On the free rotation of a molecule embedded in helium-4 clusters

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Abstract

The fact, that \(^4\)He atoms on different concentric circular paths around the axis of a quantum vortex move with identically equal angular momentum, which represents an important aspect of superfluidity of He-II, has been used to discover a model which can explain the typical nature of experimentally observed \(N\) (number of \(^4\)He atoms) dependence of the rotational constant \(B\) of the rotor part of a cluster M:He\(_N\). It reveals how exactly superfluidity is related to the said dependence of \(B\) on \(N\). We believe that this model, when used with simulation techniques, would render results that would agree closely with experiments.

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Ever since the spectroscopic study of SF$_6$ molecule embedded in superfluid helium-4 (He-II) was performed by Goyal et al. [1], rotational and vibrational dynamics of several molecules (say, OCS, CO$_2$, CO, N$_2$O, HCCCN, etc., represented by M) embedded in bulk He-II and its nano-droplets and clusters of the form M:He$_N$ (where $N = 1, 2, 3, ...$ is the number of He atoms) have been extensively investigated. While, in a breakthrough work, Grebenov et al. [2] found that OCS molecule embedded in $^4$He droplets, isolated in the normal phase of liquid $^3$He, rotates almost like a free rotor if the droplet has about 60 or more $^4$He atoms, systematic experimental study of M:He$_N$ clusters have demonstrated non-trivial dependence of their rotational constant $B$ (or moment of inertia, $I$) and vibrational frequency shift $\Delta \nu$ (of select modes of vibrations) with $N$ which concludes that superfluidity of $^4$He atoms has observable impact on $B$ of such small clusters as well [3, 4, 5, 6, 7]. A number of theoretical models, such as, (i) super-molecule model [8], (ii) two fluid model [2, 9], (iii) quantum hydrodynamic model [10], etc., have been used to explain the initial observations. Simulation techniques have also been used, recently, to explain the phenomenon but with limited success at quantitative scale [11, 12, 13, 14, 15, 16, 17, 18]. It is not surprising because all these models associate the phenomenon with the superfluidity of $^4$He atoms which by itself is not clearly understood [19, 20].

The conventional microscopic theory (CMT) [19, 20] of a system of interacting bosons (SIB), such as liquid $^4$He, uses single particle basis (SPB) for its description. It considers that particles occupy different quantum states of a single particle placed in a box of volume $V$ of the system and these states are described by plane waves ($u_k(r) = A \exp(i \mathbf{k} \cdot \mathbf{r})$ where symbols have their usual meaning); in other words a single particle represents the basic unit with an assumption that its momentum remains a good quantum number even in the superfluid state of the system. The theory concludes that: (1) the state of liquid $^4$He at a temperature ($T$) can be identified by the momentum distribution $N(p)$ of its particles where different number of particles $N_p(T)$ have different $p$, and (2) with the onset of superfluid transition, $N(p)$ does not change significantly except for the existence of a fraction of particles, $n_{p=0}(T) = N_{p=0}/N$ having $p = 0$ in LT phase. Accordingly, even the ground state (G-state) of liquid $^4$He has different number of atoms in the states of different momenta, viz., $N_{p=0}$ in $p(=\hbar k) = 0$ state and $N_{p\neq0}$ in several states of non-zero momenta, $\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, ...$ etc. (expressed in wave number). Based on different estimates by a large number of theoretical and experimental studies, $N_{p=0}$ ($p = 0$ condensate) is believed to fall around 10% [19, 21] leaving $N_{p\neq0}$ (non-condensate) to about 90%. In what follows CMT identifies that $p = 0$ condensate as the origin of superfluidity and related properties of He-II and the same viewpoint is attributed to the superfluidity exhibited by microscopic systems of $^4$He atoms (viz., droplets and clusters) too. The advances in CMT made over the last several years [20] suggest that superfluid phase of liquid $^4$He also has (in addition to $p = 0$ condensate) pair condensate (similar to Copper pair condensation in superconductors) or a composite condensate ($p = 0$ condensate, pair condensate, 3 particle condensate, etc.) as the origin of superfluidity.

