" A COMPARATIVE STUDY OF THERMAL BEHAVIORS FOR KRYPTON GAS"

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***ABSTRACT** :The basic physical idea in this approximation is that the square of the local-field operator is replaced with its mean value. The implication is that the true quantum-mechanical spectrum of this operator is replaced with a distribution around its expectation value. In this sense, SFA is an improvement on mean-field theory. This approach was used for compute the thermodynamic properties- the mean energy, pressure, entropy and heat capacity- of krypton gas in the temperature- range 120-200 K. SFA is based on deriving a closed set of nonlinear integral equations for the system. This set is solved numerically by an iteration method. The grand partition function can then be evaluated, from which thermodynamic properties follow. The main input in this approach is the HFD-B binary potential. The results are almost identical to the ideal gas and with the experimental data*

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Keywords: static fluctuation approximation; local-field operator; Internal energy; Entropy; kr.

INTRODUCTION

SFA is an improvement on mean-field theory where the operator is replaced with its expectation value [1,2]. The two-body potential representing the Kr-Kr interaction is taken in the present work as the HFD-B potential [3]. The properties of the interatomic krypton potential are:(i) The repulsive term, describes Pauli repulsion at short ranges due to overlapping electron orbitals; (ii) The attractive long-range term, describes attraction at long ranges; (iii) The interaction energy is a minimum at the equilibrium position. This is the same as in other inert atomic systems – including helium, lithium, neon, argon, among others. Several theoretical and experimental methods have been developed to investigate the properties of Kr: An ab initio potential was used in computer simulations to yield thermodynamic properties to study the thermo physical properties of krypton gas over a wide range of densities and temperatures1[4]. Measurements of P-V-T-data of krypton have been carried out in the temperature region from 0° to 150°C and at pressures up to 2900 atmospheres [5]. This work has studied the thermodynamic properties of krypton gas under certain conditions.

BASIC PRINCIPLES:

In quantum mechanics, a Hamiltonian is an operator corresponding to the total energy of the system in most of the cases. It is usually denoted by H, also Ȟ or Ĥ. Its spectrum is the set of possible outcomes when one measures the total energy of a system. Because of its close relation to the time-evolution of a system, it is of fundamental importance in most formulations of quantum theory.

By analogy with classical mechanics, the Hamiltonian is commonly expressed as the sum of operators corresponding to the kinetic and potential energies of a system in the form : $\hat{H} = \hat{H}_0 + \hat{H}_1$. (1)

Here, $\hat{H}_0$ is the kinetic-energy term:

$$\hat{H}_0 = \int d\vec{r}\,\hat{\Psi}^+(\vec{r})\left(-\frac{\hbar^2}{2m}\nabla^2\right)\hat{\Psi}(\vec{r}), \qquad (2)$$

$\hat{\Psi}(\vec{r})$ and $\hat{\Psi}^+(\vec{r})$ being the annihilation and creation field operators, respectively. $\hat{H}_1$ is the interaction term:

$$\hat{H}_1 = \frac{1}{2}\iint d\vec{r}_1 d\vec{r}_2 \hat{\Psi}^+(\vec{r}_1)\hat{\Psi}^+(\vec{r}_2)$$

$$\times V(\vec{r}_1 - \vec{r}_2)\hat{\Psi}(\vec{r}_2)\hat{\Psi}(\vec{r}_1). \qquad (3)$$

