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Title : AC-Susceptibility Study on Vortex-Molecule Lattice in Supermultilayer Cuprate HgBa₂Ca_{n-1}Cu_nO_{2n+2+δ} (n=14)

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Abstract

We investigated that in HgBa₂Ca_{n-1}Cu_nO_{2n+2+δ} [Hg-12(n-1)n], for n≥6, T_c of about 105 K, vortex melting lines remains constant and independent of number of inner plane CuO₂ layers. However the vortex dynamics of the supermultilayer system could be crucially different from that of single or double layer cuprates. Therefore we measured the fundamental and third harmonic AC susceptibility responses to see the vortex dynamics and vortex matter phase diagram of Hg-12(n-1)n (n=14), the higher member of super-

multilayered cuprate superconductors. The Lindemann type vortex melting line was estimated using onset of third harmonic susceptibility (χ_3), at very low h_{AC} (50 mOe), and represented by 3D Ginzburg Landau rescaling. Frequency dependence of out of phase susceptibility $\chi''(T)$ measured at lower fields of 500 and 1000 Oe, for 10 Hz to 10 kHz, showed resonance behaviour in the temperature region 55-68 K, suggesting rotational and translational creep dynamics of vortex molecule, even below usual melting line. We present schematic vortex phase diagram, considering the response of vortex dynamics to applied AC frequencies, taking into account the crossover temperatures obtained from critical slowing down of spin glass model and extended thermal activated model.

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Keywords : Hg-based superconductors; Vortex lattices; AC susceptibility

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1. Introduction

The homologous series of $\text{HgBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+2+\delta}$ [Hg-12(n-1)n], consist of HgBa_2O_y charge reservoir layer (CRL) and $\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n}$ infinite layers (IL), where 'n' is number of CuO_2 planes between two CRL which provides carriers to CuO_2 planes. For $n \geq 3$, it include crystallographically non-equivalent CuO_2 planes in a unit cell, outer planes (OP) with pyramidal (five) oxygen coordination and inner planes (IP) with square (four) oxygen coordination, with inhomogeneous charge distribution between OP and IP. Recently we investigated that, in Hg-12(n-1)n system, the T_c increases up to 133K at $n=3$ and decreases to 108K at $n=5$ and it remains almost constant about 105 K for $n \geq 6$ [1,2]. The samples with $n=3-5$ and $n \geq 6$, are termed as multilayer and super-multilayer cuprate superconductors, respectively. Hg-12(13)(14), $n=14$, is a higher member of super-multilayer series with $T_c = 105$ K, prepared by high pressure synthesis [1]. It was also interestingly observed that, the vortex melting lines of Hg-supermultilayer samples, $n \geq 6$, are practically the same. This vortex melting lines independent of number of IP CuO_2 plane in super-multilayer case is explained on the fact that, pancake vortices on the optimally doped OP CuO_2 layers across CRL are strongly Josephson coupled while pancake vortices on the (n-2) IP CuO_2 chain layers between CRL are magnetically coupled, which are long range weak interactions. Hence pancake vortices on OP along CRL forms *pancake vortex molecule* and the vortex cores passing through pancakes on OP and IP forms *pancake-vortex molecule lattices* [3]. The pancake molecule glass (PMG) melts into pancake molecule liquid (PML) state upon increasing the applied magnetic field or temperature. Therefore these features of constant T_c and different coupling forces on pancake vortex molecule lattice in the long crystallographic units of new Hg-super-multilayer cuprates makes it interesting to study vortex dynamics in these systems.

AC-susceptibility measurement is a unique technique to study vortex dynamics, where onset of third harmonics (χ_3) gives a vortex melting line, and frequency dependence of $\chi''(T)$ gives information on vortex mobility, flux diffusion, nature of pinning and various transitions in mixed vortex matter, and relaxation mechanism using dynamic critical exponents [4-7]. We carried AC-susceptibility measurements on grain aligned Hg-12(13)(14) samples in field cooled conditions with applied DC and AC fields in crystallographic c -direction, enabling to see dynamics on pancake vortex molecules. The frequency dependent results are analyzed by considering ‘spin glass model of critical slowing down’ and ‘extended thermal activation model’.

