Inclusion of higher order anharmonic contributions in self-consistent phonon theory

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The ansatz method of infinite summation of higher order diagrams given in Shukla and Cowley, Phys. Rev. B **58**, 2596 (1998), is extended to the self-consistent phonon theory. We demonstrate the high accuracy of this approach with respect to the first-order self-consistent and improved self-consistent (ISC) phonon theories, by comparing the results from the ansatz method with their exact counterparts. The ISC theory is then extended to include the remaining diagrams of $O(\lambda^4)$, which could not be included in its earlier formulation. This makes the ISC theory consistent, at least to $O(\lambda^4)$. This ISC theory offers a substantial improvement over the current ISC theory. The results of the equation of state for a face centered cubic nearest neighbor interaction Lennard-Jones solid from our ISC theory are shown to be in excellent agreement with the results of the classical Monte Carlo method also obtained for the same model.

I. INTRODUCTION

Anharmonic contributions to a physical property (α) arise from an infinite set of terms in the Taylor series expansion of the potential energy. Normally these terms are ignored in making the harmonic approximation where the quadratic terms are retained in the above expansion.

One method of including the anharmonic contributions to α from this infinite set is via the first-order self-consistent phonon theory (SC1) or the second-order self-consistent phonon theory (SC2). In the diagrammatic language the derivations of SC1 and SC2 have been given earlier by Choquard.[1] SC1 has also been derived by the variational method[2] and a Zubarev-type Green's function method.[3] The essence of the SC1 is that it contains only a partial sum of the infinite set of anharmonic contributions; i.e., only the *even* terms in the Taylor series of the potential energy are summed to infinity. All the *odd* terms in the series are omitted from the summation procedure. The SC2 contains these omitted terms but the numerical implementation is very difficult. However, it has been implemented numerically for a Lennard-Jones (LJ) solid by Kanney and Horton[4] and found to be divergent. In an earlier calculation, it was shown by Gillis *et al.*[5] that the numerical results obtained from SC1, again for a LJ solid, were poor. However, it appears that the improved self-consistent (ISC) phonon theory,[6] which is the SC1 free energy plus the first important correction omitted from the SC1, the cubic term in the free energy, yields results which are satisfactory except when the temperature is high.

In the numerical implementation of ISC theory the eigenvalues and eigenvectors are obtained from the SC1 theory and the third-rank tensor in the cubic term is averaged with the same density matrix as in SC1. This type of calculation using the SC1 theory was also done for the lattice thermal conductivity by Benin[7] where the cubic term was averaged with the SC1 density matrix. Recently we have shown[8] that if the averaging of the third-rank tensor in the cubic term is not done, the results of ISC and SC1 hardly differ from each other. However, in spite of the success of ISC theory over a considerable temperature range, in certain respects the theory is inconsistent. For example, four diagrams of order λ^4 are included in ISC theory, whereas four other diagrams of $O(\lambda^4)$ of similar magnitude, which are given in our earlier work,[9] are left out. The diagrams included in the total ISC free energy are 2a, 2b, 2c, and 2d along with the SC1 free energy. Diagram 2c is included via the averaging of the cubic tensor and 2d is included because the SC1 eigenvalues and eigenvectors are employed in the calculation of the cubic free energy. Diagrams 2a and 2b are naturally contained in the calculation of the SC1 free energy through ring diagram summation.

Our objective in this paper is to include the remaining four diagrams of $O(\lambda^4)$ in the implementation of the ISC theory so that the theory is consistent to at least $O(\lambda^4)$. In the notation of the earlier work[9,10] these diagrams are labeled as 2e, 2f, 2g, and 2h. Once again the total free energy now consists of the SC1 free energy, the correction from the cubic term evaluated from the SC1 eigenvalues and eigenvectors and averaged cubic force constants, and the correction from the 2e, 2f, 2g, and 2h diagrams evaluated by averaging the vertices and employing the SC1 eigenvalues and eigenvectors. We note here that diagram 2e was included in an earlier calculation to improve the results of the traditional ISC theory by Koehler.[11] Because of the extreme complexity inherent in these calculations largely due to the tensorial character of the anharmonic force constants, their Fourier transforms, multiple Brillouin zone (BZ) summations restricted by numerous Δ functions, and finally neighbor summations, we have followed the ansatz approach which was successfully employed in our recent work[10] in the summations of free energy diagrams. In this approach the higher order diagrams, e.g., many diagrams of $O(\lambda^6)$, $O(\lambda^8)$, etc., can be reduced to a simple product of the lower order diagrams, i.e., $O(\lambda^2)$ and $O(\lambda^4)$. A comparison of the numerical results, for many diagrams, from this approach with their exact numerical values shows that the ansatz approach yields very accurate answers, even better than the ISC theory in the classical limit.[8]