$V(\vec{r}_1 - \vec{r}_2)$ is taken here as a central potential, depending only on the magnitude of the interparticle separation, $|\vec{r}_1 - \vec{r}_2|$. This potential is given by [3]: $V(r) = \varepsilon V^*(x)$, (4)

$$V^*(x) = A^* \exp\left(-\alpha^* x + \beta^* x^2\right) - F(x)\sum_{j=0}^{2} c_{2j+6}/x^{2j+6} \,;$$

$$F(x) = \begin{cases} \exp -\left[\left\{\frac{D}{x} - 1\right\}^2\right], & x < D \\ 1, & x > D \end{cases} \qquad x \equiv \frac{r}{r_m};$$

r_m=4.008 Å ; A*=1.10146811; α*=9.39490495; β*=-2.32607647; D=1.28; C_6=1.08822526; C_8 = 0.53911567; C_{10} = 0.42174119; ε/k_B = 201.2K . In the usual manner of second quantization [6], the field operators are written as linear combinations of the creation and annihilation operators, $\hat{b}_{\vec{k}}$ and $\hat{b}^+_{\vec{k}}$: $\hat{\Psi}(\vec{r}) = \sum_{\vec{k}} \psi_{\vec{k}}(\vec{r})\hat{b}_{\vec{k}}$; (5)

$\hat{\Psi}^+(\vec{r}) = \sum_{\vec{k}} \psi^+_{\vec{k}}(\vec{r})\hat{b}^+_{\vec{k}}$, (6), where the coefficient $\psi_{\vec{k}}(\vec{r})$ is the single-particle wave function and $\psi^+_{\vec{k}}(\vec{r})$ is its complex conjugate, and the sum is over the complete set of compatible single-particle quantum numbers describing a specific state, $\vec{k}$ being the linear momentum of the particle. In homogeneous (infinite) systems, all physical properties must be invariant under spatial translations. This suggests periodic boundary conditions and single-particle wave functions that are plane waves: $\psi_{\vec{k}}(\vec{r}) = \frac{1}{\sqrt{\Omega}}\exp(i\vec{k}.\vec{r})$, (7), Ω being the (normalization) volume of the system. Thus,

$$\hat{H}=\int d\vec{r}\hat{\Psi}^{+}(\vec{r})\left(-\frac{\hbar^{2}}{2m}\nabla^{2}\right)\hat{\Psi}(\vec{r})$$
$$+\frac{1}{2}\int\int d\vec{r}_1 d\vec{r}_2\hat{\Psi}^{+}(\vec{r}_1)\hat{\Psi}^{+}(\vec{r}_2)V(\vec{r}_1-\vec{r}_2)\hat{\Psi}(\vec{r}_2)\hat{\Psi}(\vec{r}_1). \quad (8)$$

Integration over the spatial coordinates gives [7] : $\hat{H}=\sum_{\vec{k}}\frac{\hbar^{2}k^{2}}{2m}\hat{b}^{+}_{\vec{k}}\hat{b}_{\vec{k}}+\frac{1}{2}\sum_{\vec{k}}V(k)\hat{\rho}_{k}\hat{\rho}_{-k}$, (9), $V(k)$ being the Fourier transform of V(r) and $\hat{\rho}_k$ the density operator: $V(k)\equiv\int V(r)\exp(i\vec{k}..\vec{r})d\vec{r};\hat{\rho}_k\equiv\frac{1}{\Omega}\sum_{\vec{q}}\hat{b}^{+}_{k+q}\hat{b}_{q}$ (10).

The single-particle energy for free particles, incorporating the chemical potential μ, is

