Vortex Molecule and $i$-soliton Studies in Multilayer Cuprate Superconductors

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Abstract. We observed two dissipation peaks in the field cooled AC susceptibility measurements on aligned multilayered cuprate Superconductor, (Cu,C)Ba2Ca2Cu3Oy [(Cu,C)-1223], and demonstrated that it is the direct experimental indication of the two-component vortex matter in the multilayered cuprates. We propose that, the second loss peak at lower temperature is due to the additional degree of freedom of the rotation of ‘vortex molecule’ composed of two fractional vortices, originated at two components, mediated by an $i$-soliton bond. To probe the dynamics of this vortex molecule, we measured frequency dependence of AC susceptibility response on aligned crystallites of (Cu,C)-1223 at different temperatures and DC fields. In second peak region, it gives resonance peak. The observed frequency dependence patterns of $\chi''(T)$, for 0.5 T HDC, are rescaled with the resonating frequencies and show tail at low frequency region indicating that the dissipation is due to rotation and twisting of vortex molecule. The temperature dependence of the average relaxation time shows that the vortex molecule rotation/twisting glass state follow critical slowing down process.

1. Introduction

Multilayer cuprate superconductors with CuO$_2$ layers $\geq 3$, are novel multiband superconductors, and when the interband interaction is very weak, it can be considered as a multicomponent superconductor [1,2]. One of the consequences of two gap superconductivity could be the presence of non-Abrikosov vortices [3-8], which may be similar, from the point of view of topology, to those proposed theoretically in superfluid $^3$He [9], liquid metallic hydrogen (LMH) [10], and Bose–Einstein condensates (BEC) [11-14]. Several experimental lines of evidence on the existence of a two-component vortex matter has been obtained in a rotating spinor BEC [15], whereas nonaxisymmetric
vortices or combined spin-mass vortices and solitons have been observed in $^3$He [16-17]. Until the recent discovery by a Stanford group, on an intercomponent phase difference soliton ($i$-soliton) in artificial two-component superconductor composed of an Al bilayer [18], we were not aware of other experimental lines of evidence of two component vortex matter or non-Abrikosov vortices in high-critical temperature ($T_c$) superconductors (HTSc) or in conventional multiband superconductors. Note that we are discussing here the case of a DC magnetic field perpendicular to the superconducting planes and do not take into account Josephson vortices or tilted stacks of pancake vortices.

Independently, we observed the second dissipation peak, along with conventional high temperature dissipation peak due to flux flow and flux creep, in the out of phase susceptibility of the oriented crystallites of (Cu,C)-1223 at various DC fields. We attributed it as an experimental indication of two-component superconductor [19]. When we have two components and the intercomponent interaction is much smaller than that of an intracomponent interaction, there is an intercomponent phase-difference soliton ($i$-soliton) [20,4]. Unless we make a closed $i$-soliton loop or terminate it at the edge of the sample, the $i$-soliton should be terminated by a fractional vortex having fractional flux quanta. Vortex molecule is made of two fractional vortices and one $i$-soliton bond, so as to make the total magnetic flux to be a unit fluxoid quanta, $\Phi_0$, or zero. We attributed the second dissipation peak in $\chi''(T)$ to the possible rotational degree of freedom of the vortex molecule [2].

In low flux density, the intervortex molecular interactions could be similar to the conventional vortex tubes, and vortex molecule lattice is same as conventional hexagonal Abrikosov lattice structure. But when the vortex molecule is exposed to any orientational instability the distorted lattice can govern orthorhombic lattice structure. Such packing can be understood in the framework of the Langmuir monolayers, with respect to rotational distortion, molecular ordering within layer undergoes hexagonal to orthorhombic to herringbone lattice with degree of distortion, this also elongates the shape of the molecular backbone [21,22]. However, possible twisting is not explained in Langmuir lamellar structures. Similar distortion in the magnetic phases of spin glass systems examined in isothermal AC susceptibility measurements give the resonance peak in the frequency dependence [23]. If the rotation has the resonance frequency having temperature dependence, one should see a peak when the resonance frequency passes the frequency of the external AC field. The crucial change in the resonance frequency can be expected in the soft-mode phonon [24], or relevant critical phenomena around the phase transition or crossover [25].

In the present investigation, in order to see the properties of the vortex molecule rotation or twisting or both in response to the AC excitation field, we measured the temperature dependence of $\chi''(T)$ for frequency from 10 Hz to 10 kHz and at various DC field. Observed frequency dependences in $\chi''(T)$ are rescaled and average relaxation time is used to infer vortex molecule dynamics with decreasing temperature.

