

Mechanical measurement of vortex phase transition in $\text{YBa}_2\text{Cu}_3\text{O}_{7+\delta}$

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Using the vibrating-reed technique, the internal friction (IF) Q^{-1} and modulus as functions of temperature were measured for a $\text{YBa}_2\text{Cu}_3\text{O}_{7+\delta}$ single crystal at low applied magnetic fields up to 0.5 T. The frequency dependence of the IF related to the vortex is reported. No obvious shift of the IF peak position can be observed by varying the measurement frequency ranging from 10^2 to 10^3 Hz but the IF peak height satisfies a power law $Q^{-1} \propto \omega^{-n}$ ($1 < n < 2$). This is speculated to originate from a phase transition associated with the melting or a glass transition of the vortex lattice. These results provide a fundamental measurement of the phase diagram of vortices in high- T_c superconductors. [S0163-1829(97)03030-0]

I. INTRODUCTION

The mixed state of high- T_c superconductors has a very complicated phase diagram. Among them, the most salient feature of the phase diagram is the theoretically proposed existence of a vortex-lattice melting line which separates a vortex-liquid phase from a vortex-solid phase. The nature of the different vortex phases and the thermodynamic transitions between them are of fundamental and applicable interest, and are subjects of substantial recent theoretical and experimental efforts.¹⁻⁵ It is generally accepted that below the mean-field H_{c2} line, a vortex-liquid state persists down to a glass-transition line associated with a second-order transition to a vortex-glass or Bose glass state or a melting line associated with a first-order transition into an Abrikosov vortex lattice, which depends on the type and strength of the disorder. In highly disordered systems involving large amounts of point defects, the vortex liquid is predicted to undergo a second-order phase transition into a vortex-glass state. However, in very clean systems a first-order transition from a vortex liquid to an Abrikosov vortex lattice is predicted. Recently, Safar *et al.*⁶ reported a measurement of current voltage that the presence of first-order melting is an intrinsic, thermodynamic transition, not created by the pinning potential.

In order to investigate the physical properties of different phases of the vortex, many techniques, such as measurement of the magnetization, the susceptibility, the critical current, and the mechanical response of the vortex were developed. The first mechanical measurements developed were the audio-frequency measurements using vibrating reeds⁷ and a high- Q mechanical oscillator.⁸ These techniques involve measuring the resonance frequency and dissipation in an oscillator made of a superconductor, or which has a superconductor attached. A feature common to all the mechanical measurements of vortex motion in superconductor materials is that a dissipation peak and corresponding decreases in the stiffness of vortex line are seen below T_c when temperature is swept at constant magnetic field. There has been a contro-

versy as to the origin of this feature. While some authors considered it to be evidence for flux-lattice melting^{2,3} or some transition to a glassy state of the flux structure,⁹ others interpret the measurements in terms of thermally activated flux diffusion.¹⁰ As discussed quantitatively for vibrating superconductors by Brandt *et al.*,¹¹ the dissipation peak arises when flux diffuses through the sample in a characteristic time that matches the inverse of the oscillation frequency, without invoking a phase transition of any sort. The characteristic time is related to the geometry of the sample and the vortex pinning. For a given field, because the diffusivity changes with the temperature, this condition will be met once during a temperature sweep for each particular diffusion mode. Because the dissipation behavior is dependent on the flux diffusivity which is, in turn, dependent on the activation energy, it is a thermally activated process and the internal friction (IF) peak temperature will be frequency dependent and shifts to higher temperature with increasing measurement frequency. However, as proposed by Gammel *et al.*, "melting of the vortex" is responsible for the occurrence of the dissipation peak. Due to the characteristic of phase transition for the "melting" process, the position of the IF peak should be frequency independent but the magnitude of IF should be dependent on measurement frequency. Therefore, detecting the frequency dependence of the IF peak temperature and the magnitude of IF becomes crucial for determining the actual mechanism of the vortex dissipation at lower frequency range.

In this paper, we reported on mechanical measurement of dissipation associated with vortex motion by using the vibrating-reed technique in $\text{YBa}_2\text{Cu}_3\text{O}_{7+\delta}$ single crystals. Within our experimental errors, no obvious shift of IF peak position is observed in the audio-frequency ranging from 10^2 to 10^3 Hz. However, the inversely proportional dependence of IF Q^{-1} on ω^n ($1 < n < 2$), in which ω is the measurement frequency, is observed in our measurement. According to our experimental results, the IF peak is thought to be originated from a "melting" phase transformation of vortex, no matter whether first order or second order.