We note that SPB used in CMT not only complicates the process of finding different aspects of a SIB, e.g., the expectation value of inter-particle interaction which becomes infinitely repulsive for short distances, but also ignores the reality that the states of wave superposition assumed by the particles at low $T$ can not be described by $u_k(r)$. It is well known that: (i) two particles (say P1 and P2) in the state of their wave superposition are described by $\Psi(1, 2) = [u_k(r)u_k(r) \pm u_k(r)u_k(r)]$ which basically represents a pair of particles moving with equal and opposite momenta ($\mathbf{q}, -\mathbf{q}$) with respect to their CM which moves with momentum $\mathbf{K}$ in the laboratory frame and (ii) positions ($\mathbf{r}_1$, and $\mathbf{r}_2$), momenta ($\mathbf{k}_1$ and $\mathbf{k}_2$) and energies
(ε₁ and ε₂) of two particles as separate entities lose their meaning in this state.

While superfluidity undoubtedly has a relation with the experimental observation of the rotation of a molecule embedded in a \(^4\)He droplet and the non-trivial \(N\)-dependence of \(B\) of M:\(^4\)He\(_N\) cluster, it certainly has no relation with \(p = 0\) condensate because, as established unequivocally by one of us (Jain [22]), \(p = 0\) condensate does not exist in the superfluid phase of a SIB. Starting with an assumption that the G-state of liquid \(^4\)He has non-zero values of both, \(N_{p=0}\) and \(N_{p≠0}\), as concluded by CMT, Jain [22] finds that: (i) such an \(N(p)\) does not represent a state of minimum possible energy as expected for the G-state of every physical system, (ii) all particles in the true G-state of a SIB have identically equal energy \((ε_o = h^2/8md^2)\) with \(h\) being the Planck constant, \(m\) the mass of a particle and \(d = (V/N)^{1/3}\) and corresponding non-zero momentum \((q_o = \pi/d)\), and (iii) the real form BEC that exists in a SIB is the macroscopic condensation of bosons as \((q, -q)\) pairs in their G-state characterized by \(q = q_o = \pi/d\) and \(K = 0\).

Motivated by all such observations, one of us [23] used more realistic pair of particle basis PPB to conclude his non-conventional microscopic theory (NCMT) which emphasizes a pair as the basic unit of the system. The theory not only explains different properties of He-II at quantitative scale [23, 24, 25] but also reveals that: (i) particles (G-state) of a SIB have identically equal energy \((ε_o = h^2/8md^2)\) and corresponding non-zero \(q = q_o = \pi/d\) which agrees with a recent study by Jain [23], (ii) they constitute a kind of close packed arrangement of their representative wave packets (CPA-WP) of identically equal size \(λ/2 = h/2p = d\), (iii) they are allowed to move only coherently in order of their locations, obviously, with no relative motion and mutual collision, and (iv) they occupy phase \((φ)\) positions which differ by \(Δφ = 2nπ\) (with \(n = 1, 2, 3, ...\)). In addition, the theory finds that all the three characteristics of the G-state are retained by the superfluid phase over the entire range of temperature from \(T = 0\) to \(T_λ\) and the entire system assumes a kind of collective binding for which it behaves like a macroscopic molecule.

Since \(^4\)He atoms in M:He\(_N\) clusters are confined to a space of few Å size, it is evident that each \(^4\)He atom has non-zero energy and corresponding non-zero momentum for the confinement. This undoubtedly proves the absence of \(p = 0\) condensate in these systems and we use these aspects of Jain’s NCMT [23] to frame a model which provides a better account for the typical nature of non-trivial dependence of \(B\) on \(N\) revealed from experimental observations on selected M:He\(_N\) clusters. In this context it may be mentioned that our initial efforts [26] tried to use some simple thoughts to explain the effect by presuming that: (i) each added atom which takes the cluster from M:He\(_N\) to M:He\(_N+1\) can significantly change the positions of other \(^4\)He atoms from the axis of rotation, and (ii) with \(N\) increasing beyond its certain value (depending on several physico-chemical aspects of M), \(^4\)He atoms start occupying the second position from M (e.g. in M(zero)-\(^4\)He(first)-\(^4\)He(second)) and these atoms interact so weekly with the rotor-part of cluster (M and few \(^4\)He atoms, -at first position, which interact directly with M) that they do not follow the rotation of the rotor. In a sense the net potential seen by the rotor part of the cluster appears to remain constant with a change in the angular position of the rotor. In other words the rotor (when rotating about its axis) seems to roll over a equi-potential surface; else if there are hills and valleys in the surface, the height of hills is much lower than the energy of rotational excitation of the rotor. Although these efforts rendered a satisfying account of the phenomenon, they could not find a clear relation to superfluidity of \(^4\)He atoms.
and this motivated us in concluding this model which not only associates an important aspect of superfluid He-II with the non-trivial dependence of \( I \) on \( N \) but also clarifies how only few \( ^4 \text{He} \) atoms attached directly to \( M \) take part in the rotation and rests do not follow the rotation effectively.