II. ANSATZ FOR SC1, ISC, AND OUR ISC THEORY

In the context of the summation of free energy diagrams which are of the same order of magnitude, i.e., $O(\lambda^4)$, we have developed a simple approximation scheme or ansatz. This has enabled us to include the contributions of an infinite set of diagrams of $O(\lambda^6)$, $O(\lambda^8)$, etc., in the free energy sum. In the same spirit, here, we apply the ansatz procedure in the development of the ISC theory and *all* the corrections to this theory arising from the $O(\lambda^4)$ diagrams which are not included in the ISC theory.

The SC1 eigenvalues and eigenvectors are determined self-consistently from the three equations (1)–(3) in a recent publication[8] and there is no need to reproduce them here. However, for the implementation of the ansatz procedure the necessary equations can be derived by the method presented in Ref. 10. Once again we start with the expression for the one phonon Green's function[3]

$$G^{jj'}_{\mathbf{qq}'}(\omega)=\frac{2\omega_{\mathbf{q}j}\delta_{\mathbf{qq}'}\delta_{jj'}}{2\pi[\omega^2-\Omega^2_{\mathbf{q}j}]}, \tag{1}$$

where the renormalized phonon frequency $\Omega_{\mathbf{q}j}$ for the wave vector ($\mathbf{q}$) and branch index j is given by

$$\Omega^2_{\mathbf{q}j}=\omega^2_{\mathbf{q}j}+\frac{24\omega_{\mathbf{q}j}}{\hbar}\sum_{\mathbf{q}_1j_1}V(-\mathbf{q}j,\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}j)N_{\mathbf{q}_1j_1}+\frac{180\omega_{\mathbf{q}j}}{\hbar}\sum_{\mathbf{q}_1j_1}\sum_{\mathbf{q}_3j_3}V(-\mathbf{q}j,\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}_3j_3,-\mathbf{q}_3j_3,\mathbf{q}j)N_{\mathbf{q}_1j_1}N_{\mathbf{q}_3j_3}$$
$$+\frac{1680\omega_{\mathbf{q}j}}{\hbar}\sum_{\mathbf{q}_1j_1}\sum_{\mathbf{q}_3j_3}\sum_{\mathbf{q}_5j_5}V(-\mathbf{q}j,\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}_3j_3,-\mathbf{q}_3j_3,\mathbf{q}_5j_5,-\mathbf{q}_5j_5,\mathbf{q}j)\times N_{\mathbf{q}_1j_1}N_{\mathbf{q}_3j_3}N_{\mathbf{q}_5j_5}+\cdots. \tag{2}$$

For further details of the derivation and the meaning of the various symbols arising in Eq. (2), we refer to Ref. 3, where

$$N_{\mathbf{q}j}=\frac{\omega_{\mathbf{q}j}}{\Omega_{\mathbf{q}j}}\coth\left(\frac{1}{2}\beta\hbar\Omega_{\mathbf{q}j}\right). \tag{3}$$

In the high temperature limit ($T>\Theta_D$), where Θ_D is the Debye temperature, and with the help of Eqs. (3) and (4) of Ref. 3, the above expression for $\Omega_{\mathbf{q}j}$ reduces to