II. EXPERIMENT BASIC

The sample used in our experiment is a twinned $\text{YBa}_2\text{Cu}_3\text{O}_{7+\delta}$ single crystal. The critical temperature is 92.5 K. The vibrating reed consists of a platelet clamped at one end, while two electrodes near the free end serve to drive and detect its motion electrostatically. The platelet is a Si crystal sputtered with a metal film, with the superconducting sample glued near the free end. The configuration can be seen elsewhere in detail.¹⁰ The magnetic field is a transverse field up to 0.5 T.

It must be noted that our experiment includes a composite

$$\omega = \frac{2\pi}{\sqrt{\rho_h/M_h\kappa_h^2}} \left(\frac{\beta_1}{2l_h} \right)^2 \sqrt{1 + \eta_0(\rho_h, \rho_s, \kappa_s, \kappa_h, d_s, d_h) + \alpha \left(\frac{4l_s}{l_h} + \frac{4\sqrt{2}\Phi''''(l_h)l_s}{l_h} \right) \frac{M_s}{M_h}}, \quad (1)$$

where $\rho_h(\rho_s)$ is the density of host reed (superconductor), $M_h(M_s)$ is the modulus of the host reed (superconductor), and $\kappa_h(\kappa_s)$ is the radius of gyration of the host reed (superconductor). β_1 is a constant = 1.015, η_0 is a parameter which is independent of M_h and M_s , $\Phi''''(l_h)$ is the fourth-order derivation of the wave function of the reed at the free end, and α is given in the Appendix:

$$Q^{-1} = Q_h^{-1} + \xi \frac{l_s}{l_h} \cdot \frac{M_s}{M_h} Q_s^{-1}, \quad (2)$$

where Q_h^{-1} is the IF of host reed and Q_s^{-1} is the IF of superconductor, ξ is a shape parameter. Owing to the low IF of the Si host, about 10^{-5} – 10^{-6} , Q^{-1} is nearly proportional to Q_s^{-1} . Modifications (1) and (2) are used in our following discussion.

III. EXPERIMENT AND DISCUSSION

Figure 1 is our main result. As displayed in Fig. 1, the IF peak position does not shift obviously with varying the measurement frequency in the range $10^2 \leq \omega \leq 10^3$ Hz within our experiment errors (about 0.5 K). The frequency dependence of the dissipation of the vibrating superconductor at constant temperature and dc magnetic field can be used to test the mode of the vortex dynamics. However, it is impossible to achieve the vibrating-reed measurements in a frequency window which spans several decades. Gupta *et al.*¹³ fabricated vibrating reeds with the same thickness d but different length l . The resonance frequency of these reeds varies according to $\omega \propto d/l^2$. Thus one can expect a frequency-independent dissipation since only the product ωl^2 enters the expression for the dissipation within the frame of thermally activated diffusion.¹⁰ However, in our measurements, we regulate the vibrating frequency by varying the length of the Si host with the superconducting sample's length unchanged. Therefore, the frequency-dependent IF peak should be found if the flux motion is a thermally activated diffusion. The independence of the IF peak temperature on frequency in our measurement is not consistent with the prediction of thermally activated

diffusion theory. The independence observed here can also be explained, however, simply as the narrow frequency window spans in our experiment. Unambiguous evidence for the phase transition, in this case, is the frequency dependence of the IF magnitude displayed in Fig. 1.

The power law of the IF peak height $Q^{-1} \propto \omega^{-1}$ is obviously shown in Fig. 1. Using Eq. (2), and keeping in mind that resonance frequency ω of the composite reed is nearly inversely proportional to l_h^2 , it can be obtained that the IF of superconductor Q_s^{-1} approximately satisfies the modified power law $Q_s^{-1} \propto \omega^{-1.5}$, which indicates a characteristic of phase transformation.

