RELATIONSHIPS BETWEEN THE NONDISSIPATIVE DIAMAGNETIC CURRENTS IN THE MICROSCOPIC SIZED ATOMS AND MOLECULES AND THE SUPERCONDUCTIVITY IN THE MACROSCOPIC SIZED SOLIDS

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Abstract- We theoretically elucidate that the nondissipative diamagnetic currents in the microscopic sized atoms and molecules can be considered as superconductivity. We also suggest the guiding principle towards the syntheses of high temperature superconductivity. We can expect that any material with large $\Delta E_{\text{HOMO-LUMO,N}}$ value (more than a few eV) in which valence bands are completely occupied by electrons, which has been believed to be typical insulator in view of solid state physics and chemistry, has a possibility to exhibit high temperature superconductivity ($T_C \approx 10^4$~10^5 K) in solids.

Keywords- Bose–Einstein Condensation; Macro Superconductivity; Micro Nondissipative Diamagnetic Currents; Room Temperature Superconductivity.

I. INTRODUCTION

Historically, the conventional Bardeen–Cooper–Schrieffer (BCS) theory for superconductivity has been established by Bardeen, Cooper, and Schrieffer in 1957 on the basis of the phenomenological London theory established by London brothers in 1935, as follows. In 1935, London brothers explained the nondissipative diamagnetic currents in the closed-shell electronic structures with large energy gaps between the occupied and unoccupied orbitals in small materials such as He atoms and benzene molecules. They suggested that under the applied magnetic field, the atomic and molecular orbitals in He atoms and benzene molecules are very rigid, and thus if we assume that the canonical momentum $\boldsymbol{p}_{\text{canonical}}$ value continues to become zero even under the applied magnetic field, and the value increases according to the applied magnetic field, the nondissipative diamagnetic currents in small materials such as He atoms and benzene molecules can be explained. Furthermore, they suggested that superconductivity in the macroscopic sized solids can also be explained if we apply the London theory to the macroscopic sized superconductivity. The problem was to elucidate the mechanism how the stable electronic states with $\boldsymbol{p}_{\text{canonical}} = 0$ can be realized. On the basis of the London theory, Bardeen, Cooper, and Schrieffer explain the mechanism of the realization of the electronic structures with by considering that such stable electronic structures can be realized by electron pairing formed by two electrons with opposite momentum and spins as a consequence of the electron–phonon interactions. That is, the conventional BCS theory elucidating the mechanism of the occurrence of the macroscopic sized superconductivity has been established on the basis of the phenomenological London theory, which tries to explain the nondissipative diamagnetic currents in the microscopic sizes. On the other hand, the mechanism of the forming of the stable electronic structures with $\boldsymbol{p}_{\text{canonical}} = 0$ assumed in the phenomenological London theory in the microscopic sized atoms and molecules has not been elucidated. Even though the conventional BCS theory for the macroscopic sized superconducting materials has been established on the basis of the London theory for the nondissipative diamagnetic currents in the microscopic sized atoms and molecules, the nondissipative diamagnetic currents in the microscopic sized atoms and molecules have not been considered as superconductivity. Therefore, we try to elucidate that the nondissipative diamagnetic currents in the microscopic sized atoms and molecules can be considered as superconductivity in this article [1–3].

II. ENERGY LEVEL FOR THE ONE ELECTRON SYSTEMS

Without any applied magnetic or electric field (ground state), the energy ($E_{\text{Fermi},\boldsymbol{k}_{\text{HOCO}}\sigma}(0)$) for the fermionic states ($V_{\text{kin, Fermi},\boldsymbol{k}_{\text{HOCO}}\sigma}(0) \neq 0$) of an electron with spin σ occupying the HOCO can be expressed as $V_{\text{Coulomb,Fermi},\boldsymbol{k}_{\text{HOCO}}\sigma}(0) + V_{\text{kin,Fermi},\boldsymbol{k}_{\text{HOCO}}\sigma}(0)$, according to the conventional solid state physics, where the $V_{\text{Coulomb,Fermi},\boldsymbol{k}_{\text{HOCO}}\sigma}(0)$ value denotes the Coulomb energy between a fermionic electron

with spin □ occupying the HOCO and all another nuclei and electrons, and the $V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$ value denotes the kinetic energy for a fermionic electron with spin □ occupying the HOCO.

A. Energy for the One Electron Systems

According to Ref. [1], we can consider that an electron formed by the two electronic states with opposite momentum and spins can be considered to have bosonic properties at the ground state, and the Bose–Einstein condensation energy ($\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) from a fermionic electron to a bosonic electron with spin □ occupying the HOCO can be expressed as $V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$. The $V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$ values are usually very large (≈ 35 eV). That is, we have considered that an electron occupying an orbital in the ground state without any external fields, can become bosonic with zero total momentum ($\boldsymbol{p}_{\mathrm{canonical}}(0)=0$) and kinetic energies ($V_{\mathrm{kin,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)=0$) as a consequence of electronic state paring between two fermionic electronic states with opposite momentum and spins. Such stabilization energy of the bosonic states with respect to the two fermionic states (i.e., Bose–Einstein condensation energy) originates from the disappearance of the kinetic energy ($\boldsymbol{p}_{\mathrm{canonical}}(0)=0$ and $V_{\mathrm{kin,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)=0$), and can be estimated to be $V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$(≈ 35 eV) [1]. A boson has zero total momentum ($\boldsymbol{p}_{\mathrm{canonical}}(0)=0$) and kinetic energies ($V_{\mathrm{kin,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)=0$). Therefore, without any applied magnetic field (ground states; $\boldsymbol{p}_{\mathrm{canonical}}(0)=0$), the energy ($E_{\mathrm{Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) for the bosonic electronic states of an electron with spin □ occupying the HOCO can be expressed as $V_{\mathrm{Coulomb,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}\left(=V_{\mathrm{Coulomb,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}\right)$, where the $V_{\mathrm{Coulomb,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$ value denotes the Coulomb energy between a bosonic electron with spin □ occupying the HOCO and all another nuclei and electrons, and the $V_{\mathrm{kin,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)(=0)$ value denotes the kinetic energy for a bosonic electron with spin □ occupying the HOCO. As a consequence of the Bose–Einstein condensation, the kinetic energy ($V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) for a fermionic particle with $\boldsymbol{p}_{\mathrm{canonical}}(0)\neq 0$ has been converted to the potential energy ($V_{\mathrm{potential,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) for a bosonic particle with $\boldsymbol{p}_{\mathrm{canonical}}(0)=0$.

B. Energy for the Two Electrons Systems

Without any applied magnetic field or electric field (ground state; $\boldsymbol{p}_{\mathrm{canonical}}(0)=0$), the energy ($E_{\mathrm{Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma\boldsymbol{k}'_{\mathrm{HOCO}}\sigma'}(0)$) for the fermionic states ($V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma\boldsymbol{k}'_{\mathrm{HOCO}}\sigma'}(0)\neq 0$) of two electrons occupying the HOCO can be expressed as $2V_{\mathrm{Coulomb,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)+2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$, according to the conventional solid state physics.

