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Thermal and elastic properties of solid neon

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We apply the improved effective potential Monte Carlo (IEP) and the improved self-consistent (ISC) theories to study the thermal and elastic properties of natural solid Ne. As a first orientation, we use the (12-6) Lennard-Jones (LJ) potential for first-neighbor forces only. The two parameters in the potential are determined from the 0 K lattice spacing and the sublimation energy of the crystal. We also create a realistic interatomic potential for the Ne dimer based on our study of the existing literature. When supplemented by many-body contributions, this potential is also used with ISC and IEP. The results are then compared with the experimental data in the literature. We conclude that our realistic potential which we regard as the best currently available is not significantly superior in accounting for the experimental data to the LJ potential, though both give a decent account of the experimental data.

I. INTRODUCTION

In this paper we study selected thermal and elastic properties of natural neon and its isotopes. We make theoretical predictions and compare our results with the experimental data. We use both the improved self-consistent theory[1] (ISC) and the improved effective potential Monte Carlo theory[2] (IEP) to obtain our calculated results. Between them these two theories, ISC at the lowest temperature and IEP at all other temperatures give a reliable account of the lattice dynamics of solid neon.[3] In an earlier paper, ISC was applied to neon using a (6-12) Mie-Lennard-Jones potential at a time when the quality of the ISC results was not yet fully established.[4] That is why the additional use of IEP is an important part of the present paper. It establishes the reliability of our lattice dynamical results.

About 20 years ago, Klein and Koehler published an article[5] entitled ''Lattice Dynamics of Rare Gas Solids.'' In this article they examined how well the theory available at that time could account for the measured properties of solid neon. They concluded that section of their article with the comment that ''Much still remains to be done before we have an adequate description of solid neon at temperatures near melting.'' This is one of the topics we wish to address in order to ascertain how much progress has been made since then and to see whether (and how far) we have progressed beyond the cautious conclusion of Klein and Koehler.

We note immediately that the lattice dynamics of solid neon is conveniently divided into two distinct parts. Within the Born-Oppenheimer approximation,[6] which is clearly reliable for an insulator like neon, we need to know the electronic ground-state energy of the solid as a function of the atomic separations. We can then evaluate the partition function of the solid.

An attractive problem would be to evaluate this ground-state energy, or interatomic potential, *ab initio*. Unfortunately this is a very difficult problem because of the errors inherent in subtracting the huge energies of the separated atoms from the only slightly different energy of the interacting atoms. No such work has been carried out yet. It is, therefore, customary to proceed as follows.

One assumes a functional form for the potential energy of an isolated pair of neon atoms. This involves a number of adjustable parameters which are obtained from gas data[7] as well as some Hartree-Fock calculations for short distances. These results are then supplemented by many-body contributions to produce a crystal potential which is semiempirical. We will follow that path. There is a substantial literature on such potentials for solid Neon. We have found none of this earlier work to be entirely satisfactory, though we have used some of the results of Aziz and Slaman[8] in order to create our version of the best Neon dimer potential based on the currently available gas data. That forms the basis of the crystal potential we have set up. Once the crystal potential is known, we implemented Feynman's path integral approach[9] to evaluate the partition function. This noncausal technique has the great advantage over the older self-consistent phonon theory in that it avoids the process of successive approximations inherent in that theory which certainly fails at high temperatures and whose convergence for solid neon needs investigation. Higher-order approximations to the latter theory are too difficult, and perhaps too unrewarding, to implement numerically.

The new theory, known as the effective potential Monte Carlo theory, has been developed by a number of authors. Its most powerful and complete formulation was recently introduced by us under the title: improved effective potential theory (IEP). This theory is of such high quality that we are confident that the lattice dynamical part of calculating the thermal and elastic properties of solid neon at all temperatures and the zero-pressure volume is now in good order. This is assured because ISC theory, to which IEP reduces at

0 K is known to be reliable for solid neon at low temperatures.[2,10] We do present some ISC results to emphasize this point. Thus, given reliable interatomic forces, it should be possible to make a reliable comparison between theory and data from many experiments. That is the aim of this paper.