2. Experimental

The polycrystalline (Cu, C)-1223 multilayer cuprate superconductor samples with $T_c$ of 120K were synthesized using high-pressure technique, and the details are presented elsewhere [26]. For grain alignment, (Cu, C)-1223 was thoroughly ground, mixed with an epoxy resin in 1 : 1 weight ratio, introduced in a small cylindric container, and kept for 12 h in a high-magnetic field of 7 T. The X-ray diffraction pattern of an aligned (Cu, C)-1223 show only (00l) peaks, and is presented elsewhere [19]. A very small amount of impurity, namely, the IL compound CaCuO$_2$ was also detected.

The AC susceptibility response of (Cu, C)-1223 was measured using a Physical Properties Measurement System, PPMS Model 6000 (Quantum Design), in DC fields $\mu_0H_{DC}$ up to 14 T, with AC field amplitudes $h_{AC}$ between 50 mOe and 10 Oe and frequencies from 10 Hz to 10 kHz, both fields being parallel to the c-axis of crystals, i.e., perpendicular to the SC planes. It is important to stress here that the measured sample consists of a collection of (Cu, C)-1223 single crystals embedded into a cylindrical epoxy, and that all crystals having the c-axis parallel to the symmetry axis of the cylinder, and, under our experimental conditions, parallel to both applied DC magnetic field and excitation AC magnetic field. Also, the presence of a small amount of CaCuO$_2$ does not have any effect on vortex.
dynamics, except for providing some additional pinning centers. To minimize demagnetization effects and the possible effects of Bean–Livingston and geometrical barriers, all the measurements were performed in field-cooling conditions. The frequency dependence of $\chi''(T)$ was measured from 120K-10 K at different $\mu_0H_{DC}$ up to 14 T and $h_{AC}$ for 0.1, 1 and 10 Oe.

3. Results and Discussion
The temperature dependence of out of phase AC susceptibility response was measured on aligned (Cu,C)-1223 crystallites in different DC fields and a typical response measured at 0.5 T is presented in figure 1; the whole range of data up to 6 T is presented elsewhere [19]. At zero DC field only one small dissipation peak was observed, as expected for the oriented crystallites dispersed in epoxy matrix. Along with conventional loss peak at high temperature, the second dissipation peak starts appearing at lower temperatures from applied DC field of 500 Oe, as shown in figure 1 for 5000 Oe, and merge together at 6 T onwards to 14 T.

![Figure 1. The temperature dependence of $\chi''(T)$ on aligned (Cu,C)-1223 crystallites under DC magnetic field of 0.5 T. The higher temperature peak is assigned to the dissipation due to the translational motion of the flux lines. The lower-temperature peak is assigned to the dissipation due to the possible rotational motion of the vortex molecule.](image)

The peak in $\chi''(T)$ at higher temperature is due to the conventional dissipation due to flux-flow and flux creep. It can also be called as loss due to the translational motion of conventional flux line lattice. Our crystallographic analysis and experimental conditions confirm that the possible origin of second dissipation peak at lower temperature is not due to [27,28]; two or more superconducting phases having different $T_c$’s and different critical current densities, inter-grain dissipation like in polycrystalline samples, the angle mismatch in applied field and $c$-axis of oriented sample (zero or 90°), ‘fish-tail’ effect or peak effect in $J_c$. We consider that the multiple components give the freedom of lower temperature dissipation peak. If we consider that the Abrikosov lattice is composed of ‘Vortex Molecule’ this can be explained in simple way [2]. In the multiband type of multicomponent superconductor, an $i$-soliton relaxes the restriction of quantum phase and allows the fractional vortex having fractional flux quanta [4]. An $i$-soliton confines two fractional vortices into one vortex molecule so as to make the total flux quantum, $\Phi_0$. We observe a vortex molecule composed of fractional flux with separate normal cores glued by an $i$-soliton wall. In the weak interband interaction limit, this vortex molecule becomes stable under the balance between magnetic energy gain and a cost to create $i$-soliton [2]. The hierarchy of phase dislocations observed in multiband type of multicomponent superconductor, constituting $i$-soliton, fractional vortex and vortex molecule is shown in figure 1 of ref. [2]. The distance of the normal core inside the vortex molecule is estimated to be, about 5 times, as long as the coherence length in the case of (Cu,C)-1223. The vortex molecule has an additional rotational freedom, which is vital for giving the lower temperature dissipation peak. The energy scale of the rotation is smaller than that of the translational deformation, but thermal fluctuations can not drive it well because of its heavy mass [2], if the sample thickness is greater than...
100 μm. A microscopic external force like $J_c$ induced by the AC magnetic field is suitable for driving it. No related phenomena are observed in the DC magnetic field measurement. A translational motion like that of flux flow phenomena has been detected on both of AC and DC fields, but an applied DC field cannot drive the rotational motion. An AC field can rotate the vortex-molecule just after the vortex molecule appears, and hence the lower temperature peak at a very low field just above $H_{c1}$. We can easily understand that the resonance condition can be achieved at a specific temperature and field which makes this peak.