Furthermore, without any applied magnetic or electric field (ground states; $\boldsymbol{p}_{\mathrm{canonical}}(0)=0$), the energy ($E_{\mathrm{Bose},\pm\boldsymbol{k}_{\mathrm{HOCO}}\sigma\mp\boldsymbol{k}_{\mathrm{HOCO}}\sigma'}(0)$) for the bosonic electronic states ($V_{\mathrm{kin,Bose},\pm\boldsymbol{k}_{\mathrm{HOCO}}\sigma\mp\boldsymbol{k}_{\mathrm{HOCO}}\sigma'}(0)=0$) of two electrons can be expressed as $2V_{\mathrm{Coulomb,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$.

Therefore, we can consider that a Cooper pair formed by two electrons with opposite momentum and spins can be considered to have bosonic properties at the ground state ($\boldsymbol{p}_{\mathrm{canonical}}(0)=0$), and the Bose–Einstein condensation energy ($2\Delta E_{\mathrm{BE}}(0)$) for two electrons can be expressed as $2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)(\approx 70\ \mathrm{eV})$. That is, we can consider that two electrons occupying an orbital in the ground state without any external fields, can become bosonic with zero total momentum ($\boldsymbol{p}_{\mathrm{canonical}}(0)=0$) and kinetic energies ($V_{\mathrm{kin,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)=0$) as a consequence of electron paring between two fermionic electrons with opposite momentum and spins. Such stabilization energy of the bosonic electron with respect to the two fermionic electrons (i.e., Bose–Einstein condensation energy) originates from the disappearance of the kinetic energy ($\boldsymbol{p}_{\mathrm{canonical}}(0)=0$ and $V_{\mathrm{kin,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)=0$), and can be estimated to be $2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)(\approx 70\ \mathrm{eV})$.

III. THE HOCO–LUCO GAP FOR THE FERMIONIC ELECTRONIC STATES IN THE CONVENTIONAL SOLID STATE PHYSICS AND FOR THE BOSONIC STATES IN OUR THEORY

Let us next look into the relationships between the conventional solid state physics and our theory. Let us look into the estimation of the HOCO–LUCO gaps for the fermionic electronic states in the conventional solid state physics and for the bosonic electronic states in our theory.

According to the conventional solid state physics, an electron is considered to be a Fermi particle, which is subject to the Pauli exclusion principle. That is, in the monocationic crystals, a fermionic electron is considered to partially occupy the HOCO. The energy for the fermionic electronic state can be expressed as

$V_{\mathrm{Coulomb,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)+V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$. On the other hand, according to our theory, an electron has bosonic properties ($\boldsymbol{p}_{\mathrm{canonical}}(0)=0$) and thus condensate into the Bose–Einstein condensation states ($V_{\mathrm{kin,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)=0$). Therefore, the energy for the bosonic electronic state can be defined as $V_{\mathrm{Coulomb,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$. The energy difference ($\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) between the energy level for the HOCO occupied by one fermionic electron ($\varepsilon_{\mathrm{Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) and that occupied by one bosonic electron ($\varepsilon_{\mathrm{Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) can be estimated as $V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$. Similar discussions can be made in the case of the LUCO.

Let us next compare the HOCO–LUCO gaps ($\Delta E_{\mathrm{gap,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma,\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$) in the conventional solid states physics with those in our theory. According to the conventional solid state physics, the $\Delta E_{\mathrm{gap,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma,\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$ value can be considered as energy difference between the energy for a fermionic electron occupying the HOCO ($\varepsilon_{\mathrm{Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) and that occupying the LUCO ($\varepsilon_{\mathrm{Fermi},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$), and can be estimated as $\left\{V_{\mathrm{Coulomb,Fermi},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)+V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)\right\}-\left\{V_{\mathrm{Coulomb,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)+V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)\right\}$. On the other hand, according to our theory, the HOCO–LUCO gap ($\Delta E_{\mathrm{gap,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma,\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$) can be considered as energy difference between the energy for a bosonic electron occupying the HOCO ($\varepsilon_{\mathrm{Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$) and that occupying the LUCO ($\varepsilon_{\mathrm{Bose},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$), and can be estimated as $\Delta E_{\mathrm{gap,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma,\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)-\left\{\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)-\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)\right\}$.

In general, it is very difficult to measure the HOCO–LUCO gaps from photoelectron experiment. The $\Delta E_{\mathrm{gap,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma,\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$ values deviate from the $\Delta E_{\mathrm{gap,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma,\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$ values by $\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)-\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$, even though these values are negligible compared with the $\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$ and $\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$ values themselves. This can be rationalized as follows. We can consider that the HOCO–LUCO gaps in vacuum state (without any external forces for observation) would be the $\Delta E_{\mathrm{gap,Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma,\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$ values. On the other hand, once we try to observe the electronic states by applying the external forces such as photoinduction, the bosonic states can be easily destroyed by a photon absorption and an emitted electron is observed as a fermion with energy $\varepsilon_{\mathrm{Fermi},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$. Similar discussions can be made in an electron occupying the LUCO. Furthermore, let us consider the observation of an electron promotion from the HOCO to the LUCO. The energy for a rigid bosonic electron pair completely occupying the HOCO in the vacuum states would be $\varepsilon_{\mathrm{Bose},\pm\boldsymbol{k}_{\mathrm{HOCO}}\sigma\mp\boldsymbol{k}_{\mathrm{HOCO}}\sigma'}(0)$.

On the other hand, once we try to observe the electronic states of a rigid bosonic electron pair completely occupying the HOCO, and one of two electrons absorbs a photon, the bosonic properties of these electrons are destroyed and these electrons are observed as two Fermi particles ($2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)\neq 0$). Therefore, the energy for an electron occupying the HOCO is observed as the $\varepsilon_{\mathrm{Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$ value (not $\varepsilon_{\mathrm{Bose},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$ value). An electron in the HOCO absorbing a photon is promoted to the LUCO. During observation, this electron is interfered with the external applied forces, and thus this electron which is observed to occupy the LUCO is still a Fermi particle. Therefore, the energy for this electron is observed as the $\varepsilon_{\mathrm{Fermi},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$ value (not $\varepsilon_{\mathrm{Bose},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$ value). Only after observation has been completed, an electron can be in vacuum states, and thus an electron occupying the LUCO becomes bosonic. Therefore, the energy for an electron occupying the LUCO in the vacuum states would be $\varepsilon_{\mathrm{Bose},\boldsymbol{k}_{\mathrm{LUCO}}\sigma}(0)$.