In what follows from the experimentally observed \( N \) dependence of \( B \) (represented typically by a curve depicted in Fig.(1)), we observe the following:

(A) \( B \) decreases when \( N \) is increased from \( N = N^i \) (the lowest \( N \) for which experimental data are available) to certain \( N = N^* \) that may depend on several factors like the size and symmetry of the structure of \( M \), the strength of \( M-\text{He} \) interaction, etc.

(B) It remains nearly constant when \( N \) is increased beyond \( N^* \) only by 1 or 2 or so but increases with further increase in \( N \) up to another value, say \( N_1 \).

(C) When \( N \) is increased beyond \( N_1 \), \( B \) is observed to decrease and increase alternately over different ranges of \( N \), \( N_1 - N_2, N_2 - N_3, N_3 - N_4 \), and \( N_4 - N_5 \) and so on.

(D) \( \Delta \nu \) is observed first to increase linearly for first few \( ^4 \text{He} \) atoms (with \( N < N^* \)) but beyond this point it decreases with nearly a linear dependence on \( N \); however, the slope of this decrease has different values over the ranges, \( N^* - N_1, N_1 - N_2, N_2 - N_3, N_3 - N_4 \), and so on.

It is well known that \( \Delta \nu \) is a simple consequence of a change in potential \( V(Q_1, Q_2, ....Q_S) \) (governing all the \( S \) possible vibrational modes of \( M \)) with changing \( N \). It could be explained in terms of a small change in the related potential constant appearing in the harmonic component in the expansion of \( V(Q_1, Q_2, ....Q_S) \). Although, it is difficult to argue in favour of increase or decrease in the value of potential constant of the chosen vibration, yet, however, a simple logic indicates that \( \nu \) should increase for first few \( ^4 \text{He} \) atoms which occupy position in the closest vicinity of \( M \) since \( ^4 \text{He} \)-atoms are saturated with the electron charge density for which they would give away a small fraction of their own electron density to \( M \) which should strengthen the forces that govern its different modes of vibration and this is corroborated by experimental observation. Although, with \( ^4 \text{He} \)-atoms occupying second or third, ..., positions counting from \( M \) (at zero-th position), it is difficult to argue whether \( \nu \) would increase or decrease with \( N \), however, it is clear the effect on \( \nu \) should decrease with each added atom and this expectation agrees with decrease in slope of \( \Delta \nu \) vs \( N \) observed experimentally. In what follows from these points, the change in \( \Delta \nu \) with \( N \) has nothing to do with microscopic superfluidity of the \( ^4 \text{He} \) atoms in the cluster. Hence, in this paper, we simply concentrate to find the origin of (A), (B) and (C) and conclude a general model of the phenomenon.

1. To a good approximation, the experimental observation of decrease in \( B \) (or increase in \( I \)) for \( N^i \) to \( N^* \) can be explained by using rigid rotor picture for the cluster since this falls in line with the fact that a \( ^4 \text{He} \) atom interacts more strongly with \( M \) than with another \( ^4 \text{He} \) atom, the structure of the cluster \( M:He_N \) for \( N = N^i \) to \( N = N^* \) can, therefore, be presumed to have a rigid rotor structure for the first few rotational excitations of each cluster. We note that \( N^* \) can be different for different \( M \) (depending on its physico-chemical nature), while \( N^i \) can, in principle, be as small as 1.