2. Experimental

The polycrystalline $\text{HgBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+2+\delta}$ [Hg-12(n-1)n] ($n=14$) super-multilayer cuprate superconductor samples with T_c of 105 K were synthesized, with nominal composition, using high-pressure technique, and the details are presented elsewhere [1]. For grain alignment, Hg-12(13)(14) was thoroughly ground, mixed with an epoxy resin in 1 : 3 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 Hg-12(13)(14) show only $(00l)$ peaks. It also shows the presence of $(00l)$ peaks of Hg-12(n-1)n phases for $n = 6$ to 16.

The AC susceptibility response of Hg-12(13)(14) was measured using a Physical Properties Measurement System, PPMS Model 6000 (Quantum Design), in DC fields $\mu_0 H_{DC}$ up to 6 T, with AC field amplitudes h_{AC} of 50 mOe for determining vortex melting line with the onset temperature of χ_3 as $T_m(K)$ and corresponding DC field as $B_m(T)$, both fields being parallel to the c -axis of crystals, i.e., perpendicular to the SC planes. The detail procedure on experimental determination of vortex melting line using the onset of third harmonic susceptibility χ_3 can be found elsewhere [8,9]. It is important to note here that the measured

sample consists of a collection of Hg-12(13)(14) 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. All the measurements were performed in field-cooling conditions. Field dependence of various single frequency dependent $\chi''(T)$ was measured upto 0.8 T. The frequency dependence of $\chi''(T)$ was measured from 130K-10 K at low $\mu_0 H_{DC}$ fields of 500 and 1000 Oe and h_{AC} for 10 Oe.

3. Results and Discussion

The important factor of vortex dynamics is melting line, $B_m(T)$ or $T_m(B)$, which separates vortex glass (VG) and vortex liquid (VL) phases [10]. [Figure 1](#) shows the vortex melting line for the grain aligned sample of nominal composition Hg-12(13)(14) measured at 50 mOe h_{AC} (insuring this way very low probing current density). Analysis of the melting transition in the framework of an anisotropic three dimensional (3D) Ginzburg-Landau rescaling approach [11] gives the following temperature dependence of the melting field :

$$B_m(T) = \frac{C^2 c_L^4 \Phi_0^5}{(k_B T)^2 \lambda_{ab}^4 \gamma [\cos^2(\alpha) + \gamma^2 \sin^2(\alpha)]^{1/2}} \quad (1)$$

Where, C is a constant ($C \approx 1/4\pi^2$), c_L is the empirical Lindemann parameter (taken in the following to be 0.15), Φ_0 is the magnetic flux quantum, λ_{ab} is the penetration depth along the superconducting ab-plane (CuO_2), γ is the anisotropy factor, and α is the angle between the magnetic field lines and the ab-plane. For our preferentially oriented samples and with our experimental setup, $\alpha = 90^\circ$. The experimental melting line is fitted well using the temperature dependence of the in plane penetration depth given by the ‘two fluid model’ $\lambda_{ab}(T) = \lambda_{ab}(0)[1-(T/T_c)^4]^{-1/2}$. The one parameter fit with $\lambda_{ab}(0) = 140$ nm gives the anisotropy

values of 74. The absence of 3D-2D crossover like transition in the melting line suggest that there is a sufficient magnetic coupling in the pancake vortices of IP CuO_2 layers, which couples pancake vortex molecules on OP and pancakes on IP. The vortex melting line of Hg-12(13)(14) is experimentally same as that of super-multilayered samples up to $n = 10$.

According to vortex-glass model of Fisher et al [10,12], the vortex lines, passing through pancake vortices, interacting with random pinning undergoes a transition from vortex liquid to a truly superconducting vortex glass state with no mobile vortices. While transforming in vortex glass phase, the vortices are frozen at random, the order present in the superconducting phase is analogous to the one observed in spin glass. Also the Hg-12(13)(14) with non equivalent CuO_2 planes is a double layer structure OP-CRL-OP, would be realized as two-component superconductor like Stanford Al- AlO_x -Al bi-layer system [13]. In multiband multicomponent (Cu,C)-1223, there exists a ‘Vortex molecule’, formed by the two fractional vortices at two components and bound by a intercomponent phase difference soliton (*i*-soliton) [14]. Vortex molecule is a non-axis-symmetric or non-Abrikosov type vortex, which has rotational degree of freedom [14]. A microscopic external force like J_c induced by the AC magnetic field is suitable for driving the non-circular core vortices into translational and rotational motion. Since rotational energy scale is minimum than translational motion, when the applied AC frequency matches the rotation frequency, it should give the resonance peak [14].

