes of terms (or diagrams, in the diagrammatic formulation) to all orders. Variational formulations have been suggested from time to time to bypass this difficulty [9]. In another line of development the method of thermal Green's functions was developed by Bloch [4], Matsubara [10], and Thouless [11] and has been extensively used since by many workers [1–3] in many different contexts. There are also the cumulant methods [12,13] and the imaginary-time convolution methods [14,15] which had been mainly invoked to study the micro-macro correspondence and the high-temperature limits, and for simulating classical baths. The path-integral methods have been exploited [16,17] by computing partition function, inspired by the earlier works of Feynman [18]. More recently, there has emerged the methodology of the thermofield dynamics [19–21]. Most of these methods exploit in one form or another the striking similarity of the time-evolution operator and the statistical operator and replace the thermal trace by either an integration over a suitable measure [16,17] or a true quantum-mechanical expectation value in an expanded Fock space [19–21]. The relation between the Matsubara formalism and that of

thermofield dynamics has been explored, too [22,23].

While both the path-integral formalism [16–18] and the approach of thermofield dynamics [20,21] provide general methodologies of finite-temperature field theory, their use hitherto has been rather limited in practical applications. The path-integral formalisms were mostly confined to variational approximations with perturbative corrections; no systematic nonperturbative avenues seem to have been explored. The methods of thermofield dynamics can go beyond the perturbative regime exploiting the selective summation techniques of the Feynman-Dyson strategy, but their use for systems with many degrees of freedom is rather complicated owing to the so-called "doubling" of the degrees of freedom.

We develop and illustrate in this paper an alternative nonperturbative approach for systematically computing thermal averages. The method combines the advantages of the earlier thermodynamic perturbation theory [4–6], the more recent formalism of the thermofield dynamics [20,21], and the nonperturbative coupled-cluster theory for treating time evolutions [24–26]. Inspired by the perturbative ideas of a cumulantlike expansion of the statistical operator $\exp(-\beta H)$, we propose a *nonperturbative* cumulant expansion involving connected cluster operators (to be henceforth called "cluster cumulants"). On the other hand, in close analogy with the method of thermofield dynamics, we shall interpret the process of evaluating the Boltzmann trace as computing expectation values by invoking the apparatus of field theory. However, unlike in the traditional formulation of thermofield dynamics, we shall work with *the physical variables only* and thus shall not invoke "doubling" [19–22] of the degrees of freedom. For a convenient, efficient, and compact formulation of our development, we shall find it useful to introduce a "thermal normal ordering" of the operators, associated with "thermal contractions" and generate a "thermal Wick's theorem."

Our finite-temperature field theory can be utilized in two distinct physical situations: (a) the real-time problems involving systems in contact with a bath or transport in a nonequilibrium steady state, and (b) the imaginary-time problems as required in treating equilibrium thermal properties. It is thus quite general in its scope. In the present paper, however, we shall concentrate only on the purely imaginary-time evolution of the statistical operator, leading to the partition function. Generation of the cluster cumulants in the statistical operator will be achieved by extending our recently formulated time-dependent coupled-cluster method [24–26], generalized to "imaginary time".

In our cluster-cumulant formulation, we shall partition our Hamiltonian into a one-body unperturbed component and a perturbation and write a cluster expansion of Z/Z_0, with Z and Z_0 as the exact and unperturbed partition functions, respectively. Z/Z_0 will be written as a thermal trace of a reduced statistical operator, written in a cluster expansion. If we can introduce a thermally normal-ordered product of operators such that their thermal trace is zero, then computation of the thermal trace of the reduced statistical operator can be conveniently performed if we write the cluster expansion in terms of thermally normal-ordered products: only the number component of the operator will survive while taking the trace to get Z/Z_0. The thermal Wick's theorem, to be employed by us, will facilitate precisely such an expansion.

This paper is organized as follows. In Sec. II we shall develop the apparatus of thermal Wick expansion and study the properties of the products of operators in thermal normal order. In Sec. III, we shall formulate our nonperturbative cluster-cumulant method and establish the necessary working equations. In Sec. IV, we shall analyze the structure of the perturbative theory generated by the perturbative expansion of our nonperturbative cluster-cumulant operators in the partition function and show the natural emergence of one version of the Bloch–Balian–deDominicis thermodynamic perturbation theory [4,5]. In Sec. V, we shall apply our formalism to compute the partition function of an anharmonic oscillator with equally weighted cubic and quartic perturbation for a wide range of the coupling constant. We shall also introduce there suitable diagrammatic techniques for generating the working equations and discuss the attendant diagrammatic rules. With our numerical illustration, we shall show numerically that the free energy approaches ground-state energy in the zero-temperature limit, i.e., we verify the validity of the Kohn-Luttinger theorem for this system. Section VI contains concluding remarks.

II. NOTION OF THERMAL NORMAL ORDERING AND THERMAL WICK EXPANSION

A. Motivation

Bloch and de Dominicis [4], Matsubara [10], as well as Thouless [11] showed that the thermal average of a string of products of creation-annihilation Bose and Fermi operators of an interacting system under the time ordering can be expressed in terms of a sum of products of "unperturbed" thermal averages of pairs containing one creation and one annihilation operator:

$$\begin{aligned}\langle\langle [A_I^{(1)}(\tau_1)A_I^{(2)}(\tau_2)\cdots]\rangle\rangle &\equiv \mathrm{Tr}e^{-\beta(H_0-\mu N)}T[A_I^{(1)}(\tau_1)A_I^{(2)}(\tau_2)\cdots]/Z_0 \\ &= \sum_{\text{all pairs}}\prod(-1)^{\eta_{ij}}[\mathrm{Tr}T[e^{-\beta(H_0-\mu N)}A_I^{(i)}(\tau_i)A_I^{(j)}(\tau_j)]/Z_0] \\ &= \sum\prod(-1)^{\eta_{ij}}\langle\langle A_I^{(i)}(\tau_i)A_I^{(j)}(\tau_j)\rangle\rangle\ , \end{aligned} \tag{2.1}$$

where $Z_0=\mathrm{Tr}e^{-\beta(H_0-\mu N)}$ is the "unperturbed" grand partition function with the chemical potential μ. H_0 is the unperturbed single-particle diagonal Hamiltonian

$$H_0=\sum \varepsilon_i[a_i^\dagger a_i] . \tag{2.2}$$

The sum in Eq. (2.2) runs over all the Bose and Fermi orbitals. N is the number operator for the system. The operators $A_I^{(i)}(\tau)$, etc., are creation annihilation operators in the "interaction picture" involving either imaginary time,

$$A_I^{(i)}(\tau)=e^{H_0\tau}A^{(i)}e^{-H_0\tau} , \tag{2.3}$$

or just operators in the conventional interaction picture,

$$A_I^{(i)}(\tau)=e^{IH_0\tau}A^{(i)}e^{-iH_0\tau} , \tag{2.4}$$

and T orders the operators in increasing order of τ from right to left. There are no phase contributions in Eq. (2.1) for bosons, while for fermions the phase $(-1)^{\eta_{ij}}$ will appear for anticommuting the operators η_{ij} times to bring $A_I^{(i)}(\tau_i)$ and $A_I^{(j)}(\tau_j)$ together in the thermal average of $A_I^{(i)}$ and $A_I^{(j)}$. Unlike the ordinary Wick's theorem, *which is an operator identity*, the Bloch–de Dominicis–Matsubara theorem, Eq. (2.1), *is an equation*, which is derived by utilizing the property of the invariance under the cyclical permutation of operators in a trace and the fact that Eq. (2.3) or (2.4) generates an $A_I^{(i)}(\tau)$ as a simple multiple of $A^{(i)}$, via Eq. (2.2). Equation (2.1) is *strikingly similar* to the Wick reduction formula of the vacuum expectation value of a string of operators under time ordering. The pair averages $\langle\langle T[A_I(\tau_i)A_i(\tau_j)]\rangle\rangle$ can be naturally identified here as "thermal contractions." We shall henceforth refer to them as Matsubara contractions. The question is: can we find out an expansion of a T-ordered product in terms of some suitably defined "normal-ordered" product of operators and products of thermal averages? The thermal normal-ordered products should have the property that their thermal averages are zero and also there is commutability (or anticommutability) of the associated Bose (or Fermi) variables under this normal ordering. In that case, we may envision that the thermal average of the T-ordered product would naturally reduce to the Matsubara formula, Eq. (2.1), on taking the trace. This type of expansion, if it exists, will afford us a compact algebraic form with which to compute selective summations and similar manipulations which have been found to be useful in the analogous zero-temperature formalisms. In particular, since the time differentiation of the thermally normal-ordered operators should be rather easy; this type of expansion will allow us to generate nonperturbative cumulant expansions involving thermally normal-ordered operators. As we shall illustrate below, there is indeed a unique and natural thermally normal-ordered expansion.

B. Matsubara contractions interpreted as averages of Gaussian stochastic variables

We begin our discussion by noting that there is a close algebraic similarity between the Matsubara formula, Eq. (2.1), with the relation obeyed by the stochastic average of a product of Gaussian stochastic variables $\{x_i\}$ with zero mean:

$$\langle x_i x_2 \cdots x_{2n}\rangle=\sum_{\substack{\text{product}\\ \text{of pairs}}}\prod\langle x_i x_j\rangle . \tag{2.5}$$

Although not widely known, for such variables it is possible to define the so-called Wick powers $\{x_1^{n_1}x_2^{n_2}\cdots x_k^{n_k}\}$ uniquely [27] as symmetric functions of $x_1,x_2,\ldots,$ such that their stochastic averages are zero. The expansion expressing an ordinary monomial $x_1^{n_1}x_2^{n_2}\cdots x_k^{n_k}$ in terms of Wick powers may then be interpreted as an identity closely analogous to Wick's theorem. This suggests strongly the possibility of expressing the time-ordered products of operators in Eq. (2.1) in terms of analogous Wick powers—by interpreting the operators $A^{(i)}(\tau_i)$ as Gaussian stochastic variables and introducing appropriate generalizations of the Wick powers for quantum operators. There are nontrivial differences, of course, between the variables x_i and operators A^i: (i) the variables $A_I^{(i)}(\tau)$ are noncommuting Bose or Fermi operators and are not c numbers; (ii) there is a time ordering in the definition of averages; (iii) there are phase factors $(-1)^{\eta_{ij}}$ for the fermion variables. As we show below, despite these differences, it is possible to look upon the operators A^I as Gaussian stochastic variables: The T ordering makes the operators $A_I^{(i)}(\tau)$ commute under the T-ordered symbol (except for a sign); they thus behave either as commuting variables (for bosons) or as Grassmann variables [2,28] (for fermions).