The internal friction peak Q^{-1} developing near the first-order phase transition at low-frequency range has been stud-

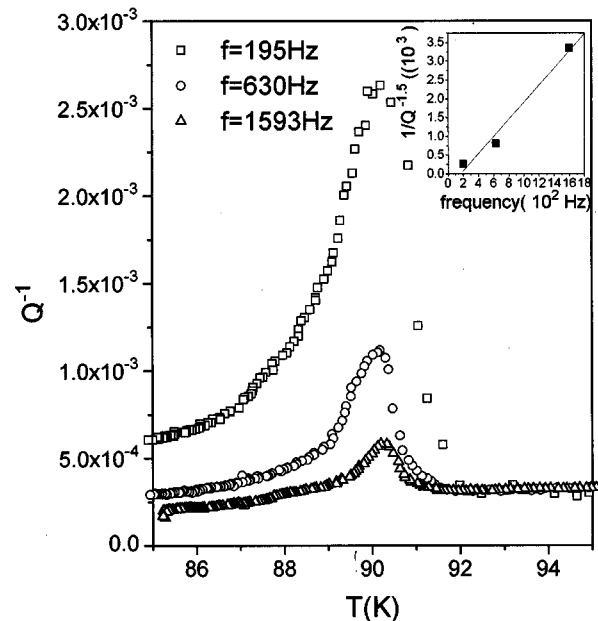


FIG. 1. Temperature dependence of the IF Q^{-1} for a different measurement frequency of the mechanical oscillator with the $\text{YBa}_2\text{Cu}_3\text{O}_{7+\delta}$ single crystal attached. The inset shows the IF peak height as a function of $1/\omega$.

ied extensively and is known to consist of two components:^{14,15}

(i) Transient component Q_t^{-1} , the internal friction measured on cooling or heating at finite rate ($\dot{T} \neq 0$) is in many cases proportional to \dot{T} and inversely proportional to frequency ω . This means the dissipation is proportional to the transformation amount of vortex per cycle.

(ii) Stationary or equilibrium component Q_e^{-1} , the isothermal measurement on step cooling or heating ($\dot{T} = 0$) is independent of frequency in the low-frequency range ($\omega < \text{kHz}$) which exhibits the characteristic of a static hysteresis loss.

Transient components are to a considerable extent characterized by the real structure of the heterophase system, i.e., by the formation and volume of a new phase. The following expression is obtained for the transient component Q_t^{-1} of internal friction:^{16–18}

$$Q_t^{-1} = K G \dot{m} / \omega, \quad (3)$$

where K is a constant, G is the shear modulus, ω is the cyclic oscillation frequency, and \dot{m} is the relative phase transition rate. This theory predicts the height of the peak Q^{-1} varies inversely proportional to the measurement frequency ω with $\dot{m} = dm/dT \cdot \dot{T} = \text{const}$. For the equilibrium component Q_e^{-1} , the mechanism of the stress-induced movement of the interface^{15,16,19,20} has been widely accepted. Most experimental results show that Q_e^{-1} is independent of measurement frequency in the low-frequency range ($\omega < \text{kHz}$).^{15,21} However, in the case of the vortex melting transition here, it is speculated that the equilibrium component Q_e^{-1} originates from the viscous dissipation of vortex. In the vicinity of the melting from a Abrikosov vortex lattice,^{22,23} the viscosity appears to diverge at this temperature. Although an actual viscosity divergence associated with continuous melting appears in the theory of melting dynamics of classical particles in two dimensions,²⁴ such a divergence of the viscosity would also result from the very long entanglement relaxation times estimated in Ref. 25. In this scenario, barriers to flux cutting would increase $L_{\text{eff}}(T)$ with increasing temperature until $L_{\text{eff}}(T) > L$, at which point the viscosity $\eta(L) \approx u_0 \tau_{\text{coll}}(L)$ is effectively infinite. Thus the viscous dissipation of the vortex at the point of melting temperature plays an important role in the dissipation of the vortex. In the vortex melting transition, the isothermal transformation would continue while $\dot{T} = 0$, so Q_e^{-1} would be attributed to the dissipation caused by the viscous movement of the vortex just melting. As indicated in Ref. 26, the viscous dissipation is inversely proportional to the measurement frequency ω . Furthermore, the amount of the vortex which leads to this dissipation is equal to the amount of vortex just transforming from vortex solid to vortex liquid per cycle, and in turns proportional to $1/\omega$. Thus, it can be obtained that the equilibrium dissipation Q_e^{-1} is inversely proportional to the square of the frequency ω .