We measured the frequency dependence of out of phase susceptibility, with higher h_{AC} of 10 Oe, as a function of temperature and applied field. [Figure 2](#) shows the frequency dependence at 0.1 T H_{DC} . The inset of [figure 2](#) shows the temperature dependence of single frequency sweeps showing the dissipation peak shifts to higher temperature with increasing frequency. Similarly resonating frequency (ω_0) shifts to lower temperature. The peak in frequency dependence is also a measure of AC field penetrating the bulk of the oriented

crystallites and interaction of this AC field causing possible translational-rotational mix dissipation of vortex molecule. Figure 3 shows the normalized scaling of frequency dependence curves, and indeed all curves follow the similar resonating like dissipation pattern. This is analogous to rotating spinors. Qualitatively this suggests that system will only attain equilibrium of vortex molecule rotation dynamics if it were left to relax for a time exceeding the longer relaxation time present in the frequency spectrum [15]. The temperature dependence of average relaxation time ($1/\omega_0$), deduced from resonating frequency, for 0.1 and 0.05 T H_{DC} fields, is plotted in figure 4, the solid lines are the fit with the relaxation function given by Eq. 2.

$$\tau = \tau_0 (T / T^* - 1)^{-x} \quad (2)$$

Where, ‘ $x = zv$ ’ is the product of dynamic critical exponent and exponent of vortex molecule correlation length, T^* is the finite critical temperature, where upon critical slowing down system attains glass state. The least square fit of the data of τ vs T , yields the parameters, (i) $T^* = 38.75$ K, $\tau_0 = 60$ μ s and $x = 16.97$ for 500 Oe, and (ii) $T^* = 34.66$ K, $\tau_0 = 70$ μ s and $x = 11.45$ for 1000 Oe. The exponent ‘ x ’ of about 15 for $Rb_2Cu_{0.782}Co_{0.218}F_4$ [15] and 11.5 for $Fe_{0.5}Mn_{0.5}TiO_3$ [16], are estimated under the assumption of critical slowing down in this rotating spin glass system. Our results are in agreement with the model.

In AC susceptibility at mixed vortex state, by varying the AC amplitude, h_{AC} , the J_c varies and by varying frequency, resistivity can be varied which arises due to flux dynamics (translational and rotational) in the sample [4]. Therefore by considering the thermal activation energy and effective pinning energy, Liu et al [17] modified the Fishers vortex glass model with extended thermal activation model, and gives the scaling behavior between the peak temperature and frequency as follows;

$$T_p = T_g + (T_c - T_g) \left(\frac{f}{f_0} \right)^{1/s} \quad (3)$$

Where, T_g is glass transition temperature, and $s = v(z+2-d)$ is product of static and dynamic critical exponent. The effective pinning energy U_{eff} is given by,

$$U_{eff} = k_B T \frac{T_c - T}{T_c - T_g} \quad (4)$$

Figure 5 shows the plot T_p vs frequency of applied AC of 10 Oe for 0.1 and 0.05 T H_{DC} . The solid lines show the data fit with Eq. 3, and the resultant fitting parameters are, (i) $T_g = 41.56K$ and $s = 15.32$ for 0.05 T, and (ii) $T_g = 34.62 K$ and $s = 11.7$ for 0.1 T. The f_0 is characteristic frequency of order of 10^9 Hz. The inset of figure 5 shows U_{eff}/k_B vs T relation and $k_B T$ fit with Eq. 4 showing how pinning force gradually act on the dynamic vortex molecule core which then slows down and attains stable vortex molecule glass state when $k_B T = T_g$.