By invoking certain auxiliary Grassmann numbers [28], it is possible to map the T-ordered products of fermion operators to a set of corresponding boson operators. Let us suppose that we replace the fermion variables $A_F^i(\tau_i)$ by a product $B_I^i(\tau_i)\gamma_i$, where B_I^i is a boson variable and γ_i is a member from a set of even number of Grassmann numbers. Similarly, $A_I^{(i)\dagger}$ gets replaced by $B_I^{(i)\dagger}\gamma_i^*$. The operator $\{B_I^i\}$ and the numbers $\{\gamma_j\}$ are assumed to commute. With this replacement, the phase η_{ij}, entering the T-ordered products involving the permutation of the Fermi operators $A_I^{(i)}$ and $A_I^{(j)}$ will be automatically taken care of by the Grassmann numbers γ_i and γ_j. This follows in a straightforward manner from the anticommuting nature of the Grassmann numbers $\{\gamma_i\}$:

$$\begin{aligned}P_{12}\{T[A_I^{(1)}(\tau_1)A_I^{(2)}(\tau_2)]\}&=\gamma_2\gamma_1 T[B_I^{(2)}(\tau_2)B_I^{(1)}(\tau_1)]\\&=-\gamma_1\gamma_2 T[B_I^{(1)}(\tau_1)B_I^{(2)}(\tau_2)]\\&=-T[A_I^{(1)}(\tau_1)A_I^{(2)}(\tau_2)] ,\end{aligned} \tag{2.6}$$

where P_{12} is the permutation operator.

Using Eq. (2.6), it is possible to replace all the fermion

variables by the corresponding boson variables and keep track of the order of appearance of the Grassmann numbers appearing in front of the time ordering, as in Eq. (2.6). Any theorem proved for the T-ordered product of Bose variables B_I can be finally mapped back to the corresponding Fermi variables by multiplying them with the associated Grassmann numbers. As an example, we verify that since

$$\langle\langle T[B_I^{(i)}(\tau_i)B_I^{(j)}(\tau_j)]\rangle\rangle = \langle\langle T[B_I^{(j)}(\tau_j)B_I^{(i)}(\tau_i)]\rangle\rangle \ ,$$

the antisymmetry under permutation of the Matsubara contraction involving Fermi operators $A_I^{(i)}(\tau_i)$ and $A_I^{(j)}(\tau_j)$ easily follows:

$$\begin{aligned}\langle\langle T[A_I^{(i)}(\tau_i)A_I^{(j)}(\tau_j)]\rangle\rangle &= \gamma_i\gamma_j\langle\langle T[B_I^{(i)}(\tau_i)B_I^{(j)}(\tau_j)]\rangle\rangle \\ &= -\gamma_j\gamma_i\langle\langle T[B_I^{(j)}(\tau_j)B_I^{(i)}(\tau_i)]\rangle\rangle \\ &= -\langle\langle T[A_I^{(j)}(\tau_j)A_I^{(i)}(\tau_i)]\rangle\rangle \ .\end{aligned} \tag{2.7}$$

Let us assume, then, that we have replaced each fermion variable by a product of boson variable and the associated Grassmann number, and henceforth we would be working with an expression containing A_I's of the original boson variables and $B_I^{(i)}(\tau)$'s for the new boson variables replacing the Fermi variables as discussed above. No phase factors appear then in the thermal average of these latter products:

$$\langle\langle T[A_I^{(1)}(\tau_1)A_I^{(2)}(\tau_2)\cdots B_I^{(i)}(\tau_i)B_I^{(j)}(\tau_j)\cdots]\rangle\rangle = \sum_{\text{all pairs}}\prod\langle\langle T[A_I^{(p)}(\tau_p)A_I^{(q)}(\tau_q)]\rangle\rangle\langle\langle T[B_I^{(i)}(\tau_i)B_I^{(j)}(\tau_j)]\rangle\rangle \ . \tag{2.8}$$

Equation (2.5) is restored by multiplying Eq. (2.8) by the appropriate product of Grassmann numbers $\gamma^i\gamma^j\cdots$, appearing in the order of appearance of the operators $B_I^{(i)}, B_I^{(j)}, \ldots$.

Since Eq. (2.8) satisfies all the properties of stochastic averages of Gaussian stochastic commuting variables, Eq. (2.5), we may introduce the "thermal normal-ordered" product $\{\cdots A_I^{(i)}(i)\ldots B_I^{(j)}(j)\ldots\}_\beta$ of the above variables which should satisfy, in close analogy with the property of Wick's power [27], the following desirable properties:

$$\text{(i)} \quad \{A_I^{(i)}\cdots B_I^{(j)}\cdots\}_\beta = P\{A_I^{(i)}\cdots B_I^{(j)}\cdots\}_\beta \tag{2.9}$$

where P is any permutation;

$$\text{(ii)} \quad \langle\langle\{A_I^{(i)}\cdots B_I^{(j)}\cdots\}_\beta\rangle\rangle = 0 \ . \tag{2.10}$$

(iii) The relation of the above normal product with the T-ordered product should not depend in any special way on the total number of variables and should be a *manifestly symmetric function* of all the variables, compatible with Eq. (2.9).

The only sensible choice for $\{C_I(\tau 1)D_I(\tau_2)\}_\beta$ for two arbitrary Bose operators $C_I(\tau_1)$ and $D_I(\tau_2)$ (which may be A_I or B_I) is the following:

$$\begin{aligned}\{C_I(\tau_1)D_I(\tau_2)\}_\beta &= T[C_I(\tau_1)D_I(\tau_2)] - \langle\langle T[C_I(\tau_1)D_I(\tau_2)]\rangle\rangle \\ &= T[C_I(\tau_1)D_I(\tau_2)] - [\overline{C_I(\tau_1)D_I(\tau_2)}]_\beta \ ,\end{aligned} \tag{2.11}$$

where the overbar is an alternative notation for the Matsubara contraction. For three operators C_I, D_I, and E_I, we can construct the normal-ordered product in terms of symmetrized products of operators, viz., from T-ordered product of three operators and $\{\}_\beta$ products of two operators, keeping the manifest symmetry of all the variables. Thus we define

$$\{C_I(\tau_1)D_I(\tau_2)E_I(\tau_3)\}_\beta \equiv \{C_ID_IE_I\}_\beta = C_1T[C_ID_IE_I] + C_2\{\overline{C_ID_I}\}_\beta E_I + C_3\{\overline{C_IE_I}\}_\beta D_I + C_4C_I\{\overline{D_IE_I}\}_\beta \ . \tag{2.12}$$

Since we want the functional definition of $\{\}_\beta$ to be independent of number of variables, Eq. (2.12) should reduce to the definition (2.11) if one of the variables in Eq. (2.12) is deleted. Thus, for example, if we delete C_I, we get

$$\{D_IE_I\}_\beta = C_1T[D_IE_I] + C_4\{D_IE_I\}_\beta \tag{2.13}$$

showing that $C_1 = -C_4 = 1$, from Eq. (2.11). From symmetry, it then follows that

$$\{C_ID_IE_I\}_\beta = T[C_ID_IE_I] - \{\overline{C_ID_I}E_I\}_\beta - \{\overline{C_ID_IE_I}\}_\beta - \{C_I\overline{D_IE_I}\}_\beta \ , \tag{2.14}$$

where $\{\overline{C_ID_IE_I}\}_\beta$ are symbols for $[\overline{C_IE_I}]_\beta D_I$. It may be easily verified that

$$\langle\langle\{C_ID_IE_I\}_\beta\rangle\rangle = 0 \tag{2.15}$$

which is consistent with Eq. (2.6).

For four variables, we should likewise have

$$\begin{aligned}\{C_ID_IE_IF_I\}_\beta = C_0T[C_ID_IE_IF_I] + (C_1[\overline{C_ID_I}E_IF_I] + (\text{other similar terms}) \\ + (C_2[\overline{C_ID_I}\,\overline{E_IF_I}] + (\text{other similar terms}) \ .\end{aligned} \tag{2.16}$$

Deleting any one of the variables should lead to Eq. (2.14), thereby showing $C_0 = 1$, and C_1 and similar coefficients

$=-1$. By demanding the expectation value $\langle\langle\{C_I D_I E_I F_I\}_\beta\rangle\rangle$ to vanish, and using Eq. (2.8) for $\langle\langle T[C_I D_I E_I F_I]\rangle\rangle$, it follows that C_2 and other similar coefficients are also -1. Proceeding upwards, it then generally follows that

$$\begin{aligned}\{C_I^{(1)}(\tau_1)C_I^{(2)}(\tau_2\cdots C_I^{(i)}(\tau_i)\cdots C_I^{(j)}(\tau_j)\}_\beta = T[C_I^{(1)}(\tau_1)\cdots] \\ -\{C_I^1(\tau_1)\cdots \overline{C_I^{(i)}(\tau_i)\cdots C_I^{(j)}(\tau_j)]_\beta}\cdots\}_\beta - \cdots \\ -\{C_I^1(\tau_1)\cdots \overline{C_I^{(i)}(\tau_i)\overline{C_I^{(k)}(\tau_k)}\overline{C_I^{(j)}(\tau_j)C_I^{(1)}(\tau_1)}}\cdots\}_\beta \\ -(\text{all possible contractions}) . \end{aligned} \tag{2.17}$$

We should note that with our "overbar" notation for contraction, only the two operators appearing on the extreme sides under the overbar are really contracted. Other operators in between are not contracted at all. If we now multiply Eq. (2.17) by the appropriate Grassmann numbers, to convert the B_I operators to the parent Fermi operators, then we have our stipulated "thermal normal product" expression of a T-ordered product:

$$\begin{aligned}T[A_I^{(1)}(\tau_1)A_I^{(2)}(\tau_2)\cdots A_I^{(i)}(\tau_i)\cdots A_I^{(j)}(\tau_j)\cdots] = [A_I^{(1)}(\tau_1)A_I^{(2)}(\tau_2)\cdots A_I^{(i)}(\tau_i)\cdots A_I^{(j)}(\tau_j)\cdots]_\beta \\ +[\{A_I^{(1)}(\tau_1)\cdots \overline{A_I^{(i)}(\tau_i)\cdots A_I^{(j)}(\tau_j)}\cdots\}_\beta + \cdots] \\ +(\text{double contractions})+(\text{triple contractions})+\cdots . \end{aligned} \tag{2.18}$$

For fermions, using the anticommuting nature of the Grassmann numbers, we have the definition

$$\{A_I^{(1)}(\tau_1)\cdots \overline{A_I^{(i)}(\tau_i)\cdots A_I^{(j)}(\tau_j)}\cdots\}_\beta = (-1)^{\eta_{ij}}[\overline{A_I^{(i)}(\tau_i)A_I^{(j)}(\tau_j)}]_\beta\{A_I^{(1)}(\tau_1)\cdots\}_\beta , \tag{2.19}$$

where $(-1)^{\eta_{ij}}$ is the phase of the permutation needed to bring $A_I^{(i)}$ and $A_I^{(j)}$ side by side and on the place indicated and the term in angular brackets on the right misses $A_I^{(i)}$ and $A_I^{(j)}$. Equation (2.18) is obviously consistent with Eq. (2.7) *by construction*.