While the agreement with experiment that we achieve is not bad, we are not completely satisfied with it. Over a century ago Lord Kelvin[11] wrote

‘‘Accurate and minute measurements seem to the nonscientific imagination a less lofty and dignified work than looking for something new. But nearly all the grandest discoveries of science have been but the rewards of accurate measurements and patient long continued labor in the minute sifting of numerical results.’’

We cite one example which started with a letter by Lord Rayleigh to Nature.[12] He wrote:

‘‘I am much puzzled by some recent results as to the density of nitrogen, and shall be obliged if any of your chemical readers can offer suggestions as to the cause. According to two methods of preparation, I obtain quite distinct values. The relative difference, amounting to about 1/1000 part, is small in itself, but it lies entirely outside the errors of experiment, and can only be attributed to a variation in the character of the gas . . . .’’

Analysis of this minute discrepancy led to the discovery of argon and all the other rare gases here on earth and the subsequent award of two simultaneous Nobel prizes—one in physics and the other in chemistry. A similar huge effort went into determining the magnetic moment of the electron, both experimentally and theoretically in the hope of finding a discrepancy. Here, however, agreement to eight significant figures convincingly confirmed the reliability of quantum electrodynamics.

About 30 years ago, Simmons and his collaborators in a brilliant investigation measured the lattice parameters of rare-gas solids as a function of temperature to seven decimal places.[13] Much effort has since gone into investigating whether lattice dynamics can account for these precise data. This paper is a contribution to that effort. Unfortunately, the best theoretical results are still far from accounting for the experimental data—the disagreements are far outside the experimental uncertainties. Now this is not like the case of metals where the failure to predict the correct lattice parameters has been traced to the inadequacy of the functional-density method.[14] We do not know the reason for the persistent disagreement. We believe that our treatment of lattice dynamics is quite reliable. And we have the very best available crystal potential. Until this disagreement is laid to rest, the possibility that some new physics is required cannot be excluded. That is what has provided a very potent challenge in this work.

II. IMPROVED EFFECTIVE POTENTIAL MONTE CARLO THEORY

The basic equations of the theory[2] start most conveniently with the partition function

$$Z=\left(\frac{m}{2\pi\beta\hbar^2}\right)^{3N/2} e^{-\beta(\Delta F-\Delta F_{class})}\int d^{3N}R e^{-\beta V_{eff}(R)}, \tag{1}$$

whose structure accounts for the name given to the theory. The integral in Eq. (1) can be evaluated by classical Monte Carlo methods since quantum effects are included in V_{eff}. The expression for V_{eff} is

$$V_{eff}(R)=K(R)-\frac{M\alpha_a f_a^2}{\beta^2\hbar^2}+\frac{1}{\beta}\sum_a \ln\left(\frac{\sinh f_a}{f_a}\right), \tag{2}$$

where

$$K(R)=\frac{1}{2}\sum_I\sum_{J\neq I}\int d^3x\frac{e^{-\vec{x}^T[D^{IJ}]^{-1}\vec{x}}}{(\pi^3\det D^{IJ})^{1/2}}\phi(\vec{R}_I-\vec{R}_J+\vec{x}), \tag{3}$$

is the potential and the remaining expressions are the phonon terms.

The Gaussian width is given by

$$(D^{IJ})_{\alpha\beta}=(U_{I\alpha,a}-U_{J\alpha,a})\alpha_a(U_{I\beta,a}-U_{J\beta,a}), \tag{4}$$

where

$$\alpha_a=\frac{\beta\hbar^2}{2mf_a}(\coth f_a-1/f_a), \tag{5}$$

and

$$f_a=\beta\hbar\omega_a/2. \tag{6}$$

The secular equation is

$$U^T_{a,I\alpha}K_{I\alpha,J\beta}U_{J\beta,b}=m\omega_a^2\delta_{ab} \tag{7}$$

with the U's representing polarization vectors and the dynamical matrix given by

$$K_{I\alpha,J\beta}=\frac{\partial^2K(R)}{\partial R_{I\alpha}\partial R_{J\beta}}. \tag{8}$$

