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Consideration for the development of room-temperature ambient-pressure superconductor (LK-99)

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Abstract This paper examines the way of thinking and limitations of physicists regarding the phenomenon of superconductivity and outlines how room-temperature and ambient-pressure superconductors can be developed through the statistical thermodynamic background of the liquid state theory. In hypothesis, the number of electron states should be limited by confining them to a state close to one-Dimension. Simultaneously, the electron-electron interactions should be frequent enough for the electrons to have liquid-like properties. As an example of implementing the hypothesis, our team reports the development of room-temperature and ambient-pressure superconductivity of a material named LK-99 (superconducting compound name developed in the research), whose structure was revealed through numerous experiments with a clue found by chance.
Moreover, we summarize the theoretical and experimental basis for the characteristics and discovery of the world's first superconducting material surpassing the critical temperature of 97o C at atmospheric pressure.

Key words Room-temperature and ambient-pressure superconductivity, Liquid-like property, Critical temperature, One Dimension, Electron-electron interactions

Consideration for the development of normal temperature and pressure superconductor (LK-99)

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Abstract In this paper, the flow and limitations of the thoughts of physicists looking at the existing superconductivity phenomenon are reviewed, and the theoretical background presented from the perspective of the statistical thermodynamic liquid theory outlines that normal temperature and normal pressure superconductors can be developed. The way to achieve this is to have an electronic state close to 1-Dimension, where the number of states in which electrons can move is significantly limited, and electron-electron interactions must be frequent enough that electrons in that state can exhibit liquid properties. is to do As an example of this practice, we report the development data of LK-99 (the name of the normal-temperature normal-pressure superconductor developed in this study), whose structure has been revealed through numerous experiments by getting a clue by chance . The properties of superconducting materials and the theoretical and experimental basis for their discovery are summarized.

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1. Introduction

Since the discovery of superconductivity by Onnes [1] in 1911, this phenomenon has passed over a century as an iterative process in which new theoretical developments continue after the experimental discovery. As always, physicists who value scientific systems must be careful and conservative in their approach to this phenomenon, and it is natural that they stick to their systems until a phenomenon that clearly deviate from their expectations is discovered. As briefly summarized in the previous article by Professor Emeritus Oh Keun-ho [2], this phenomenon is not progressed through experimental proposals and proofs by physicists, but rather the development of discoveries by inorganic chemists or material engineers who understand physics is a new breakthrough [3]. ,4] in most cases. If so, what are the limitations of the theoretical approach of physicists currently accepted as a scientific system, and examining the reason will be a basic matter that can increase the possibility of developing a new normal-temperature normal-pressure superconductor. Unfortunately, general physicists' expectations about the development of normal

temperature and pressure superconductors are basically 'skeptical'. The first reason is that only repulsive force exists in the electron-electron interaction, and the only way to mediate this in a solid material is the lattice vibration (phonon), and the lattice vibration increases exponentially as the temperature approaches room temperature. , because it is too high an energy state for electron-electron mediation. Second, it is because of the viewpoint of band theory, which can explain most of the conductivity from the microscopic point of view to various physical phenomena in describing the state of electrons. Basically, this theory can be said to be an electron gas theory that combines fermi-Dirac statistics, which are statistics that half spin (s = ±½), which is a quantum characteristic of electron spin, and Boltzmann statistics from a thermodynamic point of view. However, the main thing in the explanation of superconductivity in the early days was that the superconducting state was an electron-liquid, fermi-liquid [5,6] state.

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It was because it was similar to [7].

A microscopic setting that satisfies these two conditions is the cooper pair theory of BCS [8,9]. The microscopic setting of this theory is as follows. (1) The problem of repulsion due to electron-electron collision can be solved by mediating the energy amount (phonon) of the vibration mode between lattices. (2) The spin of two electrons can be solved by treating it as a Boson, where the sum of each ±½ is 0. Summarizing these two things, the lattice energy mediates the two electrons, so that the electron-specific ±½ s pin is paired within the energy space (k-space). The comparison of electron energy distributions between general metals and superconductors is shown in Fig. It can be conceptually organized as in 1.

The proof of pairing between two electrons in the energy space is experimentally confirmed that the following relational expression is established between the isotope lattice of different masses and the critical temperature [10,11].

$$T_c\ M\ddot{y}\ \ddot{y}\ \ \ddot{y}\ddot{y}1/2 \qquad (1)$$

Later, in the superconducting-insulator-superconducting junction (SIS junction) tunneling experiment by Josephson [12], the default value (magnetic flux, ÿ0) of the voltage increase per quantized electron was predicted from the classical point of view calculated by London [13]. Through experimental proof [14,15] that according to equation (3), which is half of the magnetic flux defined by equation (2), the assumption that two electrons are involved is established as an established theory.

$$\ddot{y}0\ \ddot{y}\ddot{y}\ \frac{2\ddot{y}h}{\text{It is}}\ \frac{h}{\text{It is}} = 4.135\ 10\ddot{y}1\ \text{gauss cm2} \qquad (2)$$

$$\ddot{y}0 = \frac{h}{2e} \qquad (3)$$

However, it is argued that the calculation of the magnetic flux value of rotational motion by 4ÿ periodicity, which is a mathematical process related to translation, rotation, and vibration of charged particles with half-spin, also shows the same value.