Around the second-order phase transition, the ultrasonic attenuation α is proportional to ω^2 at high frequency ($> \text{MHz}$) for the relaxation of the order parameter. So, the λ -type peak appearing in the MHz range cannot be detected in the kHz range. Otherwise, owing to the absence of the

nucleation process and coexistence of new and parent phases, all theories consider that there is no internal friction peak related to the second-order phase transition in the lower frequency range. But Wang *et al.*²⁰ reported that there exists an IF peak in the kHz range around the second-order phase transition temperature T_c in the $\text{La}_{1-x}\text{Nd}_x\text{P}_5\text{O}_{14}$ (LNPP) crystal. The height of this peak decreases with increasing frequency. They attributed it to the fluctuation of a new phase around the phase-transition temperature and the appearance of a preferential state of the transient new phase caused by an external field which destroys the equilibrium state. Although it does not have a stable nucleus, owing to the fluctuation near T_c , the new phase may appear instantaneously in some small region of the parent phase. When no external field is applied, the various domains with different orientation appear or disappear with equal probability. However, since the new phase is accompanied by spontaneous strain, the applied stress will destroy the equilibrium state and those favorable domains will appear more frequently than others, which results in nonelastic strain and then IF. They also deduced the IF expression related to the second-order phase transition as a form:

$$Q_{2\text{nd}}^{-1} \propto dF/dt \cdot \omega^{-n} \quad \text{with } n > 0, \quad (4)$$

where n is a constant, dependent on material and characteristic of phase transition. dF/dt is the quantity of fluctuation per unit time, which exhibits a peak around T_c , and thus leads to the IF peak near T_c . In the present context, the fluctuation of the transition can be interpreted as a measure of the typical spatial extent of the domains of a vortex liquid which are present in the vortex solid. The amount of these domains controls the mechanical dissipation behavior of the system.²⁷ As analyzed above, it is considered that the inverse proportion of the IF Q^{-1} to frequency ω and the frequency-independent IF peak position, measured at finite rate \dot{T} , is a demonstration for the phase transformation of the vortex, no matter whether first order or second order.

Figure 2 gives the IF versus temperature in different vibrating amplitudes. It can be seen the IF is amplitude dependent (hysteric loss) below the IF peak temperature and amplitude independent (viscous loss) above the IF peak temperature. The same results were also obtained by other authors and were attributed to the couple-decouple of grains in samples. We thought that this may originate from a stress-induced depinning before the flux-line-lattice (FLL) is thoroughly melting. Real superconductors exhibit always many pins per flux line and not many flux lines per pin. Therefore, an ideally rigid infinite FLL cannot be pinned at all by dense randomly positioned pins since all pinning forces average to zero when there is no correlation between the position of flux and pins.²⁸ By extending the collective-pinning theory of Larkin and Ovchinnikov to finite temperature,²⁹ Feigel'man and Vinokur³⁰ have argued that the thermal fluctuation of the vortex position $\langle u^2 \rangle \propto T$ increases the effective range $r_p \approx \xi$ of the pinning forces to $r_p \approx (\xi^2 + \langle u^2 \rangle)^{1/2}$. This thermal motion smooths the effective pinning potential and in turn reduces the collective-pinning force. Below the temperature of the IF peak, effective pinning potential U exists and the flux line can see a pinning potential so that the amplitude-dependent dissipation as a result of stress-induced depinning

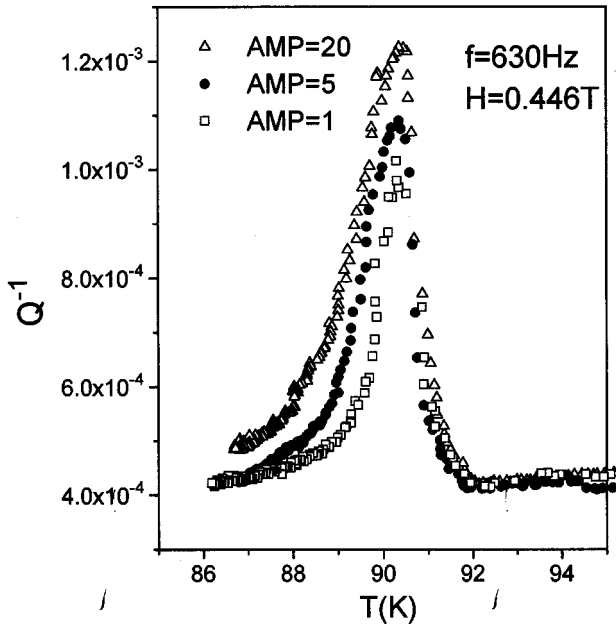


FIG. 2. Temperature dependence of IF Q^{-1} at different vibrational amplitudes.

can be measured, however, above the IF peak temperature, the effective pinning potential is smoothed by thermal fluctuation and the flux line cannot see a pinning potential, and thus can move viscously which leads to an amplitude-independent dissipation.

