MACROSCOPIC QUANTUM STATE AND HIGH-TEMPERATURE SUPERCONDUCTIVITY IN SEMI-LOCALIZED 2D ELECTRON SYSTEM WITH CIRCULAR MOLECULAR ORBITS

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Recently new type of high temperature superconductors is found which are characterized by the existence of circular molecular orbits in each unit site of 2D s/p electron system. In view of the characteristic, a new model of superfluidity is studied based on the coherent state where the zero-point oscillation of toroidal wave function causes a macroscopic quantum state. This model gives an estimation of the superfluidity transition temperature: $T_c \approx 52-117$ K for fcc C_{60}, and $T_c \approx 50-150$ K for hole-doped MgB_2.

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I. INTRODUCTION

The cuprate-oxide high T_c superconductivity are known to be caused by 2D *d*-electron system in CuO_2 network where the probability amplitude of *d* electron extends crosswise from each Cu ion. Recently new trend of high temperature superconductivity [1,2] is attracting attention where (i) non-*d* electron system seems to be responsible to superconductivity, and where (ii) crystal structure appears to possess $\|c$ 2D network composed of the inter-connection of circular molecular orbits. In the report of Schön *et al.* [1] the surface of fullerene (C_{60}) fcc crystal was hole-doped by field effect, where $T_c \approx 52-117$ K was found when lattice constant is 1.415-1.445 nm. The 2D conduction surface in the system is formed by the inter-connection of the circular molecular orbit of each spherical C_{60} molecule. On the other hand, Nagamatsu et al. [2] found that MgB_2 shows superconductivity with $T_c \approx 39$ K. The crystal structure of MgB_2 is composed of 2D network of B hexagons, which has similarity to the 2D structure of graphite (2D network of C hexagons) except the existence of centripetal attraction of electron by the field of Mg^{2+} ion.

In this paper we study the macroscopic quantum state of the 2D electron system in a network which is composed of the inter-connected molecular units with circular molecular orbits [3], where each of the 2D electrons is supposed to be semi-localized in the annular potential well of the respective molecular orbit [4].

In section II is considered the macroscopic quantum state originated from the zero-point oscillation of toroidal wave function. In Section III is given the estimation of superfluidity threshold temperature of the materials based on the result of Section II.

II. GROUND STATE OF ONE PARTICLE AND MANY PARTICLE SYSTEM

We consider a 2D system of charged particle carriers (charge Q_0 and mass M_0) in a network, which is composed of the inter-connected molecular units each of which has a circular particle orbit with the following condition. (i) Each of the 2D particles is supposed to be semi-localized in the annular potential well of the respective molecular unit. (ii) In the annular well the ground state wave function ψ_0 of each particle has toroidal amplitude distribution with null angular momentum (or $\psi_0(x,y)$ is real function like a caldera). (iii) ψ_0 makes radial zero-point oscillation of expansion and contraction by frequency ω (iv) Thermal excitation of oscillation is forbidden by the condition

$\hbar\omega >> k_B T$.

According to the assumption, we select the ground state wave function of the particle to be a caldera-like real function

$$\psi_0(m,\zeta)=\text{const}\times|\zeta|^m \exp(|\zeta|^2/4) \quad \text{with } m=1,2,3 \ldots \quad (1)$$

where $\zeta=(x\pm iy)/l$, and $l=\sqrt{\hbar/M_0\omega}$. The functional form of (1) is the same as the ground state wave function for charged particle in magnetic field without phase factor $\exp(im\theta)$. The wave function size $\sqrt{2ml}$ may be equated to the effective radius r_0 of the molecular unit. The wave function (1) is found to be the solution of Hamiltonian

$$H(x,y) = -\frac{\hbar^2}{2M_0}\left(\frac{\partial}{\partial x^2}+\frac{\partial}{\partial y^2}\right)+V(x,y)-\frac{\omega}{2}m\hbar\omega \quad , \quad (2)$$

where

$$V(x,y)=\frac{M_0\omega^2}{8}(x^2+y^2)+\frac{\hbar^2 m^2}{2M_0}\frac{1}{x^2+y^2} \quad (3)$$

$$H\psi_0(m)=(\hbar\omega/2)\psi_0(m). \quad (4)$$

V(x,y) with annular valley is the coulomb potential well made by the ions of molecular unit (or "lattice"). (2) and (3) can be rewritten in the following expression using "vector potential"

$$H(x,y)=\frac{1}{2M_0}\left[\left(\frac{\hbar}{i}\frac{\partial}{\partial x}-Q_0A_x\right)^2+\left(\frac{\hbar}{i}\frac{\partial}{\partial y}-Q_0A_y\right)^2\right]$$