$$\Omega^2_{\mathbf{q}j}=\omega^2_{\mathbf{q}j}+\frac{1}{2\beta N}\sum_{\mathbf{q}_1j_1}\frac{\Phi(-\mathbf{q}j,\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}j)}{\Omega^2_{\mathbf{q}_1j_1}}+\frac{1}{8(\beta N)^2}\sum_{\mathbf{q}_1j_1}\sum_{\mathbf{q}_2j_2}\frac{\Phi(-\mathbf{q}j,\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}_2j_2,-\mathbf{q}_2j_2,\mathbf{q}j)}{\Omega^2_{\mathbf{q}_1j_1}\Omega^2_{\mathbf{q}_2j_2}}$$
$$+\frac{1}{48(\beta N)^3}\sum_{\mathbf{q}_1j_1}\sum_{\mathbf{q}_2j_2}\sum_{\mathbf{q}_3j_3}\frac{\Phi(-\mathbf{q}j,\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}_2j_2,-\mathbf{q}_2j_2,\mathbf{q}_3j_3,-\mathbf{q}_3j_3,\mathbf{q}j)}{\Omega^2_{\mathbf{q}_1j_1}\Omega^2_{\mathbf{q}_2j_2}\Omega^2_{\mathbf{q}_3j_3}}+\cdots. \tag{4}$$

The solutions for $\Omega_{\mathbf{q}j}$ from this equation are to be obtained self-consistently because $\Omega_{\mathbf{q}j}$ arises in the various terms in the denominator on the right hand side of Eq. (4). An approximate self-consistent solution can be obtained by substituting

$$\Omega^2_{\mathbf{q}j}=\omega^2_{\mathbf{q}j}(1+X) \tag{5}$$

in Eq. (4), where the quantity X is defined by

$$X=\frac{1}{3N}\sum_{\mathbf{q}j}X_{\mathbf{q}j}, \tag{6}$$

where $X_{\mathbf{q}j}$ is defined as the sum of the second, third, and fourth terms of the right hand side of Eq. (4) divided by $\omega^2_{\mathbf{q}j}$.

The final expression for X can then be expressed in terms of the following expressions for the free energy diagrams F_{1a}, F_{2a}, and F_{3a}:

$$F_{1a}=\frac{1}{8N\beta^2}\sum_{\mathbf{q}_1j_1}\sum_{\mathbf{q}_2j_2}\frac{\Phi(\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}_2j_2,-\mathbf{q}_2j_2)}{\omega^2_{\mathbf{q}_1j_1}\omega^2_{\mathbf{q}_2j_2}}, \tag{7}$$

$$F_{2a}=\frac{1}{48N^2\beta^3}\sum_{\mathbf{q}_1j_1}\sum_{\mathbf{q}_2j_2}\sum_{\mathbf{q}_3j_3}\frac{\Phi(-\mathbf{q}_2j_2,\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}_3j_3,-\mathbf{q}_3j_3,\mathbf{q}_2j_2)}{\omega^2_{\mathbf{q}_1j_1}\omega^2_{\mathbf{q}_2j_2}\omega^2_{\mathbf{q}_3j_3}}, \tag{8}$$

$$F_{3a}=\frac{1}{384N^3\beta^4}\sum_{\mathbf{q}_1j_1}\sum_{\mathbf{q}_2j_2}\sum_{\mathbf{q}_3j_3}\sum_{\mathbf{q}_4j_4}\frac{\Phi(-\mathbf{q}_2j_2,\mathbf{q}_1j_1,-\mathbf{q}_1j_1,\mathbf{q}_3j_3,-\mathbf{q}_3j_3,\mathbf{q}_4j_4,-\mathbf{q}_4j_4,\mathbf{q}_2j_2)}{\omega^2_{\mathbf{q}_1j_1}\omega^2_{\mathbf{q}_2j_2}\omega^2_{\mathbf{q}_3j_3}\omega^2_{\mathbf{q}_4j_4}}. \tag{9}$$

Therefore, in terms of F_{1a}, F_{2a}, F_{3a}, etc., the expression for X is given by

$$X=\frac{4\beta}{3N}\frac{F_{1a}}{1+X}+\frac{6\beta}{3N}\frac{F_{2a}}{(1+X)^2}+\frac{8\beta}{3N}\frac{F_{3a}}{(1+X)^3}, \quad (10)$$

$$X=\frac{4\beta}{3N}\left(\frac{F_{1a}}{1+X}\right)\left[1+\frac{1}{2}\frac{C}{1+X}+\cdots\right], \quad (11)$$

where

$$C=\frac{3F_{2a}}{F_{1a}}. \quad (12)$$

The series on the right hand side of Eq. (11) can be summed approximately in closed form, and the final expression for X is given by

$$X=\frac{2\beta}{3N}\left(\frac{2F_{1a}}{1+X}\right)\left(\frac{1+X}{C}\right)[e^{C/(1+X)}-1], \quad (13)$$

from which the solution for X is obtained iteratively from the knowledge of only two free energy diagrams F_{1a} and F_{2a}.