We can consider that the electronic states in the vacuum ground states without any external applied forces can be bosonic. On the other hand, we cannot observe these bosons because we must apply some external forces to these electronic states for observation. Once we try to observe them by applying some external forces, the bosonic properties of these electrons are destroyed and thus we cannot observe bosons. Therefore, we usually observe electrons as Fermi particles.

We can consider that even an electron, which has been considered as a Fermi particle in the conventional solid state physics, has bosonic properties, and is in the Bose–Einstein condensation state. That is, one electron as well as Cooper pairs formed by two electrons have bosonic properties, and are more stable than those considered in the conventional solid state physics by the Bose–Einstein condensation energies $\Delta E_{\mathrm{BE}}(0)$. On the other hand, in the conventional solid state physics, one electron has been considered as Fermi particles.

Therefore, the energy for all electronic states in our theory are smaller (more stable) than that in the conventional solid state physics by the Bose–Einstein condensation energies $\Delta E_{\mathrm{BE}}(0)$. However, since all electrons considered in our theory are smaller (more stable) than that in the conventional solid state physics, all physical parameters estimated in the conventional solid state physics are similar to those estimated in our theory. Furthermore, as we described above, we cannot usually observe a bosonic electron but can usually observe the fermionic electron. Therefore, we usually observe the physical parameters estimated on the basis of the conventional solid state physics.

IV. ENERGY GAP FORMING IN THE SUPERCONDUCTIVITY

Let us next look into the energy gap formed by electron–phonon interactions in superconductivity. According to the conventional BCS theory, we consider that two fermionic electrons are condensated into one bosonic Cooper pair as a consequence of electron–phonon interactions, and at the same time, the Bose–Einstein condensation occurs. This interpretation contradicts the equation of the BCS theory itself. Obeying this interpretation, the energy difference ($\Delta E_{\mathrm{gap,NM-SC}}(0)$) between them can be expressed as

$$\Delta E_{\mathrm{gap,e-ph}}(0)+\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma\boldsymbol{k}'_{\mathrm{HOCO}}\sigma'}(0).$$

In such a case, the $\Delta E_{\mathrm{gap,NM-SC}}(0)$ values are very large, and are not estimated to be the same with the $\Delta E_{\mathrm{gap,e-ph}}(0)$ values estimated on the base of the conventional BCS theory.

The $\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma\boldsymbol{k}'_{\mathrm{HOCO}}\sigma'}(0)$ values are usually very large (≈ 70 eV), on the other hand, the $\Delta E_{\mathrm{gap,NM-SC}}(0)$ and $\Delta E_{\mathrm{gap,e-ph}}(0)$ values are usually observed to be very small (≈ 10^{-2}~10^{-3} eV). This is because unstable two electrons before the electron–phonon interactions in the normal metallic states are considered to be fermionic while the stable two electrons after electron–phonon interactions are considered to be bosonic. If we consider that the stable two electrons after electron–phonon interactions as well as the unstable two electrons before the electron–phonon interactions are fermionic, the $\Delta E_{\mathrm{gap,NM-SC}}(0)$ values are estimated to be the same with the $\Delta E_{\mathrm{gap,e-ph}}(0)$ values. This is the equation appearing in the conventional BCS theory and actually observed physical parameters $\Delta E_{\mathrm{gap,NM-SC}}(0)=\Delta E_{\mathrm{gap,e-ph}}(0)$. That is, estimation and interpretation of the physical parameters for $\Delta E_{\mathrm{gap,NM-SC}}(0)$ and $\Delta E_{\mathrm{gap,e-ph}}(0)$ are somewhat ambiguous in the conventional BCS theory. Furthermore, the $\Delta E_{\mathrm{gap,NM-SC}}(0)$ and $\Delta E_{\mathrm{gap,e-ph}}(0)$ values have been called the "Bose–Einstein condensation energy" in the conventional BCS theory. It is also ambiguous definition because the $\Delta E_{\mathrm{gap,NM-SC}}(0)$ and $\Delta E_{\mathrm{gap,e-ph}}(0)$ values are not related to the conversion from the fermionic states to the bosonic states, but are related to the energy difference between the stable fermionic two electrons after electron–phonon interactions and the unstable fermionic two electrons before electron–phonon interactions, which are actually observed. We should consider that the Bose–Einstein condensation occurs after electron–phonon interactions are completed, and the Bose–Einstein condensation energy can be expressed as $2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)\approx 70$ eV in the BCS theory. We should reconsider the interaction of the BCS theory.

The $\Delta E_{\mathrm{gap,NM-SC}}(0)$ and $\Delta E_{\mathrm{gap,e-ph}}(0)$ values appearing in the BCS do not denote the Bose–Einstein condensation energy but denote the stabilization energy of the two fermionic electrons in the closed-shell electronic structure after electron–phonon interactions with respect to the two fermionic electrons in the opened-shell electronic structure before electron–phonon interactions. We should consider that after electron–phonon interactions are completed, the Bose–Einstein condensation can occur.

In such a case, the Bose–Einstein condensation energy $\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma\boldsymbol{k}'_{\mathrm{HOCO}}\sigma'}(0)$ denotes the stabilization energy of a bosonic Cooper pair in the closed-shell electronic structure after electron–phonon interactions with respect to the two fermionic electrons in the closed-shell electronic structure after electron–phonon interactions.

On the other hand, according to our theory, the already condensated two bosonic electrons are converted to the already condensated one bosonic Cooper pair as a consequence of the electron–phonon interactions ($\Delta E_{\mathrm{gap,e-ph}}(0)$). Our theory can well explain the equation in the BCS theory. According to our theory, two electrons before electron–phonon interactions in the normal metallic states are also bosonic, and we can estimate the $\Delta E_{\mathrm{gap,NM-SC}}(0)$ values as follows. We consider that the already condensated two bosonic electrons are converted to

the already condensated one bosonic Cooper pair as a consequence of the electron–phonon interactions ($\Delta E_{\mathrm{gap,e-ph}}(0)$). In this case, the $\Delta E_{\mathrm{gap,NM-SC}}(0)$ value becomes the same with the $\Delta E_{\mathrm{gap,e-ph}}(0)$ values. This is because two electrons in the normal metallic states as well as the superconducting states are considered to be bosonic, and the $\Delta E_{\mathrm{BE},\boldsymbol{k}_{\mathrm{HOCO}}\sigma\boldsymbol{k}'_{\mathrm{HOCO}}\sigma'}(0)$ values for the unstable two bosonic electrons before electron–phonon interactions are the same with that for the stable bosonic Cooper pair after electron–phonon interactions. However, as we described above, we cannot usually observe bosonic electron but can usually observe fermionic electron, in particular, when the orbitals occupied by electrons are changed. Therefore, we usually observe the physical parameters estimated on the basis of the Fermi particles in the conventional solid state physics. Let us next look into the observation of the electron–phonon interaction processes. The process from the destruction of the already condensated two bosonic electrons (formation of the two fermionic electrons) in the opened-shell electronic structure to the formation of the two fermionic electrons) in the closed-shell electronic structure as a consequence of the electron–phonon interactions is considered in the BCS theory, and is usually observed. When we try to observe the electron–phonon interaction processes, we cannot usually observe the bosonic metallic states before electron–phonon interactions and the bosonic superconducting states after electron–phonon interactions.