Figure 6 shows the plot of T^* and T_g obtained from both the models, plotted in the actual melting line (ML) of Hg-12(13)(14). The characteristic temperature T^* and T_g obtained with both the models are same, and common line drawn from these point is referred as T_{CR} . This is not a thermodynamic phase change boundary, but a ‘crossover’ temperature which separates mixed state in two regions (I) and (II), based on the vortex mobility and pinning strength. Above T_{CR} (region-I), we have seen high dissipation in frequency dependence of AC-susceptibility with high vortex mobility of the form of rotational and translational motion, when excited with the higher AC amplitudes. It can also be resembled as region of low pinning strength regime (I) (hence high mobility). Below T_{CR} (region-II), vortex mobility undergoes critical slowing down as pinning energy is strong, which further takes system in glassy state, when vortex mobility completely stop. Therefore, T_{CR} is the cross-over from high vortex mobility to low vortex mobility crossover. Similar weak to strong pinning crossover has been seen in Hg-1201 [18]. Similar Also coupling in pancake vortices on CuO_2

IP might have increased at this low temperature. Since the applied AC amplitude was as high as 10 Oe, this line can also be close to the irreversibility line.

4. Conclusions

Vortex melting line of Hg-12(n-1)n (n=14) suggest that it follows the pancake vortex molecule model having considerable magnetic coupling energy in the CuO₂ inner planes, similar to other Hg-supermultilayer samples with n=6-10. The resonating scaling in frequency dependence of out of phase susceptibility depicts the possible rotational dissipation of the vortex molecule. The frequency dependent vortex dynamics at higher AC excitations follows the ‘critical slowing down of spin glass model’ and ‘extended thermal activation model’; and give rise to the crossover temperatures, which separates vortex molecule dynamic region in B-T phase with vortex mobility and pinning strength. The long IP CuO₂ stacks give more flexibility to vortex molecule dynamics.

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Figure Captions

Fig. 1. Experimentally determined vortex melting line of Hg-12(13)(14) measured at 50 mOe of h_{AC} and 10kHz frequency. Experimental data fits with the $\lambda_{ab}(T)$ from two-fluid model, and *one parameter best fit* gives anisotropy value, $\gamma = 74.07$.

Fig. 2. Frequency dependence of $\chi''(T)$ measured at $H_{DC} = 0.1$ T. Inset shows the trend of temperature dependence of out of phase susceptibility χ'' at different single frequencies.

Fig. 3. The normalized scaling of frequency dependence of $\chi''(T)$ at DC field of 0.1 T. For the applied frequency range of 10 Hz to 10kHz the resonance peak is observed in $\chi''(T)$ in the temperature range of 55 K to 67 K.

Fig. 4. Average relaxation time τ vs T at the applied DC field of 500 and 1000 Oe. The solid line shows the fitting of the data by Eq. 2. The resulting fitting parameters are; (i) $T^* = 38.75K$, $\tau_0 = 60\mu s$ and $x = 16.97$ for 0.05T, and (ii) $T^* = 34.66K$, $\tau_0 = 70 \mu s$ and $x = 11.45$ for 0.1T.

Fig. 5. Frequency dependence of T_p of AC response for Hg-12(13)(14) sample at 0.1 T and 0.05 T. The solid line is a fit with Eq. 3. The fitting parameters are, (i) $T_g = 41.56$ K and $s = 15.32$ for 0.05 T, and (ii) $T_g = 34.62$ K and $s = 11.7$ for 0.1 T. The f_0 is characteristic frequency of order of 10^9 Hz. Inset shows the temperature dependence of effective pinning energy for Hg-12(13)(14) sample at 0.1 T. The solid line is a fit with Eq. 4. And the dotted line is the thermal activation energy in unit of K.

Fig. 6. Schematic B-T Phase diagram of Hg-12(13)(14). Only T_m and $T_{CR} (=T^*, T_g)$ are to the scale and other lines are schematic.

VPP-7_ISS2007_Shivagan_Fig. 1

VPP-7_ISS2007_Shivagan_Fig. 2

VPP-7_ISS2007_Shivagan_Fig. 3

VPP-7_ISS2007_Shivagan_Fig. 4

VPP-7_ISS2007_Shivagan_Fig. 5

VPP-7_ISS2007_Shivagan_Fig. 6