$$+V'(x,y)+\hbar\omega/2 \quad (5)$$

$$(A_x,A_y)=(iM_0\omega x/2Q_0,\ iM_0\omega y/2Q_0) \quad (6)$$

$$V'(x,y)=\frac{1}{2M_0(x^2+y^2)}\hat{S}^2 \quad (7)$$

$$\hat{S}(x,y) = i\left[x\left(\frac{\hbar}{i}\frac{\partial}{\partial x} - Q_0 A_x \right) + y\left(\frac{\hbar}{i}\frac{\partial}{\partial y} - Q_0 A_y \right)\right], \quad (8)$$

$$\hat{S}\,\psi_0(m,\zeta) = m\hbar\psi_0(m,\zeta) \qquad m=1,2,3 \ldots \quad (9)$$

The vector potential (6) gives the dynamic expression of the electric force exerted by the lattice when the charged particle goes outward. In the displacement u from its equilibrium position, the particle feels lattice charge density ρ with dielectric constant ε by the equation

$$M_0 \frac{d^2}{dt^2} u = -Q_0 \rho u/\varepsilon - \rho = \omega^2 \varepsilon / Q_0 ,$$

ρ makes the following electric field **E** and vector potential **A**

$$div\mathbf{E} = div\left(-\frac{\partial \mathbf{A}}{\partial t}\right) = -i\omega\, div\mathbf{A} = \rho/\varepsilon = \omega^2 M_0 / Q_0$$

which leads to (6). The physical meaning of (8) and (9) is explained by the quantization of oscillation energy exchanged between the particle and the lattice:

$$\begin{aligned} & x\times dp_x/dt + y\times dp_y/dt \\ &= i\omega\left[x\left(\frac{\hbar}{i}\frac{\partial}{\partial x} - Q_0 A_x \right) + y\left(\frac{\hbar}{i}\frac{\partial}{\partial y} - Q_0 A_y \right)\right] \\ &= m\hbar\omega \ . \end{aligned} \quad (10)$$

The potential energy (7) expresses the repulsive polarization potential energy $Q_0\mu/4\pi\varepsilon r^2$, where μ is the effective electric dipole moment formed in the lattice in the inward particle motion. The potential may balance with the kinetic energy

$$Q_0\mu/4\pi\varepsilon r^2 = p_2/2M_0 = (rp)^2/2M_0 r^2 = \hbar^2 m^2/2M_0 r^2,$$

where quantization (10) is considered.

We must note that H in (5) expresses only the particle energy. The interaction energy ΔE caused by the exchange of oscillation energy $m\hbar\omega$ between the particle and lattice system is estimated using (6) and (10) as follows.

$$\Delta E = -\langle |Q_0\omega\, \mathbf{A}\cdot\mathbf{r}|^2 \rangle / m\hbar\omega = -m\hbar\omega/2 = -(\omega/2)S. \quad (11)$$

Now we consider a 2D system of N semi-localized particles. The ground state Ψ_N of the system is described using (1) and (5) as follows:

$$E_N\Psi_N = H_N\Psi_N, \quad (12)$$

$$H_N = \sum_{j-1}^{N} H(x_j, y_j) \quad (13)$$

$$\Psi_N = \prod_{j-1}^{N} \psi_0(m,\zeta_j) \quad (14)$$

We find the zero-point energy

$$E_{N0} = N\hbar\omega/2 \quad (15)$$

and interaction energy by (11)

$$\Delta E_N = -N m\hbar\omega/2. \quad (16)$$

In (13) $(x_j\ ,\ y_j)$ are the relative coordinate from the coordinate $(X_j\ ,\ Y_j)$ of the center of the molecular unit j. $(X_j\ ,\ Y_j)$ does not appear in the Hamiltonian because particle energy is independent of it in our assumption. In case of electron (or fermion) system, one may consider a Slater determinant in place of (14). In the following we show that the N-particle Laughlin state Ψ_{LN} can have higher stability than Ψ_N state employing H_N of (13).

It is known that the Laughlin function [5] describes well the 2D quantum state of fractional quantum Hall effect (FQHE) [5,6].

$$\Psi_{LN} = const\times \prod_{j>k} (\zeta_j - \zeta_k)^m \exp\left(-\sum_l |\zeta_l|^2/4\right). \quad (17a)$$

Since the phase of the wave function does not have direct physical meaning in FQHE [7], we study Ψ'_{LN} instead of Ψ_{LN}

$$\Psi'_{LN} = const\times \prod_{j>k} (\zeta_j - \zeta_k)^m \exp\left(-\sum_l |\zeta_l|^2/4\right) \quad (17b)$$

where Ψ'_{LN} is obtainable by a gauge transformation from Ψ_{LN}.