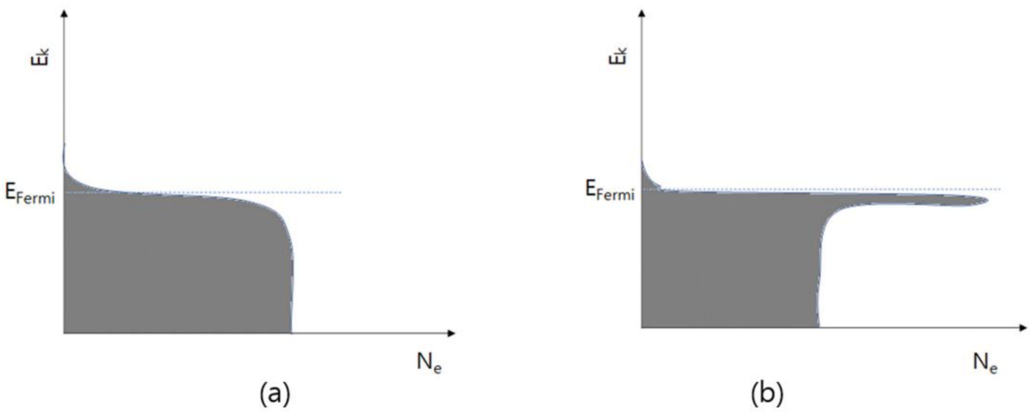


Fig. 1. Schematic (a) metallic and (b) superconducting energy diagram.

Although it had its meaning [16], it was limited to the mathematical and physical domain and did not become a mainstream explanation for superconductivity. This is a value due to the 2ÿ rotation of two paired electrons in classical space, or a quantum physical quantity that appears when one electric charge rotates 4ÿ (2 rotations in real space) on the half spin space, which is a virtual space. It is judged that there is no way to make a difference [17]. In this case, it is natural that the BCS theory, which has succeeded in constructing a new system from a microscopic point of view that can explain all areas of physics related to conductivity, establishes itself as an orthodox theory. This superconducting state is transitioned as the kinetic energy of electrons increases (temperature rise), and this temperature is the critical temperature (Tc) of superconductivity . Then, if superconductivity is viewed from this

point of view, what innovative method can be used to increase the critical temperature at atmospheric pressure? As mentioned earlier, it is 'skeptical'. This can be seen from the fact that the upper limit of the critical temperature (Tc) of a superconductor calculated at the time of the establishment of the BCS theory is about 30 K (ÿ243o C). A study on the extension of this approach is a recently published material that exhibits room-temperature superconductivity under a pressure of 10,000 atmospheres [18] and previous studies related to it [19,20]. This is based on Ashcroft [21]'s prediction that the critical temperature can rise extremely if the effect of the lattice is limited by pressing with ultra-high pressure. However, even in this case, there are reports that explain that electron-electron interaction is a larger factor than phonon mediation by significantly limiting the lattice energy [22,23].

A new turning point in the 'skeptical' discussions of physicists in the early days of superconductivity research is high-Tc superconductivity. Since Bednorz and Müller [3] experimentally discovered LaBaCuO in 1986, which exceeded the critical temperature of 30 K, which was pointed out as the limit of the existing theory, the rise in the critical temperature of high-temperature superconductors was noticed by chemical/material engineers for the next 10 years. Critical temperature rise I brought [24,25]. This means that relatively inexpensive liquid nitrogen can be used instead of expensive liquid helium as a refrigerant. Of course, physicists' approach to explain this has to be based on the concept of electron pairs, which are bosons in which superconducting electrons have the same energy state in k space, and attention has been focused on what substance mediates electron pairs at high temperatures.

On the other hand, with the advent of ARPES (Angle-resolved photoemission spectroscopy), an advanced technology that can map the electronic energy state at ks pace of a superconducting single crystal into a 3D state, the distribution of electron energy in a superconducting state and a non-superconducting state can be distinguished. As a result, it has come closer to confirming the essential phenomenon of superconductivity [26,27]. However, since this experimental method is obtained through the Fourier transform of the spectroscopic measurement values, it is possible to check how much the electron energy distribution in the crystal is in k space, but it is not possible to obtain information on the position of electrons in the lattice. .

Nevertheless, it seems that the MIT research team [28] found information about the position of electrons and new hints about the spatial structure. It was confirmed that graphene, a structure of single carbon, showed superconductivity when it was overlapped at a magic angle (1.1o) . It has been found experimentally along with calculations that a constant energy state is displayed near the level. The prediction that a stable energy state exists near or just below the Fermi-level is consistent with the ARPES measurement results, and accordingly, it is possible to predict the existence of a recently hidden superconducting band [26] or theoretically propose its basis [29]. New theorists are emerging one after another.

Based on statistical thermodynamics, the author has three waves with clear physical definitions of the gaseous state (fluid) including the liquid state.