Since the Green's function, given by Eq. (1), is of the same mathematical structure as the Green's function derived in the harmonic approximation, the Helmholtz free energy (F) can be written as

$$F=U_{\text{static}}+\frac{1}{\beta}\sum_{\mathbf{q}j}\ln\left[2\sinh\left(\frac{1}{2}\beta\hbar\Omega_{\mathbf{q}j}\right)\right], \quad (14)$$

where U_{static} is the static energy contribution to F. We note here that it has been possible to write the above expression from the standard harmonic expression for F with $\omega_{\mathbf{q}j}$ replaced by $\Omega_{\mathbf{q}j}$ because $\Omega_{\mathbf{q}j}$ is independent of the applied frequency ω. For yet another method of the derivation of Eq. (14) see Ref. 3. However, compared to a simple harmonic vertex (representing the force constant) here we have a renormalized vertex or force constant with ambiguities in the interpretation of the ring and the insert in the ring in the renormalization process for the lowest order diagrams which happens to be of first order in the SC1 theory. Therefore, certain contributions of the first-order free energy like F_{1a}, F_{2a}, etc., with appropriate numerical factors have to be subtracted from the above expression for F. Hence, the correct free energy in the SC1 theory is given by

$$F=U_{\text{static}}+\frac{1}{\beta}\sum_{\mathbf{q}j}\ln\left[2\sinh\left(\frac{1}{2}\beta\hbar\Omega_{\mathbf{q}j}\right)\right]-G(X), \quad (15)$$

where

$$G(X)=\frac{F_{1a}}{(1+X)^2}+\frac{2F_{2a}}{(1+X)^3}+\frac{3F_{3a}}{(1+X)^4}+\cdots. \quad (16)$$

We note here that in the usual expression for the SC1 free energy, the sum total of all these subtracted terms is represented by the average of $U_{\text{static}}-(\hbar\Omega_{\mathbf{q}j}/4)\coth(\frac{1}{2}\beta\hbar\Omega_{\mathbf{q}j})$.

In the high temperature or the classical limit the terms independent of $\hbar$ in the second term of Eq. (15) can be isolated with the help of the standard product representation of $\sinh x/x$, with $x=\frac{1}{2}\beta\hbar\Omega_{\mathbf{q}j}$, where $\Omega_{\mathbf{q}j}^2=\omega_{\mathbf{q}j}^2+D_4(\mathbf{q}j)+D_6(\mathbf{q}j)+D_8(\mathbf{q}j)$. The symbols $D_4(\mathbf{q}j)$, $D_6(\mathbf{q}j)$, and $D_8(\mathbf{q}j)$ introduced here represent the second, third, and fourth terms, respectively, on the right hand side of Eq. (4). The above procedure yields

$$F=U_{\text{static}}+\frac{3N}{2\beta}\ln(1+X)-G(X). \quad (17)$$

Now we replace in the expression for $G(X)$ every $1+X$ by $\alpha+X$, where α is a positive parameter. The derivative of $G(X)$ with respect to X yields the following:

$$\frac{dG(X)}{dX}=\frac{-2F_{1a}}{(\alpha+X)^3}\left[1+\frac{C}{\alpha+X}+\cdots\right], \quad (18)$$

which can be approximately summed in the form

$$\frac{dG(X)}{dX}=\frac{-2F_{1a}}{(\alpha+X)^3}\exp\left(\frac{C}{\alpha+X}\right). \quad (19)$$

Integration of this equation with the boundary condition $G\to 0$ as $\alpha\to\infty$ determines the constant of integration. The resulting sum for $G(X)$, correct to $O(\lambda^4)$, is then given by

$$G(X)=\frac{2F_{1a}}{C^2}\left[\exp\left(\frac{C}{\alpha+X}\right)\left(\frac{C}{\alpha+X}-1\right)+1\right]. \quad (20)$$