$$\varepsilon(k)\equiv\frac{\hbar^{2}k^{2}}{2m}-\mu \quad (11)$$

This is equivalent to the particle being in a potential well with depth $-\mu$. It follows that $\hat{H}=\sum_{\vec{k}}\left(\frac{\hbar^{2}k^{2}}{2m}-\mu\right)\hat{b}^{+}_{\vec{k}}\hat{b}_{\vec{k}}+\frac{1}{2}\sum_{\vec{k}}V(k)\hat{\rho}_{k}\hat{\rho}_{-k}$ (12) The Heisenberg representation of a creation operator $\hat{b}^{+}_{k}(\tau)$, for a Hamiltonian $\hat{H}$, is given b $\hat{b}^{+}_{k}(\tau)=\exp(\tau\hat{H})\hat{b}^{+}_{k}(0)\exp(-\tau\hat{H})$ (13) $\tau\equiv$ it. The equation of motion for $\hat{b}^{+}_{\vec{k}}(\tau)$ in this representation can be written as $\frac{d\hat{b}^{+}_{k}}{d\tau}=\left[\hat{H},\hat{b}^{+}_{k}(\tau)\right]$ (14) In SFA, it is assumed that the Hamiltonian can be expressed as a linear combination of $\hat{E}_k$ and the number-of-particles operator $\hat{n}_{\vec{k}}=\hat{b}^{+}_{\vec{k}}\hat{b}_{\vec{k}}$, such that [8] $\hat{H}=\sum_{k}\hat{E}_{k}\hat{n}_{\vec{k}}$, (15), where $\hat{E}_k$ is hermitian and is assumed to commute with $\hat{b}_{\vec{k}}$ and $\hat{b}^{+}_{\vec{k}}$, which obey the well-known relations for a Bose system: $\left[\hat{b}_{k},\hat{b}^{+}_{q}\right]=\delta_{kq};\quad\left[\hat{b}^{+}_{k},\hat{b}^{+}_{q}\right]=0$ (16)

From Eqs. (14) and (15):

$$\frac{d\hat{b}^{+}_{k}}{d\tau}=\left[\hat{H},\hat{b}^{+}_{k}(\tau)\right]=\hat{E}_{k}\hat{b}^{+}_{k}. \quad (17)$$

The closed set of nonlinear integral equations describing any infinite homogeneous Bose system is obtained as follows [8,9] $\hat{E}_{\vec{k}}=\varepsilon(k)+\frac{1}{\Omega}\sum_{q}W(k,q)\,\hat{n}_{q}$, $W(k,q)=V(0)+V\left(\left|\vec{k}-\vec{q}\right|\right).$ (18) As already mentioned, in the mean-field approximation, $\hat{E}_k$ *is* replaced with its mean value: $\hat{E}_{\vec{k}}\cong\left\langle\hat{E}_{\vec{k}}\right\rangle$. (19) The implicit assumption is that the fluctuations in this operator are negligible. On the other hand, in SFA, it is the square of the operator that is replaced with its mean value:

$$\hat{E}^{2}_{\vec{k}}\cong\left\langle\hat{E}^{2}_{\vec{k}}\right\rangle=\varphi^{2}_{\vec{k}}. \quad (20)$$

The desired long-range (generating) equation is given by [8]

$$\left\langle\hat{n}_{\vec{k}}\hat{A}\right\rangle=\eta_{0}(\vec{k})\left\langle\hat{A}\right\rangle+\eta_{1}(\vec{k})\left\langle\Delta\hat{E}_{\vec{K}}\hat{A}\right\rangle, \quad (21)$$

where $\eta_{0}(\vec{k})\equiv\frac{1}{2}\left(\frac{1}{\exp\left(\frac{\langle E_k\rangle+\varphi_k}{\beta}\right)-1}+\frac{1}{\exp\left(\frac{\langle E_k\rangle-\varphi_k}{\beta}\right)-1}\right)$ (22)

$$\eta_{1}(\vec{k})\equiv\frac{1}{2\varphi_k}\left(\frac{1}{\exp\left(\frac{\langle E_k\rangle+\varphi_k}{\beta}\right)-1}-\frac{1}{\exp\left(\frac{\langle E_k\rangle-\varphi_k}{\beta}\right)-1}\right) \quad (23)$$

Putting $\hat{A}=1$ in Eq. (21) and since $\left\langle\hat{E}_{\vec{k}}\right\rangle=0$, thanks to the symmetry involved, we have $\left\langle\hat{n}_{\vec{k}}\right\rangle=\eta_{0}(\vec{k})$. (24)

In terms of the deviations of $\hat{n}_k$, defined as

$$\Delta\hat{n}_{k}\equiv\hat{n}_{k}-\left\langle\hat{n}_{k}\right\rangle, \quad (25)$$