Finally,

$$\Delta F=\frac{-\hbar^2}{3NM^3}\sum_{123}\frac{\Delta(\vec{q}_1+\vec{q}_2+\vec{q}_3)}{\omega_1\omega_2\omega_3}|\psi_{123}|^2W_{123}, \tag{9}$$

with

$$W_{123}=\frac{n_1n_2+n_2n_3+n_3n_1+n_1+n_2+n_3+1}{\omega_1+\omega_2+\omega_3}+\frac{3(n_2n_3+n_3n_1-n_1n_2+n_3)}{\omega_1+\omega_2-\omega_3} \tag{10}$$

and

$$\psi_{123}=\sum_\rho e^{i(\vec{q}_1+\vec{q}_2+\vec{q}_3)\cdot R_{\rho/2}}S_\alpha(1)S_\beta(2)S_\gamma(3)\langle\psi_{\alpha\beta\gamma}(\vec{R}_\rho)\rangle, \tag{11}$$

where the sum is over lattice vectors and

$$S_\alpha(1)=\sin(\vec{q}_1\cdot\vec{R}_{\rho/2})e_\alpha(\vec{q}_1j_1),\text{ etc.} \tag{12}$$

Here $\vec{q}$ is a wave vector in the first Brillouin zone, $n=(e^{\beta\hbar\omega}-1)^{-1}$ is the average occupation number,

$\langle \psi_{\alpha\beta\gamma}(\vec{R}_\rho)\rangle$ are the smeared third-order force constants and e_α and ω_α are the normal mode vectors and frequencies.

We note that

$$\Delta F_{class} = \frac{-4}{3Nm^2\beta^2}\sum_{123}\frac{\Delta(\vec{q}_1+\vec{q}_2+\vec{q}_3)}{\omega_1^2\omega_2^2\omega_3^2}|\psi_{123}|^2. \tag{13}$$

From the structure of Eqs. (1) through (13) it is clear that at $T=0$ K, IEP reduces to ISC, and we know that ISC is reliable at low temperatures. In the high-temperature limit, never reached by solid neon, these IEP equations are exact classically and actually reproduce the first term in the Wigner expansion.[15]

In order to implement the theory, these equations must be solved iteratively. To make this process manageable, we shall use the low coupling approximation (LCA) in which the changes of α, U, and the cubic correction from their equilibrium values are neglected. One of the virtues of IEP is that, when used with the LCA (which becomes exact at high and low temperatures) there is a cancellation of errors leading the extremely reliable results.[10] To summarize, IEP and ISC theory used together can be trusted to predict the thermodynamics of solid neon reliably once the interatomic forces are known.

The IEP expressions for the physical quantities we will calculate, using Eq. (1), for comparison with available experimental data are

$$U = \frac{\partial}{\partial\beta}\ln Z = (\Delta F - \Delta F_{class}) + \beta\frac{\partial}{\partial\beta}(\Delta F - \Delta F_{class}) + \frac{3N}{2\beta} + \langle V_{eff}\rangle + \beta\left\langle \frac{\partial}{\partial\beta}V_{eff}\right\rangle, \tag{14}$$

$$P = \frac{1}{\beta}\frac{\partial}{\partial V}\ln Z = -\frac{\partial}{\partial V}(\Delta F - \Delta F_{class}) + \frac{N}{\beta V} - \left\langle \frac{\partial}{\partial V}V_{eff}\right\rangle, \tag{15}$$

$$C_V = \left(\frac{\partial U}{\partial T}\right)_V = -k\beta^2\left[2\frac{\partial}{\partial\beta}(\Delta F - \Delta F_{class}) + \beta\frac{\partial^2}{\partial\beta^2} \times (\Delta F - \Delta F_{class})\right] + \frac{3Nk}{2} - k\beta^2\left\langle 2\frac{\partial V_{eff}}{\partial\beta} + \beta\frac{\partial^2 V_{eff}}{\partial\beta^2}\right\rangle + k\beta^2\left\{\left\langle\left(V_{eff} + \beta\frac{\partial V_{eff}}{\partial\beta}\right)^2\right\rangle - \left\langle V_{eff} + \beta\frac{\partial V_{eff}}{\partial\beta}\right\rangle^2\right\}, \tag{16}$$