Using the replacement

$$M x_+ = \left(\sum_{j-1}^{N} x_j\right)/\sqrt{N}, \quad y_+ = \left(\sum_{j-1}^{N} y_j\right)/\sqrt{N}\ , \quad (18a)$$

$$p_{x,+} = \left(\sum_{j-1}^{N} p_{jx}\right)/\sqrt{N},\ p_{y,+} = \left(\sum_{j-1}^{N} p_{jy}\right)/\sqrt{N}\ , \quad (18b)$$

$$x_{jk} = (x_j - x_k)/\sqrt{N}\ ,\ y_{jk} = (y_j - y_k)/\sqrt{N}\ , \quad (18c)$$

$$p_{x,jk} = (p_{jx} - p_{kx})/\sqrt{N}\ ,\ p_{y,jk} = (p_{jy} - p_{ky})/\sqrt{N}\ , \quad (18d)$$

we can rewrite (17b) into a "diagonalized" form Ψ

$$\begin{aligned} \Psi'_{LN} &= const\times \exp\left(-\sum_l |\zeta_l|^2/4\right) \\ &\times \prod_{j>k} |\zeta|_{jk}^m \exp\left(-\sum_l |\zeta_l|^2/4\right) \end{aligned} \quad (19)$$

where is used the equality

$$\begin{aligned} \sum_{j-1}^{N} |\zeta_l|^2 &= \frac{1+(N-1)}{Nl^2}\sum_{j-1}^{N}(x_j^2 + y_j^2)^2 \\ &= \frac{\left(\sum_{j-1}^{N} x_j\right)^2 + \left(\sum_{j-1}^{N} y_j\right)^2}{Nl^2} + \\ &+ \sum_{j>k} \frac{(x_j - x_k)^2 + (y_j - y_k)^2)}{Nl^2} \\ &= |\zeta_+|^2 + \sum_{j>k} |\zeta_{jk}|^2 \ . \end{aligned} \quad (20)$$

Considering (1)~(9), we know that the function (19) is the zero-point solution of the Hamiltonian

$$H_{LN} = H_+ + \sum_{j>k} H_{jk} \quad (21)$$

$H_+ = H(x_+, y_+)$ $\quad (S_+=0)$

$H_{jk} = H(x_{jk}, y_{jk})$ $\quad (S_{jk} = \hbar m)$

with eigen-energy

$$E_{LN0} = [1+N(N-1)/2]\,\hbar\omega/2 \quad (22)$$

and interaction energy by (11)

$$\begin{aligned} \Delta E_{LN} &= -(\omega/2)(S_+ + \sum_{j>k} S_{jk} \\ &= -mN(N-1)\,\hbar\omega/4 \end{aligned} \quad (23)$$

Using (18) and referring to the equality (20) and other equalities

$$\begin{aligned} & x_+ p_{x,+} + y_+ p_{y,+} + \sum_{j>k} (x_{jk} p_{x,jk} + y_{jk} p_{y,jk}) \\ &= \sum_{j-1}^{N} (x_j p_{jx} + y_j p_{jy}) \end{aligned}$$

$$[m^2\hbar^2/(x^2+y^2)]\ \psi_0(m,\zeta) = -(p_x^2 + p_y^2)\ \psi_0(m,\zeta).$$

we find the operator correspondence between (13) and (21)

$$H_{LN} = H_N \quad (24)$$

with

$$\hat{S}_+ + \sum_{j>k} \hat{S}_{jk} = \sum_{j>k} \sum_{j-1}^{N} \hat{S}_j \quad (25)$$

Based on the correspondence (24) and (25), and considering the interaction energy, we compare the system energy between the states Ψ_N and Ψ_{LN} using (15), (16), (22) and (23)

$$\Delta E_t = (E_{LN0})+\Delta E_{LN}) - (E_{N0}+\Delta E_N)= \\ = - [(N^2 - 3N)(m\text{-}1)\text{-}2]\, \hbar\omega/4 \quad (26)$$

ΔE_t becomes negative when $m \geq 2$ and $N \geq 4$.

