Experimental realities refuting the existence of p=0 condensate in a system of interacting bosons : II. Spectroscopy of embedded molecules

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Abstract

Experimental observation of superfluidity in a microscopic cluster, $M : (^4He)_x$, of a molecule (M) and x number of ^4He atoms (with x ranging from 1 to many) is qualitatively analyzed. It concludes that: (i) each ^4He atom in the cluster has to have non-zero momentum for its confinement to a space of size (< the size of the cluster), (ii) superfluidity does not require atoms with zero momentum ($p = 0$), and (iii) while all ^4He atoms in the cluster cease to have relative motions (hence the inter-atomic collisions), they retain a freedom to move coherently in order of their locations on a closed path around the rotor (M plus few nearest ^4He atoms which follow the molecular rotation for their relatively strong binding with M). The analysis also identifies the basic arrangement of ^4He atoms which allows the rotor to have free rotation in the cluster.

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Statistical analysis of a *system of non-interacting bosons* (SNIB) [1] and Bogoliubov's field theoretical study of a system of weakly interacting bosons [2] provided foundations for a popular belief that particles in the ground state (G-state) of a *system of interacting bosons* (SIB) (such as liquid ^4He [3] and trapped dilute gases [4]) are distributed over the states of different momenta, $k = 0, k_1, k_2, k_3, ...$ *etc.* (in wave number unit); bosons with zero and non-zero momenta, respectively, constitute what we call Bose Einstein condensate (BEC) [or zero momentum (p $(= \hbar k) = 0$) condensate] and non-condensate. The momentum distribution of particles in the G-state of a SNIB (Fig.1(A)) is compared with the above said distribution in a SIB (Fig.1(B)) for their better understanding. While the latter has been believed to exist in superfluid SIB at all temperatures, $T < T_c$ (the critical T for the onset of superfluidity in a SIB), as the origin of superfluidity and related properties for the last seven decades, we recently discovered [5] that this distribution by no means represents the G-state of a SIB because it does not constitute a state of lowest possible energy as expected; in other words the laws of nature (demanding the G-state of a physical system to have minimum possible energy) forbids the said momentum distribution of bosons ($p = 0$ condensate + non-condensate) in the G-state of a SIB. Our study [5] also discovered the true form of energy/momentum distribution of particles in the G-state of a SIB, -accordingly, all particles in this state have to have identically equal energy $\varepsilon_o = h^2/8md^2$ ($\equiv$ momentum $p = h/2d$ depicted in Fig.1(C)) which not only represents the G-state energy (momentum) of a particle trapped in a cavity formed by nearest neighbors but also underlines the fact that not even a single particle has $p = 0$; consequently, the question of macroscopically large number of particles having $p = 0$ does not arise. However, the distribution (Fig.1(B)) seems to have strong bias in its favor, possibly, because of prolonged belief of people in it. A shift from this belief, naturally, not only demands a theoretical foundation as discovered in [5] but also seeks strong experimental support for the distribution (Fig.1(C)) concluded in [5]. In this context, we identify several physical realities and experimental observations which not only support the G-state represented by Fig.1(C) but also refute the possibility of existence of $p = 0$ condensate in a SIB. We prove these points in our recent paper [6] (the first of a series of papers on this issue) by using the physical reality of the existence of an *electron bubble* (EB) in liquid helium. Similarly, this paper (the second of the series) uses the experimentally observed high resolution ro-vibrational spectra of molecules embedded in different clusters of ^4He atoms since these spectra provide strong evidence for the resistance free rotations of the embedded molecule in the clusters.

Ever since a systematic study [8] of the high resolution spectra of OCS molecule embedded in liquid ^4He droplets concluded that superfluidity is exhibited even by a drop having fewer (as low as 60) ^4He atoms, high resolution ro-vibrational spectra of different molecules, M (OCS, $/N_2O$, *etc.*) embedded in ^4He clusters or droplets ($M :^4 He_x$ where the number of ^4He atoms, x, changes from 1 to many), have been reported [9-12]. In what follows $M :^4 He_x$ clusters even with fewer ^4He atoms (say 6 or so) exhibit superfluidity and the effective moment of inertia (I^*) of the molecule has non-trivial dependence on x; as expected, it first increases with increasing x but beyond certain x (depending on the embedded M) it starts decreasing with increasing x and with further increase in x, it has a kind of periodic (*nearly*) increase and decrease. Undoubtedly, these observations reveal resistance free rotational motion of the rotor (M or M attached with a few ^4He atoms) which underlines the fact that a set of ^4He atoms in each cluster assume the state of superfluid for which they do not follow the rotations of the rotor. Although, numerous efforts have been made to understand the phenomenon, it is still not clear whether $p = 0$ condensate of ^4He atoms exists in these clusters as the origin of the phenomenon in agreement with conventional belief and this motivated us to examine this issue in this paper.