Eq. (21) becomes

$$\left\langle\hat{n}_{\vec{k}}\hat{A}\right\rangle=\eta_{1}(\bar{k})\left\langle\Delta\hat{E}_{\vec{K}}\hat{A}\right\rangle \quad (26)$$

Putting $\hat{A}=\Delta\hat{n}_{q}$ in this equation, one has (the index c denoting 'correlations')

$$\left\langle\Delta\hat{n}_{k}\Delta\hat{n}_{q}\right\rangle_{c}=\frac{\eta_{1}(k\ )}{\Omega}\sum_{\vec{p}\neq\vec{k}}V(\vec{k}-\vec{p})\left\langle\Delta\hat{n}_{p}\Delta\hat{n}_{q}\right\rangle$$
$$=\frac{\eta_{1}(k)}{\Omega}\left(V(k-q)\left\langle(\Delta\hat{n}_{q})^{2}\right\rangle\right)+ \quad (27)$$
$$\frac{\eta_{1}(k\ )}{\Omega}\sum_{\vec{p}\neq\vec{k}}V(\vec{k}-\vec{p})\left\langle\Delta\hat{n}_{p}\Delta\hat{n}_{q}\right\rangle_{c}$$

To determine the unknown value $\left\langle(\Delta\hat{n}_{q})^{2}\right\rangle$ appearing in Eq. (27), we use the identity $\left\langle\hat{b}^{+}_{q}(\beta)\hat{b}_{q}\hat{A}\right\rangle=\left\langle\hat{b}_{q}\hat{A}\hat{b}^{+}_{q}\right\rangle$. Putting $\hat{A}=\hat{n}_{q}$ in this identity, we obtain

$$\left\langle\hat{b}^{+}_{q}(\beta)\hat{b}_{q}\hat{n}_{q}\right\rangle=\left\langle\hat{b}_{q}\hat{n}_{q}\hat{b}^{+}_{q}\right\rangle \quad (28)$$

With the commutation relation $[\hat{n}_{q},\hat{b}^{+}_{q}]=\hat{b}^{+}_{q}$, this equation becomes $\left\langle\hat{b}^{+}_{q}(\beta)\hat{b}_{q}\hat{n}_{q}\right\rangle=\left\langle\hat{b}_{q}\hat{n}_{q}\hat{b}^{+}_{q}\right\rangle=\left\langle1+2\hat{n}_{q}+\hat{n}^{2}_{q}\right\rangle$ (29) Since

$\hat{E}_q(k)$ commutes with $\hat{b}_{\vec{k}}$ and $\hat{b}^{+}_{\vec{k}}$, Eq. (28) assumes the form

$$\left\langle \hat{b}^{+}_{q}(\beta)\hat{b}_{q}\hat{n}_{q}\right\rangle = \left\langle \exp\left(\beta\hat{E}_{q}(q)\right)\hat{b}^{+}_{q}\hat{b}_{q}\hat{n}_{q}\right\rangle \qquad (30)$$

From Eqs. (29) and (30):

$$\left\langle \hat{n}_q^2\right\rangle = \left\langle \frac{1+2\hat{n}_q}{\exp\left(\beta\hat{E}_q(k\ )\right)-1}\right\rangle \qquad (31)$$

$$= \left\langle \hat{n}_q\right\rangle\left(1+2\left\langle \hat{n}_q\right\rangle\right) - 2\eta_1(q\ )\left\langle \Delta\hat{n}_q\Delta\hat{E}_q(q)\right\rangle$$

Substituting $\hat{A} = \Delta\hat{E}_k$ in Eq. (25), we have

$$\eta_1(k)\varphi_k^2 = \frac{1}{\Omega}\sum_{\vec{p}} V(\vec{k}-\vec{p})\left\langle \Delta\hat{n}_k\Delta\hat{n}_p\right\rangle_c \qquad (32)$$