V. RELATIONSHIPS BETWEEN THE FERMIONIC AND BOSONIC PROPERTIES IN TWO ELECTRONS SYSTEMS

Let us look into the two electrons with opposite momentum and spins occupying the same orbitals in the closed-shell electronic structures with large energy gaps between the occupied and unoccupied orbitals in various small sized materials such as the neutral He atom and benzene. There are two possible electronic properties, i.e., (i) two fermionic intrinsic insulating particles states (the most probable from the point of view of the conventional solid state physics), and (ii) one bosonic superconducting particle states (Cooper pair) (predicted from our theory). According to the conventional solid state physics, the two fermionic intrinsic insulating particles states are predicted, on the other hand, we usually observe the nondissipative diamagnetic current states (one bosonic superconducting particle states). This is the problem we must solve in this article.

A. Two Fermionic Intrinsic Insulating Particles States
Let us first look into the two fermionic intrinsic insulating particles states, which obey the Pauli exclusion principle. In the electronic states, there are independent two fermionic electrons with opposite momentum and spins occupying the same orbitals in the closed-shell electronic structures with large energy gaps between the occupied and unoccupied orbitals. Furthermore, these electronic states are not changed even under the applied magnetic field H_{applied}, and the magnetic fields are not expelled by these two electrons. This electronic state can be predicted from the point of view of the conventional solid state physics since it is reasonable to consider that the total momentum of two fermionic electrons in the closed-shell electronic structure with large energy gaps between the occupied and unoccupied orbitals cannot be easily changed by the applied magnetic field H_{applied}. However, we usually observe the nondissipative diamagnetic current states (not intrinsic insulating states).

B. One Bosonic Superconducting Particle States
Let us next look into the one bosonic superconducting particles states (Cooper pairs). In the electronic states, there is a bound particle formed by two electrons with opposite momentum and spins occupying the same orbitals in the closed-shell electronic structures with finite energy gaps between the occupied and unoccupied orbitals. These electronic states are changed under the applied magnetic field H_{applied}, and the magnetic fields are expelled by these two electrons. On the other hand, this electronic state cannot be predicted from the point of view of the conventional solid state physics since it is reasonable to consider that the total momentum states of two electrons in the closed-shell electronic structure with large energy gaps between the occupied and unoccupied orbitals cannot be easily changed by the applied magnetic field H_{applied}. However, we usually observe the nondissipative diamagnetic current states (not intrinsic insulating states).

VI. THE MECHANISM OF THE BOSE-EINSTEIN CONDENSATION IN TWO ELECTRONS SYSTEMS

A. Two Fermionic Intrinsic Insulating Particles States
Let us next discuss the reason why two electrons with opposite momentum and spins occupying the same orbital in the closed-shell electronic structures with large energy gaps between the occupied and unoccupied orbitals can be in the Bose–Einstein condensation. We first consider that two electrons with opposite momentum and spins occupying the same orbital in the closed-shell electronic structures with large energy gap between the occupied and unoccupied orbitals move independently as two

Fermi particles. When very small magnetic field (H_{applied}) is applied, the energy ($E_{\text{Fermi, IN, }\boldsymbol{k}_{\text{HOCO}}\sigma\boldsymbol{k}'_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$) for the two fermionic electron systems can be expressed as $2V_{\text{Coulomb, Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}\left(H_{\text{applied}}\right)$ $+ 2V_{\text{kin,Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}\left(H_{\text{applied}}\right)$ $+ \mu_0 H^2_{\text{applied}} v_{\text{applied}} / 2$, where the $\varepsilon_{\text{Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma\boldsymbol{k}'_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ value denotes the energy level for two electrons, and the $V_{\text{Coulomb,Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}\left(H_{\text{applied}}\right)$ and $V_{\text{kin, Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}\left(H_{\text{applied}}\right)$ values denote the Coulomb energy and the kinetic energy for an electron occupying the HOCO, respectively, and furthermore, the μ_0 denotes the magnetic permeability in vacuum, and the v_{applied} denotes the volume of the space where the magnetic field H_{applied} is applied. In the two fermionic systems, two electrons are not equivalent, and these two electrons independently behave as two Fermi particles, and thus we must consider the $V_{\text{kin, Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}\left(H_{\text{applied}}\right)(\neq 0)$ values as well as the $V_{\text{Coulomb,Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}\left(H_{\text{applied}}\right)$ values.

B. Superconducting States as a Consequence of the Bose–Einstein Condensation

In a similar way, the energy ($E_{\text{Bose,SC,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$) for the one bosonic electron systems under the applied magnetic field H_{applied} can be expressed as $V_{\text{Coulomb,Bose,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ $+ V_{\text{kin, Bose,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ $= 2V_{\text{Coulomb, Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}(0)$ $+ V_{\text{emf,Bose,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ $+\mu_0 \left\{H_{\text{applied}} v_{\text{applied}} / \left(v_{\text{applied}} - v_{\text{SC}}\right)\right\}^2$ $\left(v_{\text{applied}} - v_{\text{SC}}\right) / 2$, where the $V_{\text{Coulomb,Bose,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ and $V_{\text{kin, Bose,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ values denote the Coulomb and kinetic energies for two electrons occupying the HOCO, respectively, and furthermore, v_{SC} denotes volume of the superconductor. The $V_{\text{emf,Bose,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ value denotes the kinetic energy of an electron pair as a consequence of the electromotive forces.

C. Energy Difference between the Two Fermionic Insulating States and the One Bosonic Superconducting States

Let us next compare the $E_{\text{Fermi, IN, }\boldsymbol{k}_{\text{HOCO}}\sigma\boldsymbol{k}'_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ values with the $E_{\text{Bose,SC,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ values. The energy difference ($\Delta E_{\text{SC}-\text{IN}}\left(H_{\text{applied}}\right)$) between the two fermionic insulating particle states ($E_{\text{Fermi, IN, }\boldsymbol{k}_{\text{HOCO}}\sigma\boldsymbol{k}'_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$) and the one bosonic superconducting states ($E_{\text{Bose,SC,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$) under the applied magnetic field H_{applied} can be expressed as $\mu_0 H^2_{\text{applied}} v_{\text{SC}} / 2 - 2V_{\text{kin, Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}\left(H_{\text{applied}}\right) << 0$

The $2V_{\text{kin,Fermi,}\boldsymbol{k}_{\text{HOCO}}\sigma}\left(H_{\text{applied}}\right)$ values are usually very large (≈ 70 eV), and thus are much larger than the $\mu_0 H^2_{\text{applied}} v_{\text{SC}} / 2$ values (usually in the order of 10^{-2}~10^{-3} eV).