$$B_T = -V\left(\frac{\partial P}{\partial V}\right)_T = V\frac{\partial^2}{\partial V^2}(\Delta F - \Delta F_{class}) + \frac{N}{\beta V} + V\left\langle\frac{\partial^2 V_{eff}}{\partial V^2}\right\rangle - \beta V\left\{\left\langle\left(\frac{\partial V_{eff}}{\partial V}\right)^2\right\rangle - \left\langle\frac{\partial V_{eff}}{\partial V}\right\rangle^2\right\}. \tag{17}$$

III. INTERATOMIC FORCES IN GASEOUS AND SOLID NEON

Since no *ab initio* calculations of the ground-state electronic energy of the neon dimer as a function of nuclear separation have been presented,[14] we have turned to the extensive literature[16] on heuristic potentials to give us a starting point for evaluation of the partition function of the solid as a function of volume and temperature. We have also relied on the Born-Oppenheimer approximation[6] since the band gap in an insulator like neon is about 25 eV. Many citations to earlier work can be found in the paper by Aziz and Slaman.[8]

The neon dimer potential must exhibit van der Waals attractive dispersion behavior at large separations and then go over to strong short-range repulsion at small separation. In addition, it must account for a wide range of diverse properties including second virial coefficients, viscosity, thermal conductivity, diffusion, spectroscopic data, differential, and high-energy total scattering cross sections and link up with the small separation calculated united atom perturbation results. In addition, when supplemented by many-body forces, it must account for the 0 K binding energy and lattice spacing of the solid.

We have found that none of the published potentials satisfy these requirements. This is our conclusion in spite of the suggestion of the authors of one potential that their[17] ESMSV-III potential ‘‘must be very close to the real neon pair potential.’’ In particular we have studied the HFB-B potential of Aziz and Slaman carefully and found that it could not account for the lattice spacing of solid neon at 0 K, and suffered from unacceptable discontinuities in its higher derivatives.

One of the problems that earlier authors ran into was the fact that different sets of data were relevant at different nuclear separations and that the interatomic potential had a different analytical behavior at various separations. This led earlier authors to piece together different sections of the potential which were joined by requiring the potential and its derivative to be continuous throughout. Thus the so-called ESMSV III potential is spliced together out of no less than five different pieces. At an early stage it was Barker[19] who recognized the failings of this approach for applications to the lattice dynamics of solid neon, a highly anharmonic solid. Higher derivatives of the potential which play an important role are not well represented by the interpolation procedure. There is a related problem in subsequent work which relied on a single analytical potential but uses a cutoff function[18] to attenuate the dispersion forces at small separations. Unfortunately this cutoff also introduces unphysical potentials derivatives into the formalism although it was quoted as recently[7] as 1996.

A. Tang-Toennis-type neon potential

We have, therefore, preferred to base our potentials for the neon dimer on the work of Tang and Toennis.[20] This gives the following result for solid neon:

$$\phi(r) = Ae^{-\alpha r} - \sum_{n\geqslant 3}^{\infty}\left[1 - \sum_{k=0}^{2n}\frac{(\alpha r)^k}{k!}e^{-\alpha r}\right]\frac{C_{2n}}{r^{2n}}, \tag{18}$$

where

TABLE I. Contributions to the binding energy (all in J/mole).

Static energy	-2637
ZPE	648
DDD	63
$1+2+3$	-1926
Experiment[a]	-1933 ± 8
DDQ[b]	13
QQD[b]	2

[a] Reference 25.