CALCULATIONS: Most of the aggregate thermodynamic variables of the system, such as the total energy, free energy, entropy, and pressure, can be expressed in terms of the partition function Q or its derivatives, where Q is:

$$Q\ = \mathrm{Tr}\left(\exp(-\beta\hat{H}\ )\right) = \sum_{n_p}\exp\left[-\beta\sum_{p}\hat{E}_P\hat{n}_p\right]$$

$$= \Pi_{p}\sum_{n_p}\exp\left(-\beta\hat{E}_P\hat{n}_p\right). \qquad (33)$$

It is simpler to take the logarithms of both sides:

$$\ln Q = \ln\left(\Pi_{\vec{p}}\sum_{n_p}\exp\left(-\beta\hat{E}_P\hat{n}_p\right)\right)$$

$$= \sum_{\vec{p}}\ln\left(\sum_{n_p}\exp\left(-\beta\hat{E}_P\hat{n}_p\right)\right)$$

$$= \sum_{\vec{p}}\ln\left(\frac{1}{1-\exp\left(-\beta\hat{E}_P\right)}\right)$$

$$= -\sum_{\vec{p}}\ln\left(1-\exp\left(-\beta\hat{E}_P\right)\right) \qquad (34)$$

By using the identity $B\left(a+b\Delta\hat{E}_K\right) \equiv \eta_0(k)+\eta_1(k)\Delta\hat{E}_k$, one has

$$\ln Q = -\sum_{\vec{p}}[q_0(p)+q_1(p)\Delta\hat{E}_P] \qquad (35)$$

Taking into account the symmetry of the two eigenvalues of the operator $\Delta\hat{E}_P$, one finally gets $\ln Q = -\sum_{\vec{p}} q_0(p)$

(36)

$$q_0(p) = \frac{1}{2}\ln\left[\begin{array}{l}\left(1-\exp\left(-\beta\left(\left\langle \hat{E}_P\right\rangle+\varphi_P\right)\right)\right)\\ \times\left(1-\exp\left(-\beta\left(\left\langle \hat{E}_P\right\rangle-\varphi_P\right)\right)\right)\end{array}\right] \qquad (37)$$

(1) The mean internal energy U

$$U = -\left(\frac{\partial \ln Q}{\partial\beta}\right) \qquad (38)$$

so that

$$U = \sum_{\vec{p}}\left\langle \hat{n}_p\hat{E}_p\right\rangle = \sum_{\vec{p}}\left(\left\langle \hat{E}_p\right\rangle\left\langle \hat{n}_p\right\rangle + \varphi_p^2\eta_1(p)\right) \qquad (39)$$

(2) The pressure P

$$P = k_B T\frac{\ln Q}{\Omega} \qquad (40)$$

(3) The entropy S

$$S = \frac{1}{T}(U+P\Omega-\mu N) \qquad (41)$$

(4) The heat capacity at constant volume C_v

$$C_V = \left(\frac{\partial U}{\partial T}\right)_{\Omega} \qquad (42)$$

In the thermodynamic limit, the summations in Eqs. (18), (27) and (32) can be converted to integrals. Performing the angular integrals, one gets

$$\left\langle \hat{E}_k\right\rangle = \varepsilon(k) + \frac{1}{2\pi^2}\int_0^\infty V(k,q)\left\langle \hat{n}_q\right\rangle q^2 dq \qquad (43)$$

$$\left\langle \Delta\hat{n}_k\Delta\hat{n}_q\right\rangle_c = \frac{\eta_1(k)}{2\pi^2}\int_0^\infty V(k,p))\left\langle \Delta\hat{n}_p\Delta\hat{n}_q\right\rangle_c p^2 dp \qquad (44)$$

$$\eta_1(k)\varphi_k^2 = \frac{1}{2\pi^2}\int_0^\infty V(k,p)\left\langle \Delta\hat{n}_k\Delta\hat{n}_p\right\rangle_c p^2 dp \qquad (45)$$