Therefore, it is clear from the calculated results that the $E_{\text{Bose,SC,}\pm\boldsymbol{k}_{\text{HOCO}}\sigma\mp\boldsymbol{k}_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ values are much smaller than the $E_{\text{Fermi, IN, }\boldsymbol{k}_{\text{HOCO}}\sigma\boldsymbol{k}'_{\text{HOCO}}\sigma'}\left(H_{\text{applied}}\right)$ values, and thus the one bosonic superconducting particle states are much more stable than the two fermionic insulating states. It can be also understood from the fact that the superconducting states are stable and thus the stable one bosonic superconducting states are not converted into the unstable two fermionic intrinsic insulating states via photon emission (Joule's heats) even under the constant applied magnetic field H_{applied}.

Therefore, the nondissipative diamagnetic currents in the microscopic sized molecules as well as the macroscopic sized superconductivity can be explained by one bosonic superconducting states (Cooper pairs).

It should be noted that since there are equivalent two electrons with opposite momentum and spins, the total momentum of a Cooper pair can be also zero ($\boldsymbol{k}_{\text{canonical, HOCO}} = \left(+\boldsymbol{k}_{\text{HOCO}}\uparrow\right) + \left(-\boldsymbol{k}_{\text{HOCO}}\downarrow\right) = 0$; $\boldsymbol{p}_{\text{canonical, HOCO}} = 0$), and thus the $V_{\text{kin, Cooper pair, }N}\left(+\boldsymbol{k}_{\text{HOMO}}\uparrow, -\boldsymbol{k}_{\text{HOMO}}\downarrow\right)$ value can be zero. That is, the kinetic energy, which generally reduces the destruction energy of the bosonic Cooper pairs, and is compensated by the Coulomb energy ($V_{\text{Coulomb,}N}\left(\boldsymbol{k}_{\text{HOMO}}\sigma, \boldsymbol{k}_{\text{HOMO}}\sigma'\right)$), does not play a role in the decision of the

destruction energy of the bosonic Cooper pairs. This is because the kinetic energy for fermionic state is converted to the potential energy for bosonic state. The Cooper pair can be formed by the Coulomb interactions between two electrons with opposite momentum and spins occupying the same orbitals via the positive charges of the nuclei. This is the reason why the $E_{\mathrm{Bose,one,SC},N}\left(H_{\mathrm{applied}}\right)$ values are much smaller than the $E_{\mathrm{Fermi,two,IN},N}\left(H_{\mathrm{applied}}\right)$ values, and the reason why the bosonic particle is much more stable than two fermionic particles ($\Delta E_{\mathrm{SC-IN,\,stabilized},N}\left(H_{\mathrm{applied}}\right) >> 0$). Furthermore, this is the reason why the Bose–Einstein condensation occurs in the closed-shell electronic structures with finite energy gaps between the occupied and unoccupied orbitals, and thus the reason why we usually observe the nondissipative diamagnetic currents in the microscopic sized atoms and molecules as well as the macroscopic sized superconductivity.

These calculated results can be also understood from the fact that we usually observe the supercurrent states, rather than the intrinsic insulating states. Furthermore, we can also say that the intrinsic insulators formed by two fermionic particle states in the closed-shell electronic structures with finite energy gaps between the occupied and unoccupied orbitals, which has been believed to exist from the point of view of solid state physics, do not usually exist.

We can conclude that the kinetic energy $2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}(0)$ is converted to the internal potential energy, as a consequence of the Bose–Einstein condensation, by which the bosonic states with zero kinetic energy become much more stable than the fermionic states with large kinetic energy. The bosonic states are the resonance states (wave characteristics) formed composed from the various fermionic states (particle characteristics).

VII. RELATIONSHIPS BETWEEN THE APPLIED MAGNETIC FIELD AND ELECTRONIC STATES

A. Comparison of the One Bosonic Superconducting Particle States with the Two Fermionic Intrinsic Insulating Particles States

Let us next look into the relationships between the H_{applied} values and the electronic properties in order to investigate the possibility that we can observe two fermionic intrinsic insulating particles states. When the H_{applied} value increases, and at $H_{\mathrm{applied}} = H_{\mathrm{c,BE}}$, the one bosonic superconducting particle states would be converted to the two fermionic intrinsic insulating particles states, theoretically, $\mu_0 H_{\mathrm{c,BE}}^2 v_{\mathrm{SC}} / 2 - 2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}\left(H_{\mathrm{c,BE}}\right) = 0$. Therefore, the $H_{\mathrm{c,BE}}$ values can be estimated as $H_{\mathrm{c,BE}} = 2\sqrt{V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}\left(H_{\mathrm{c,BE}}\right)/\left(\mu_0 v_{\mathrm{SC}}\right)}$. The $H_{\mathrm{c,BE}}$ values are very large because of the large $2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}\left(H_{\mathrm{c,BE}}\right)$ values of approximately 70 eV, and thus the conversion from the one bosonic superconducting particle states to the two fermionic intrinsic insulating particles states would not be usually realistic.

B. Destruction of the One-Bosonic Superconducting Particle States

When the H_{applied} value increases, the one-bosonic superconducting particle states would be destroyed by the critical field $\min\left(H_{\mathrm{c,singlet}}, H_{\mathrm{c,BE}}\right)$ as follows. Usually, the $\Delta E_{\mathrm{gap,\,e-ph}}\left(H_{\mathrm{applied}}\right)$ ($\approx 10^{-2}$~10^{-3} eV) and $\Delta E_{\mathrm{gap,\,HOMO-LUMO}}\left(H_{\mathrm{applied}}\right)$ values (< 10 eV) are much smaller than the $2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}\left(H_{\mathrm{c,BE}}\right)$ values ($\approx$ 70 eV), and thus the transition from the one-bosonic superconducting particle states to the two one-bosonic normal metallic particles states occurs before that from the one-bosonic superconducting particle states to the two-fermionic insulating particles states occurs.

That is, the $H_{\mathrm{c,singlet}}$ value is much smaller than the $H_{\mathrm{c,BE}}$ value. Therefore, the transition from the superconducting states to the normal metallic states can be usually observed at the critical magnetic field $H_{\mathrm{c,singlet}}$.

This is the reason why we usually observe the transition from the superconducting states to the normal metallic state and not to the intrinsic insulating states.

On the other hand, if the $\Delta E_{\mathrm{gap,\,HOMO-LUMO}}\left(H_{\mathrm{applied}}\right)$ values are larger than the $2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}\left(H_{\mathrm{applied}}\right)$ values, we can observe the transition from one-bosonic superconducting particle states to the two-fermionic intrinsic insulating particles states at the critical magnetic field $H_{\mathrm{c,BE}}$, theoretically. However, even in the neutral He atoms, the $\Delta E_{\mathrm{gap,\,HOMO-LUMO}}\left(H_{\mathrm{applied}}\right)$ values of 48.2 eV is

much smaller than the $2V_{\mathrm{kin,Fermi},\boldsymbol{k}_{\mathrm{HOCO}}\sigma}\left(H_{\mathrm{applied}}\right)$ values of 77.7 eV. Therefore, it would be very difficult to observe the two-fermionic intrinsic insulating particles states predicted from the band theory in the textbooks in the conventional solid state physics.

