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Terahertz oscillations in mercury cuprate superconductors

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Abstract. It has been recently reported that the three-dimensional Bose–Einstein condensation of the quasi-particles is valid for the mercury cuprates at liquid helium temperature. In this study, the validity of the interlayer theory in three dimensions has been investigated for optimally oxygen-doped mercury cuprates at the temperature interval of 0–15 K. Furthermore, some thermodynamic and electrodynamics parameters of mercury cuprates have been calculated for both the under-doped and the over-doped samples at the vicinity of 4.2 K. Moreover, it has been determined that the superconducting system behaves as a terahertz wave cavity regardless of the oxygen doping concentration.

Keywords. Mercury cuprates; terahertz oscillations; interlayer theory; Bose–Einstein condensation.

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1. Introduction

The mercury cuprate family superconductors, which have the highest Meissner transition temperature [1–3], high critical magnetic fields and critical current density values, have a great importance for both theoretical investigations and technological applications. As the result of theoretical labouring for explaining the electrical conductivity mechanism of high-temperature superconductors, it has been recently pointed out that the interlayer theory (ILT) [4,5] is valid for optimally oxygen-doped $HgBa_2Ca_2Cu_3O_{8+x}$ mercury cuprates at liquid helium temperature [6]. According to ILT, electron pairing in the superconducting state makes the transport process along the c-axis to be coherent via Josephson (Lawrence–Doniach-like) coupling between the superconducting copper oxide layers. Moreover, in order to provide the coherent transport mechanism along the c-axis, the Josephson coupling energy,

ε_J, must be equal to the superconducting condensation energy, E_b [4]. This requirement of ILT leads to the formation of a plasma resonance mode in the system that results in a three-dimensional Bose–Einstein condensation (BEC) [7] that can be represented by the symmetrical quantum wave function. In this point of view, the two-dimensional BEC, carried out by the electron pairs in ab-plane of superconducting copper oxide layers, has been extended to all layers along the *c*-axis via quasi-particle tunnelling at the Josephson plasma resonance. In this context, the optimally oxygen-doped $HgBa_2Ca_2Cu_3O_{8+x}$ (Hg-1223) superconductor exhibits three-dimensional BEC via Josephson coupling at the Josephson plasma resonance frequency, f_p, of 83.3×10^{12} Hz at the liquid helium temperature. Therefore, Hg-1223 superconductors can also be considered as a coherent electromagnetic wave cavity [6,7].

Scientists have exerted many efforts to figure out a solid state system containing quasi-particles that can fulfil the necessary conditions for BEC since 1962 [8]. It has been reported that there has been an increasing number of experiments on spontaneous Bose-coherence of excitons and polaritons [9]. Recently, it has been reported that CdTe-based semiconductor microcavities exhibit BEC at temperatures reachable by standard cryogenic techniques [10]. In the last few years, experimentalists have tried to determine the limit of the symmetric BEC state in the case of adding little fermionic content, i.e. antisymmetric state doping, to the system [11]. In this study, Hg-1223 cuprate superconductors with different oxygen doping rates have been investigated in detail by means of ILT and BEC at the vicinity of liquid helium temperature of 4.2 K via magnetic hysteresis curves obtained by superconducting quantum interference device (SQUID) as explained in §3. Especially, it is thought that investigation of the limit of the oxygen doping on optimally oxygen-doped mercury cuprate superconductors, which are represented by a symmetrical quantum wave function, can help to determine the BEC limit for the antisymmetric state doping to the system.

2. Theoretical

Copper oxide ceramic high-temperature superconductors have a common structure in which superconducting copper oxide layers are separated by a thin insulating layer. Copper oxide layers are coupled together by Josephson tunnelling between adjacent layers. According to the experimental evidences, cuprates such as $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2Ca_2Cu_3O_{10}$ and $YBa_2Cu_3O_{7-x}$ behave like stacks of superconductor–insulator–superconductor intrinsic Josephson junctions (IJJ) [12].