In the hitherto found new type of high temperature superconductors [1,2], the filling factor ν_s of carriers per 1 molecular unit may be $\nu_s \geq 1$. On the other hand the ground Laughlin state of the single electron (or fermion) system appears at the filling factor $\nu_s = 1/m = 1/3, 1/5, \ldots$. Therefore it may be difficult to find there the possibility of stability of Laughlin state. The situation is different in case of electron pair (or boson) system, where the ground Laughlin state appears at the pair filling factor $\nu_p = 1/2, 1/4, 1/6, \ldots$. Suppose an 2D electron system of $\nu_s = 1$. In pairing the system has pair filling factor $\nu_p = 1/2$ where $m=2$, and where $\Delta E_t < 0$ when $N \geq 4$.Of course we must consider the "Hubbard energy" increase $E_H \approx 10$eV per 1 pair. However the energy increase is overcome by the energy decrease given by (26) when the pair number N_p of the 2D system is large enough to satisfy

$$N_p \geq 3+4E_H/\hbar\omega = N_{pc}. \quad (27)$$

The pair oscillation frequency is supposed to be $\omega_p = 2\omega$. When we consider the general rule that the system with even number of electrons per 1 molecular unit becomes insulator, we must say that the Laughlin state can be realized in a 2D system with odd number of electrons (or holes) per 1 molecular unit if (27) is satisfied.

III. SUPERFLUIDITY

It is known that superfluidity is one of the properties of the system in the coherent state $|\Psi_\theta\rangle$ with definite phase Θ expressed by

$$|\Psi_\theta\rangle = \sum_{N=0}^{\infty} \sqrt{w_N}\, \exp(iN\Theta)\, |\Psi_N\rangle \quad (28)$$

where $|\Psi_N\rangle$ is the particle-number-definite macroscopic quantum state with N particles, and

$w_N = \langle N\rangle^N \exp(-\langle N\rangle)/N!$

where $\langle N\rangle$ is the mean particle number. Laughlin state Ψ'_{LN} is a particle-number-definite macroscopic quantum state $|\Psi_N\rangle$. Therefore a coherent state may be composed by the superposition of many Laughlin states with different N. The least uncertainty condition $\Delta N \Delta\Theta = \Delta p \Delta r/\hbar = 1/2$ is satisfied in the coherent state, realizing the minimization of kinetic and potential zero-point energy. Therefore we can expect the stable appearance of a superfluidity based on the coherent state in the situation where many Laughlin states of $\nu_p = 1/2$ with different pair number N_p appear as quantum fluctuation.

Such a fluctuating situation may be expected in the multilayer crystal of 2D circular molecular networks when the filling factor deviates from $\nu_p = 1/2$ by a small quantity $\Delta\nu$ by carrier doping. In case of mono-layer system, the deviation $\Delta\nu$ makes quasiparticle excitations without N fluctuation. In multilayer system, however, the existence of inter-layer particle exchange leads to the N fluctuation, the strength of which is determined by the inter-layer tunneling probability.

It is well known in the experiment of quantum Hall effect that the localization causes "plateau" where a Laughlin ground state is stabilized over a finite width of filling factor. Therefore to observe ideal superfluidity, one may completely remove the localization using "ideal" multilayer crystal and set the filling factor in the de viation $\nu_p = 1/2 \pm \Delta\nu$. If one increases localization keeping the filling factor $\nu_p = 1/2 \pm \Delta\nu$ starting from the ideal state, the macroscopic quantum state may smoothly change from superfluidity type $|\Psi_\theta\rangle$ to Laughlin type $|\Psi_N\rangle$, so long as the localization potential is not too strong to destroy even $|\Psi_N\rangle$.

Equating $l = r_0$, we find

$$\hbar\omega = \hbar^2/M_0 r_0^2 \quad (29)$$

where r_0 is the effective radius of a molecular unit. In order to consider the thermal effect, we use the thermal decoherence length

$$l_{th} = \eta\, \hbar v_F/k_B T = \eta(\hbar^2/k_B T M_0)(3\pi^2 n)^{1/3} \quad (30)$$

where η is a coefficient of $1 \geq \eta \geq 1/\pi$. Fermi velocity is supposed to be given by 3D free carrier model as $v_F = (\hbar/M_0)(3\pi^2 n)^{1/3}$ for 3D carrier density n. Quantum coherence may extend over the area l_{th}^2, where the pair number is

$$\Delta N_{th} = (1/2\pi_0 r_0^2) l_{th}^2 \quad (31)$$

Supposing the fluctuating appearance of the Laughlin states when ΔN_{th} exceed N_{pc} given by (27), we can determine the threshold temperature of the onset of superfluidity using (29)-(31)

$$T_c = (\eta\, \hbar v_F/2\sqrt{\pi}\ k_B r_0)/\sqrt{1.5 + 2E_H/\hbar\omega} \quad (32)$$