VIII. GUIDING PRINCIPLE TOWARDS HIGH-TEMPERATURE SUPERCONDUCTIVITY

A. The Role of Waters and Hydrogen Plasma in the Occurrence of Superconductivity

It was reported that room temperature superconducting behavior is observed in the graphite powder treated by water or exposed to the hydrogen plasma. This means that we can consider that hydrogen atoms and water molecules play an essential role in the occurrence of superconducting behavior in these carbon materials.

Let us discuss how hydrogen atoms and water molecules play an important role in the electronic structures in these carbon materials. We define the $p_{\mathrm{saturated}}$ value denoting the ratio of the number of carbon atoms with saturated sp^3 hybrid orbitals to that of the total carbon atoms located in the area directly related to the superconductivity.

In untreated virgin graphite powder ($p_{\mathrm{saturated}} = 0$), there are some impurities. Therefore, there are some holes in valence bands in untreated virgin graphite powder. Furthermore, in untreated virgin graphite powder ($p_{\mathrm{saturated}} = 0$), there are many □-conjugated carbons originating from sp^2 hybrid orbitals. Therefore, the $\Delta E_{\mathrm{HOMO-LUMO},N}$ values for materials with N carbon atoms are very small (≈ 0.0 eV). In such a case, the electronic states can be in the normal metallic states. This is the reason why the superconductivity has not been observed in the untreated virgin graphite powder ($p_{\mathrm{saturated}} = 0$).

On the other hand, in the graphite powder treated by water or exposed to hydrogen plasma, first, hydrogen atoms and part of water molecules (for example, hydrogen atoms and hydroxyl group) can be completely terminated with carbon framework ($p_{\mathrm{saturated}} = 0$). Therefore, the valence bands are completely occupied by electrons. If completely terminated □-conjugated graphite compounds ($p_{\mathrm{saturated}} = 0$) are formed, the valence bands are completely occupied by electrons, on the other hand, the $\Delta E_{\mathrm{HOMO-LUMO},N}$ values are still very small (≈ 0.0 eV) with a grain size of several tens of micrometers. If many hydrogen atoms and part of water molecules are absorbed in the virgin graphite powder, there is a possibility that the alkane parts originating from the sp^3 hybrid orbitals as well as the alkene parts originating from the sp^2 hybrid orbitals can be formed. The $p_{\mathrm{saturated}}$ value increases with an increase in the number of these additional materials combined with carbon framework. The $\Delta E_{\mathrm{HOMO-LUMO},N}$ value increases with an increase in the $p_{\mathrm{saturated}}$ value. Therefore, the $\Delta E_{\mathrm{HOMO-LUMO},N}$ value increases with an increase in the number of these additional materials combined with carbon framework. In the virgin graphite powder exposed to the hydrogen plasma, the only hydrogen atoms are surely combined with the carbon framework in the graphite powder. On the other hand, in the water-treated graphite powder, whether water molecules, hydrogen atoms, hydroxyl group, or oxygen atoms can be combined with carbon framework are not clarified. However, we can at least say that the $p_{\mathrm{saturated}}$ value increases with an increase in the number of these additional materials (whatever they are) combined with carbon framework.

In summary, absorption of many additional materials with carbon framework by water treatment and expose to hydrogen plasma in the virgin graphite powder plays an important role in the complete occupation of electrons in the valence bands, and furthermore, plays an important role in the enlargement of the $\Delta E_{\mathrm{HOMO-LUMO},N}$ values as a consequence of the increase of the sp^3-hybrid orbitals.

B. Superconducting Critical Temperatures as a Function of the Valence–Conduction Band Gaps

Let us next discuss the critical temperatures $T_{\mathrm{c,intra},N}$ at which the intragranular diamagnetic supercurrents can disappear in □-conjugated sp^2-type graphene hydrocarbons and sp^3-type alkane hydrocarbons with N carbon atoms, on the basis of recent experimental and our previous theoretical researches.

According to our previous researches, in the materials with large $\Delta E_{\mathrm{HOMO-LUMO},N}$ values, the closed-shell electronic structures with large $\Delta E_{\mathrm{HOMO-LUMO},N}$ values are formed by the quantization of orbitals by nature, and the attractive interactions between two electrons with opposite momentum and spins occupying the same orbitals can be realized by the Coulomb interactions between all nuclei and electrons. That is, in the materials with large $\Delta E_{\mathrm{HOMO-LUMO},N}$ values, the Cooper pairs are formed by the large $\Delta E_{\mathrm{HOMO-LUMO},N}$ values

as a consequence of the quantization of the orbitals by nature, and by the attractive Coulomb interactions between two electrons with opposite momentum and spins occupying the same orbitals via the positively charged nuclei. The energy levels of the HOMO ($\varepsilon_{\mathrm{HOMO},N}$) in the neutral 6an, 10ac, and 14ac are about –10 eV, and the attractive Coulomb interactions between two electrons occupying the HOMO ($V_{\mathrm{Coulomb,HOMO},N}$) in the neutral 6an, 10ac, and 14ac were calculated to be about –70 eV in our previous researches.

On the other hand, according to the recent experimental research, the Cooper pairs have been observed at room temperatures in the neutral 6an, 10ac, 14ac, and coronene. The energies of the double-photoionization in the neutral 6an, 10ac, and 14ac observed from the photoemission experiments are about 20~25 eV, which are similar to about the $-2\varepsilon_{\mathrm{HOMO},N}(\approx 20\ \mathrm{eV})$ values, and the destruction energies of the Cooper pairs in the neutral 6an, 10ac, and 14ac observed from the photoemission experiments are about 70 eV, which are similar to the $-V_{\mathrm{Coulomb,HOMO},N}(\approx 70\ \mathrm{eV})$ values.

The energies for photoionization (approximately half of the double photoionization ($-\varepsilon_{\mathrm{HOMO},N}\left(\sim \Delta E_{\mathrm{HOMO-LUMO},N}(\approx 10\ \mathrm{eV})\right)$)) are related to the $T_{\mathrm{c,intra},N} \sim \Delta E_{\mathrm{HOMO-LUMO},N}$ $\left(\approx 10\ \mathrm{eV} \approx 10^4 \sim 10^5\ \mathrm{K}\right)$ values, and the destruction energies of the Cooper pairs (70 eV) are closely related to the $-V_{\mathrm{Coulomb,HOMO},N}(\approx 70\ \mathrm{eV})$ values. That is, our prediction in our theoretical researches can be well confirmed by the recent experimental research, and our previous theory can be reasonably applied to the explanation of the mechanism of the occurrence of the granular high temperature superconductivity in carbon materials under consideration in this study.