We can also verify that a generalized Wick expansion also holds good:

$$\begin{aligned}T[\{C_I D_I\cdots\}_\beta\{F_I G_I\cdots\}] = \{C_I D_I\cdots F_I G_I\cdots\}_\beta \\ +(\text{all possible contractions between the operators from the first group} \\ \text{and those from the second group}) . \end{aligned} \tag{2.20}$$

The expressions in Eq. (2.18) and (2.20) above will be our *central building blocks* in formulating the thermal-cluster-cumulant expansion. We should note that, unlike the ordinary normal products, in the thermal normal ordering *we do not reorder the operators with destruction at the right.* Also, we have generally *nonzero values* for both $[\overline{a_{iI}^\dagger a_{jI}}]_\beta$ and $[\overline{a_{jI}a_{iI}^\dagger}]$. The values of the contractions are

$$\overline{a_{iI}^\dagger(\tau_1)a_{jI}(\tau_2)} = \delta_{ij}\exp[\varepsilon_i-\mu)(\tau_1-\tau_2)]\times \begin{cases} n_i & \text{for } \tau_1>\tau_2 \quad (2.21a) \\ 1\mp n_i & \text{for } \tau_1<\tau_2 \quad (2.21b) \end{cases}$$

with upper (lower) signs for fermions (bosons). n_i's are the unperturbed occupation probabilities and are given by

$$n_i = 1/[e^{\beta(\varepsilon_i-\mu)}\pm 1] . \tag{2.22}$$

ε_i are the unperturbed energies of the system:

$$H_0 = \sum \varepsilon_i a_i^\dagger a_i . \tag{2.23}$$

Moreover, by setting the variables $\tau_1,\ldots,\tau_n$ as $\tau_n=0$, $\tau_{n-1}=\delta$, $\tau_{n-2}=2\delta,\ldots$, and letting $\delta\to 0+$, we may derive—if desired— the thermal Wick expansion for products of time-independent variables.

We should mention here that the notion of a thermal normal product expansion was also explored by Balian and Veneroni [29] who considered a recursive definition of the thermally normal-ordered product.

III. THE THERMAL-CLUSTER-CUMULANT METHOD FOR THE STATISTICAL OPERATOR

Let us assume that the system Hamiltonian H has a one-body unperturbed Hamiltonian H_0 and a perturbation V:

$$H = H_0 + V . \tag{3.1}$$

The grand partition function Z for the system is defined as

$$Z = \mathrm{Tr}\exp[-\beta(H-\mu N)] , \tag{3.2}$$

where μ is the chemical potential of the system. The analogous unperturbed partition function Z_0 is given by

$$Z_0 = \mathrm{Tr}\exp[-\beta(H_0-\mu N)] . \tag{3.3}$$

Introducing the operator $U\equiv U(\beta)$ as $\exp[-\beta(H-\mu N)]$, we can write the "imaginary-time" Heisenberg equation for U in the "interaction picture"

[2] as

$$-\frac{\partial U_I(\tau)}{\partial \tau} = V_I(\tau) U_I(\tau) , \tag{3.4}$$

where U_I and V_I are given by

$$U_I(\tau) = \exp[\tau(H_0 - \mu N)] \exp[-\tau(H - \mu N)] , \tag{3.5}$$

$$V_I(\tau) = \exp[\tau(H_0 - \mu N)] V \exp[-\tau(H_0 - \mu N)] . \tag{3.6}$$

Equation (3.4) leads to the following Feynman-Dyson expression for $U_I(\beta)$:

$$U_I(\beta = T\left[\exp\left[\int_0^\beta - V_I(\tau) d\tau\right]\right] . \tag{3.7}$$

This leads, via Eq. (3.5), to the expression for Z/Z_0:

$$Z/Z_0 = \mathrm{Tr}\{\exp[-\beta(H_0 - \mu N)] U_I(\beta)\} / \mathrm{Tr} \exp[-\beta(H_0 - \mu N)] , \tag{3.8}$$

which we may compactly rewrite as

$$Z/Z_0 = \langle\langle U_I(\beta) \rangle\rangle = \left\langle\left\langle T \exp \int_0^\beta [-V_I(\tau) d\tau] \right\rangle\right\rangle \tag{3.9}$$

using the definition of the thermal average.

If we expand the T-ordered exponential in Eq. (3.9) in powers of perturbation, we will get then a perturbative expansion of Z/Z_0. The difficulty with such an expansion is that, any truncated expression will lack the multiplicative separability of Z for an ensemble of weakly interacting subsystems, and as a result, the logarithm of Z—related to the free energy—will not be additively separable. Such truncations thus *violate the property of extensivity* of the computed extensive thermodynamic quantities. In the many-body parlance, this feature is expressed as the appearance of disconnected terms in $\ln(Z/Z_0)$. Using the cumulant expansion [12,13], it is possible to devise approximations which satisfy the additive separability of $\ln(Z/Z_0)$, which implies the *presence of connected quantities only*. These connected entities—denoted as cumulants—can be generated by first writing

$$Z/Z_0 = \exp(-\beta C) . \tag{3.10}$$

and expanding C in order of V_I. By equating the nth-order term of Z/Z_0 from Eq. (3.10) to the corresponding term of Eq. (3.9), the nth-order cumulant $C^{(n)}$, may be obtained. This procedure, in fact, forms the cornerstone of the Kubo-Fox perturbative operator cumulant expansion [12,13] and is also conceptually very close to the Bloch–Balian–de Dominicis thermodynamic perturbation theory [4–6]. Perturbative cumulants also appear implicitly in many path-integral formulations [16,17].

In our formulation, too, we express $\ln(Z/Z_0)$ as a connected entity, which we want to evaluate in a nonperturbative manner. If we expand the T-ordered product in Eq. (3.9) using our thermal Wick's theorem expounded in Sec. II [Eq. (2.18) and (2.20)], and regroup all the connected operators together in an entity to be denoted as $A_I(\beta)$, then by combinatoric reasoning $U_I(\beta)$ can directly be written as a thermally normal-ordered exponential of $A_I(\beta)$:

$$U_I(\beta) = \{\exp[A_I(\beta)]\}_\beta . \tag{3.11}$$

In the expansion, Eq. (3.11) above, the thermal normal ordering prevents contractions between the various $A_I(\beta)$ operators in $U_I(\beta)$. In what follows, we shall derive the equations for determining the various components of $A_I(\beta)$.

Each of the components of $A_I(\beta)$ has a specific particle rank dictated by the number of creation and annihilation operators. Broadly speaking, these components may be divided into two distinct categories. In one, we have "diagonal" operators in which there is *no change* in occupancy before and after the scattering. They thus induce scattering from any function to the same function. We shall call such operators as "closed" from now on, and shall denote then as $X_I(\beta)$. The rest of the operators are nondiagonal in the sense that they scatter from one function to a different function, and induce *at least one* change of occupancy after its action on a function. We denote these operators as $S_I(\beta)$ and call them as "external." The external operators are of excitation or deexcitation types, depending on whether they induce transitions to higher-energy states or to lower-energy states.

We illustrate the above classification of the operators by a specific example. Let us assume that we have a Hamiltonian describing a bosonic system involving only one type of boson (see, e.g., our example application in Sec. V). Denoting the creation and annihilation operators for the boson as $b^\dagger$ and b, the "closed operators" $X_I(\beta)$ for the problem are of the

$$X_I(\beta) = \sum_{n=0}^{\infty} [1/(n!)^2] X_I^n(\beta) b^{\dagger n} b^n \equiv \sum_{n=0} X_{n1}(\beta) , \tag{3.12a}$$

where each $X_I(\beta)$ induces a diagonal scattering involving no change in occupancy of the bosons. Similarly, the "external" operators $S_1(\beta)$ can be written as

$$S_1(\beta) = \sum_{\substack{m,n \\ m \neq n}} S_I^{m,n}(\beta) \equiv \sum_{\substack{m,n \\ m \neq n}} [1/(m!n!)] S_I^{m,n}(\beta) b^{\dagger m} b^n \equiv \sum_{\substack{m,n \\ m \neq n}} S_{m,n1}(\beta) \tag{3.12b}$$

which induce changes in the occupancy. The $m > n$ type of operators are excitation types, while the $m < n$ type of

operators are deexcitation types. For more than one single-particle states, there is an obvious generalization.

With the above classification in mind, let us write Eq. (3.11) as

$$U_I(\beta)=\{\exp[A_I(\beta)]\}_\beta = \{\exp[S_1(\beta)+X_1(\beta)]\}_\beta . \tag{3.13}$$

Our intention now is to derive differential equations for $S_I(\tau)$ and $X_I(\tau)$, which can be integrated to yield expressions for $S_I(\beta)$ and $X_I(\beta)$.

Substituting Eq. (3.13) into Eq. (3.4), and utilizing the fact that operators commute, apart from a possible phase factor under thermal normal ordering, we derive

$$-\left\{\left[\frac{\partial S_I(\tau)}{\partial\tau}+\frac{\partial X_I(\tau)}{\partial\tau}\right]\exp[S_I(\tau)+X_I(\tau)]\right\}_\beta = V_I(\tau)\left\{\exp[S_I(\tau)+X_I(\tau)\right\}_\beta . \tag{3.14}$$

The advantage of using a thermal normal-ordered ansatz for U_I is evident, since we have a rather simple expression for the "time derivative" of U_I. The S_I and X_I operators with boson variables commute with U_I under $\{\}_\beta$, while for fermions there are always even number of fermion variables in S_I and X_I and thus they again commute with U_I under $\{\}_\beta$. Using again the generalized thermal Wick's theorem [Eq. (2.20)] on the right side of Eq. (3.14), we have

$$-\left\{\left[\frac{\partial S_I(\tau)}{\partial\tau}+\frac{\partial X_I(\tau)}{\partial\tau}\right]\exp[S_I(\tau)+X_I(\tau)]\right\}_\beta = \{\overline{V_I(\tau)\{\exp[S_I(\tau)+X_I(\tau)]\}}\{\exp[S_I(\tau)+X_I(\tau)]\}\}_\beta , \tag{3.15}$$

where $\overline{V_I\exp(S_I+X_I)}$ involves thermal contractions between V_I and various powers of S_I and X_I from the exponential, omitting contractions among S_I and X_I. Since the various powers of S_I and X_I are *linearly independent*, it follows that [24–26]

$$-\frac{\partial S_I(\tau)}{\partial\tau}=\{\overline{V_I\exp[S_I(\tau)+X_I(\tau)]}\}_{\rm ex} , \tag{3.16a}$$

$$-\frac{\partial X_I(\tau)}{\partial\tau}=\{\overline{V_I\exp[S_I(\tau)+X_I(\tau)]}\}_{\rm cl} , \tag{3.16b}$$

where we have equated the external and closed components of $\{\overline{V_I\exp(S_I+X_I)}\}_\beta$ to $-\partial S_I(\tau)/\partial\tau$ and $-\partial X_I(\tau)/\partial\tau$, respectively. In Eqs. (3.16), those operators of the composite $\{\overline{V_I\exp(S_I+X_I)}\}_\beta$ are external which involve at least one change of occupancy. Similarly, those operators of the composite $\{\overline{V_I\exp(S_I+X_I)}\}_\beta$ are closed which involve no change in occupancy. The subscripts ex and cl, respectively, stand for the external and closed components of an operator.