For central potentials, by virtue of spherical symmetry, one can use the familiar simple form of the relative partial-wave expansion:

$$V\left(\vec{k},\vec{p}\right) = \sum_{\ell(\mathrm{even})}(2\ell+1)V_\ell(k,p)P_\ell\left(\hat{k}.\hat{p}\right) \qquad (46)$$

where $V_\ell(k,p)$ is the Fourier-Bessel transform of potential:

$$V_\ell(k,p) \equiv 4\pi\int_0^\infty V(r)j_\ell(kr)j_\ell(pr)r^2 dr \qquad (47)$$

$j_\ell(x)$ being the spherical Bessel function of order ℓ and argument x.This transform was calculated using a program originally constructed by Ghassib and coworkers for interhelium potentials [10]. The set of nonlinear integral Eqs. (43), (44) and (45) were solved numerically by Gaussian quadrature [11,12,13].

RESULTS AND DISCUSSION:

Figures 1-8 show the thermodynamic properties of Kr gas. Figs.1-4showT-dependenceat $n=2\times10^{25}$ atoms/m^3,with the ideal gas results; Figs. 5-8 concern with n- dependence at different T. Temperature is an indicator of internal energy, U is proportional to the T, as shown in Fig.1; This is because repulsive forces increase with increasing T. The slope of the curve (U-T) was about 12.15 J/K... The physical meaning of this slope is Cv/NkB ; then this value close to the ideal gas value

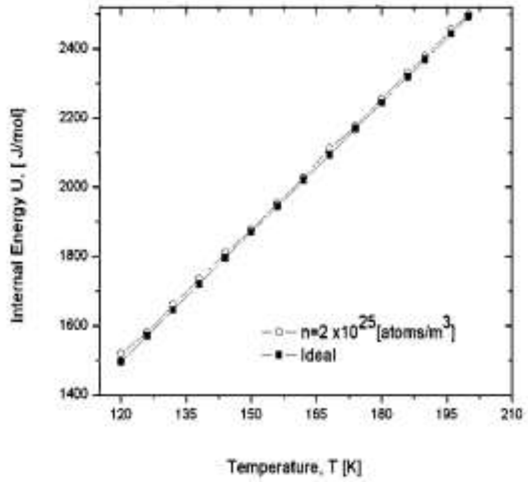

Figure 1: The internal energy U [J/mol] varies with temperature[K]at n= 2x10^{25}atoms/m^3.

In Fig.2 Plotting the pressure (P) against the absolute temperature (T) gives a straight line. This shows the pressure of the gas is directly proportional to the absolute temperature of the gas; as the temperature increases, the molecules in the gas move faster, so the kinetic energy increase the pressure**.**

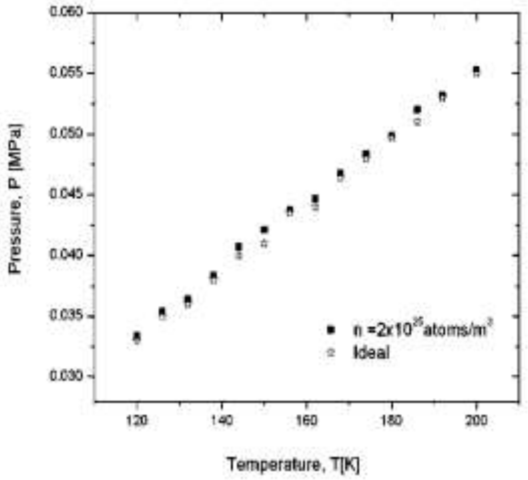

Figure 2: The pressure P[MPa] varies with temperature T [K]at n=2 ×10^{25}atoms/m^3.

Entropy is a measure of how much the energy of atoms and molecules become more spread out in a process. Fig.3 shows changes in temperature will lead to changes in entropy. The higher the temperature the more thermal energy the system has; the more thermal energy the system has, the more ways there are to distribute that energy; the more ways there are to distribute that energy, the higher the entropy. Increasing the temperature will increase the entropy.