As is known, the main Josephson plasma excitation modes in weakly Josephson coupled layered superconductors are the longitudinal (along the *c*-axis) and transversal (in the ab-plane) plasma modes. Whereas the transverse-mode plasma oscillations can be converted into electromagnetic waves at the boundary of the junctions [13], the longitudinal plasma propagation modes in an array of IJJ do not lead the electromagnetic wave radiation, since the electromagnetic wave has only transverse mode [14].

Tachiki *et al* [15] have theoretically proved that if the Josephson plasma is excited in some way in high T_c superconductors, the excited plasma decays by emitting an electromagnetic wave. The simulation results of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) have shown that when an external current is applied to the sample, the voltage due to the fluxon flow in the system excites the Josephson plasma with terahertz frequency [16,17]. The frequency of the electromagnetic waves emitted at any bias voltage in the fluxon flow regime of the junctions gets increasing attention due to the coherent, continuous and tunable character [14].

In this work, mercury-based superconductors have been considered as an array of IJJ, as well. Since the ILT is valid at low temperatures for optimally oxygen-doped mercury cuprates, all copper oxide layers along the c-axis of the system are in the resonating state with a Josephson plasma frequency of 83.3×10^{12} Hz [6,7]. This type of Josephson plasma resonance mode cannot be attributed to the fluxon flowing mechanism, since the working magnetic field interval never exceeds the lower magnetic field of H_{c1}. Hence, the magnetic flux is totally expelled from the superconductor. The phenomenon is displayed as a perfect diamagnetic response on SQUID data. Ultimately, the terahertz plasma oscillations determined in the system provide the bulk mercury cuprate superconductor as a coherent terahertz wave source without any bias voltage. As is known, terahertz waves have various advanced technological applications including medical diagnoses, security, and information technology. From this point of view, this work is devoted to examine low-temperature (at the vicinity of the liquid helium temperature) electrodynamics parameters of the mercury cuprate superconductors in the context of both ILT and terahertz wave plasma resonance.

3. Experimental

The $HgBa_2Ca_2Cu_3O_{8+x}$ (Hg-1223) superconductors were synthesized using the two-step procedure. In the first step, the nominal composition of $Ba_2Ca_2Cu_3O_x$ (reactant material) was prepared using four nine purity compounds of BaO, CaO, CuO, and heating them at 1340°C for 10 h in oxygen environment. The reactant material and the high-purity HgO, which was put in a smaller diameter test tube, were both placed in the first tube, which was finally evacuated and sealed. In the second step, the sealed tube was placed horizontally in a furnace and heated at a rate of 1.5°C/min to 940°C for 3 h. In order to obtain the optimally doped Hg-1223 superconducting material, the sample was annealed in oxygen atmosphere at 300°C, well below the decomposition temperature of HgO, for 10 h. Eventually, optimally over- and under-oxygen-doped Hg-1223 superconductors have been obtained by repeating the oxygen annealing process for several times. Regarding the temperature dependence of the susceptibility data, the Meissner transition temperatures of the samples have been determined as 137.3 K [6], 126 K [18] and 123 K [2] for optimally over- and under-oxygen-doped $HgBa_2Ca_2Cu_3O_{8+x}$ superconductors, respectively.

Some thermodynamic and electrodynamics parameters such as thermodynamic critical magnetic field, H_c, superconducting condensation energy E_b, Josephson coupling energy E_J, Josephson penetration depth λ_J and Josephson plasma frequency f_P of the optimally, over- and under-doped samples have been calculated via magnetic critical current density J_c by Ginzburg–Landau theory, Lawrence–

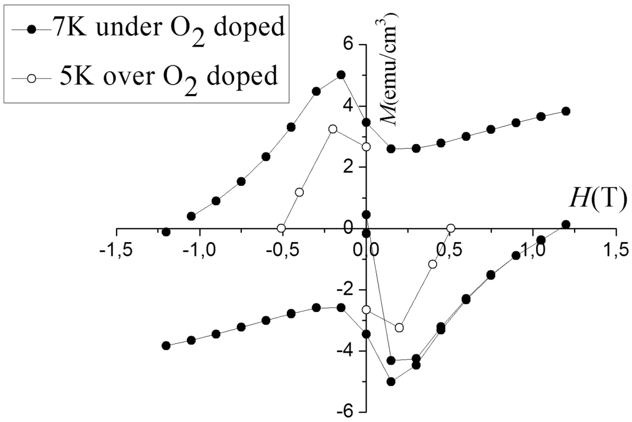

Figure 1. Magnetization vs. applied magnetic field (M–H) for under- and over-oxygen-doped Hg-1223 samples at 7 and 5 K, respectively.