Equations (3.16) are the principal working equations of our cluster-cumulant theory. The connectedness of the right-hand side of the equations indicates that $S_I(\beta)$ and $X_I(\beta)$ will be connected operators, justifying the term "cluster cumulant" used to denote them. Any truncations of the right side of Eq. (3.16) *would still preserve the connectedness of* the S_I and X_I. Since Z/Z_0 is the thermal average of U_I, only the zero-body (number) component of U_I will survive in Z/Z_0, owing to the thermal normal ordering present in the expression of U_I, Eq. (3.13); this in turn implies that only the *zero-body part* of the cluster-cumulant operators will contribute to Z/Z_0. This, of necessity, will be a closed operator, which we may denote as X_I^0. The advantage of thermal normal ordering is again evident, since otherwise we would have had contractions from the operators S_I and X_I in Z/Z_0. Thus we have

$$Z/Z_0=\exp(X_I^0) . \tag{3.17}$$

We can now generate all the thermodynamic quantities from Eq. (3.17). In particular, the free energy of the ensemble is given by

$$F=F_0-X_I^0/\beta . \tag{3.18}$$

The unperturbed free energy F_0 is to be computed from the knowledge of the unperturbed partition function. Since X_I^0 is a connected quantity [even when computed via some truncated series of Eq. (3.16)], it follows that $\Delta F=F-F_0$ is also a connected quantity and hence is an extensive quantity. The chemical potential μ should be determined from the condition

$$\begin{aligned}\bar{N}&=\langle\langle NU_I\rangle\rangle/\langle\langle U_I\rangle\rangle\\&=[\langle\langle N(1+X_I^{(1)})\rangle\rangle\exp(X_I^{(0)})]/[\exp(X_I^0)]\\&=\langle\langle N(1+X_I^{(1)})\rangle\rangle .\end{aligned} \tag{3.19}$$

In Eq. (3.19) above, only the one-body component of the diagonal operator $X_I^{(1)}$ can contribute to $\langle\langle NU_I\rangle\rangle$, and only the zero-body component $X_I^{(0)}$ can contribute to $\langle\langle U_I\rangle\rangle$. From the value of the observed average particle number $\bar{N}$, μ can be found out by a self-consistent solution of the set of equations (3.16) and (3.19).

Since there are both exponentially growing and decaying terms in U_I, a direct solution of Eq. (3.16) might entail potential "stiffness" and hence numerical instability. To obviate this, it seems better (unlike the real-time theories) to revert to the "Schrödinger picture," and solve the corresponding cluster operators $S(\tau)$ and $X(\tau)$, defined as

$$S(\tau)=\exp[-\tau(H_0-\mu N)]S_I(\tau)\exp[\tau(H_0-\mu N)] , \tag{3.20a}$$

$$X(\tau)=\exp[-\tau(H_0-\mu N)]X_I(\tau)\exp[\tau(H_0-\mu N)] . \tag{3.20b}$$

The cluster-cumulant equations for $S(\tau)$ and $X(\tau)$ are given by

$$-\frac{\partial S(\tau)}{\partial \tau}=[H_0-\overline{\mu N S(\tau)}-\overline{S(\tau)[H_0-\mu N]}+\{\overline{V\exp(S+X)}\}_{\rm ex}\ , \tag{3.21a}$$

$$-\frac{\partial X(\tau)}{\partial \tau}=[\overline{H_0-\mu N]x}-\overline{x[H_0-\mu N]}+\{\overline{V\exp(S+X)}\}_{\rm cl}\ . \tag{3.21b}$$

In our numerical applications, we shall solve for $S(\tau)$ and $X(\tau)$ first from Eq. (3.21), then go over to $S_I(\tau)$ and $X_I(\tau)$ via the inverse transforms of Eq. (3.20) and finally compute Z/Z_0 from Eq. (3.17). We should mention that Altenbokum *et al.* [30] had suggested earlier a coupled-cluster approach for thermal averages, which requires the knowledge of the eigenspectra of H, and is thus structurally very different from our more direct formulation.

We conclude this section by emphasizing that the thermal field theory developed by us in Secs. II and III never explicitly used the concept of a thermal vacuum nor introduced auxiliary variables [19–22] for computing the thermal averages. The thermal Wick's theorem derived by us requires only the knowledge of contractions like $[\overline{A_I^{(i)}A_I^{\dagger(j)}}]_\beta$ and $[\overline{A_I^{+(j)}A_I^{(i)}}]_\beta$. While this is sufficient for developing the thermal-cluster-cumulant theory, one may envisage another possible realization of the thermal field theory where there is an explicit appearance of a thermal vacuum and of normal ordering in the traditional sense. We have shown in Appendix A, there is indeed such an alternative representation which is related to the traditional formulation of thermofield dynamics [18–20].

IV. PERTURBATIVE SOLUTION OF THERMAL-CLUSTER-CUMULANT EQUATIONS: EMERGENCE OF A VARIANT OF BLOCH–BALIAN–de DOMINICIS PERTURBATION THEORY

In this section, we shall indicate a way to generate a perturbative solution of our cluster-cumulant equations. The resulting equations are the analogs of one of the variants of the perturbative cumulant results of Bloch, Balian, and de Dominicis [4,5], which defines a perturbative series for the free energy as powers of V_I and the unperturbed occupation probabilities n_i [see, e.g., Eq. (2.22)]. We shall illustrate our perturbative approach in Sec. V by taking as a concrete example the case of the anharmonic oscillator.

To effect the perturbative construction, it is more convenient to start with the thermal-cluster-cumulant equations in the interaction picture, and expand the cluster cumulants S_I and X_I in orders of perturbation. This leads to a perturbative solution for S_I and X_I at order n, ${}^{(n)}S_I$, and ${}^{(n)}X_I$, starting from solutions for S_I and X_I at lower orders. Since the relevant contractions involved in the cluster-cumulant equations refer to the unperturbed occupation probabilities n_i [e.g., Eq. (2.21)], this mode of solution would entail a power-series expansion of S_I and X_I in terms of V_I and unperturbed occupation probabilities.

At the first order, we have

$$-\frac{\partial^{(1)}S_I(\tau)}{\partial \tau}=\{V_I(\tau)\}_{\rm ex}\ , \tag{4.1}$$

$$-\frac{\partial^{(1)}X_I(\tau)}{\partial \tau}=\{V_I(\tau)\}_{\rm cl}\ . \tag{4.2}$$

At second order, likewise, we have

$$-\frac{\partial^{(2)}S_I(\tau)}{\partial \tau}=\{\overline{V_I(\tau)^{(1)}S_I(\tau)}\}_{\rm ex}+\{\overline{V_I(\tau)^{(1)}X_I(\tau)}\}_{\rm ex} \tag{4.3}$$

$$-\frac{\partial^{(2)}X_I(\tau)}{\partial \tau}=\{\overline{V_I(\tau)^{(1)}S_I(\tau)}\}_{\rm cl}+\{\overline{V_I(\tau)^{(1)}X_I(\tau)}\}_{\rm cl}\ . \tag{4.4}$$

Clearly, the contribution to Z/Z_0 comes only from the zero-body component X_I^0, which has a nonvanishing contribution starting at second order. This is because ${}^{(1)}X_I^0$, from Eq. (4.2), involves the completely contracted component of $[V_I(\tau)]_{\rm cl}$, which is zero owing to the normal ordering of V_I.

Writing in long hand, we have, for ${}^{(1)}S_I$:

$${}^{(1)}S_I(\tau)=-\int_0^\tau [V_I(\tau_I)]_{\rm ex}d\tau_I =-\int_0^\tau [e^{L_0\tau_1}V]_{\rm ex}d\tau_1\ , \tag{4.5}$$

where L_0 is the Liouvillian Superoperator: $L_0V\equiv[H_0,V]$.

Substituting this value in Eq. (4.4), we find

$${}^{(2)}X_I^0(\beta)=\int_0^\beta e^{L_0\tau_1}\left[\overline{V\,d\tau_1\int_0^{\tau_1}e^{L_0\tau_2}V}\right]^0 d\tau_2 =\int_0^\beta e^{L_0\tau_1}(L_0^{-1})[\overline{V(e^{L_0\tau}1-1)V}]^0 d\tau_1\ , \tag{4.6}$$

where the quantity $[\ ]^0$ indicates the zero-body (number) component. This procedure can be repeated to generate higher order ${}^{(n)}X_I^0$'s. In our perturbative solution, $S_I(\tau)$ and $X_I(\tau)$ are obtained first as integrals with time integrations from 0 to τ in Eqs. (4.3) and (4.4). $X_I(\beta)$'s are then obtained as integrals with time going from 0 to β. At second order, we thus have two time integrations over τ_1 and τ_2 with $\tau_1>\tau_2$. At higher order n, ${}^{(n)}X_I^0$ will involve multiple integrals with limits $\beta\geq\tau_1\geq\tau_2\geq\tau_3\cdots\tau_n\cdots\geq\tau_n$. In the literature, there exists several versions of the thermodynamic perturbation theory involving series expansion in V_I and unperturbed occupation probabilities, which differ in ways the time integrations are carried out. In some variants, the time integrations are integrated with restrictions such as $\tau_1>\tau_2$, etc. Our perturbative solution corresponds precisely to such a development. There are other variants where the time arguments are independently integrated from 0 to β; the restrictions $\tau_1\geq\tau_2$ are enforced via suitable Heavy-

side step functions [4,5]. This way of rewriting the expression under the integrals for the perturbative cumulants is convenient to study the zero temperature, or the Kohn-Luttinger limit. There are also related formulations, where the integrations involving step functions are performed using suitable contours in the complex time plane—yielding formulas strikingly similar to the zero-temperature stationary Brückner Goldstone theory.

Balian, Bloch, and de Dominicis [6] had formulated another resummed version of the perturbative formulation, which involves "perturbed" or exact occupation probabilities. This is very useful for treating translationally invariant systems. Our present cluster-cumulant formulation does not naturally generate this perturbation series. By introducing a suitable self-consistent one-particle potential, we may, however, generate such a resummed version. This will also be very useful in studying the zero-temperature limit. We shall discuss the formal aspects of the convergence of the stationary coupled-cluster theory in the zero-temperature limit in a forthcoming publication [31], using the resummed version of the cluster-cumulant theory.

V. ILLUSTRATIVE APPLICATIONS: THE ANHARMONIC OSCILLATOR WITH CUBIC AND QUARTIC PERTURBATIONS

A. An optimal zeroth-order description: The thermal Hartree function

We shall apply the thermal-cluster-cumulant method to compute the partition function of an anharmonic oscillator with a cubic-plus-quartic perturbation. The Hamiltonian for our problem is given by

$$H=a^{\dagger}a+\tfrac{1}{2}+2^{-3/2}\gamma(a^{\dagger}+a)^3+\lambda/4(a^{\dagger}+a)^4 . \tag{5.1}$$

This has been used earlier for studying the eigenspectra of anharmonic oscillators by coupled-cluster methods [32,33]. The unperturbed frequency is taken as unity. In our numerical application, we shall ultimately take $\gamma=\lambda$. This equally weighted cubic-plus-quartic perturbation produces very asymmetric potentials as λ increases and the spectra of H differ significantly from that of $H_0=a^{\dagger}a+\frac{1}{2}$ in the large-coupling regime. Since the entire spectrum of H contributes to Z, the anharmonic oscillator with an asymmetric perturbation should turn out to be a stringent testing ground of our formalism. The case of symmetric perturbations, such as quartic and sextic terms in the potential, have been found to be somewhat better behaved in the earlier numerical studies of the partition function [17,34], presumably because the unperturbed and the perturbed Hamiltonians have the same symmetry. In contrast, the cubic-plus-quartic perturbation destroys the inversion of the system, exerting greater demands on the theory.