Thus by setting $\alpha=1$ in Eq. (20), the ansatz for the SC1 free energy is

$$F=U_{\text{static}}+\frac{3N}{2\beta}\ln(1+X)-\frac{2F_{1a}}{C^2}\left[\exp\left(\frac{C}{1+X}\right)\left(\frac{C}{1+X}-1\right)+1\right]. \quad (21)$$

We have evaluated the equation of state using Eqs. (13) and (21), and find that it is indeed a very good approximation to the full SC1 results. This is not in itself important since the SC1 calculation is not particularly demanding, but it does show that the type of approximation used here is valid. We can also sum the effect of adding any number of loops to the two vertices in diagram 1b. Diagram 2c is the lowest order example. We can write the result as

$$\frac{F_{1b}}{(1+X)^3}\exp\left(\frac{D}{1+X}\right),$$

where $D=F_{2c}/F_{1b}$. We can expect that, if we add this contribution to the SC1 free energy, given by Eq. (21), but leave the equation for X, Eq. (13), unchanged, we shall mimic the equations of the ISC approximation, and this turns out to be the case. The equation of state calculated in this way is in excellent agreement with the results of the full ISC calculation.

For the new ISC theory, we add the following corrections from diagrams 2e, 2f, 2g, and 2h:

$$\frac{F_{2e}}{(1+X)^4}\left[\exp\left(\frac{C}{1+X}\right)\right]^2,$$

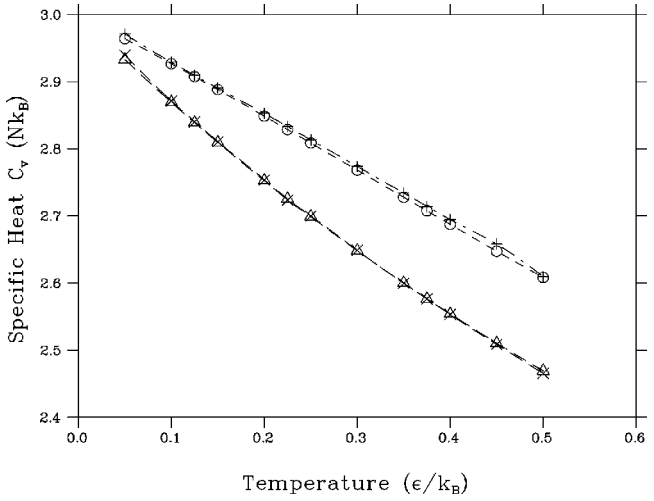

FIG. 1. Specific heat at constant volume (C_v). Symbols are ×, SC1; △, ansatz SC1. +, ISC; ○, ansatz ISC.

$$\frac{F_{2f}}{(1+X)^6}\left[\exp\left(\frac{D}{1+X}\right)\right]^2,$$

$$\frac{F_{2g}}{(1+X)^5}\exp\left(\frac{C}{1+X}\right)\exp\left(\frac{D}{1+X}\right),$$

$$\frac{F_{2h}}{(1+X)^6}\left[\exp\left(\frac{D}{1+X}\right)\right]^2.$$

When these contributions are added to the previous ISC equations, with X given by Eq. (13), we have our ISC theory.

As a final point, we can add to the expression for X a contribution proportional to the smeared 1b and include this in the iteration procedure. This approximates a *second-order* self-consistent phonon scheme (SC2). However, we find that the iterative procedure diverges for temperatures above about half of the melting temperature (i.e., above about 0.25). There have been other indications that this is a correct result. Kanney and Horton implemented a version of SC2 and also reported a divergence.[4] Cowley, Horton, and Leese made a rather different type of calculation which should have been equivalent to SC2 and also found a divergence.[12] The difficulties arise from the inclusion of the cubic shift contribution in the the quantity X, which is physically equivalent to including it in the calculation of the smearing width. The cubic shift is negative and this reduces the value of $(1+X)$. This, in turn, increases the effect of the smearing and this makes the cubic shift even more negative. Above a certain temperature the effects escalate and the iteration process diverges. While we have used an apparently crude set of numerical approximations, we believe the conclusion to be correct.