2. However, the non-trivial dependence of \( B \) on \( N \) represented by the observations that \( B \) remains nearly unchanged when \( N \) changes by one or two \( ^4 \text{He} \)-atoms beyond \( N^* \) and there
after it follows cycles of its increase from $N^* + \Delta N$ to $N_1$, $N_2$ to $N_3$, ... and decrease from $N_1$ to $N_2$, $N_3$ to $N_4$, ... . This unexpected observation, naturally indicates its relation with superfluidity of $^4$He-atoms. Consequently, we try to explain it in terms of an important aspect of superfluid $^4$He exhibited by it under the influence of its rotation.

The fact, that different atoms on different concentric circles around the axis of a quantum vortex in He-II move coherently in order of their locations in a manner that they have no difference in their angular momentum [27] implies that their angular velocity ($\omega$) changes as $r^{-2}$ where $r$ is the distance of the atom from the axis of the vortex. This differs from atoms of a rigid body rotor where all atoms move around the axis of rotation with identically equal $\omega$.

In the following we consider the example of a set of $^4$He atoms moving on two concentric circles around the axis of rotation (as shown in Fig.2A) under the condition of: (i) constant angular velocity and (ii) constant angular momentum. To this effect we evaluate the kinetic energy of the set by using,

$$E = \sum_{i} \frac{n_i}{2} mr_i^2 \omega_i^2 + \sum_{j} \frac{n_j}{2} mr_j^2 \omega_j^2,$$

where we have $n_i = 1, 2, 3, ... 6$ and $n_j = 1, 2, 3, ... 12$ with indices $i$ and $j$ to identify different atoms on orbits 1 and 2, respectively. Presuming that the radii of orbits 1 and 2 satisfy

$$r_2 = 2r_1 = 2r_o$$

and all atoms move as a single rigid body with

$$\omega_1 = \omega_2 = \omega_o,$$

we have

$$E = \frac{1}{2} \left[ 6mr_o^2 \omega_o^2 + 48mr_o^2 \omega_o^2 \right] = \frac{1}{2} \left[ 54mr_o^2 \omega_o^2 \right] = \frac{1}{2} I \omega_o^2 \quad \text{or} \quad I = 54mr_o^2.$$  

Since the condition of constant angular momentum, applied to two atoms moving on different orbits of radius $r_1$ and $r_2$ (cf., Fig.2B) renders

$$mr_1^2 \omega_1 = mr_2^2 \omega_2 = C \quad \text{(constant)} \quad \text{or} \quad \omega \propto r^{-2},$$

which implies that corresponding linear velocity $v = r\omega$ changes as $r^{-1}$. As expected, this agrees with the well known dependence of $v$ on $r$ in a quantum vortex observed in superfluid $^4$He [27]. Using Eqn.(5) in Eqn.(1), we get

$$E = \frac{1}{2} \left[ 6mr_1^2 \omega_1^2 + 12mr_2^2 \omega_1^2 \left( \frac{r_1}{r_2} \right)^4 \right]$$

which for the orbits satisfying Eqn.(2) and $\omega_1 = \omega_o$ renders

$$E = \frac{1}{2} \left[ 6mr_o^2 \omega_o^2 + 12mr_o^2 \omega_o^2 \frac{1}{4} \right] = \frac{1}{2} \left[ 9mr_o^2 \omega_o^2 \right]; \quad \text{with} \quad I = 9mr_o^2.$$  

Eqn.(6) clearly reveals that the contribution to $I$ from an atom at a distance $r_2$, under the condition of constant angular momentum (Eqn. 5), gets reduced by a significant factor of $(r_1/r_2)^4$ (since $r_1 < r_2$) in comparison to that found under the condition of constant angular
velocity (Eqn.(4)). As an example, the contribution to \( I \) by an atom added to an orbit satisfying \( r_2 = 2r_1 \), Eqn.(2) gets reduced to a value as low as 1/16 (\( i.e. \approx 6.3\% \)) and for the orbit satisfying \( r_2 = 3r_1 \), the said contribution is as low as 1/81 (\( \approx 1.3\% \)). This speaks of the smallness of the contribution of an added atom to the \( I \) of the cluster when it goes to an orbit of higher \( r \) and evinces that the said atom has a +ve contribution indicating that \( I_{N+1} > I_N \).