The IF is thought to be written in the following form:

$$Q_s^{-1} = Q_T^{-1} + Q_e^{-1} + Q_D^{-1}, \quad (5)$$

where Q_T^{-1} is the transient component of the IF associated with phase transition which is proportional to $1/\omega$, Q_e^{-1} is the stationary component of the IF which is proportional to $1/\omega^2$, Q_D^{-1} is the IF as a result of stress-induced depinning of the flux line. Q_D^{-1} which resulted from the motion of a single flux line is much smaller than Q_T^{-1} and Q_e^{-1} , then Q_s^{-1} is mainly attributed to the IF associated with the phase transformation of the vortex, thus proportional to ω^{-n} in which $1 < n < 2$.

This interpretation is supported by the modulus measurement as shown in Fig. 3. Figure 3 gives the temperature dependence of the resonance frequency at a magnetic field 0.433 T. Using Eq. (1), it is obtained that the modulus defect $\Delta M_s/M_s$ of the superconductor has the following form:

$$\frac{\Delta M_s}{M_s} = \alpha(4 + 4\sqrt{2}\Phi_{(l_h)}''') \frac{l_h}{l_s} \frac{2\Delta\omega}{\omega} \frac{M_h}{M_s}. \quad (6)$$

It is noted that except for l_h , all other parameters in Eq. (6) remain unchanged with varying the frequency and keeps in mind that frequency ω is proportional to $1/l_h^2$, thus it can be obtained $\Delta M_s/M_s \propto \Delta\omega/\omega^{3/2}$. As shown in Fig. 3, the total frequency variation $\Delta\omega$ is independent of frequency ω , thus the modulus defect is consequently also inversely proportional to $1/\omega^{3/2}$.

According to the thermally activated relaxation theory (Debye relaxation), the modulus defect is expressed as follows:²⁶

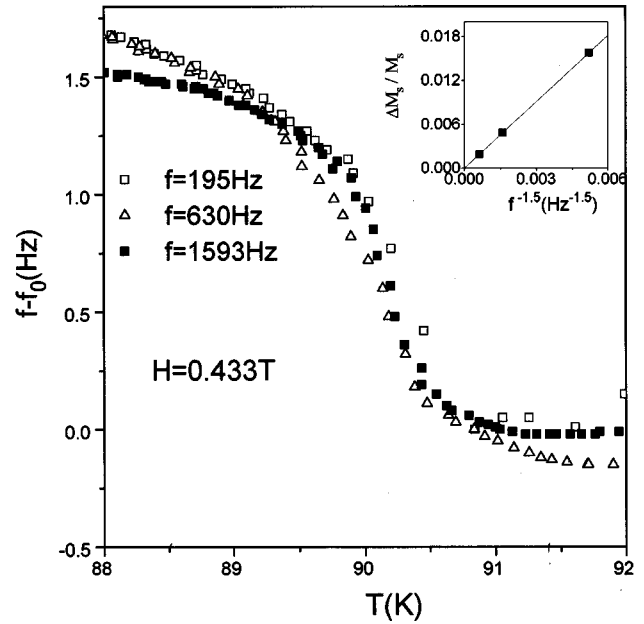


FIG. 3. Temperature dependence of the frequency shift for different measurement frequency. The inset shows the jump of modulus defect $\Delta M/M$ as a function of $1/\omega$.