[b] Reference 8.

$$C_{2n+4}=\left(\frac{C_{2n}+2}{C_{2n}}\right)^{3} C_{2n-2}, \tag{19}$$

so that C_6, C_8, and C_{10} are required and the remainder are generated by Eq. (19). The required C's were taken from the work of Aziz and Slaman.[8] Following the work of Tang and Toennis[20] on the argon dimer who found that their form of the potential required an increase in the parameter A compared with other potentials by about 16%, we have multiplied the Aziz and Slaman value of A by 1.161 948 5. That value was chosen to give the correct experimental lattice spacing and a binding energy of -1925.614 J/mole, which is just inside the experimental error. As far as possible, we have taken the parameter values from Aziz and Slaman, to take advantage of their careful fitting to a wide range of properties. We used the following parameters:

$$A=\epsilon A^{*}\times 1.161\,948\,5,$$

$$\epsilon=42.25\ \mathrm{K},$$

$$A^{*}=895\,717.95,$$

$$\alpha=4.726\ \text{per angstrom unit},$$

$$C_6=6.447\ \mathrm{au},$$

$$C_8=96.5\ \mathrm{au},$$

$$C_{10}=1520\ \mathrm{au}. \tag{20}$$

Although this Tang-Toennis-type neon dimer potential is consistent with all the known experimental and theoretical information available on neon, both are still subject to refinement which leaves room for both small changes in these parameters and the possible modification of the structure of this potential in the future. This potential, and all its derivatives are continuous. In any case the heuristic nature of our potential must always be kept in mind though we do believe it to be the best available at the present time.

In order to adapt the dimer potential for use with solid neon at atmospheric pressure, we must include many-body forces. It is commonly assumed that the most significant of these is the triple dipole contribution.[19,8] To illustrate the convergence of the contributions we have summarized the 0 K results in Table I. The coefficient that leads to the triple dipole term we quote in that table is 11.95 Hartree-(Bohr)^{9}.[21]

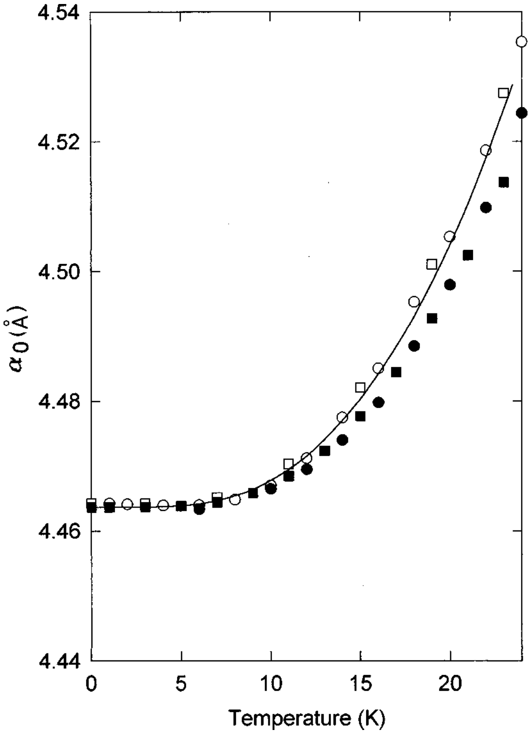

FIG. 1. The lattice spacing α_0. Hollow circles are IEP results for the Lennard-Jones potential, full circles are ISC results for the Lennard-Jones potential, hollow squares are IEP results for the TT potential, and full squares are ISC results for the TT potential. The line is the experimental curve. Error bars are smaller than the thickness of the line. The lattice spacing of solid neon at 0 K is $(4.463\,68\pm 0.000\,008)$ Å.

B. The nearest-neighbor Mie-Lennard-Jones potential

In spite of much criticism focused on its many shortcomings, the nearest-neighbor (6-12) Mie-Lennard-Jones potential continues to be used. It provides continuity as well as a first orientation enabling comparison with a whole range of earlier work. This potential is entirely *ad hoc* and compensates for the lack of many-body forces through the choice of the two potential parameters. To provide a reference, we have done all our calculations using the following potential:

$$\phi(r)=4\epsilon\left[\left(\frac{\sigma}{r}\right)^{12}-\left(\frac{\sigma}{r}\right)^{6}\right] \tag{21}$$

with

$$\epsilon=72.09\times 10^{-16}\ \mathrm{ergs}, \tag{22}$$

and

$$\sigma=2.7012\times 10^{-8}\ \mathrm{cm}. \tag{23}$$

These parameters correctly reproduce the sublimation energy and the 0 K lattice spacing of the crystal.