In what follows the above stated inferences, we can expect a small increase or almost no change in \( I \) for each added atom to the cluster with \( N = N^* \). However, it gives no clue for the experimental observations of \( I_{N+1} < I_N \) for \( N > N^* \). To this effect our critical thinking reveals that the phenomenon is possible only if the added atom reduces the distance of 4He atoms (all the \( N \) atoms or a few of them) in M:4He\(_N\) cluster from the axis of rotation to an extent that contribution to \( I \) by the added single atom is over compensated by the decrease in \( I_N \). It is also possible if the added atom transforms the structure of M:4He\(_N\) cluster in a manner that one atom from first orbit of radius \( r_1 \) moves to the orbit of radius \( r_2 \) of the added atom (as shown in Fig.2C); in this case the net change \( \Delta I = I_{N+1} - I_N \) becomes

\[
\Delta I = 2mr_o^2 \frac{r_1^4}{r_2^2} - mr_1^2 = mr_1^2 \left[ 2\frac{r_1^2}{r_2^2} - 1 \right]
\]

which assumes a -ve value for \( r_2 > \sqrt{2}r_1 \) indicating that \( I_{N+1} < I_N \) when \( r_2 > \sqrt{2}r_1 \). Using this possibility for \( N \) increasing beyond \( N^* \) by 1 atom, we have

\[
\Delta I = -0.5mr_o^2
\]

by using Eqns.(2) and (8). Presuming further that another atom moves similarly from orbit-1 to orbit-2 when an added atom to the cluster occupies orbit-2 as shown in Fig.2D, we have

\[
\Delta I = 4mr_o^2 \frac{r_1^4}{r_2^2} - 2mr_1^2 = mr_1^2 \left[ 4\frac{r_1^2}{r_2^2} - 2 \right] = 1.0mr_o^2
\]

for orbits satisfying Eqn.(2). Generalizing Eqns.(8) and Eqn.(10), we have

\[
\Delta I = mr_1^2 \left[ 2n_c \frac{r_1^2}{r_2^2} - n_c \right] = mr_o^2 \left[ \frac{2n_c}{\alpha^2} - n_c \right]
\]

which represents the change in \( I \) when \( n_c \) atoms (above \( N^* \)) added to orbit-2 (making total \( N = N^* + n_c \)) induce \( n_c \) atoms from orbit-1 to jump to orbit-2. Eqn.(11) reveals that \( I \) has no change if \( \alpha(= r_2/r_1) = \sqrt{2} \), it decreases by \( \Delta I = -0.5n_cmr_o^2 \) for \( r_2 = 2r_1 = 2r_o \) (\( i.e., \alpha = 2 \)) and by a maximum of \( \Delta I = -n_cmr_o^2 \) for \( r_2 >> r_1 (= r_2) \). Such changes in \( I \) for \( n_c = 1, 2, 3, ... \) for different \( \alpha = r_2/r_1 \) are depicted in Fig.3 for their better perception.

For a possible situation where no atom jumps from orbit-1 to orbit-2 when \((n_c + 1)\)-th atom is added to orbit-2, it is evident that the added atom increases \( I \) by \( mr_o^2/\alpha^2 \). We have

\[
\Delta I = mr_o^2 \left[ \frac{2n_c}{\alpha^2} - n_c \right] + mr_o^2 \frac{1}{\alpha^2}.
\]

which again means \( I_{N+1} > I_N \) provided the added atom makes no change in \( \alpha \). However, if it does and changes in \( r_2 \) and \( r_1 \) are such that \( \alpha \) increases to \( \alpha^* = \alpha + \Delta \alpha \), then by using Eqn.(12), we find

\[
\delta(\Delta I) = \Delta I(\alpha^*) - \Delta I(\alpha) = -mr_o^2 \frac{2(2n_c + 1)\Delta \alpha}{\alpha^3}.
\]
We note that this -ve change in $I$ can overcompensate the increase in $I$ by $mr^2/\alpha^2$ if

$$\Delta \alpha \geq \frac{\alpha}{2(2n_c + 1)} \left( \frac{1}{2} \frac{r_2}{r_1} \right)$$