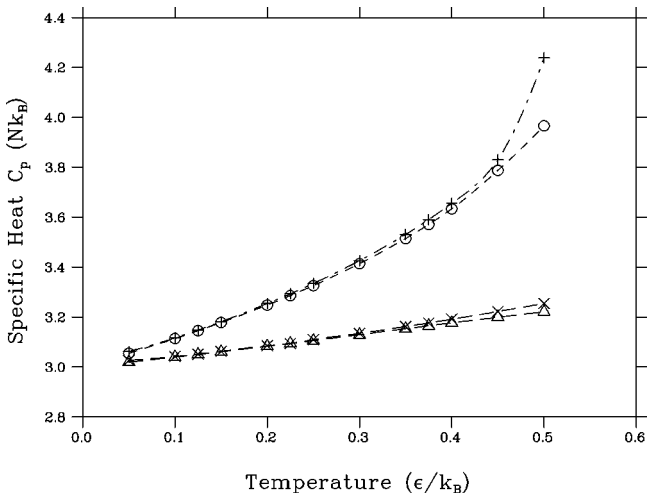

FIG. 2. Specific heat at constant pressure (C_p). Symbols are as in Fig. 1.

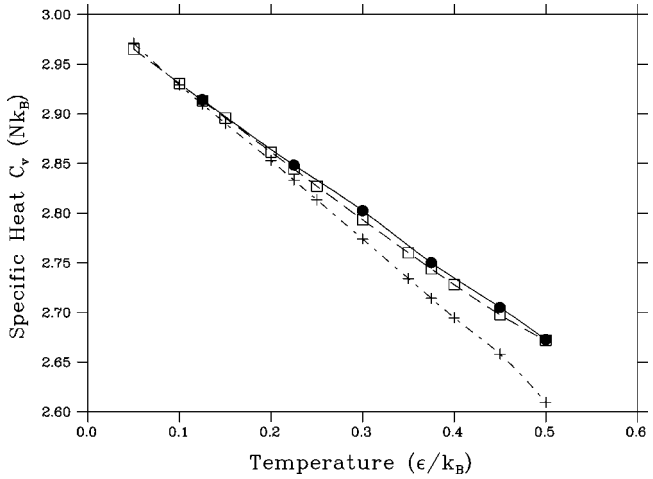

FIG. 3. Specific heat at constant volume (C_v). Symbols are ●, Monte Carlo; □, ISCX; +, ISC.

III. RESULTS AND DISCUSSION

It is clear from the ansatz-type theory presented in Sec. II that the calculations for the SC1, ISC, and our ISC theory require knowledge of the two λ^2 free energy diagrams, viz., F_{1a} and F_{1b}, and the six $O(\lambda^4)$ free energy diagrams, viz., F_{2a}, F_{2c}, F_{2e}, F_{2f}, F_{2g}, and F_{2h} as a function of the temperature (T) and volume (V). The other two free energy diagrams of $O(\lambda^4)$, viz., F_{2d} and F_{2b}, are already included through the normalization of phonon lines in F_{1b} and the SC1 free energy. Hence as a first step in the calculation, we

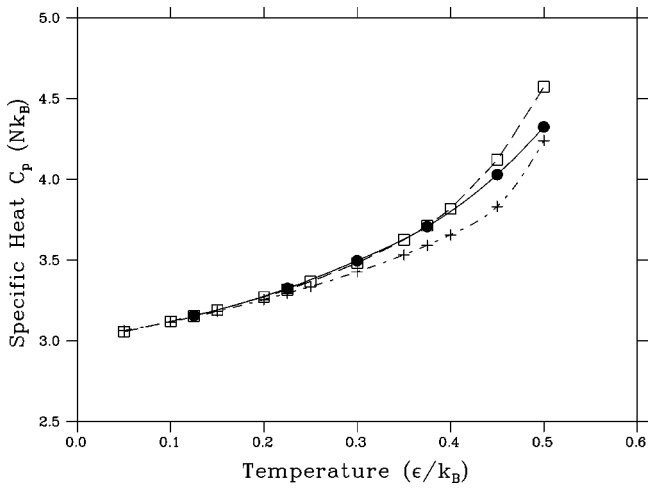

FIG. 4. Specific heat at constant pressure (C_p). Symbols as in Fig. 3.