IV. CALCULATIONAL METHODS AND EXPERIMENTAL DATA

We have divided our IEP simulations into three parts. The first part, the static calculation, involves calculating the static contribution to each thermodynamic quantity, namely the en-

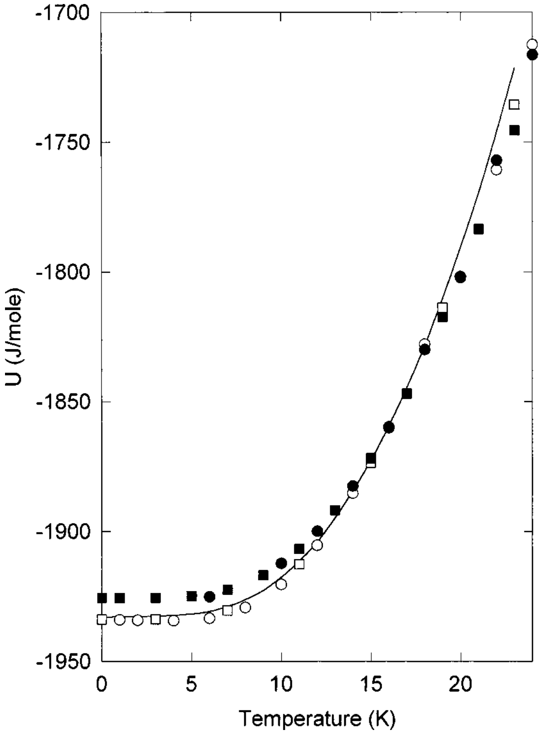

FIG. 2. The internal energy. The meaning of the symbols is same as in Fig. 1.

ergy, pressure, and specific heat at constant volume and the bulk modulus. Since this is the least costly of all three calculations, we included 112 nearest shells in the calculation corresponding to 7010 atoms.

The second part, the phonon part, involves sums over normal modes. For this part we used 1331 atoms, giving us 3993 normal modes which was sufficient for accurate evaluation of the harmonic phonon contribution. Finally, the third part was the Monte Carlo simulation itself. Here, we used 125 atoms with periodic boundary conditions. We ascertained that finite-size effects were negligible. Of course, we subtracted out the static equilibrium portion of the averaged quantities. Using IEP, at each of the 12 temperatures, we calculated the quantities given by Eqs. (14)–(17) at the zero pressure experimental volume. As expected, the calculated pressure is not zero as our potential is not perfect. So we obtained the zero-pressure theoretical lattice spacing by using the bulk modulus to make the required corrections. For the ISC calculation which are inherently faster, the quantities in Eqs. (14)–(17) were calculated directly. All the various sums involved were done to a high degree of accuracy on a PC.

The experimental data we quote come from several sources. The lattice spacings were measured by Batchelder *et al.*[22] The bulk modulus was measured by Batchelder *et al.*[22] using x-ray diffraction as well as by Anderson and Swenson[23] by extrapolation from high pressure P-V data. There is an almost 30% difference between these two sets of results at 20 K which is unresolved. The internal energy U was taken from Somoza and Fenichel[24] who integrated their smoothed C_p values (which may be too high at high temperatures thus overestimating U). The specific heats were taken from the compilation of data due to Korpiun and Lüscher.[25] We note that these are smoothed data, whereas