(14)

which is obtained by equating RHS of Eqn.(13) to $mr^2/\alpha^2$. This indicates that $I$ of the cluster can have continuous decrease with increase in $N$ (possibly from $N^*$ to $N_1$) if $\alpha = r_2/r_1$ increases by an appropriate value of $\Delta \alpha$ with each added atom. In other words an agreement between theory and experiment can be seen by using $n_c$, $\alpha$, and $\Delta \alpha$ as adjustable parameters. Note that increase in $\alpha$ is possible both by decrease in $r_1$ and increase in $r_2$ when an atom is added to the cluster. However, it appears that desired increase in $r_2$ is more probable than decrease in $r_1$. As revealed by Eqn.(14), decrease in $I$ is possible if $\Delta \alpha/\alpha$ increases by more than 16.6%, 10% and 7.1%, respectively, in case of $n_c = 1$, 2, and 3. These aspects are depicted in Fig. 4 for their better understanding.

Summing up the possible explanation for the phenomenon in the light of our preceding analysis, we may mention that:

(1) With increasing $N$ from $N^i$ to $N^*$, $^4$He atoms in M: $^4$He$_N$ cluster (for $N \leq N^*$) seem to have reasonably strong binding with M for which the cluster as whole represents a rigid rotor, to a good approximation, and its $I$ increases (or corresponding $B$ decreases) with $N$ in agreement with experiments (cf. Fig.1, for $N \leq N^*$).

(2) For nearly no change in $I_N$ from $I_{N^*}$, when $N$ is set to have a value $N^* + \Delta N$ (where $\Delta N$ has only small value such as 1 or 2 or so), it appears that each of the $\Delta N$ atoms go to orbit 2 for which $\Delta I (= I_{N^*+1} - I_{N^*})$ is $\approx 6.3\%$ of the contribution of an atom in orbit 1. Such a small increase can be easily compensated if the contribution to $I_{N^*+1}$ due to each of the $N^*$ atoms in orbit 1 gets reduced by $6.3/N^*\%$ presumably due to small decrease, $\Delta r$, in the distance of atoms from the axis of rotation of M:$^4$He$_{N^*}$ and this does not demand necessarily a decrease in M:$^4$He bond length; a decrease by $\Delta r$ in the projection of the bond on the plane $\perp$ to the axis of rotation would suffice and this can be estimated to fall around $6.3/2N^*\%$ of the said projection which equals to $\approx 0.8\%$ if $N^* = 4$, or $0.5\%$ if $N^* = 6$ or $\approx 0.4\%$ if $N^* = 8$. Such a small change can easily be expected as a possible effect of an atom added to M:$^4$He$_{N^*}$ or another added to M:$^4$He$_{N^*+1}$; this naturally explains the said observation.

(3) For the remaining part of $I_N$ vs. $N$ curve (i.e., for $N > N^* + \Delta N$) where $I_N$ is observed to have significant decrease with increase in $N$, changes in $\alpha = r_2/r_1$ along with the jump of an atom from orbit 1 to orbit 2 seem to take place when an atom is added to orbit 2. Depending on the physico-chemical nature and size of M, the decrease in $I_N$ for $N$ changing from $N = N^* + \Delta N$ to $N = N_1$ can be explained by choosing $n_c = 1$ and 2 etc. (in different steps) clubbed with appropriate values of $\alpha$ and $\Delta \alpha$. This speaks of the sensitivity of the changes in $I_N$ on $\alpha$ and $\Delta \alpha$ as well as $\Delta r$ (change in the said projection of M:$^4$He bond length). This naturally simplifies the basis our understanding of the observed increase and decrease in $I$ (Fig.1) with $N$. (4) The decrease in $B$ (increase in $I$) for $N$ increasing from $N_1$ to $N_2$ is as per normal expectation. However, each atom added to the cluster in this range contributes only very small fraction of the contribution to $I$ by an atom in orbit 1. Guided by this fact it appears that value of $\alpha$, $\Delta \alpha$ and $\Delta r$ should explain the $N-$ dependence of $I_N$ not only in this range but for all values of $N > N_1$.