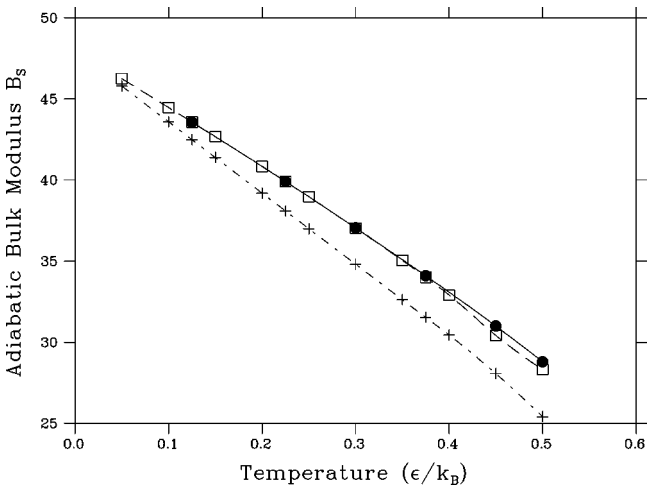

FIG. 5. Adiabatic bulk modulus (B_S). Symbols as in Fig. 3.

have evaluated these diagrams for a range of volumes, and then evaluated the thermal properties for each of the above-listed theories by suitable interpolations and differentiations. Because our calculations are done in the high temperature limit, the free energies of $O(\lambda^2)$ and $O(\lambda^4)$ have temperature factors of T^2 and T^3, respectively.

We have calculated all the thermodynamic properties from the ansatz procedure presented in Sec. II and for comparison purposes the same properties from the exact formulations of the SC1 and ISC theories. How successful the ansatz approach is in reproducing the exact results for the SC1 and ISC theories can be seen from the results for C_v (the zero pressure specific heat at constant volume) and C_p (the specific heat at constant pressure) presented in Fig. 1 and 2, respectively. Clearly the ansatz results are in excellent agreement for both the SC1 and ISC theories.

Another check of the ansatz for our ISC theory is obtained by adding the correction of only one diagram of $O(\lambda^4)$, viz., the 2e free energy diagram. An exact calculation for this case was done by Koehler[11] and our ansatz results for this case are in excellent agreement with that of Ref. 11.

The results from our ISC theory (labeled as ISCX) for C_v and C_p and the adiabatic bulk modulus (B_s) are presented in Fig. 3–5, respectively, along with the exact ISC and Monte Carlo (MC) results,[13] instead of the real experimental data because we recognize that the Lennard-Jones potential may not provide an adequate representation for the real solid. We have found a slight error in the previous calculation of C_v at the reduced temperature of $T^*=0.5$ for the exact ISC. The new value is 2.60 rather than 2.65 as reported in our earlier work.[8] This value is used in both the Figs. 1 and 3 for ISC.

Clearly the C_v results show a considerable improvement from this ISC theory because as T^* approaches 0.5, our ISC results keep up with the MC results whereas the ISC results fall below the MC values.

In the case of C_p the results from our ISC theory are in exact agreement with the MC values right up to $T^*=0.4$ and then deviate slightly for $T^*=0.45$ and 0.5 where they move higher than the MC values. On the other hand, the C_p values from the ISC theory first start falling below the MC values from $T^*=0.25$, and then go through a slight dip around $T^*=0.45$, and finally creep up to approach the MC value at $T^*=0.5$.

As we see from the results presented in Fig. 5 the B_s values from our ISC theory are in exact agreement with the MC up to $T^*=0.4$ and show a very slight deviation at $T^*=0.45$ and $T^*=0.5$. On the other hand, the B_s values from the ISC theory show a consistent trend of falling below the MC values in the entire temperature range.

IV. CONCLUSIONS

We have accomplished the goal set out in this paper of extending the ansatz for summing the contributions of the higher order diagrams in SC1 and ISC theories. The ISC theory now includes the contributions to the free energy from *all* the self-energies in the Green's function of $O(\lambda^4)$ and beyond through the propagator and vertex renormalizations. The results of the ansatz calculations agree extremely well with the exact SC1 and ISC results. For most of the thermodynamic properties in the entire temperature range, the results from our ISC theory are substantially in better agreement with the classical MC results than the existing ISC theory.

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