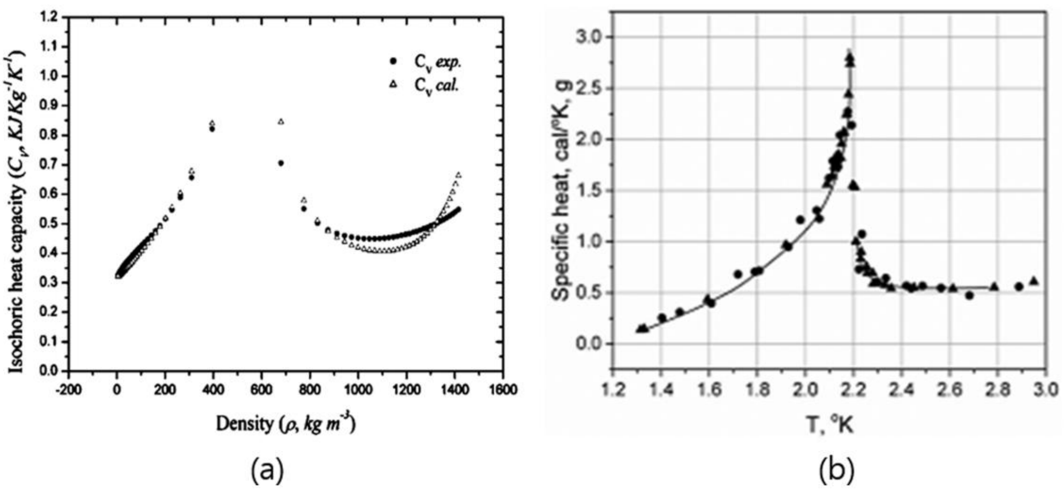


Fig. 2. Heat capacity ÿ-transition near critical point (a) Isochoric heat capacity (Cv) of Ar (b) Isobaric heat capacity (Cp) of ^{4}He.

A generalized van der Waals equation of state was proposed through modeling [30], which describes it as accurately as possible using lametta, and based on this, it was derived that the viscosity of the liquid is a state function due to the state of the material, up to the Corresponding State [31] has presented a new perspective on viscosity. This is a departure from Henry Eyring's model approach to liquids [32-34], and it presents a new alternative to solve various fluid problems that have been delayed due to difficulties in modeling in the meantime from the point of view of state functions.

Based on this experience, I revisited the early ideas of superconducting research that superconducting electrons have liquid-like properties. In fact, at the time of the initial discussion of superconductivity, it is regrettable that I could not participate in the discussion of superconductivity or superfluid phenomena due to the limitations of the liquid theory that was not fully established. This is because the heat capacity characteristics of superconducting electrons, which BCS cannot explain, coincide with the ÿ-transition, which is the heat capacity change characteristic at the critical point of a material (Fig. 2) [35,36].

1.1. Fluid analysis of the critical temperature (Tc) and heat capacity (Cp) of a substance

In general, when looking at the state in which gases and liquids coexist from the point of view of particles, gases have degrees of freedom in the xyz three directions that each particle has, but in liquids, clusters are formed by van der Waals forces between particles, and within the gravitational field In , it is condensed on the bottom as 2-Dimension, and the number of 3-Dimension particles increases when the surface tension is large. Of course, in the zero-gravity field, pseudo-2D particles and dense 3D particles due to the surface tension of the clusters are mixed. Then, what is the state of the particles at the critical temperature (Tc) of the substance? According to the author's thesis , it means the point where Nce = 0, where the cell state modeling the liquid state completely disappears, and the physical quantity that can clearly see this state is the point at which the heat capacity dissipates to infinity. It means to make a transition.

1.2. State Equation, Viscosity, and Electrical Resistance

Among the characteristics of fluids, the main issue regarding viscosity that can be seen in everyday life is whether viscosity related to the flow of matter can be described as a state function related to the state of matter. This is because viscosity becomes a criterion for judging whether it is simply a dynamic property or a thermodynamic property, which is an inherent property of a material. If the general formula of viscosity [31] representing the degree of fluid flow can be created, it can be said to prove that viscosity is an intrinsic thermodynamic property of matter.

all. The astrology theory based on the break theory of Prof. Dongsik Choi and Prof. Wonsoo Kim [37] describes the form of this general formula. This viscosity theory is a phenomenological theory applied to real fluids derived by assuming a new concept that the sum of the absolute values of the attractive force and the repulsive force is the absolute pressure when calculating viscosity as a state function. This ^{3}He,^{4} research led to the calculation of properties such as the viscosity of the He superfluid at the time [38], and it was intuitively possible to see the possibility that it could be applied to electronic fluids as well. This is because, in the case of superconducting transition or metal conductivity, the changes in electrical resistance and isopoise viscosity line are similar, and the phase transition of heat capacity has the same pattern, ÿ-transitio, on both sides [13]. At the time, it was a hypothesis and a problem that could not be mathematically proven, but the feasibility of this approach was confirmed by an experiment by Levitov and Flakcovich in 2016 [39].