Since each cluster studied in these experiments has to have stable structure under their physical conditions, its constituents (M and ^4He atoms) have sufficiently strong binding (originating from their mutual interactions) that does not allow them to escape the cluster. Further since their mutual interaction at short distances has infinitely strong repulsive character, no constituent is expected to share its position coordinate with others. Thus each constituent in the G-state of the cluster exclusively occupies certain space of size, $d < R$ with R being the size of the cluster which agrees with [5, 6]; obviously, such a ^4He atom is expected to behave like a trapped particle and for this reason has reasonably high non-zero momentum $q = \pi/d$, certainly $> \pi/R$. Evidently the question of a ^4He atom having zero momentum or the cluster having $p = 0$ condensate does not arise. It may be noted that, long before in 1973, Kleban [13] indicated that the existence of $p = 0$ condensate in superfluid ^4He contradicts excluded volume condition which states that each hard core particle, such as ^4He atom, occupies certain volume in the fluid exclusively. However, our analysis, reported in this paper and in [6], is in exact agreement with excluded volume condition [13].

The resistance free rotation of the molecule in a cluster (identified as a proof of superfluid state of ^4He atoms) has no relation with $p = 0$ condensate because it does not exist. This agrees exactly with our recent study [5] which concludes that the laws of nature, that demand the G-state of a SIB (microscopic or macroscopic) to have lowest possible energy, forbid the existence of $p = 0$ condensate in the state.

Since this holds true for the G-state where superfluid density (ρ_s) equals the total density (ρ) of the system, possibility of the existence of $p = 0$ condensate at non-zero T where $\rho_s < \rho$ does not arise. In what follows from this observation and our study [5], superfluidity of a cluster of ^4He atoms and the bulk of superfluid ^4He has a common origin. Accordingly, it is a property which comes into play when all bosons assume localized positions with a possibility to move coherently in the order of their locations.

When different atoms have different momenta, they have relative motions which render inter-atomic collisions which are expected to impede the rotations of the rotor in the cluster. Hence the observation of free rotation of an embedded molecule in a cluster is a proof for the absence of relative motions of its constituents (M and ^4He atoms). Evidently, a set of such ^4He atoms would move (if they do) coherently in order of their locations and it is well known that atoms in superfluid ^4He really have coherence of their motion.

As an important property of a fluid, its constituents move freely on a surface of constant potential unless they suffer mutual collisions. For a cluster of ^4He atoms, surrounding a molecule, a surface (*path*) of constant potential can be a closed shell (*closed orbit on a equi-potential shell*) with the molecule at their center. Naturally ^4He atoms located on such a shell or an orbit would not affect a molecular rotation provided they do not suffer collisions which is only possible when every set of ^4He atoms move coherently in order of their location with identically equal momentum and two such orbits do not cross each other because in such a case atoms would have finite probability to collide.

We believe that the physical situation offered by the cluster to its rotor is not much different from the situation of a molecule trapped at the center of a cage formed by a set of ^4He atoms in their ground state (where they have their localized positions with certain amount of position and momentum uncertainty) and the interactions between the molecule and ^4He atoms are such that the molecule sees no change in its potential energy with a change in its angular posture with respect to stationary cage. Naturally when the molecule is made to rotate by its excitation it would rotate like a free molecule. To be more realistic it is possible that the potential energy of the molecule changes with a change in its angular posture but with a peak value much lower than the energy of first rotational excitation. The shape, size and structure of the cage depend on the number of ^4He atoms which constitute a part of the rotor for their strong binding with the molecule due to their nearest neighbor positions. This number may, obviously, depend on the size of the embedded molecule (larger is this size, larger should be the number of ^4He atoms taking positions as its nearest neighbors). When the number of ^4He atoms available for the structure of the cage are very low, it could be a simple ring around the axis of rotation, however, with increase in the number of such ^4He atoms the ring may spread into a first shell around the rotor and with further increase in x it may grow into several shells (second, third, ...). We believe that : (i) minimum number of ^4He atoms available to form the said cage should be two, and (ii) each ^4He atom added to the cluster not only change the size, shape and structure of the cage, it also affects shape, size and structure of the rotor; of course the impact on the rotor should, obviously, diminish with growing size of the cage particularly after the completion of first shell. We note that these observations identify the basic arrangement of ^4He atoms which allows the embedded molecule to rotate like a free rotor. In a recent paper [14], we used this picture to explain the x dependence of non-trivial changes in I^* of N_2O and $HCCCN$ molecules embedded in clusters of ^4He atoms.