Since we want to employ our formalism well into the strongly nonperturbative regime, i.e., $\lambda\gg1$, it is convenient to transform the unperturbed ground state $|0\rangle$ of H_0 to a shifted Gaussian with an altered width and fix the shift as well as the width in some optimal manner. We would then have the advantage of starting our computation from a good zeroth-order description. To achieve this, we induce a Bogoliubov transformation of the boson variables $a/a^{\dagger}$ to generate another (shifted) Gaussian function $|\phi\rangle$ [32,33]:

$$|\phi\rangle\sim\exp(sa^{\dagger}+\tfrac{1}{2}ta^{\dagger 2})|0\rangle \tag{5.2}$$

and introduce new boson operators $b/b^{\dagger}$ for which $|\phi\rangle$ is the vacuum. b and $b^{\dagger}$ are given by

$$b=(1-t^2)^{-1/2}(a-ta^{\dagger}-s) , \tag{5.3a}$$

$$b^{\dagger}=(1-t^2)^{-1/2}(a^{\dagger}-ta-s) , \tag{5.3b}$$

where we restrict ourselves only to real parametrizations of $|\phi\rangle$. We note that s and t are, respectively, related to the shift and width parameter of the Gaussian.

For a finite-temperature application, there is no unique way of choosing the optimal values of s and t. We may, e.g., in the spirit of the zero-temperature formalism, choose them to minimize the ground-state expectation value $\langle\phi|H|\phi\rangle/\langle\phi|\phi\rangle$. This leads to the so-called Hartree Gaussian function $|\phi\rangle$ [32]. We propose to choose instead *a different starting point* which is optimal in a certain sense in the finite-temperature formalism. We demand that s and t should be chosen in such a way that the unperturbed, i.e., mean-field, free energy is a minimum. The minimizing equations show that the Gaussian function, thus determined, leads to the minimum value for the thermally averaged energy $\langle\langle H\rangle\rangle$ as well. We may thus call this Gaussian function as the "thermal Hartree function." We have found in the actual numerical implementation that the thermal Hartree function is a much better starting point as compared to the ordinary (zero-temperature) Hartree function—particularly in the large coupling regime.

To determine the optimal s and t parameters, we first rewrite H in terms of the boson variables $b/b^{\dagger}$, and bring it to the thermal normal order. The rearranged Hamiltonian is

$$\begin{aligned}H=&\langle\langle H\rangle\rangle+\Omega\{b^{\dagger}b\}_{\beta}+\omega^{-3/2}\{(8\lambda\omega)\omega'^3+(3\sqrt{2}\gamma\omega)\omega'^2+[\omega+6\lambda(2n+1)]\omega'+(3\gamma/2\sqrt{2})(2n+1)\}\{b^{\dagger}+b\}_{\beta}\\&-(2\omega)^{-2}[\omega^3-(1+6\sqrt{2}\gamma\omega'+24\lambda\omega'^2)\omega-6\lambda(2n+1)]\{b^{\dagger 2}+b^2\}_{\beta}\\&+\{[\gamma/(2\omega)^{3/2}]+[4\sqrt{2}\lambda\omega'/(2\omega)^{3/2}]\}\{b^{\dagger 3}+b^3+3b^{\dagger 2}b+3b^{\dagger}b^2\}_{\beta}\\&+[\lambda/(2\omega)^2]\{b^{\dagger 4}+b^4+4b^{\dagger 3}b+4b^{\dagger}b^3+6b^{\dagger 2}b^2\}_{\beta} ,\end{aligned} \tag{5.4}$$

where

$$\Omega=(2\omega)^{-1}[\omega^2+(1+6\sqrt{2}\gamma\omega'+24\lambda\omega'^2)+(6\lambda/\omega)(2n+1)]\ , \tag{5.5a}$$

$$\langle\langle H\rangle\rangle=\tfrac{1}{2}+[(1+\omega^2)/2\omega]n+[(3\gamma\omega'/\sqrt{2}\omega)+(6\lambda\omega'^2/\omega)](2n+1)$$
$$+(3\lambda/\omega^2)n(n+1)+(3\lambda/4\omega^2)+(1-\omega)^2/4\omega+\omega'^2+2\sqrt{2}\gamma\omega'^3+4\lambda\omega'^4\ , \tag{5.5b}$$

and ω and ω' are given by

$$\omega=(1-t)/(1+t)\ , \tag{5.6a}$$

$$\omega'=s/(1-t)\ . \tag{5.6b}$$

In what follows, we shall take ω and ω' to be our variational parameters rather than s and t. Let us note that the transformed Hamiltonian has a different frequency Ω in Eq. (5.4). In defining the thermal normal order, we have assumed that the occupation probability n has the value $[\exp(\beta\Omega)-1]^{-1}$. Z_0 has been computed with this new frequency, Ω. Since this is a number nonconserving Hamiltonian, $\mu=0$.

We take the new unperturbed Hamiltonian H_0 to be given by

$$H_0=\langle\langle H\rangle\rangle+\Omega\{b^\dagger b\}_\beta\ . \tag{5.7}$$

Z_0 for this H_0 is given by

$$Z_0=\{\exp[-\beta(\langle\langle H\rangle\rangle-\Omega n)]/(\exp(\beta\Omega)-1)\}[\exp(\beta\Omega)]\ . \tag{5.8}$$

From this Z_0, we derive the expression for the unperturbed free energy F_0 as

$$F_0=-\frac{1}{\beta}\ln Z_0\equiv\langle\langle H\rangle\rangle+\frac{1}{\beta}[n\ln n-(n+1)\ln(n+1)]$$
$$\equiv\langle\langle H\rangle\rangle-\frac{1}{\beta}S_0 \tag{5.9}$$

where S_0 is the unperturbed entropy.

The minimizing condition of F_0 with respect to ω can be written as

$$\frac{\partial F_0}{\partial\omega}=\left.\frac{\partial F_0}{\partial\omega}\right|_n+\frac{\partial F_0}{\partial n}\frac{\partial n}{\partial\omega}=0\ . \tag{5.10}$$

Now, $\langle\langle H\rangle\rangle$ contains terms with various powers of n stemming from the complete contractions of $b^\dagger/b$ operators entailed in $\langle\langle H\rangle\rangle$. The quantity $\Omega\{b^\dagger b\}$ in H_0 comes from those terms of H where one contraction remains to be done to reach complete contraction. It then follows that

$$\frac{\partial\langle\langle H\rangle\rangle}{\partial n}=\Omega\ . \tag{5.11}$$

Using Eq. (5.11), we find

$$0=\frac{\partial\langle\langle H\rangle\rangle}{\partial\omega}+\Omega\frac{\partial n}{\partial\omega}-\frac{1}{\beta}\frac{\partial S_0}{\partial n}\frac{\partial n}{\partial\omega}\ . \tag{5.12}$$

From the expression for S_0 in Eq. (5.9), we find that

$$\frac{1}{\beta}\frac{\partial S_0}{\partial n}=\Omega\ . \tag{5.13}$$

The minimization condition for F_0 with respect to ω then reduces to

$$\frac{\partial F_0}{\partial\omega}=\frac{\partial\langle\langle H\rangle\rangle}{\partial\omega}=0\ . \tag{5.14}$$

This condition is the same as minimizing the average energy with respect to ω. Equation (5.14) can be explicitly written as

$$\omega^3-(1+6\sqrt{2}\gamma\omega'+24\lambda\omega'^2)\omega-6\lambda(2n+1)=0\ . \tag{5.15}$$

Although there are several local minima for $\langle\langle H\rangle\rangle$, we always choose the global minimum corresponding to the positive frequency ω. This minimum is rather unique and can be reached by a Newton-Raphson iterative strategy.

Using Eq. (5.15), the expression for Ω in Eq. (5.5a) can be drastically simplified as

$$\Omega=\omega\ . \tag{5.16}$$

It then follows that S_0 in Eq. (5.9) is a function only of ω. Minimizing F_0 with respect to ω', we have

$$\frac{\partial F_0}{\partial\omega'}=\frac{\partial\langle\langle H\rangle\rangle}{\partial\omega'}=0 \tag{5.17}$$

which again shows that the minimization is essentially with respect to the average energy. Equation (5.17) can be explicitly written as

$$(8\lambda\omega)\omega'^3+(3\sqrt{2}\gamma\omega)\omega'^2+[\omega+6\lambda(2n+1)]\omega'$$
$$+(3\lambda/2\sqrt{2})(2n+1)=0\ . \tag{5.18}$$

From now on we shall call the ω at the minimizing point as the thermal Hartree frequency and the optimal Gaussian thus determined as the thermal Hartree function.

The average thermal energy, $\langle\langle H\rangle\rangle$ for the thermal Hartree function is given by

$$\langle\langle H\rangle\rangle=\tfrac{1}{2}+[(1+\omega^2)/2\omega]n+[(3\gamma\omega'/\sqrt{2}\omega)+(6\lambda\omega'^2/\omega)](2n+1)$$
$$+(3\lambda/\omega^2)n(n+1)+(3\lambda/4\omega^2)+(1-\omega)^2/4\omega+\omega'^2+2\sqrt{2}\gamma\omega'^3+4\lambda\omega'^4 \tag{5.19}$$

and H can be simplified to

$$H=\langle\langle H\rangle\rangle+\omega\{b^\dagger b\}_\beta+\eta\{b^{\dagger 3}/3!+b^3/3!+b^{\dagger 2}b/2!+b^\dagger b^2/2!\}_\beta$$
$$+\alpha\{b^{\dagger 4}/4!+b^4/4!+b^{\dagger 3}b/3!+b^\dagger b^3/3!+b^{\dagger 2}b^2/2!2!\}_\beta\ , \tag{5.20}$$

where $\eta=[\gamma+4\sqrt{2}\lambda\omega'][6/(2\omega)^{3/2}]$ and $\alpha=6\lambda/\omega^2$.

The unperturbed Hamiltonian is $H_0=\langle\langle H\rangle\rangle+\omega\{b^\dagger b\}_\beta$.

Let us note that we have no terms containing $b^\dagger$, b, $b^{\dagger 2}$, and b^2 in the thermally normal-ordered expression for H in Eq. (5.19) when b and $b^\dagger$ are defined with respect to the thermal Hartree function. *This is a consequence of the minimizing nature of the thermal Hartree function* and may be interpreted as the "thermal Brillouin condition" [35]. A general proof of this relation is given in Appendix B.

With respect to the renormalized frequency ω, the relevant contractions are given by

$$\overbrace{b_I^\dagger(\tau_1)b_I(\tau_2)}=\exp[\omega(\tau_1-\tau_2)]\times\begin{cases}n & \text{for } \tau_1>\tau_2 \quad (5.21a)\\ (n+1) & \text{for } \tau_1<\tau_2\ .\end{cases}$$
(5.21b)

B. Classification of operators:

The closed operators for this problem are all of the form $\{b^{\dagger m}b^m\}$ for all powers of $m\geq 0$. The $m=0$ term is a c number. Any other operator of the form $\{b^{\dagger m}b^n\}$ for $m\neq n$ is external. We may thus write S and X in the "Schrödinger picture" as

$$S(\tau)=\sum_{\substack{m,n\\ m\neq n}}[1/(m!n!)]s_{m,n}(\tau)\{b^{\dagger m}b^n\}_\beta$$
$$\equiv\sum_{\substack{m,n\\ m\neq n}}S_{m,n}(\tau)\ , \tag{5.22a}$$

$$X(\tau)=\sum_{m=0}^{\infty}[1/(m!)^2]x_m(\tau)\{b^{\dagger m}b^m\}_\beta\equiv\sum_m X_m(\tau)\ . \tag{5.22b}$$

Since the thermal contractions of the generators of the Hamiltonian (i.e., the terms appearing in H) produce new higher-body operators not contained in the Hamiltonian, the time-ordered exponential $T[\exp\int_0^\beta -V_I(\tau)d\tau]$ defining $U_I(\beta)$ will generate higher and higher body connected operators S_I and X_I on expansion in normal order and regrouping and, as a result, would entail a countably infinite number of external and closed-cluster operators of all possible ranks. For any practical implementation, the cluster cumulants in S_I and X_I, or equivalently S and X in the "Schrödinger picture" must, therefore, be truncated. A fruitful way of generating systematic approximation schemes is to include in successive higher-order calculations, cluster operators of higher particle ranks. Thus our cluster-cumulant approach affords a *natural systematics* of including higher-order interactions.