1.3. How to raise the critical temperature: 1-Dimensional electron structure & electron correlation

What is noticeable in the new approach based on the above-mentioned basis is that it is important to have a low-dimensional (2D, 1D, etc.) electronic structure that severely limits the number of electron states in order to increase the critical temperature under normal pressure. Theoretically, as the critical temperature of a material decreases in dimension, the following ratio was calculated [40].

$$3D:2D:1D = 1:1.767:4.5 \qquad (3)$$

$$2D:1D = 1:2.546 \qquad (4)$$

From this point of view, when 28 K, the highest critical temperature of a superconducting alloy with electrons having a three-dimensional degree of freedom, is substituted into Equation (3), the upper limit of the critical temperature in the second dimension is about 50 K, and the upper limit of the critical temperature in the first dimension is about 126 K. According to Equation (4), substituting Tc = 138 K for $HgBa_2Ca_2Cu_3O_{8+ÿ}$, which has the highest critical temperature among superconductors with two-dimensional characteristics , reaches 351 K. Of course, this is a schematic result that does not consider the pressure effect in the bonding structure, such as chemical pressure, and qualitatively means that the critical temperature of a material with a one-dimensional electronic structure has a possibility of about 126 to 350 K. only Of course, as mentioned above, the distribution of electrons in the position where electron-electron interaction is possible must exist in high

2. Experimental method

With this theoretical background and considering commercialization, four elements (Pb, Cu, S, P = 1:1:1:0.2) among the most abundant materials on Earth

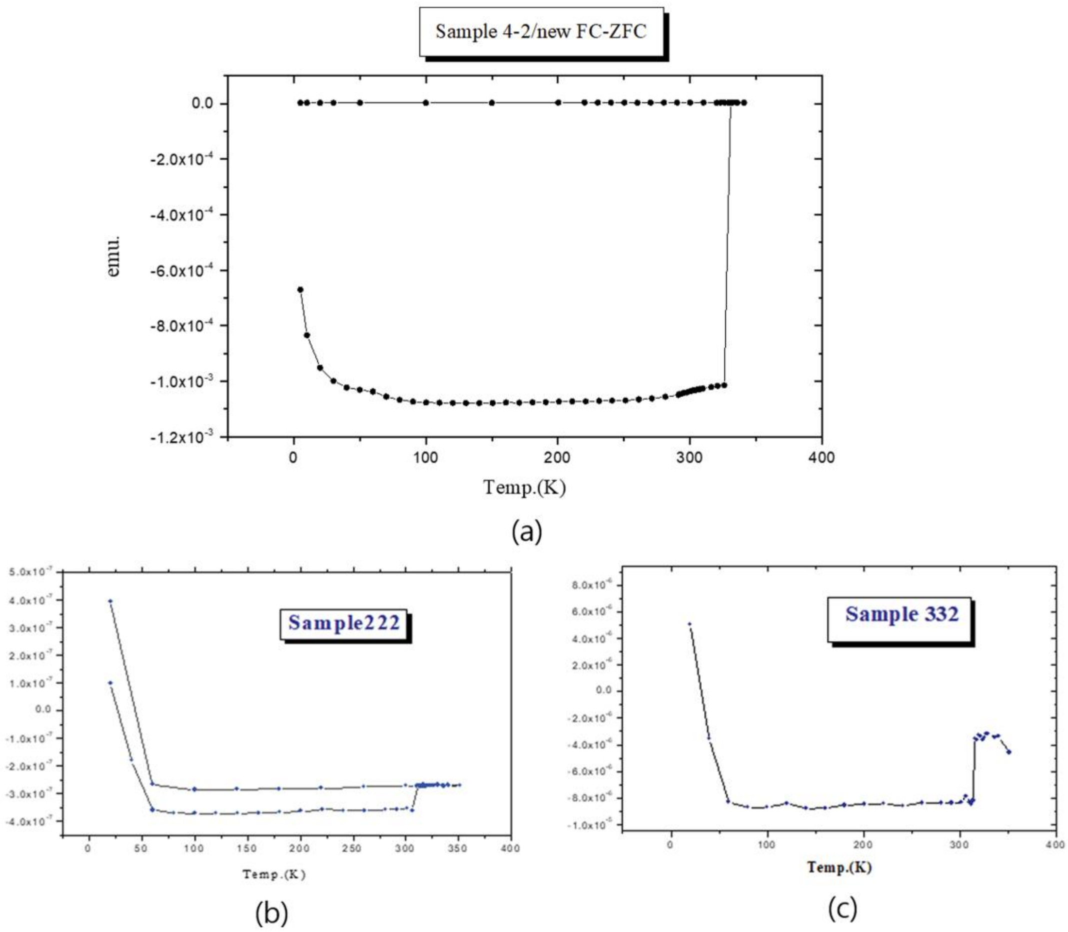


Fig. 3. Abnormal magnetic hysteresis behavior under Zero-Field-Cooling and Field-Cooling. (a) Transition temperature is about 323 K (50o C) (b), (c) Reproducibility results of same experiment procedure.

In 1999, a new attempt was made in a simple system by selecting . The manufacturing method is to sufficiently grind the selected elements with a pestle and subdivide them into 1g each in a 10 mm diameter quartz tube, and then use a vacuum pump to keep the inside of the quartz tube in a vacuum at 10ÿ3 Torr to propane to a length of about 15 cm . Seal the quartz tube with an oxygen torch. Thereafter, the quartz tube taken out of the furnace is cooled by heating in a furnace (Furnace, A-Jeon Industrial Co. Ltd) at 900 ° C. for 24 hours, and a sample is taken. It was well ground and sampled, and the magnetic susceptibility was measured using SQUiD (Superconducting quantum used interference device, MPMS-XL, Institute of Basic Science). Magnetic Property Fig. The same results as in Fig. 3a could be confirmed, and the reproducibility of the critical temperature through repeated experiments at the time. 3b and 3c were confirmed. However, due to a manufacturing method problem, it

was not possible to find even the structure of a characteristic material that appears to have a critical temperature of over 300 K, which is assumed to be in a very small amount. While studying the superconductivity phenomenon amidst continuous suspicion, in 2017, based on these data, repeated experiments were conducted in earnest, and cases where the quartz tube was destroyed due to internal pressure during rapid cooling or reaction were found.