Table 1. Critical current density and some critical magnetic fields for optimally-doped (opt. dop.), over-doped (ov. dop.), and under-doped (und. dop.) Hg-1223 at 4.2, 5 and 7 K, respectively.

Temperature T (K)	J_c at H_{c1}(A/m^2)	H_{c1} (T)	H_{c2} (T)	H_c (T)
4.2 (opt. dop)	1.0×10^{12}	0.35	3	1.024
5 (ov. dop.)	1.58×10^{12}	0.20	0.51	0.318
7 (und. dop.)	1.43×10^{11}	0.25	1.18	0.543

Doniach model and ILT as had already been expressed in our previous works [6,7]. Critical current densities have been deduced from the dynamic magnetization (M) vs. magnetic field (H) (hysteresis) curves (figure 1) of under- and over-oxygen-doped Hg-1223 samples at the vicinity of 4.2 K.

The experiments have been performed by the MPM-5S model SQUID susceptometer. In all the measurements, the magnetic field has been applied along the c-axis and hence the critical current has flowed in the ab-plane of the specimens. The J_c and the related parameters of the optimally oxygen-doped Hg-1223 sample have also been taken from refs [6,7].

J_c values have been calculated by Bean critical state model [19] at the lower critical magnetic field H_{c1}. The calculated J_c and the values of lower and upper critical magnetic fields H_{c1} and H_{c2} together with H_c have been listed for under-, optimally- and over-oxygen-doped Hg-1223 cuprates in table 1.

4. Results

In this paper, the validity of ILT has been examined for under-, optimally- and over-oxygen-doped Hg-1223 samples by the comparison of E_b and E_J energies. E_J and E_b energies per unit area have been calculated using eqs (1) and (2), respectively.

Table 2. Superconducting condensation and Josephson coupling energies per unit area for various oxygen-doped Hg-1223 cuprates.

Temperature T (K)	E_b (J/m^2)	E_J (J/m^2)
4.2 (opt. dop)	3.285×10^{-4}	3.29×10^{-4}
5 (ov. dop.)	3.173×10^{-4}	5.199×10^{-4}
7 (und. dop.)	9.262×10^{-5}	4.706×10^{-5}

$$E_J = J_c \Phi_0 / 2\pi, \tag{1}$$

$$E_b = H_c^2 s / 2\mu_0. \tag{2}$$

In eqs (1) and (2), $s = 7.887 \times 10^{-10}$ m is the average spacing between CuO_2 layers and Φ_0 is the magnetic flux quantum. The calculated energies are given in table 2. In this context, the comparison of E_b and E_J values for three samples has shown that the ILT has proved to be valid for only the optimally O_2-doped Hg-1223 superconductor at the liquid helium temperature of 4.2 K. As is known by Hall measurements, the oxygen-doped mercury cuprate superconductors display hole type of conductivity. In particular, optimally oxygen doping process results in the coupling of the uncoupled electrons in the system. Hence, extra oxygen doping to the system in over-doping procedure works as adding some antisymmetric contamination of the symmetric state.

According to table 2, the copper oxide layers are in a maximum Josephson coupling at the liquid helium temperature, that leads the emanation of three-dimensional BEC in the optimally oxygen-doped Hg-1223 superconducting system. The temperature interval at which the ILT is valid, has been determined for optimally O_2-doped Hg-1223 cuprate by the calculation of E_b and E_J values [6] from the $E_b = f(T)$ and $E_J = f(T)$ functions which have been illustrated in figure 2.