In fcc C_{60} with 3 holes in a molecular unit, where we suppose that 2 holes make filled band and one hole is in carrier state. Then we find $T_c \approx 70$-200 K from (32) with $v_F \approx 4\times10^5$m/s $r_0 \approx 0.5$nm and $\hbar\omega \approx 0.3$eV. The value is in the same order as $T_c \approx 52$-117 K reported in Ref. 1. With respect to the MgB_2, the field made by Mg^{2+} disturbs the formation of stable potential valley for π electrons in the branches of the 2D hexagon network. Concerning hole carriers on the network, however, the repulsive Mg^{2+} field makes stable potential valley just on the branch. A hole (a defect of σ electron) may appear in a hexagon neighboring the hexagon where new π electron creation is made through the lowering of π electron potential via attractive Mg^{2+} field. In order to reduce zero-point energy, the hole may take an outside larger orbit around the hexagon of the new π electron, feeling lattice potential valley and with some attraction from the newly created electron. Such hole state may be realized at auto-doping ratio $\beta = 1/13$ (1 hole per 13 hexagons). Concerning the 2D system of the doped holes, we find $v_F \approx 4.4\times10^5$m/s, $r_0 \approx 0.62$nm, $\hbar\omega/e \approx 0.2$eV and get $T_c \approx 50$-150 K which is in the same order to the observation 39 K.

IV. DISCUSSION AND CONCLUSION

We propose a new model of the high temperature superconductivity in crystal with 2D plane, which is composed of the planar connection of circular molecular orbits. Assuming the semi-localization of carriers in each molecular unit, we find the following. (i) boson (or carrier-pair) type Laughlin state with filling factor $\nu_p=1/2$ is the most stable state. (ii) By the superposition of many Laughlin states, a coherent state with superfluidity appears in "good" crystal when $\nu_p=1/2$. (iii) An estimation of the superfluidity threshold temperature gives $T_c \approx 70\text{-}200$ K for fcc C_{60}, and $T_c \approx 50\text{-}150$ K for MgB_2, which are respectively in the same order to the reported onset temperature of superconductivity 52-117 K and 39 K.

It must be noted that there exists some discrepancy between the assumption used in the model and the crystal property of the referred experiments. In case of fcc C_{60}, only the circular molecular orbits ($\|xy$) which are in parallel with the conduction plane is effective, and other orbits ($\|yz$ and $\|zx$) have no contribution to the model. If hole carrier is stabilized in $\|xy$ orbit, electron carrier must be stabilized in $\|yz$ and/or $\|zx$ orbits. It is interesting that $T_c \approx 52\text{-}117$ K is observed in 3 hole doping per 1 molecule, but lower T_c in 3 electron doping. Concerning MgB_2, we now have not enough information of its carrier state. Anyway it is difficult in the attractive field of Mg^{2+} to consider stable annular potential well for the graphite-like π electrons existing on the hexagon network branches. In order to apply our model, we must suppose the existence of localized holes at the self-doping ratio $\beta=1/13$. However the potential well of a localized hole has connection paths between the neighboring wells, which results in the incompleteness of "semi-localization" condition.

We must also note that the possibility of the paired superfluid state in FQHE has been theoretically proposed in the case of the filling factor ν_S with even denominator. Greiter et al. [8] discussed the *p*-wave pairing between the complex fermions in Pfaffian state via interaction mediated by vector potential at $\nu_S=1/2$, where the solution of the Hamiltonian of the system in BCS approximation leads to a gap equation having large pairing energy at relative angular momentum $L_z=h$. Ho pointed out [9] that the Pfaffian state and the Ψ_{331} state respectively correspond to A phase and A_1 phase in superfluid ^{3}He [10]. Ψ_{331} may be the ground state of 2 layer FQHE system with $\nu_S=1/4$ in each layer when the inter-layer distance $d_s \cong 1.5l$ [11]. Morf pointed out [12] that the ground state of coulomb-interacting $\nu_S=5/2$ system is a spin-polarized state with large overlapping with Pfaffian (or pairing) state.

These pairing states are interesting and deserve consideration with respect to the electronic state of the new trend high T_c superconductors in multi-layer crystal with interconnected networks of 2D circular molecular orbits. We suppose, however, that the effective filling factor of the superconductors is $\nu_s \cong 1$ which may exclude the possibility of above pairing states.

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