It has been discovered that an amorphous form of material of unknown origin has unique magnetic properties. (2018) The magnetism of foreign matter was not typical of superconductivity, but showed superparamgnetism characteristics. Regarding this characteristic, research professor Kwon Yeong-wan of Ku-KiST Graduate School of Convergence, Korea University, gold-plated DNA, which is the research subject of electron transfer through EPR. Following the similarity between the relationship between state and magnetic susceptibility [41] and the opinion that the superconductivity phenomenon could also be regarded as EPR, the evaluation of EPR characteristics was conducted from 2017, but the structure of the material was clearly secured and the structure It was difficult to see progress with EPR-only methods until the manufacture of high-purity materials containing a large amount was successful. In order to clarify this process, firstly, the process of finding a structure showing the above characteristics in 2019 and the conjecture contents and results, and secondly, in 2021, it succeeded in separating the substance and structural analysis was possible. The results of the synthesis of normal-temperature and normal-pressure superconductivity were summarized and applied for, and a third patent application was filed in

2022 with more detailed claim reinforcement [42]. The manufacturing method of this material (LK-99) [43] identified so far is summarized as follows.

Lanarkite and Cu_3P were uniformly mixed at a molar ratio of 1:1 in a mortar and pestle. The sample was placed in a reaction tube, sealed with a vacuum of 10ÿ3 Torr, and reacted at 925o C for 5 to 20 hours. Re-evaluate the dark gray ingot after the reaction

It is now possible and then shaped into a thin cuboid for electrical measurements. For other analyses, the remaining samples were pulverized and used in powder form. In the case of the former, lanarkite and Cu_3P as raw materials were prepared by mixing PbO and $PbSO_4$ powder in a mortar

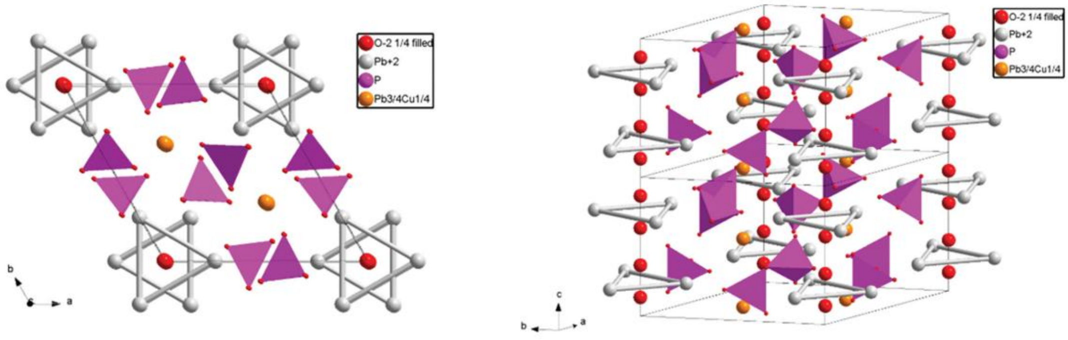
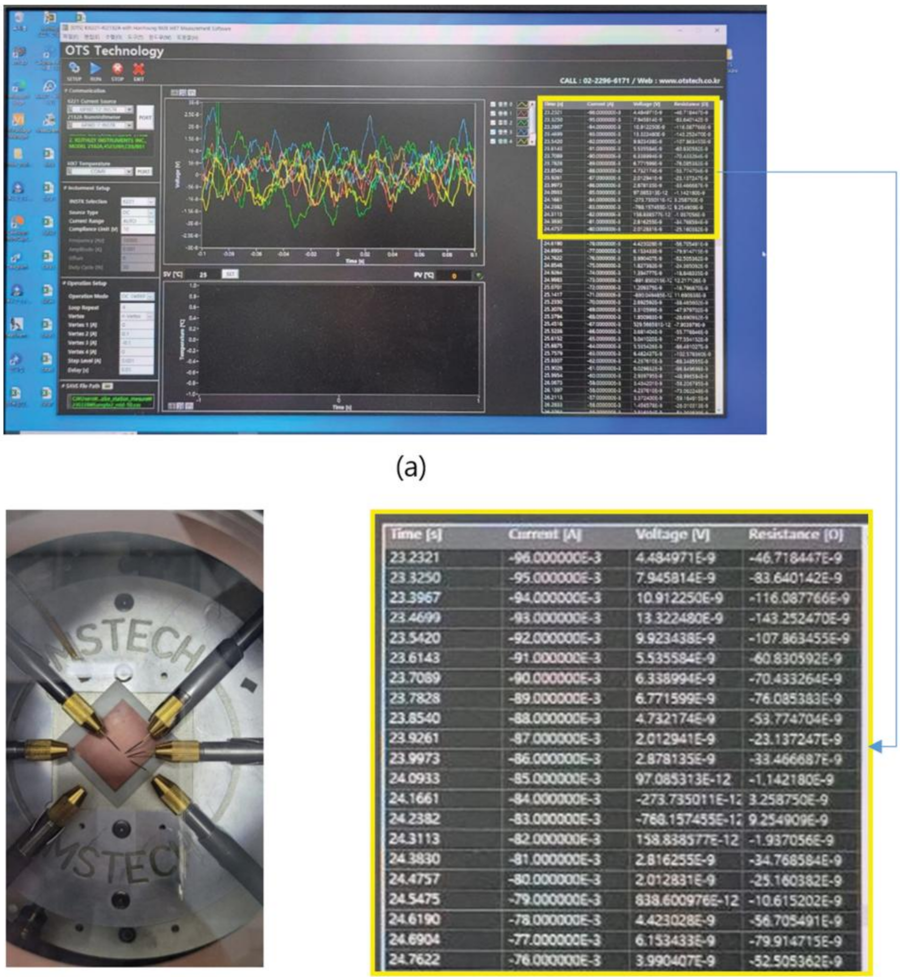