The graphical representation of the calculated energy values from the fitting functions given in figure 2 with respect to temperature has demonstrated that both ILT and three-dimensional BEC are valid for the temperature interval of 3–7 K (figure 3). Due to the exhibition of the resonance and the three-dimensional BEC in the system, the transport of the electrical current through all copper oxide layers is to be without loss at low temperatures. In this point of view, the dilemma that goes on about the flow of supercurrent of the high-temperature superconductors whether intra- or intergrain has been resolved for the Hg-1223 superconductor at the temperature interval of 3–7 K.

Josephson plasma frequency, f_p, which is one of the crucial electrodynamics parameters to form resonance state in the system, has been calculated by

$$f_p = c / 2\pi\lambda_J, \tag{3}$$

where c is the velocity of light and λ_J is the Josephson penetration depth ($\lambda_J = (\Phi_0 / 2\pi J_c \mu_0 s)^{1/2}$). The Josephson plasma frequencies of under- and over-doped Hg-1223 samples at the vicinity of liquid helium temperature (table 3) correspond to mid-infrared radiation of the electromagnetic spectrum.

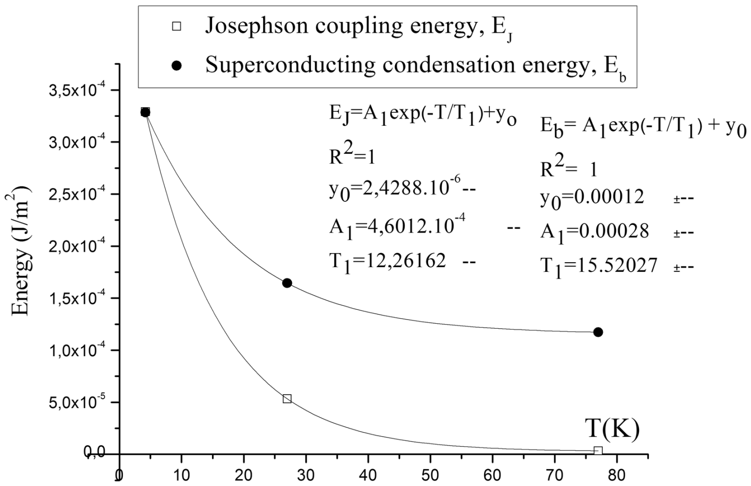

Figure 2. E_{b} and E_{J} energies vs. temperature graphics performed by Origin Lab 7.5 program for optimally oxygen-doped Hg-1223 superconductor at a temperature interval of 4.2–77 K.

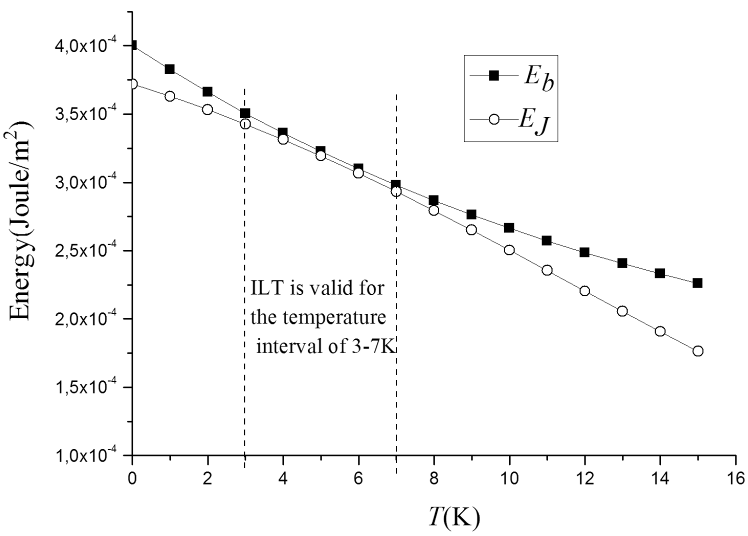

Figure 3. The temperature dependence of the derived Josephson coupling and superconducting condensation energy as obtained from fitting functions in figure 2 for optimally oxygen-doped Hg-1223 sample in the temperature range of 0–15 K.