In the present case, we propose to include in the cluster cumulants all operators up to total rank of four. Thus our closed operators are I, $\{b^\dagger b\}_\beta$, $\{b^{\dagger 2}b^2\}_\beta$; our external operators are $\{b^\dagger\}_\beta$, $\{b\}_\beta$, $\{b^{\dagger 2}\}_\beta$, $\{b^2\}_\beta$, $\{b^{\dagger 2}b\}_\beta$, $\{b^\dagger b^2\}_\beta$, $\{b^{\dagger 3}\}_\beta$, $\{b^3\}_\beta$, $\{b^{\dagger 4}\}_\beta$, $\{b^4\}_\beta$, $\{b^{\dagger 3}b\}_\beta$, and $\{b^\dagger b^3\}_\beta$. We should note that we follow the convention of throwing all the "time" dependence on the cluster amplitudes: $S_{m,n}(\tau)\equiv(1/m!)(1/n!)s_{m,n}(\tau)\{b^{\dagger m}b^n\}_\beta$, etc.; the $b^\dagger/b$ operators have no "time" dependence.

C. Diagrammatics

The generation of the thermal-cluster-cumulant equations for S and X is very conveniently accomplished by resorting to diagrammatics. We depict the various n-body operators in H by unfilled circles as vertices, with incoming and outgoing lines signifying the b and $b^\dagger$ operators. Some typical H diagrams are displayed in Fig. 1. Since we have contractions of both the types $b_I^\dagger(\tau_1)b_I(\tau_2)$ and $b_I(\tau_1)b_I^\dagger(\tau_2)$, with $\tau_1>\tau_2$, it is necessary to indicate by arrows on the line the nature of an operator, viz., whether it is a b or $b^\dagger$. A line entering a vertex (arrow towards the vertex) is a b operator. Likewise, a line with an arrow emanating from a vertex is a $b^\dagger$ operator. We have shown in Fig. 2 the above two cases of thermal contractions. The vertices of the operators are placed on a diagram in increasing order of the "time" argument from right to left. S and X vertices are depicted as filled circles, some typical terms of which are shown in Fig. 3. As long as we indicate on the diagrams for S and X the $b^\dagger$ and b operators by appropriate arrows, it does not matter in what way we orient them.

The cluster-cumulant equations for S and X are generated by drawing the composite diagrams obtained by joining H_0 and V vertices with arbitrary numbers of S and X vertices in all possible ways, with the restriction that H_0 is always joined to the S and X vertices from both left and right, while V is joined only from the left. This special status for H_0 is a consequence of the presence of both $\overbrace{H_0S}$ (or $\overbrace{H_0X}$) and $\overbrace{SH_0}$ (or $\overbrace{XH_0}$) contractions in the cluster-cumulant equations in the "Schrödinger picture," *viz.*, Eqs. (3.21). Having drawn all the connected composite diagrams, we classify all the composites having the same disposition of ingoing and

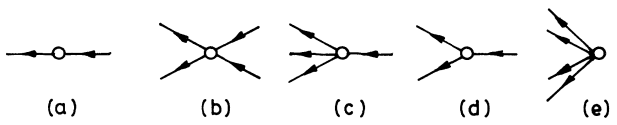

FIG. 1. Typical H diagrams are shown here.

FIG. 2. Two cases of thermal contractions are being depicted.

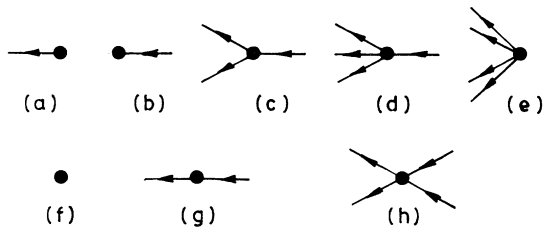

FIG. 3. The cluster-cumulant operators S and X are shown with filled circles. (a)–(e) are external operators S, since they involve change in occupancy, while (f)–(h) depict closed operators X involving no change in occupancy. The vertices (a)–(e) are, respectively, $S_{1,0}$, $S_{0,1}$, $S_{2,1}$, $S_{3,1}$, and $S_{4,0}$. Similarly, (f) (g), and (h) depict X_0, X_1, and X_2.

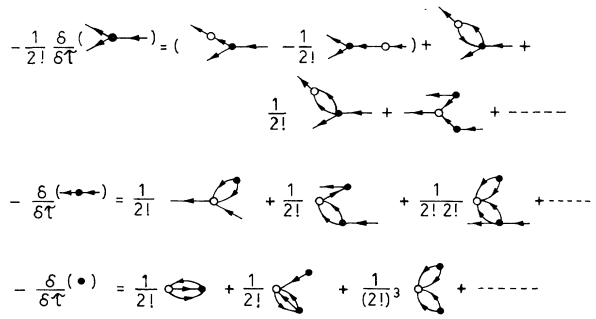

FIG. 4. Diagrammatic depiction of some terms entering the cluster-cumulant equations for $S_{2,1}$, X_1, and X_0.

outgoing lines together and equate these diagrams to the negative "time" derivative of the cluster operator having the same shape. Depending on whether these composites are external or closed types, we equate them to the negative time derivative of an S or an X. For our applications, in constructing the diagrams, we have included at most two cluster-cumulant vertices. We have found that this suffices even for large λ. As examples, we have shown the diagrammatic depiction of some terms entering the cluster-cumulant equations for $S_{2,1}$, X_1, and X_0 in Fig. 4. The rules for evaluating the diagrams are collected in Appendix C.

The cluster-cumulant equations have been solved by Runge-Kutta initiation followed by predictor-corrector method. For a wide range of λ, starting from $\lambda=0.1$ to $\lambda=1000$, the equations are very robust and show no erratic or unstable behavior. The $S_I^{m,n}$ and X_I^m operators in the interaction picture are generated from the $S_{m,n}$ and X_m operators via

$$S_I^{m,n}(\tau)\{b^{\dagger m}b^n\}_\beta=\exp-[(m-n)\omega\tau]S_{m,n}(\tau)\{b^{\dagger m}b^n\}_\beta\,, \tag{5.23a}$$

$$X_I^m(\tau)\{b^{\dagger m}b^m\}_\beta=X_m(\tau)\{b^{\dagger m}b^m\}_\beta\,, \tag{5.23b}$$

and finally Z/Z_0 is evaluated as $\exp(X_I^0)$.

For generating the perturbative solution, let us note that, in the first order, ${}^{(1)}S_I(\beta)$'s of the forms $S_I^{2,0}, S_I^{0,2}, S_I^{1,0}, S_I^{0,1}$ are zero, since the corresponding V_I's are zero as a consequence of the thermal Brillouin condition. In Eq. (4.2), the first-order contribution is obtained only for the operator $X_I^{(2)}$. Thus, we have no first-order contribution to X_I^0. At second order, ${}^{(2)}X_I^0$ thus receives contributions from ${}^{(1)}S_I^{4,0}$, ${}^{(1)}S_I^{0,4}$, ${}^{(1)}S_I^{3,1}$, ${}^{(1)}S_I^{1,3}$, and ${}^{(1)}X_I^2$. As examples of concrete results, we quote below the expression for ${}^{(2)}X_I^0$:

$$\begin{aligned}{}^{(2)}X_I^0=&(\eta^2/2\omega)[-n^3/9+(n+1)^3/9-n^2(n+1)+n(n+1)^2]\beta\\&+(\alpha^2/12\omega)[-n^4/8+(n+1)^4/8-n^3(n+1)+n(n+1)^3]\beta\\&+(\eta^2/\omega^2)\{(n^3/54)(e^{3\beta\omega}-1)+[(n+1)^3/54](e^{-3\beta\omega}-1)+[n^2(n+1)/2](e^{\beta\omega}-1)+[n(n+1)^2/2](e^{-\beta\omega}-1)\}\\&+(\alpha^2/\omega^2)\{(n^4/384)(e^{4\beta\omega}-1)+[(n+1)^4/384](e^{-4\beta\omega}-1)+[n^3(n+1)/24](e^{2\beta\omega}-1)\\&+[n(n+1)^3/24]\times(e^{-2\beta\omega}-1)\}+(\alpha^2/8)n^2(n+1)^2\beta^2\,.\end{aligned} \tag{5.24}$$

These results coincide exactly with a variant of the Bloch–Balian–de Dominicis theory [4,5], where the arguments for the "time" variables are in the range $\beta\geq\tau_1\geq\tau_2\geq\tau_3\cdots\geq\tau_n\geq 0$ for the nth-order diagram. The series generated involve "unperturbed" occupation probabilities n with respect to the renormalized frequency, Ω.

D. Results and discussions

In Table I, we show the values of Z and Z_0 and the free energy F for various values of $T=1/\beta$ and λ. The corresponding "exact" values are also displayed for assessing the performance of the method. They have been computed by using the eigenspectrum of H, obtained by diagonalizing H in the harmonic-oscillator basis of the unperturbed $H_0=a^\dagger a+\frac{1}{2}$. Basis functions up to the quantum number 19 have been used for $\lambda=0.1$ and 1.0, up to the quantum number 30 for $\lambda=10$ and 100, and up to quantum number 40 for $\lambda=1000$. From a perusal of Table I it is clear that the value of Z obtained for even very large values of λ are quite good *vis-à-vis* the exact results. Since we have included the cluster-cumulant operators only up to rank four, the performance of the cluster-cumulant method is very satisfactory.

Bloch, Balian, and de Dominicis [4–6] have discussed

TABLE I. The thermal-cluster-cumulant (TCC) results for the anharmonic oscillator with cubic-plus-quartic perturbation (cluster operators up to rank 4 are used). The entry for $T=0$ corresponds to the Kohn-Luttinger limit, which is the ground-state energy. ω corresponds to the thermal Hartree frequency (see text for details). F corresponds to the free energy computed from Z.