Fig. 4. Molecular unit cell structure of LK-99 (a) projection view from c-axis (b) double unit cell following c-axis.



Time [s]	Current [A]	Voltage [V]	Resistance [Ω]
23.2321	-96.000000E-3	4.484971E-9	-46.718447E-9
23.3250	-95.000000E-3	7.945814E-9	-83.640142E-9
23.3967	-94.000000E-3	10.912250E-9	-116.087766E-9
23.4699	-93.000000E-3	13.322480E-9	-143.252470E-9
23.5420	-92.000000E-3	9.923438E-9	-107.863455E-9
23.6143	-91.000000E-3	5.535584E-9	-60.830592E-9
23.7089	-90.000000E-3	6.338994E-9	-70.433264E-9
23.7828	-89.000000E-3	6.771599E-9	-76.085383E-9
23.8540	-88.000000E-3	4.732174E-9	-53.774704E-9
23.9261	-87.000000E-3	2.012941E-9	-23.137247E-9
23.9973	-86.000000E-3	2.878135E-9	-33.466687E-9
24.0933	-85.000000E-3	97.085313E-12	-1.142180E-9
24.1661	-84.000000E-3	-273.735011E-12	3.258750E-9
24.2382	-83.000000E-3	-768.157455E-12	9.254909E-9
24.3113	-82.000000E-3	158.838577E-12	-1.937056E-9
24.3830	-81.000000E-3	2.816255E-9	-34.768584E-9
24.4757	-80.000000E-3	2.012831E-9	-25.160382E-9
24.5475	-79.000000E-3	838.600976E-12	-10.615202E-9
24.6190	-78.000000E-3	4.423028E-9	-56.705491E-9
24.6904	-77.000000E-3	6.153433E-9	-79.914715E-9
24.7622	-76.000000E-3	3.990407E-9	-52.505362E-9

Fig. 5. Zero resistivity measurement of thin film sample (a) measurement window picture at R0 experiment (Keithley 6221 & 2182A) (b) uploaded thin film on copper plate in probe station (MSTech) (c) Measured data of LK-99 thin film (under ± 3 × 10ÿ8 V) Zoom in data of yellow box.

were uniformly mixed with a pestle at a molar ratio of 1:1. Then, the sample was transferred to an alumina crucible and reacted in a furnace at 725o C for 24 hours. After completion of the reaction, a white sample was obtained and pulverized with a mortar. In the latter case, Cu and P powders were mixed in respective compositional ratios. Then, the sample was transferred to a quartz tube. A thick quartz tube was sealed with a thickness of about 20 cm to 1 g of sample in a vacuum of 10ÿ3 Torr, and reacted in a furnace at 550° C for 48 hours. After taking it out of the tube, a dark gray ingot was obtained and pulverized. Reagents used in the solid phase reaction were PbO (JUNSEI, GR), PbSO4 (KANTO, GR), Cu (DAEJUNG, EP) and P (JUNSEI, EP).

3. Results and discussion

3.1. Superconducting properties of LK-99

(1) Apatite structure based on lead (Pb)

About 1 out of 10 lead Pb in (Pb10(PO4)6) is replaced with copper (Cu), resulting in shrinkage (Fig. 4) (Pb10ÿxCux(PO4)6, 0.9 < x < 1.1) (Space group: P 63/m(no. 176), Unit cell dimensions: a = 9.8430 Å, c = 7.4280 Å) [42].

(2) Resistance 0 characteristics: international standard in current-voltage characteristics in a narrow range when materials are thermally deposited to produce high-purity thin films Data in the range of 10ÿ8 ~ 10ÿ9 V/cm below 1.0ÿ6 V/cm (International Electrotechnical Commission standards) were secured (Fig. 5). (3) Current-critical temperature

characteristics: When measuring the resistivity of an ingot sample based on ± 30 mA, which can be measured stably using the 4-probe method, the critical temperature (Tc) is 104 degrees C (approximately 377 K) [42], when resistance was measured based on ± 60 mA, it was confirmed that the critical temperature (Tc) was 97 degrees Celsius (approximately 370 K) (Fig. 6a, Fig. 6b). As the temperature rises, which is a general superconductivity characteristic in current-voltage characteristics from room temperature to almost 400 K, the critical current decreases, and even at 400 K, microcurrents of less than 10 mA are maintained without superconducting-conducting