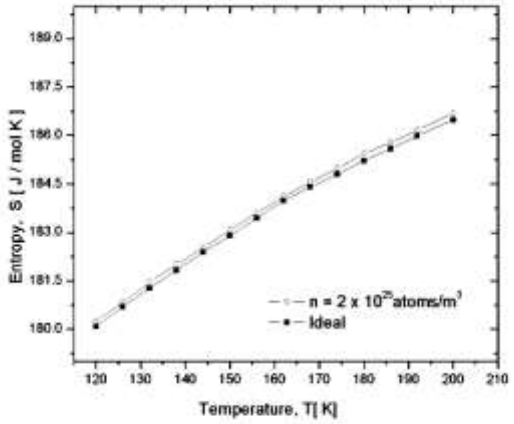

Figure 3: The entropy S [J/mol K] varies with temperature T [K] at n= 2×10^{25} atoms/m^3.

Further, in Fig. 4 C_v remains constant with increasing T,(C_V~ 12.5 J/mole K) and the gas behave qualitatively like an ideal gas, as expected on physical grounds.

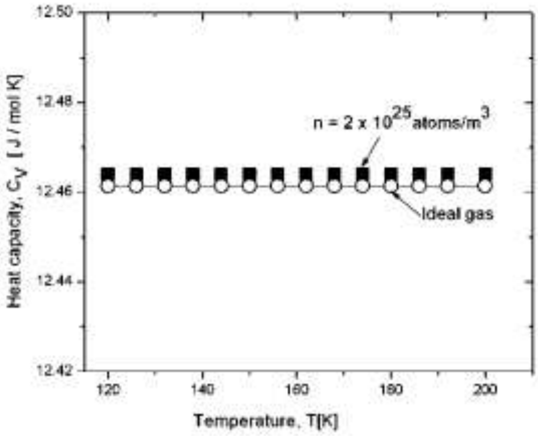

Figure 4:The heat capacity CV [J/mol K] varies with temperature T [K] at n= 2×10^{25} atoms/m^3.

The functional dependence on T of the above thermodynamic quantities can be straightforwardly obtained by fitting procedures. The results are:

1. $U \propto T^{0.9}$;
2. $P \propto T^{1.00008}$;
3. $S \propto T^{0.085}$;
4. C_v is nearly constant.

This T-dependence is the same as for the ideal system; the small deviation arises from the weak interactions in Kr gas. Also, P and U both increased with increasing *n*, as shown in Figs. 5 and 6, respectively. When density is higher, the gas contains more gas atoms. The repulsive forces increase; therefore the increase in n will result in the increase of U and P.

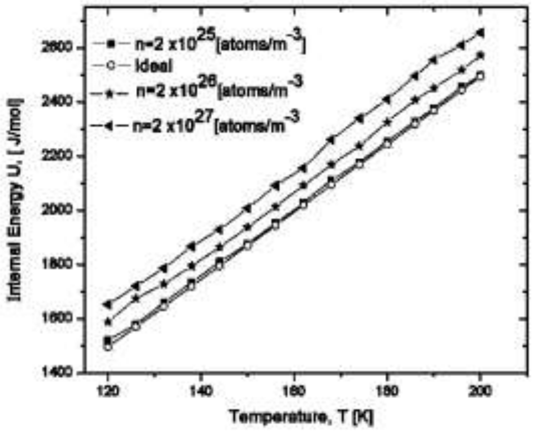

Figure 5: The internal energy U [J/mol] varies with temperatures T [K] at different n .