λ	$T=1/\beta$	ω	ω'	Z_0	Z TCC Exact[a]	Z Second-order cumulant	F TCC Exact[a]	F Second-order cumulant
	0.0						0.553 52	0.553 16
							[0.553 52]	
	0.4	1.2228	−0.062 88	0.262 19	0.263 71	0.264 28	0.533 16	0.532 30
					[0.263 80]		[0.533 02]	
0.1	0.5	1.2366	−0.065 52	0.360 79	0.362 98	0.363 98	0.506 70	0.505 32
					[0.363 17]		[0.506 44]	
	0.8	1.2851	−0.074 04	0.632 56	0.637 43	0.640 65	0.360 25	0.356 22
					[0.638 17]		[0.359 32]	
	1.0	1.3177	−0.079 19	0.799 44	0.806 45	0.811 76	0.215 12	0.208 55
					[0.807 76]		[0.213 49]	
	0.0						0.721 54	0.715 66
							[0.720 46]	
	0.8	1.9692	−0.153 93	0.440 10	0.450 25	0.459 70	0.638 36	0.621 75
					[0.452 64]		[0.634 12]	
1.0	1.0	2.1085	−0.155 26	0.560 08	0.573 06	0.587 50	0.556 76	0.531 88
					[0.576 99]		[0.549 92]	
	1.25	2.1021	−0.1567	0.698 01	0.714 51	0.735 83	0.420 21	0.383 45
					[0.720 68]		[0.409 45]	
	1.6	2.1989	−0.158 48	0.875 55	0.896 81	0.928 07	0.174 25	0.119 44
					[0.906 33]		[0.157 37]	
	0.0						0.793 74	0.782 83
							[0.791 30]	
	1.0	2.3689	−0.176 78	0.494 15	0.508 53	0.524 34	0.676 22	0.645 61
					[0.512 78]		[0.667 90]	
2.0	1.25	2.4502	−0.176 78	0.617 50	0.635 25	0.658 39	0.567 16	0.522 45
					[0.641 87]		[0.554 21]	
	1.6	2.5606	−0.176 78	0.774 57	0.796 82	0.830 62	0.363 40	0.296 94
					[0.807 05]		[0.343 00]	
	2.0	2.6773	−0.176 78	0.939 02	0.966 11	1.0117	0.068 95	−0.023 33
					[0.980 44]		[0.039 51]	
	0.0						0.835 39	0.790 63
							[0.825 35]	
	1.6	3.7173	−0.229 66	0.641 25	0.672 85	0.722 88	0.633 98	0.519 21
					[0.687 87]		[0.598 65]	
10	2.0	3.8595	−0.225 49	0.762 47	0.797 63	0.861 49	0.452 23	0.298 18
					[0.817 40]		[0.403 25]	
	2.5	4.0323	−0.221 12	0.899 67	0.938 93	1.0186	0.157 53	−0.046 12
					[0.964 35]		[0.097 64]	
	4.0	4.4849	0.212 29	1.2591	1.3093	1.4264	−1.0778	−1.4206
					[1.348 4]		[−1.195 6]	
	0.0						−3.7836	−3.745 9
							[−3.7742]	
	4.0	11.679	−0.4294	2.5064	2.9646	2.844 3	−4.3470	−4.1813
					[2.9770]		[−4.3637]	
100	5.0	11.213	−0.41264	2.2139	2.6712	2.259 9	−4.9127	−4.7753
					[2.6666]		[−4.9041]	
	8.0	9.6749	−0.325 16	2.0876	2.3241	2.845 2	−6.7468	−8.3652
					[2.500 4]		[−7.3314]	
	0.0						−82.292	−82.281
							[−82.291]	
	10.0	44.986	−0.508 21	3705.7	3803.5	3798.0	−82.437	−82.422
					[3807.3]		[−82.447]	
1000	20.0	44.422	−0.503 01	67.774	70.268	69.783	−85.046	−84.908
					[71.065]		[−85.274]	
	25.0	43.969	−0.498 81	32.045	33.634	33.228	−87.889	−87.584
					[34.362]		[−88.432]	

[a]Quantities within brackets indicate exact values.

thoroughly the problem of going to the zero-temperature limit for the free energy for a number conserving H. In this case $\mu \neq 0$. They have shown that F approaches the ground-state energy E_{gr} when the energy levels are discrete. When the system is of infinite extension but of finite density, the limit has to be taken with much care, and one should use perturbed occupation probabilities [6] (or, in an alternative version certain quasiparticle occupation probabilities, as in [36]) to approach the limit. For the problem studied by us here, $\mu=0$ and the energy levels are discrete, so that the zero-temperature limit for F should be reached quite smoothly:

$$\Delta E = E_{gr} - E_0 = \lim_{\beta \to \infty} (F - F_0)$$

$$= \lim_{\beta \to \infty} \left[-\frac{1}{\beta} \ln Z / Z_0 \right] . \tag{5.25}$$

This relation is known as the Kohn-Luttinger theorem [37]. Using Eq. (3.18), we have

$$\Delta E = \lim_{\beta \to \infty} -X_I^0/\beta . \tag{5.26}$$

Table I displays the total energy E_{gr} as a function of λ, which agrees very well both with the exact values and with the corresponding results obtained with the zero-temperature version of the stationary coupled-cluster theory [33] using the cluster operators up to rank 4. We conclude that our theory verifies numerically the Kohn-Luttinger limit and reduces to the corresponding zero-temperature version of the stationary coupled-cluster theory for the anharmonic oscillator. An analytical treatment of the Kohn-Luttinger limit and the use of perturbed occupation probability will be the subject of our future paper [31]. In Table I, we have also displayed the second-order perturbative cumulant results, obtained by using Eq. (5.25). A perusal of these results shows that the second-order cumulant results are generally rather inaccurate as compared to the thermal cumulant results.

We should mention that, beyond the value of $\lambda=1.78$, the potential generates a second minimum away from the coordinate origin, which gets deeper with further increase of λ. The thermal Hartree functions obtained by us are centered closer to the deeper minimum. All the excited states are thus also centered around this minimum. The presence of double minima should make the prediction of the high-temperature behavior of the partition function particularly difficult, since at the higher temperatures the shallow minimum at the origin should make its presence felt via higher excited states which should be better described as bound state localized at the origin rather than at the deeper minimum. The qualitative feature of the potential at $\lambda=100$ and 1000 are very similar. However, its relative depth is more for $\lambda=1000$, making the description of the bound state relatively better as compared to $\lambda=100$. This is reflected in the increased accuracy of the computed Z and F for $\lambda=1000$ relative to $\lambda=100$.

VI. CONCLUDING REMARKS

We have presented in this paper a nonperturbative cluster-cumulant approach for computing thermal averages by combining the best features of thermodynamic perturbation theory involving cumulants, certain ideas of thermofield dynamics and the imaginary-time generalization of the time-dependent coupled-cluster theory. We have introduced connected quantities—called cluster cumulants by us—which are the appropriate nonperturbative analogues of perturbative cumulants. We have developed a finite-temperature field theory involving only the physical variables, by introducing the concepts of thermal normal ordering, thermal Wick expansion, and thermal contractions. Our formulation is shown in Appendix A to be unitarily equivalent to the traditional formulation of thermofield dynamics, but is more suitable for numerical implementation since we have no "doubling" of the degrees of freedom. The thermally averaged quantities appear as c-number entities as a consequence of the validity of the thermal Wick's theorem. The performance of the method has been illustrated by computing the partition function of a cubic-plus-quartic anharmonic oscillator at wide ranges of temperature $T=1/\beta$ and the coupling constant λ. The method has been found to be both robust and stable, provided we start from an unperturbed Gaussian as the vacuum for the creation and annihilation operators which minimize the thermally averaged energy. This function has been termed the thermal Hartree function by us. We have also verified numerically that the ground state of the anharmonic oscillator is obtained in the low-temperature limit. The cluster-cumulant equations are conveniently set up using diagrammatic techniques, which are illustrated by citing specific examples.

ACKNOWLEDGMENTS

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APPENDIX A: THERMOFIELD DYNAMIC ANALOG OF THE THERMAL WICK EXPANSION

The thermofield dynamics was originally developed to extend the zero-temperature quantum-field theory to finite temperature [19–22], while retaining an underlying wave-function approach. The thermal average of any observable at the inverse temperature β is written as an *expectation value* over a "thermal vacuum" $|\bar{0}_\beta\rangle$. To achieve this explicitly, one introduces *extra quantum-mechanical variables* acting in another Fock space, called the tilde space, and the physical Fock space is expanded to generate a direct-produce Fock space of the physical Fock space and the tilde Fock space. The functionals of the physical variables $F(A)$ are replaced by $F(A_\beta)$ with the "thermal" operators [19–22] A_β given by

$$A_\beta = U_{\beta B}^\dagger A U_{\beta B} = A\cosh\theta - \tilde{A}^\dagger \sinh\theta \quad \text{(bosons)}, \tag{A1a}$$

$$A_\beta = U_{\beta F}^\dagger A U_{\beta F} = A\cos\theta - \tilde{A}^\dagger \sin\theta \quad \text{(fermions)}, \tag{A1b}$$

where $\tilde{A}$, etc., are the operators for the tilde variables. The transformations in Eqs. (A1) are induced by two unitary operators $U_{\beta B}$ and $U_{\beta F}$ for the bosonic and fermionic cases, respectively. $|\bar{0}_\beta\rangle$ is chosen as the simultaneous vacuum for both the physical and the tilde variables:

$$|\bar{0}_\beta\rangle \equiv |0,\tilde{0}\rangle \ . \tag{A2}$$

The thermal average of a functional $F(A, A^\dagger)$ is given by

$$\langle\langle [F(A, A^\dagger)] \rangle\rangle = \langle \bar{0}_\beta | F(A_\beta, A_\beta^\dagger) | \bar{0}_\beta \rangle \tag{A3}$$

and the angle θ in Eqs. (A1) is chosen in such a way that Eq. (A3) reproduces the required expressions for the thermal average. In particular, using the expression of the number operator $a^\dagger a$ for the functional $F(a,a^\dagger)$, we have

$$\sinh\theta = \{\exp[\beta(\omega_0-\mu)]+1\}^{-1/2} \quad \text{(bosons)}, \tag{A4}$$

$$\sin\theta = \{\exp[\beta(\omega_0-\mu)]-1\}^{-1/2} \quad \text{(fermions)}. \tag{A5}$$

Thus, in thermofield dynamics, one first induces a Bogoliubov transformation, Eq. (A1), to generate a new expression from $F(A, A^\dagger)$, *viz.*, $F(A_\beta^\dagger, A_\beta^\dagger)$, involving $A/A^\dagger$ and $\tilde{A}/\tilde{A}^\dagger$, and then works out a field theory—which becomes a zero-temperature formalism—using $|\bar{0}_\beta\rangle$ as the new vacuum. The Hamiltonian gets mapped onto a new Hamiltonian via the Bogoliubov transformation. The evolution takes place in an expanded Fock space since all the degrees of freedom are doubled. This increases the complexity of the problem. Our thermal field theory, in contrast, involves only the physical variables and in this sense seems more physically motivated for the numerical implementation.