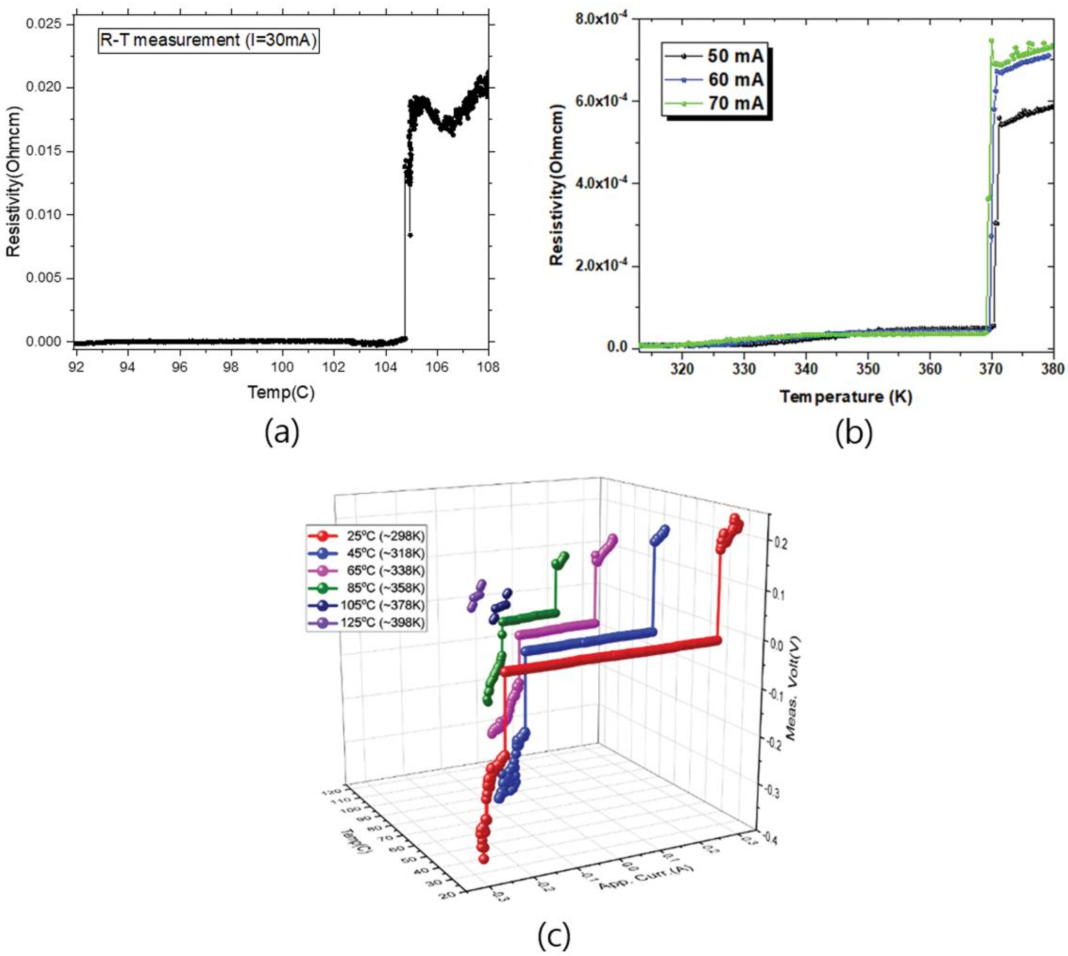


Fig. 6. LK-99 Ingot sample results of 4-probe measurement for Transition temperature (Tc) following applied Currents (a) ± 30 mA (from patent: 10-2021-0112104) (b) ± 50, ± 60, ± 70 mA, respectively. (c) Current-voltage measurement results following tempera ture variation (T) (Keithley 228A, 182, 2000).

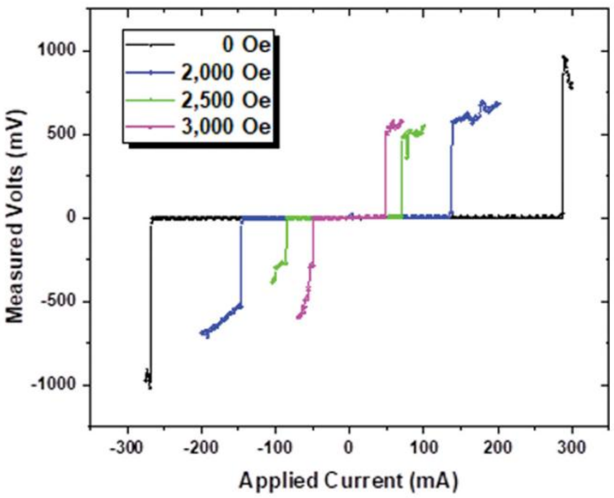


Fig. 7. Current-voltage measurement results following external magnetic field (H) variation (Keithley 228A, 182, 2000).