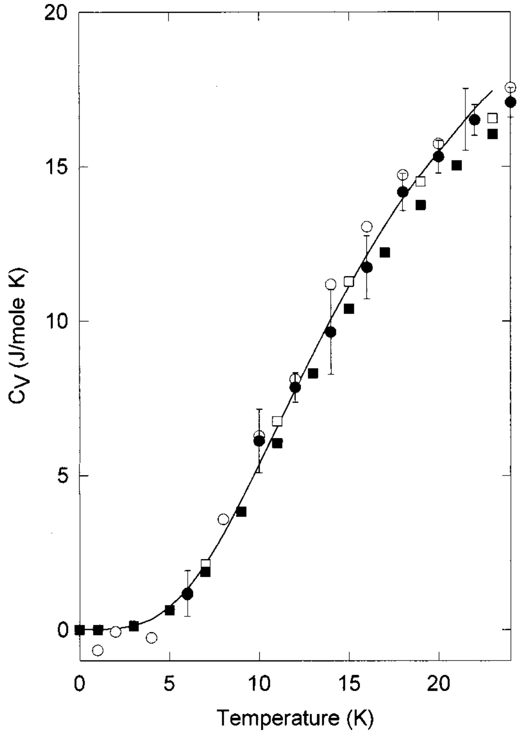

FIG. 3. The heat capacity C_v. The meaning of the symbols is same as in Fig. 1. An estimated error bar is shown on the experimental line at high T.

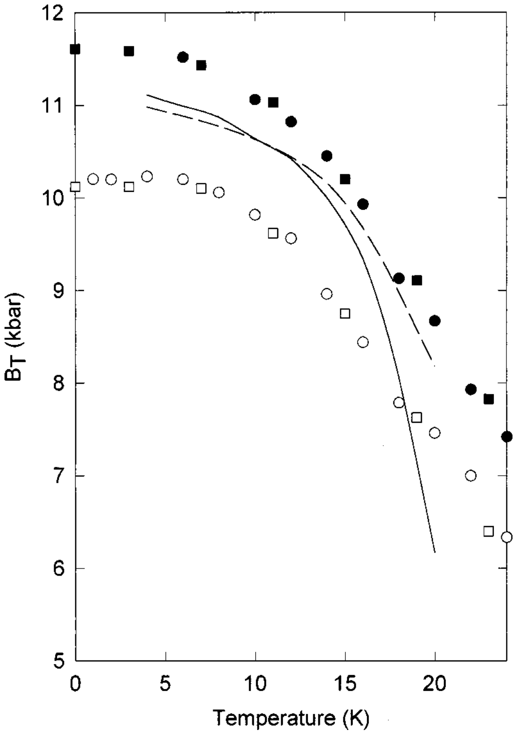

FIG. 4. The bulk modulus B_T. The meaning of the symbols is same as in Fig. 1. The solid line shows the experimental results of Batchelder *et al.*, the dashed line is the experimental results of Anderson *et al.*

our calculated values have not been smoothed. The isobaric specific heat C_p was measured and the isochoric specific heat C_v was calculated from

$$C_p - C_v = B_T V T \beta^2, \tag{24}$$

where B_T is the isothermal bulk modulus, V is the volume, T is the temperature, and β is the thermal-expansion coefficient. Since we have already noted the substantial uncertainty in B_T at high temperatures, it follows from Eq. (24) that C_v may also be uncertain by as much as 1 J/mole K.

V. CONCLUSION

We are again astonished how well the heuristic nearest-neighbor Mie-Lennard-Jones potential, Eq. (21), accounts for the experimental data we have chosen to focus on in Figs. 1–4. It is also clear that our TT-type potential for neon while reasonably satisfactory, is far from the last word on the subject. No doubt it may give a better overall fit of the experimental data if a wider range of experiments is considered, especially if volume dependences are more fully explored. The assumption that many-body forces are adequately represented by the triple-dipole term should also be re-examined. We also note that the ISC theory for Ne is quite satisfactory except near melting and is clearly superior, as expected, at the very lowest temperatures where the Monte Carlo method leads to significant scatter, including some negative C_v's, to IEP theory. The two theories clearly complement each other.

We expect that IEP theory will lead to significantly different results from those of ISC for solid argon which melts close to its debye temperature. We will show that in a subsequent paper.

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