We, however, show that the conventional thermofield dynamics can be transcribed into our formulation. Since the functional F are an algebraic expression of $A/A^\dagger$, we may rewrite $F(A_\beta, A_\beta^\dagger)$ as

$$F(A_\beta, A_\beta^\dagger) = U_\beta^\dagger F(A, A^\dagger) U_\beta \tag{A6}$$

from Eq. (A1). The actual form of U_β:$U_{\beta B}$ or $U_{\beta F}$, should be chosen depending on the situation. If we now define $|0_\beta\rangle$ as a new thermal "base state" by the relation

$$|0_\beta\rangle = U_\beta |\bar{0}_\beta\rangle \ , \tag{A7}$$

then the thermal average, Eq. (A3) above may be equivalently expressed as

$$\langle \bar{0}_\beta | F(A_\beta, A_\beta^\dagger) | \bar{0}_\beta \rangle \equiv \langle 0_\beta | F(A, A^\dagger) | 0_\beta \rangle \ . \tag{A8}$$

Since $A_\beta/A_\beta^\dagger$ are linear combinations of $A/A^\dagger$ and $\tilde{A}/\tilde{A}^\dagger$, we may write $F(A_\beta, A_\beta^\dagger)$ directly *in ordinary normal order* with respect to $|\bar{0}_\beta\rangle$. Thus, if $F(A_\beta, A_\beta^\dagger)$ is a time-ordered product of operators, we have

$$T[A_{I\beta}^{(1)}(\tau_1) A_{I\beta}^{(2)}(\tau_2)\cdots] = \{a_{I\beta}^{(1)}(\tau_1) A_{I\beta}^{(2)}(\tau_2)\cdots\} + (\text{single contractions}) + (\text{double contractions}) + \cdots \ . \tag{A9}$$

Let us premultiply and postmultiply Eq. (A9) by U_β and $U_\beta^\dagger$, and get

$$T[A_I^{(1)}(\tau_1) A_I^{(2)}(\tau_2)\cdots] = U_\beta\{A_{I\beta}^{(1)}(\tau_1) A_{I\beta}^{(2)}(\tau_2)\cdots\} U_\beta^\dagger + U_\beta\{(\text{single contraction})\} U_\beta^\dagger + \cdots \ . \tag{A10}$$

If we define now a *thermal normal ordering* by

$$\{A_I^{(1)}(\tau_1) A_I^{(2)}(\tau_2)\cdots\}_\beta = U_\beta\{A_{I\beta}^1(\tau_1) A_{I\beta}^{(2)}(\tau_2)\cdots\} U_\beta^\dagger \ , \tag{A11a}$$

$$\{A_I^{(1)}(\tau_1)\cdots\{\overbrace{A_I^{(i)}(\tau_i)\cdots A_I^{(j)}(\tau_j)}\}_\beta\cdots\}_\beta = U_\beta\{A_{I\beta}^{(1)}(\tau_1)\cdots\overbrace{A_{I\beta}^{(i)}(\tau_i)\cdots A_{I\beta}^{(j)}(\tau_j)}\cdots\} U_\beta^\dagger \ , \ \text{etc.} \ , \tag{A11b}$$

then we have

$$T[A_I^{(1)}(\tau_1) A_I^{(2)}(\tau_2)\cdots] = \{A_I^{(1)}(\tau_1) A_I^{(2)}(\tau_2)\cdots\}_\beta + \{(\text{single contraction})\cdots\}_\beta + \cdots \ . \tag{A12}$$

Since Eq. (A12) is a unitary transformation of the original expansion, Eq. (A9), the thermal normal ordering introduced above satisfies all the desirable properties of a normal order, and the average $\langle 0_\beta|\{\ \}_\beta|0_\beta\rangle$ are zero automatically:

$$\langle 0_\beta|\{\ \}_\beta|0_\beta\rangle = \langle \bar{0}_\beta| U_\beta^\dagger U_\beta \{\ \} U_\beta U_\beta^\dagger |\bar{0}_\beta\rangle \langle \bar{0}_\beta|\{\ \}|\bar{0}_\beta\rangle = 0 \ . \tag{A13}$$

Equation (A12) then relates the thermal normal ordered Wick expansion stipulated by us, by the consideration of the Gaussian stochastic nature of the Bose-Fermi operators in a T product with respect to thermal averaging, with the one generated as appropriate unitary transforms of thermal field operators from the more conventional thermofield dynamics. The transcription of our representation to the traditional formulation is completed if we now identify $\langle\langle F(A, A^\dagger)\rangle\rangle$ as $\langle 0_\beta|F(A, A^\dagger)|0_\beta\rangle$. The thermofield dynamic version of the cluster cumulant theory, with the thermal normal ordering as in Eq. (A12), has been introduced recently by us [35] along with a pilot, numerical illustration. We note that the thermofield dynamical treatment, although shown to be unitarily equivalent here, is operationally very different. In particular, *we shall never need to know* the detailed form of either U_β or $|0_\beta\rangle$. In our formulation, the tilde variables do not appear at all, and the original Hamiltonian H governs the evolution of U. One may also show, although we do not illustrate it here, that the usual Kubo-Martin-Schwinger relations [38] are satisfied by our formulation.

APPENDIX B: THERMAL HARTREE VS THERMAL BRILLOUIN CONDITIONS

In this appendix, we show formally that enforcing the thermal Hartree minimizing condition on the unperturbed starting "vacuum" function $|\phi\rangle$ automatically implies the validity of associated thermal Brillouin conditions.

Let us assume that the Hamiltonian H has been written in thermal normal order. The "unperturbed" function $|\phi\rangle$ which is the lowest state of the unperturbed Hamiltonian H_0' can be parametrized, in terms of certain generators of H. Let us write H in thermal normal order:

$$H=h_0+\sum_j h_j\{e_j\}_\beta \ , \tag{B1}$$

where h_0 is just $\langle\langle H\rangle\rangle$, and $\{e_i\}_\beta$'s are the generators in thermal normal order, accompanying the coefficients h_j.

Let us assume that an arbitrary Hartree state $|\phi'\rangle$ can be written in terms of the vacuum as

$$|\phi'\rangle \sim \left\{\exp\left[\sum_\alpha t_\alpha e_\alpha\right]\right\}_\beta |\phi\rangle \ , \tag{B2}$$

where e_α's are certain specific generators of H used to parametrize $|\phi\rangle$; $e_\alpha^\dagger$'s annihilate $|\phi\rangle$. Using this parametrization, any infinitesimal variation in $|\phi\rangle$, $|\delta\phi\rangle$, can be similarly written as

$$|\delta\phi\rangle \sim \sum_\alpha \delta t_\alpha \{e_\alpha\}_\beta |\phi\rangle \ . \tag{B3}$$

Let us assume that the excited states of H_0 can be generated by the action of combination of certain operators $\{e_p^*\}$ on $|\phi\rangle$:

$$|k\rangle = W_k^\dagger|\phi\rangle = \sum_p d_{pk}\{e_p^*\}_\beta|\phi\rangle \ . \tag{B4}$$

We also write $|\phi\rangle = W_0|\phi\rangle$; $W_0 = W_0^\dagger = 1$.

The thermal Hartree energy $\langle\langle H\rangle\rangle$ can then be expressed as

$$\begin{aligned}\langle\langle H\rangle\rangle &= \sum_{k=0}^{\infty}\langle\phi|W_k e^{-\beta H_0} H W_k^\dagger|\phi\rangle \Big/ \sum_{k=0}^{\infty}\langle\phi|W_k e^{-\beta H_0} W_k^\dagger|\phi\rangle \\ &\equiv \sum_{k=0}^{\infty}\langle\phi|W_k e^{-\beta H_0} H W_k^\dagger|\phi\rangle / Z_0 \ .\end{aligned} \tag{B5}$$

We then write $\delta\langle\langle H\rangle\rangle$ as

$$\delta\langle\langle H\rangle\rangle = \left[\sum_{k=0}^{\infty}\langle\delta\phi|W_k e^{-\beta H_0} H W_k^\dagger|\phi\rangle/Z_0 - \sum_{k=0}^{\infty}\langle\delta\phi|W_k e^{-\beta H_0} W_k^\dagger|\phi\rangle\langle\langle H\rangle\rangle/Z_0 + \text{H.c.}\right] . \tag{B6}$$

Using the relation (B3), and using the fact that $\{e_\alpha^\dagger\}_\beta$ and W_k both contain destruction operators, we find that

$$\begin{aligned}\langle\delta\phi|W_k e^{-\beta H_0} W_k^\dagger|\phi\rangle/Z_0 &= \sum_\alpha \langle\phi|W_k\{e_\alpha^\dagger\}_\beta e^{-\beta H_0} W_k^\dagger|\phi\rangle\delta t_\alpha \Big/ Z_0 \\ &\equiv \sum_\alpha \delta t_\alpha \langle\langle\{e_\alpha^\dagger\}_\beta\rangle\rangle = 0 \ ,\end{aligned} \tag{B7}$$

since the thermal average of a thermally normally ordered operator is zero. We can similarly rewrite the first term in Eq. (B6) as

$$\sum_{k=0}^{\infty}\langle\phi|\left[\sum_\alpha\{e_\alpha^\dagger\}_\beta\delta t_\alpha\right]W_k e^{-\beta H_0} H W_k^\dagger|\phi\rangle\Big/Z_0 = \sum_\alpha\langle\langle\{e_\alpha^\dagger\}_\beta H\rangle\rangle\delta t_\alpha \ .$$

Since both $\{e_\alpha^\dagger\}_\beta$ and H are in thermal normal order, it follows that at the minimizing point of $\langle\langle H\rangle\rangle$, we should have

$$\delta\langle\langle H\rangle\rangle = 0 = \sum_\alpha \langle\langle\{e_\alpha^\dagger\}_\beta\{e_\alpha\}_\beta\rangle\rangle h_\alpha \delta t_\alpha + \text{H.c.}, \ , \tag{B9}$$

which shows that h_α's must be vanishing. Thus, if $|\phi\rangle$ can be parametrized by the generators $\{e_\alpha\}$, then the coefficients $\{h_\alpha\}$ should be vanishing for the thermal Hartree function. For a general anharmonic oscillator, $|\phi\rangle$ can be parametrized by $b^\dagger$ and $b^{\dagger 2}$; as a result the coefficients of $b^\dagger$ and $b^{\dagger 2}$ (and their adjoints) will be zero when H is written in thermal normal order with respect to the thermal Hartree function as the underlying unperturbed function $|\phi\rangle$.

APPENDIX C: DIAGRAMMATIC RULES FOR EVALUATING COMPOSITE DIAGRAMS

We consider here for simplicity the case of $\mu=0$. (i) Any composite diagram consists of an H vertex and any number of $S(\tau)$ and $X(\tau)$ vertices joined to H. (ii) We draw only the topologically distinct diagrams. For any

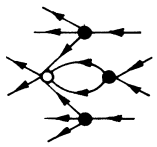

FIG. 5. A typical diagram, used for demonstrating the rules for evaluating the factors associated with any diagram.

contraction in which the arrow goes from right to left, we associate a factor $(n+1)$. For a contraction where the arrow goes from left to right, we associate a factor n. ω is the unperturbed frequency of the thermal Hartree function. (iii) For a diagram with each set of k equivalent outgoing and l equivalent ingoing lines, we associate a topological weight of $1/(k!l!)$. (iv) For any set of equivalent r internal lines, we associate a factor $1/r!$. (v) For each set of p equivalent vertices of S or X, we associate a further weight of $1/p!$.

We illustrate the use of the above diagram rules by evaluating the factors of the diagram shown in Fig. 5. There are three pairs of equivalent outgoing and one pair of equivalent ingoing lines, contributing the factor $1/(2!)^3 2!$. There is a pair of equivalent internal lines, stemming from X, giving another factor of $1/2!$. Two S vertices are equivalent, giving an additional factor of $1/2!$.

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