Experimental observations of the resistance free rotation of a molecule in a cluster of ^4He atoms clearly refutes the existence of $p = 0$ condensate of ^4He atoms. We find that : (i) no ^4He atom in the cluster has zero momentum since it assumes non-zero energy and equivalent non-zero momentum for its confinement and this leaves no possibility for the ^4He atoms to constitute what we define as $p = 0$ condensate, and (ii) superfluidity of ^4He atoms in the clusters has nothing to do with $p = 0$ condensate; it is a simple property which comes in to play because particles (molecule and ^4He atoms) in the cluster cease to have relative (collisional) motions (the main reason for the viscosity of a fluid) and retain the possibility to move in order of their locations on closed paths. Since our microscopic theory of a SIB identifies exactly these factors as the origin of superfluidity of the bulk of liquid ^4He, it finds strong experimental support from the observation of superfluidity in the said clusters and so is particularly true for its conclusion about the momentum/ energy distribution (*cf.*, Fig.1(C)) of particles in the G-state of a SIB. With the same objective, we would study experimentally observed quantum evaporation of ^4He atoms from superfluid ^4He and the Stark effect of roton transition seen in microwave absorption in our forthcoming papers.

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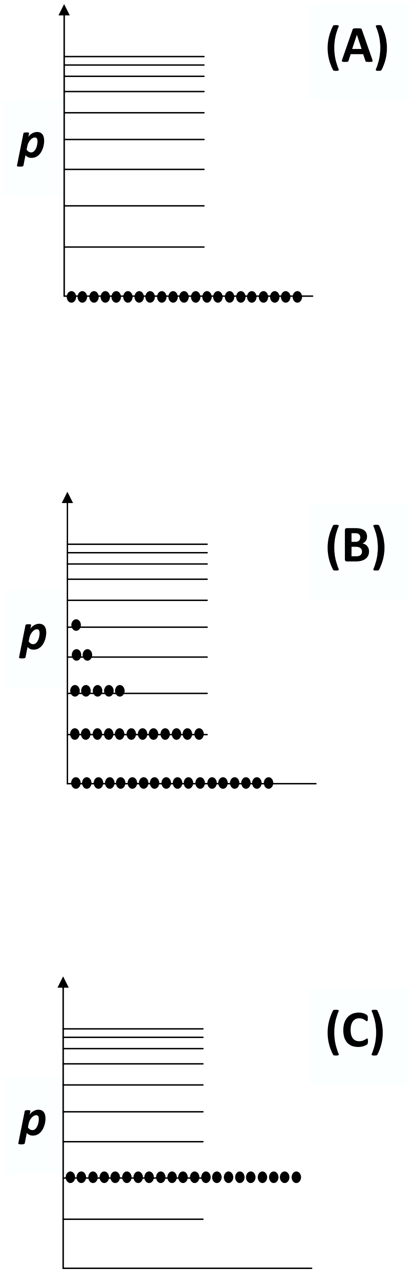

Fig.1 : Schematic of distribution of N bosons in their ground state. (A) All the N particles occupy $p = 0$ state in a system of non-interacting bosons, (B) depletion of $p = 0$ condensate (*i.e.* only a fraction $n_{p=0} = N_{p=0}/N$ of N bosons occupy $p = 0$ state) in weakly interacting boson system as predicted by Bogoliubov model [2], and (C) all the N particles occupy a state of $p = p_o = \hbar q_o = h/2d$ and $\hbar K = 0$ as concluded by this study and our recent analysis [5].