It has been determined that the infrared Josephson plasma frequency is an intrinsic property of three copper oxide layered Hg-1223 superconductors at the vicinity of 4.2 K. It has been revealed that this infrared characteristic of the plasma

Table 3. Josephson penetration depth and Josephson plasma frequency for various oxygen-doped Hg-1223 superconductors at the vicinity of liquid helium temperature.

Temperature T (K)	λ_J (μm)	f_p (Hz)
4.2 (opt. dop)	0.575	83.3×10^{12}
5 (ov. dop.)	1.499	32.9×10^{12}
7 (und. dop.)	1.52	31.4×10^{12}

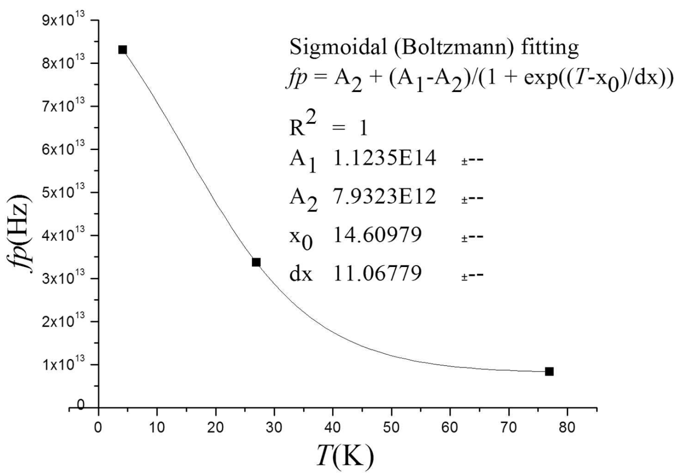

Figure 4. Josephson plasma frequency vs. temperature graphics for optimally oxygen-doped Hg-1223 superconductor.

frequency of mercury cuprates at low temperatures is not affected by the oxygen doping rate. Furthermore, the maximum value of the infrared frequency of 83.3×10^{12} Hz has been achieved for the optimally-doped sample which exhibits three-dimensional BEC. In this case, this frequency may be defined as a critical resonance frequency that allowed the system to condense into the lowest state of existence in three dimensions. Moreover, it has already been reported that as the temperature increases from 4.2 K to 77 K, the plasma frequency decreases one order of magnitude [6] in the optimally-doped Hg-1223 cuprate. This characteristic shift indicated that the system behaves as a terahertz wave cavity above about 57.8 K which has been determined from the fitting function of $f_p = f(T)$ in figure 4 from the data given in ref. [6]. The same terahertz shift tendency has been determined for the over-oxygen-doped samples at about 45 K [20]. Moreover, since 1–10 THz frequency interval is used for bioimaging, the determined plasma frequency of 8.3 THz at liquid nitrogen temperature for optimally-doped Hg-1223 [6] has revealed that the mercury cuprate superconductors can be used as terahertz wave source for the purposes of bioimaging.

5. Discussion and conclusions

This study intended to give an answer to the questions such as "What would happen if the bosons (Cooper pairs) had a little fermionic content, i.e. antisymmetric contribution (antisymmetric state doping) to the symmetric superconducting state?" In this context, it has been well understood that both the confirmation of the ILT and the realization of three-dimensional BEC are very sensitive to oxygen doping concentration and only the optimally oxygen-doped Hg-1223 satisfies ILT theory and the three-dimensional BEC at the vicinity of 4.2 K. Furthermore, we believe that the realization of three-dimensional BEC in mercury cuprate family superconductors provides a crucial contribution to the BEC investigations and represents an important step towards a survey of the promising compact solid-state devices that are based on the controllable macroscopic quantum effects.

It is also predicted that the mercury cuprate superconductors can be considered as promising candidates for the advanced terahertz technological applications including bioimaging, sensing and spectroscopy.

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