On the other hand, the second-order perturbation effect as a consequence of one electron promotion from the HOMO to the LUMO becomes more important with a decrease in the $\Delta E_{\mathrm{HOMO-LUMO},N}$ value. In such a case, the conventional BCS theory should be applied, and the electron–phonon interactions play an essential role in the forming of energy gap and the decision of the $T_{\mathrm{c,intra},N}$ value (10^0~10^2 K).

The critical temperature $T_{\mathrm{c,intra},N}$ at which the intragranular diamagnetic supercurrents in carbon powder under consideration can disappear can be estimated to be larger than $T_{\mathrm{c,inter,upper}} \approx 10^4$ K. The critical temperature $T_{\mathrm{c,intra},N}$ values for hydrocarbon with N carbon atoms can be defined as, $T_{\mathrm{c,intra},N} = T_{\mathrm{c,intra,HOMO-LUMO},N} \propto \Delta E_{\mathrm{HOMO-LUMO},N}$ for large $\Delta E_{\mathrm{HOMO-LUMO},N}$, and $T_{\mathrm{c,intra},N} = T_{\mathrm{c,intra,BCS},N} \propto e^{-1/\lambda_N}$ for small $\Delta E_{\mathrm{HOMO-LUMO},N}$. The $T_{\mathrm{c,intra,HOMO-LUMO},N}$ values (10^4~10^5 K) for the materials with large $\Delta E_{\mathrm{HOMO-LUMO},N}$ values are estimated to be much larger than the $T_{\mathrm{c,intra,BCS},N}$ values (10^0~10^2 K) for the materials with small $\Delta E_{\mathrm{HOMO-LUMO},N}$ values.

C. sp^2-Type Alkene Hydrocarbons

Let us discuss the $T_{\mathrm{c,intra},N}$ values for sp^2-type alkene hydrocarbons. In small sized molecules, the large energy gaps between the HOMO and LUMO (10^4~10^5 K) are formed by nature, by which the electron pairs formed by attractive Coulomb interactions between two electrons occupying the same orbital with opposite momentum and spins via the positive charges of nuclei become stabilized. On the other hand, the $\Delta E_{\mathrm{HOMO-LUMO},N}$ value decreases with an increase in molecular size. Actually, the $\Delta E_{\mathrm{HOMO-LUMO},N}$ values for 10ac, 14ac, 18ac, pentacene (22ac), and hexacene (26ac) are estimated to be 7.97, 6.93, 5.89, 5.35, and 2.98 eV, respectively, those for nanosized graphene are estimated to be about 0.35 eV, and furthermore, those for macroscopic sized graphite become nearly 0.0 eV. Therefore, the second-order perturbation effect as a consequence of one electron promotion from the HOMO to the LUMO becomes more important with an increase in the molecular size. In such a case, the conventional BCS theory should be applied, and the electron–phonon interactions play an essential role in the forming of energy gap (10^0~10^2 K).

On the other hand, the $\Delta E_{\mathrm{HOMO-LUMO},N}$ value should be larger than the experimental $T_{\mathrm{c,inter,upper}}$ values of 10^4 K (a few eV). Therefore, according to our calculations and experimental results, the material sizes for the □-conjugated sp^2-type graphene hydrocarbons must be smaller than the nano-size in order that the $T_{\mathrm{c,intra},N}$ values for these materials are larger than the experimental values of $T_{\mathrm{c,inter,upper}} \approx 10^4$ K. On the other hand, in such a case, very large number of Josephson links ($\approx N_{\mathrm{CH}}$) is needed in each grain.

D. sp^3-Type Alkane Hydrocarbons

Let us next discuss the $T_{c,intra,N}$ values for sp^3-type alkane hydrocarbons. The $\Delta E_{HOMO-LUMO,N}$ values are estimated to be 12.11, 11.47, 10.97, 11.00, 10.62, and 9.28 eV (140530, 133103, 127301, 127649, 123239, and 107589 K) for 2alka, 3alka, 4alka, 5alka, 5dia, and 14dia, respectively. Furthermore, the $\Delta E_{HOMO-LUMO,N}$ value for diamond with even macroscopic size is very large, 5.47 eV (63476 K). Therefore, the $\Delta E_{HOMO-LUMO,N}$ value does not significantly decrease with an increase in material size in sp^3-type alkane hydrocarbons and diamonds.

The $\Delta E_{HOMO-LUMO,N}$ values are very large (5.47 eV) and the energy levels of the occupied orbitals are always negative ($\varepsilon_{orbital,N,j} < 0$) in diamonds, and thus the Coulomb energy between two electrons occupying the same orbitals via the positive charges of the nuclei can become large negative. Therefore, by analogy with the supercurrents and discovery of forming of the Cooper pairs in the microscopic sized molecules such as 6an, 10ac, 14ac, and coronene with large $\Delta E_{HOMO-LUMO,N}$ values, if we consider diamond as a macroscopic sized molecule with large $\Delta E_{HOMO-LUMO,N}$ values, we can expect that the superconductivity can be induced in the macroscopic sized diamonds, the valence bands of which are completely occupied by electrons, at 300 K, as described in the previous researches. Therefore, the $T_{c,intra,N}$ values for any size of sp^3-type alkane hydrocarbons can be larger than $T_{c,inter,upper} \approx 10^4$ K.

E. Guiding Principle towards Higher Relative Superconducting Masses

Let us consider the case where there are N_{grain} grains, and there are N_{CH} molecules in each grain. We can consider that there are about N_{CH} Josephson links between two neighboring molecules in each grain, about N_{grain} Josephson links between two neighboring grains, and totally about $N_{CH}N_{grain}$ Josephson links in solids under consideration. As described above, if each grain is formed by only □-conjugated graphenes, we must consider that there are many small (smaller than nano-sized molecules) sized □-conjugated molecules with large $\Delta E_{HOMO-LUMO,N}$ values (larger than a few eV, at the smallest) in each grain. In such a case, very large numbers of nanosized □-conjugated hydrocarbons and Josephson links are needed in each grain with the size of several tens of micrometers, and thus the N_{CH} values should be very large. Such large number of Josephson junctions ($\approx N_{CH}$) would cause the small relative superconducting masses.