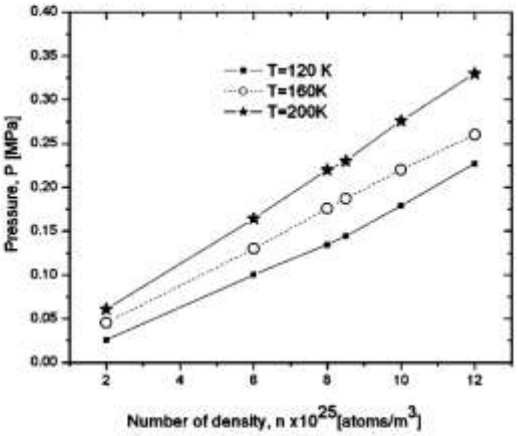

Figure 6:The pressure P [MPa] as a function of number density n×10^{25} [atoms/m^3] at different temperatures T [K].

If we increased density then we increased the number of atoms, which causes an increase in the randomness/disorder of the system; therefore the increase in n will result in the increase S as shown in Fig. 7.

On the other hand, C_v almost is independent on n.

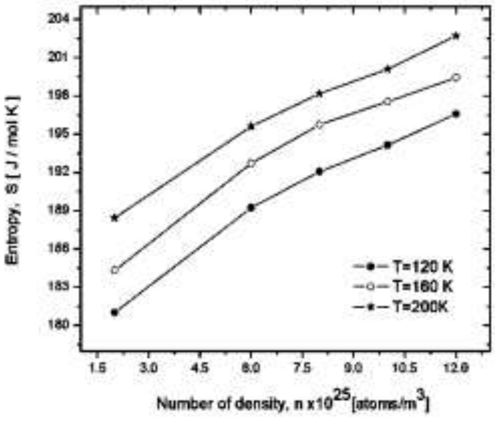

Figure 7:The entropy S [J/mol K] varies with number density n×10^{25} [atoms/m^3] at different temperatures [K].

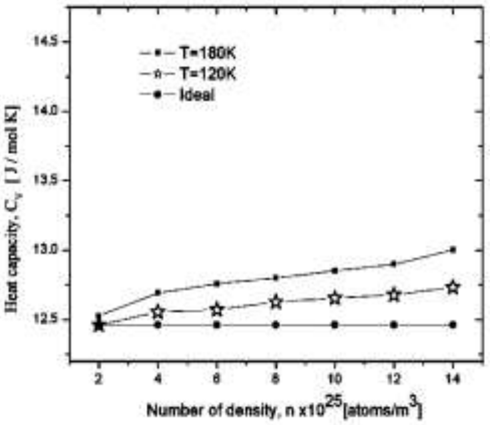

Figure 8: The heat capacity C_v [J/mol K] as a function of number density n × 10^{25} [atoms/m^3] at different temperatures T [K].

Tables 1,2 show our work for P and C_v values respectively and the experimental values [14] at the same conditions, i.e., there was very good agreement.

Table 1:Comparison of our work and experimental pressure values [MPa] for different number densities n[atoms.m^{-3}] and temperatures T.

T[K]	n	$P_{exp.}$	Our work	ΔP%
124	8.5×10^{25}	0.13966	0.146	4
128	10×10^{25}	0.18487	0.181	2
132	10×10^{25}	0.24037	0.198	17.6

Table 2:Comparison of our work and experimental heat capacity values[J/moleK] for different number densities n[atoms.m^{-3}] and temperatures T.

T[K]	n	C_{vexp}	Our work	ΔCv%
124	8.5×1025	13.6	12.72	6
128	10×1025	13.86	12.88	4
132	10×1025	14.09	12.89	8.5

CONCLUSION

In this paper the internal energy, pressure, entropy and heat capacity of the Kr gas were calculated within the SFA framework at temperature range (120-200)K using the HFD-B potential. The results were compared with those of an ideal gas the small deviation arises from the weak interactions in Kr gas. Our results are compared in Table 12 to previous experimental results. U, P and S are proportional to T and n. This is because repulsive forces increase with increasing T and increasing n, On the other hand the dependence of the specific heat on the temperature and density is weak and the gas becomes more and more ideal with decreasing n. As expected on physical grounds, the noble gases such as Kr gas are nearly ideal gases. Our results are compared in Table 1,2 to previous experimental results, the results of our work are in good agreement with the experimental data.

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