Identifying the ranges, $0 - N_1$, $N_1 - N_2$, $N_2 - N_3$, etc. (Fig. 1) as cycles of increase and
decrease in \( I_N \), it appears that M:\(^4\)He\(_N\) cluster has different shells of \(^4\)He atoms around M and each cycle represents the completion of one shell. While \(^4\)He atoms in the first shell have direct bond with M, those in second, third, ... shells are separated from M, respectively, by 1, 2, ... \(^4\)He atoms in between. The maximum number of \(^4\)He atoms, in a particular shell increases in proportion of \( R^2 \) (where \( R \) is the radius of the shell which changes from one shell to next shell in units of the diameter of the sphere which represents the shape and size of \(^4\)He atom; however it also depends on the shape and size of M. Assuming that M has a shape and size of a \(^4\)He atom, a rough estimate reveals that the number of \(^4\)He atoms in first, second and third shells, should be around 6, 18 and 40, respectively. However, it may be emphasized that these numbers agree approximately with experimental values because M may have linear or a complex structure. Further, it may also be mentioned that it is not the length of M-\(^4\)He bond which changes much with atoms added to the cluster but the projection of this bond on the plane \( \perp \) to the axis of the rotor which should be considered to explain the changes in \( I_N \) with added \(^4\)He atoms that we observe through experiments.

The experimental observations seem to indicate that the part of M:\(^4\)He\(_N\) cluster which rotates, to a good approximation as a rigid rotor, has fewer than \( N^* \) \(^4\)He atoms in clusters of \( N \gg N^* \). This agrees with our suggestion that \( n_c (= 1 \text{ or } 2, \text{ or so}) \) atoms move from orbit 1 to orbit 2 with increasing \( N \) beyond \( N^* \). However, the physics of this possibility is not yet very clear.

In the light of the fact that \(^4\)He atoms in superfluid state make a close packed arrangement of their wave packets (CPA-WP) supported by a number of experimental observations such as the observation of Stark effect of roton transition seen through microwave absorption [28] and the unequivocal conclusions of a number of theoretical studies [22, 23, 24], the rotor part of the cluster in CPA-WP type arrangement of \(^4\)He atoms may in certain cases experience a kind of low energy potential barrier with an axial symmetry of the order \( n \) (as shown in Fig.5). A theoretical analysis for such a case [29] reveals that the effective \( I \) of the rotor has a lower value that depends on the height and symmetry of \( V_n \) (cf., Fig. 5). This renders an additional reason for a small decrease in \( I \).

As concluded by Jain’s NCMT of a system of interacting bosons such as liquid \(^4\)He [23], atoms in He-II not only move coherently in order of their locations but also have identically equal angular momentum when they move on different concentric paths around the axis of a quantum vortex; in fact this theory for the first time answers a question raised by Wilks [27] in relation to Feynman’s account for the origin of quantized circulation [30]. Wilks has rightly argued that Feynman’s basis for quantum vortices in He-II is equally valid for He-I but the latter does not show any quantum vortex. Using a basic aspect of quantum vortices observed in He-II, we discover a model which has enough potential to explain qualitatively the typical nature of experimentally observed \( N \) dependence of the rotational constant \( B \) of the rotor part of the cluster M:He\(_N\). Naturally, the question, how exactly superfluidity is related to the said dependence of \( B \) is answered with utmost clarity. We hope that this model, when used with simulation techniques on individual cluster, would render results that would agree closely with experiments. This would not only help in improving the model but also for having a clear understanding of the phenomenon. However, we could not take up this task for the want of facilities of computer simulations at our end.
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Figure 1: Typical nature of the $N$ dependence of rotational constant (B) of the rotor in a $M$:He cluster.
Figure 2: (A) Two concentric circular paths of a vortex on which atoms move in order of their locations, (B) two atoms moving on different concentric paths, (C) jump of an atom from inner path to outer path with an added atom in the cluster, (D) jump of two atoms from inner path to outer path with two added atoms in the cluster.
Figure 3: Dependence of $\Delta I$ on $\alpha = \frac{r_2}{r_1}$ (Eqn.11) for $n_c = 1, 2, \text{ and } 3$
Figure 4: Dependence of $\delta(\Delta I)$ on $\Delta \alpha$ (Eqn.13) for $n_c = 1$ and different values of $\alpha$
Figure 5: Variation of n fold symmetric potential with rotation angle $\theta$. 

$V_n(\theta)$