Therefore, we concentrated on the temperature dependence of resonating condition. Figure 2 shows the frequency dependence of $\chi''(T)$ measured in low temperature peak range, for frequencies from 10 Hz to 10 kHz of 10 Oe $h_{AC}$ at applied DC field of 0.5 T. We can see that frequency dependence indeed give the clear resonating peaks, and that, resonating frequencies becomes lower with temperature. At lower temperatures, the penetration depth shrinks, also the vortex-molecule lattice becomes closely packed. Therefore vortex molecule interactions make the resonance frequency become lower. Two effects should be taken into account here, one, the potential on flux tube and second the critical fluctuation related to coherence length of orientational order.

![Figure 2](image1.png)

**Figure 2.** Frequency dependence of $\chi''(T)$ at DC field of 0.5 T, in the temperature range of lower temperature dissipation peak, giving resonance peak.

![Figure 3](image2.png)

**Figure 3.** Scaling of frequency dependence of $\chi''(T)$ at DC field of 0.5 T in the temperature range of second dissipation peak, using individual resonance frequencies.
For the applied frequency range of 10 Hz to 10 kHz, the frequency dependence of $\chi''(T)$ gives resonating peak only in the temperature range of 80 K to 90 K when $H_{DC}$ was 0.5 T. This temperature dependence of the frequency measurements are rescaled with individual resonance frequencies and presented in figure 3. All patterns merge to give the same profile. This is analogous to rotating spinors. Qualitatively this suggests that system will only attain equilibrium of vortex molecule rotation if it were left to relax for a time exceeding the longer relaxation time present in the frequency spectrum [29]. Also, it can be noticed that, the peak profile has a tail at lower $\omega\tau$. This can be explained with the fact that, as frequency of excitation is low, although this excitation energy is insufficient to rotate the vortex molecule, it lead twisting or cutting and reconnects of vortex molecules. Vortex molecule size is few nanometers (7.5 nm), but disorder induced by it, such as twisting, reaches about 0.1 $\mu$m. So the tail might be the dissipation due to such perturbations. By reducing the applied DC field, it can be brought back to the critical rotating state of vortex molecule.

The temperature dependence of average relaxation time, deduced from resonating frequency, is plotted in figure 4, and it can be noticed that the relaxation time increases almost exponentially as the temperature is lowered. This indicates that the rotation/twisting of vortex molecule undergo critical slowing (relaxation) down as the temperature of the system is lowered and then becomes vortex molecule glass. The relaxation time can be described by,

$$\tau = \tau_0 (T / T^* - 1)^{-x}$$

(1)

Where, ‘$x$’ is the product of dynamic critical exponent and exponent of vortex molecule correlation length, $T^*$ is the finite critical temperature. The solid line in the figure 4 shows the least square fit of the data of $\tau$ vs $T$, that yields the parameters, $\tau_0 = 0.0053$ sec, $T^* = 40.9$ K and $x = 22.8$. As, $\log(\tau) = \log(\tau_0) - x \log(T/T^* - 1)$, plot of $\log(\tau)$ vs $-\log(T/T^* - 1)$ is straight line as show in inset of figure 4. The exponent ‘$x$’ of about 15 is estimated for Rb$_2$Cu$_{0.782}$Co$_{0.218}$F$_4$ [29], under the assumption of critical slowing down in this rotating spin glass system. The higher value of exponent in our experiment is attributed to the presence of vortex molecule rotation and twisting states.

By considering the various magnetic flux densities and the orientation domains of vortex molecule, we exclusively present, the phase diagram of Vortex molecule lattice in multilayer cuprate superconductor, as a separate article [30].

4. Conclusions

The out-of phase susceptibility measurements $\chi''(T)$ on preferred oriented (Cu,C)-1223 crystallites gives second dissipation peak due to the multi-component nature of these multilayer superconductor. The origin of second dissipation peak is explained on the basis of vortex-molecule model.
frequency dependence of $\chi''(T)$ suggest that this second peak is due to the combination of vortex molecule rotation and twisting, which undergoes critical slowing down with decrease in temperature, with the equilibrium finite critical temperature of 40.93 K at 0.5 T $H_{DC}$.

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Because of a problem with the Digital-Analog converter of the superconducting magnet controller, the magnitude of the DC applied field was indicated as two times larger than the real value. Therefore the magnitude of the applied field is actually half of the described value. The conclusion is not altered by this correction.