On the other hand, it is also possible that there is only one sp^3 diamond-type alkane hydrocarbon ($N_{CH} = 1$) with large $\Delta E_{HOMO-LUMO,N}$ value of 10^4~10^5 K in each grain with the sizes of the order of several tens of micrometers. Furthermore, the $p_{saturated}$ value increases with an increase in the number of additional materials combined with carbon framework. The $\Delta E_{HOMO-LUMO,N}$ value for each molecule increases with an increase in the $p_{saturated}$ value. The needed number of Josephson links N_{CH} in the hydrocarbons in each grain decreases with an increase in the $p_{saturated}$ value as a consequence of the increase in the number of additional materials combined with carbon framework. If the $p_{saturated}$ value becomes large enough ($p_{saturated} \geq 0.35$), only one hydrocarbon with sp^2- and sp^3-type hybrid characteristics with $\Delta E_{HOMO-LUMO,N}$ values in the order of 10^4~10^5 K is needed in each grain ($N_{CH} = 1$).

Even though we cannot clearly show which kind of structure or stoichiometry the superconducting grain part has, it is natural to conclude that the hydrocarbons with significant sp^3 characteristics rather than the sp^2 characteristics, the $\Delta E_{HOMO-LUMO,N}$ values of which are larger than a few eV, and the valence bands of which are completely occupied by electrons, are formed by water treatment or expose to the hydrogen plasma in the virgin graphite powder.

Let us next consider how to reduce the number of intergranular Josephson links N_{grain} between two neighboring grains in solids. If the $p_{saturated}$ value becomes large enough, only one hydrocarbon with $\Delta E_{HOMO-LUMO,N}$ values in the order of 10^4~10^5 K is needed in the macroscopic sized solids. For example, the virgin diamond treated by water or exposed to the hydrogen plasma ($N_{CH} = 1$ and $N_{grain} = 1$) in a macroscopic sized grain, in which the valence bands are completely occupied by electrons, has a possibility to exhibit high-temperature superconductivity.

In general, regardless of the kinds of additional materials combined with carbon framework, the macroscopic sized materials with larger $\Delta E_{HOMO-LUMO,N}$ values than a few eV, in which the valence bands are completely occupied by

electrons (on the other hand, conduction bands can be also partially occupied by electrons), have a possibility to exhibit room temperature superconductivity ($N_{\mathrm{CH}} N_{\mathrm{grain}} = 1$).

F. Guiding Principle towards High Temperature Superconductivity

Let us next summarize the guiding principle towards the syntheses of high temperature superconductivity. We can expect that any material with large $\Delta E_{\mathrm{HOMO-LUMO},N}$ value (more than a few eV) in which valence bands are completely occupied by electrons, which has been believed to be typical insulator in view of solid state physics and chemistry, has a possibility to exhibit high temperature superconductivity ($T_C \approx 10^4$~10^5 K) in solids. In these cases, it is also possible that the conduction bands are somewhat occupied by electrons even though electrons occupying not the conduction bands but the valence bands play an essential role in supercurrent. If some appropriate additional materials are combined enough with these materials so that the valence bands are completely occupied by electrons, these materials can exhibit high temperature superconductivity in solids. Furthermore, in such cases, finite magnetic field for intergranular tunneling effects is not needed ($h_{\mathrm{c,inter}}(0) = 0$). Discovery of appropriate combination of additional materials with the macroscopic sized main materials, by which the $\Delta E_{\mathrm{HOMO-LUMO},N}$ value becomes larger than a few eV, and by which the valence bands are completely occupied by electrons, is awaited.

IX. CONCLUDING REMARKS

We theoretically elucidate that the nondissipative diamagnetic currents in the microscopic sized atoms and molecules can be considered as superconductivity. We also suggest the guiding principle towards the syntheses of high temperature superconductivity.

We investigated the reasonable mechanism of the occurrence of granular high temperature superconductivity in the graphite powder treated by water or exposed to the hydrogen plasma, discovered by Esquinazi et al., on the basis of our previous theoretical works described above, which can be well confirmed by the recent experimental work. We also suggested the guiding principle towards larger relative superconducting mass in these materials. Furthermore, we also suggested the general guiding principle towards high temperature superconductivity.

We first try to elucidate the essential role of water treatments and expose to the hydrogen plasma in decision of the electronic structures in untreated virgin graphite powder. We showed that absorption of many additional materials with carbon framework by water treatment and expose to hydrogen plasma in the virgin graphite powder plays an important role in the complete occupation of electrons in the valence bands, and furthermore, plays an important role in the enlargement of the $\Delta E_{\mathrm{HOMO-LUMO},N}$ values as a consequence of the increase of the sp^3-hybrid orbitals.

We next investigate the critical temperature for the intragranular diamagnetic supercurrents, on the basis of the recent experimental and our previous theoretical researches. According to these researches, in the materials with large $\Delta E_{\mathrm{HOMO-LUMO},N}$ values, the closed-shell electronic structures with large band gaps between the occupied and unoccupied orbitals are formed by the quantization of orbitals by nature, and the attractive interactions between two electrons with opposite momentum and spins occupying the same orbitals can be realized by the Coulomb interactions between all nuclei and electrons. That is, in the materials with large $\Delta E_{\mathrm{HOMO-LUMO},N}$ values, the Cooper pairs are formed by large $\Delta E_{\mathrm{HOMO-LUMO},N}$ values as a consequence of the quantization of the orbitals by nature, and by the attractive Coulomb interactions between two electrons with opposite momentum and spins occupying the same orbitals via the positively charged nuclei. These results can be well confirmed by the recent experimental results that the Cooper pairs have been observed in the neutral 6an, 10ac, 14ac, and coronene. We showed that if the $p_{\mathrm{saturated}}$ value becomes large enough, only one hydrocarbon with sp^2- and sp^3-type hybrid characteristics with the $\Delta E_{\mathrm{HOMO-LUMO},N}$ values of a few eV is needed in each grain. Even though we cannot clearly show which kind of stoichiometry the superconductivity grain part has, it is natural to consider that the hydrocarbons with significant sp^3 characteristics rather than the sp^2 characteristics, the valence–conduction band gaps of which are larger than a few eV, and the valence bands of which are completely occupied by electrons, are formed by water treatment or expose to the hydrogen plasma in the virgin graphite powder.

We next suggest the guiding principle towards larger relative superconducting mass. For example, the virgin diamond treated by water or exposed to the hydrogen plasma in a macroscopic sized grain, in which the valence bands are completely occupied by electrons, have a possibility to exhibit high-

temperature superconductivity. In general, regardless of the kinds of additional materials combined with carbon framework, the macroscopic sized materials with larger $\Delta E_{\mathrm{HOMO-LUMO},N}$ values than a few eV, in which the valence bands are completely occupied by electrons, have a possibility to exhibit high temperature superconductivity. Discovery of appropriate combination of additional materials with the main carbon materials, by which the $p_{\mathrm{saturated}}$ value becomes large enough, is awaited.

Finally, we suggest the guiding principle towards the syntheses of high temperature superconductivity. We suggest that any material with large $\Delta E_{\mathrm{HOMO-LUMO},N}$ value (more than a few eV), in which valence bands are completely occupied by electrons, which has been believed to be typical insulator in view of solid state physics and chemistry, has a possibility to exhibit high temperature superconductivity in solids.

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