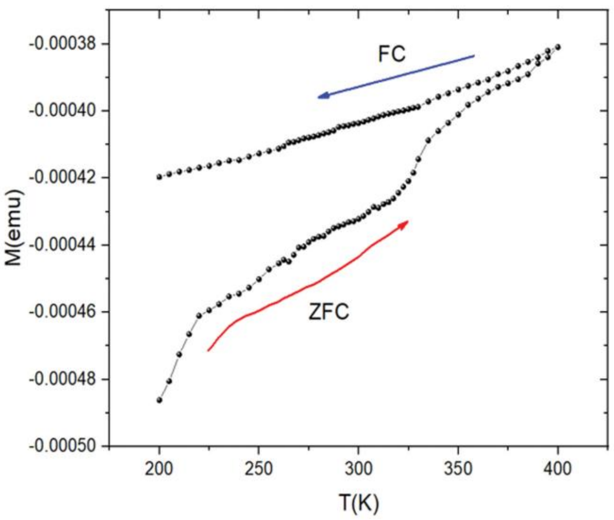


Fig. 8. Hysteresis results through zero field cooling and field cooling of LK-99.

Confirmed. (4)

Current-critical magnetic field measurement: When measuring the ingot sample and testing the current-voltage characteristics under an external magnetic field, the critical current drop phenomenon according to the increase in the external magnetic field was confirmed (Fig. 7). (5) Magnetic susceptibility: In the magnetic susceptibility measurement according to the temperature of the thin film sample, the hysteresis of ZFC (zero field cooling)-FC (field cooling) up to 400 K, which is the limit of SQUID, was confirmed (Fig. 8) (KARA : MPMS 3-evercool).

(6) Calculation result of heat capacity (Cp/T) through precise voltage-temperature measurement based on current application : The characteristic change of heat capacity at the critical temperature (Tc) has the characteristics of ÿ-transition as mentioned above. . The temperature is constantly raised within the limits that the measuring equipment can measure, and a constant current is applied in units of 400 msec, based on the results of measuring the voltage and temperature of the sample.

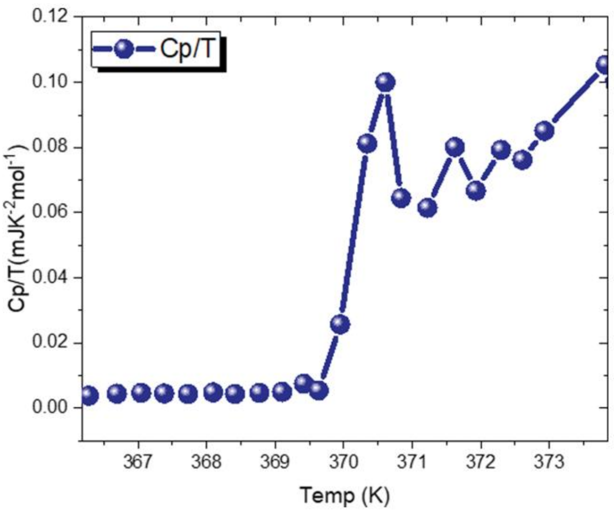


Fig. 9. Calculated specific heat capacity over Temperature (Cp/T) of LK-99 at ± 60 mA current near Tc. It shows the charac teristic form of ÿ-transition near Tc.

The heat capacity was calculated as, Fig. As shown in Fig. 9, characteristics very similar to ÿ-transition were confirmed around the critical temperature. A detailed analysis

of the precise structure and electronic structure of this, and an interpretation of the measurement results of the magnetic susceptibility curve (MH) according to the controversial external magnetic field will be summarized and published in an international journal.

Apatite structure is uniquely a material with an anisotropic electronic structure with different physical properties along the c-axis and ab-axis, and is a complex material with an electronic structure close to 1D. The condensation of the crystal structure during impurity doping on this structure was detailed in a previous study [44] by Professor Emeritus Oh Geun-ho. In this paper, this material (LK-99), which has an electronic structure close to 1D, has a critical temperature measured over 400 K, that is, it provides surprising data that maintains the superconductivity phenomenon beyond room temperature and exceeds 100 degrees Celsius under normal pressure. In order to help understand the physical meaning and interpretation of this material, the difference between the approach of physicists from the existing perspective and the statistical thermodynamic logic of Professor Dong-shik Choi, who seems new but old, has been outlined. . In addition, due to the nature of venture companies, the approach was shared to help scholars who will have academic discussions about it at the time of patenting and disclosure.

In fact, these approaches are based on A. Mourachkine's [45] book, 'Room Temperature Superconductivity', which suggests the possibility of normal temperature and pressure superconducting materials, which do not seem to be accepted as orthodoxy among physicists, and a new material based on iron (Fe). It can be confirmed that Dr. Hosono [46] of Japan, who opened the series of superconducting materials, presented the possibility of superconducting 1D materials having the same apatite structure. and

Based on their experience, it can be seen that LK-99 meets the standard of normal temperature and pressure superconductor. The emergence of

normal temperature and normal pressure superconductors that can solve all kinds of current environmental and energy problems, as summarized in Professor Oh Geun-ho's contribution [2], will lead to the future environment of mankind (quantum computers, efficient use of electrical energy, quantum wireless It is judged that it can be used for communications, ultra-high efficiency motors based on high magnetic fields, precision instruments including MRI, hyper loops, etc.). Due to the nature of electronic materials called superconductors, precise analysis is preceded based on single-crystal materials, and this paper is submitted after judging that intensive research on materials and various attempts are needed.

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