$$\frac{\Delta M}{M} = \Delta_M \frac{1}{1 + (\omega\tau)^2}, \quad (7)$$

where Δ_M is the relaxation strength, ω is the frequency, and τ is the relaxation time satisfying the Arrhenius equation: $\tau = \tau_0 e^{U/kT}$. In the case of $\omega\tau \gg 1$, the modulus defect $\Delta M/M \rightarrow 0$; in the other case of $\omega\tau \ll 1$, the modulus defect $\Delta M/M \rightarrow \Delta_M$. Therefore the jump of modulus defect $\Delta M/M$ becomes equal to Δ_M which is independent of the frequency. Our results show the jump of modulus defect $\Delta M/M$ is inversely proportional to frequency which cannot be explained in the framework of thermally activated diffusion. As known to all, the nonelastic strain can be expressed in the form of: $\varepsilon'' = \varepsilon_1'' + i\varepsilon_2''$, in which the imaginary part ε_2'' of the nonelastic strain ε'' causes internal friction, and the real part ε_1'' of the nonelastic strain ε'' results in a modulus defect. Owing to the results that the IF and modulus defect are both inversely proportional to the frequency, the nonelastic strain thus satisfies the law $\varepsilon'' \propto \omega^{-1.5}$, which is consistent with the phase-transformation theory that the nonelastic strain is proportional to the amount of transformation from parent phase to new phase per oscillation cycle, in turn inversely proportional to frequency ω .^{15,20} In the vortex phase-transformation scenario, the IF results from the difference of the amount between favorable and unfavorable variants transforming from vortex-solid state to vortex-liquid state and the corresponding viscous dissipation caused by the vortex just melting.

How to obtain the phase diagram has concentrated extensive interest on experiment.³¹ Numerous experimental results reported the frequency dependence of the ‘‘transition line’’ (TRL) when measured by ac susceptibility.³² The temperature of the peak in the out-of-phase first harmonic χ_{ac} response shifts with varying the frequency, which indicates it is not actually a measurement of the TRL. Deak *et al.*³³ reported the frequency-independent of the vortex-glass phase

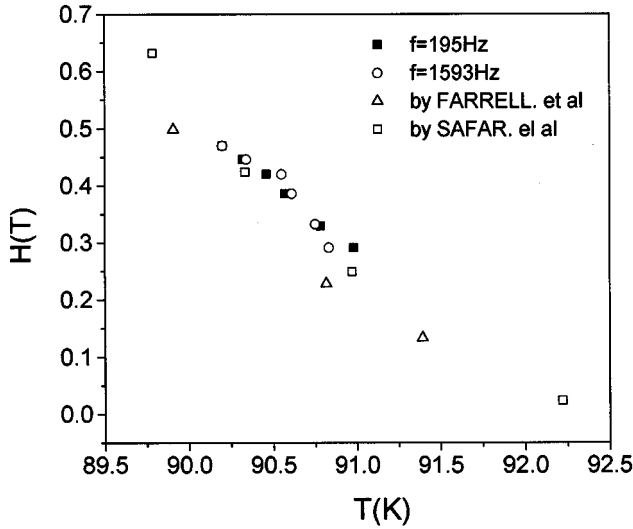


FIG. 4. The magnetic phase diagram for our sample. For comparison, the data of Farrell *et al.* and Safar *et al.* are also shown.

transition in a $\text{YBa}_2\text{Cu}_3\text{O}_{7+\delta}$ thin film in the frequency range from 10^{-1} to 10^5 Hz by resistance measurement. They thought their results provide a fundamental measurement of TRL. Because our measurement of the mechanical response of the vortex shows a characteristic of ‘‘phase transformation,’’ we think our results also provide a method to determine the phase diagram of the vortex. Figure 4 displays the phase diagram in which the transition temperature is determined from the temperature of the IF peak. According to the melting theory,³⁴ an approximated power law $B_m(T) = B_0(1 - T/T_c)^n$ is expected, where $n < 2$. The best fit for our result leads to $n = 1.9$ which is consistent with the theory. For comparison, the data of Farrell *et al.*³⁵ and Safar *et al.*³⁶ are also shown in Fig. 4. To within our errors, our phase line appears to agree with that obtained by Farrell *et al.* as measured with a low-frequency ($\omega < 1$ Hz) mechanical oscillator and by Safar *et al.* as measured by static current-voltage ($\omega = 0$ Hz) on a Y-Ba-Cu-O single crystal. The coinciding data show that mechanical dissipation behavior is determined by the vortex phase transformation.

IV. CONCLUSION

In summary, we present results about the frequency dependence of mechanical dissipation of a vortex in twinned $\text{YBa}_2\text{Cu}_3\text{O}_{7+\delta}$. The inverse proportion of the IF peak to frequency ω^n ($1 < n < 2$) indicates that the mechanical dissipation peak originates from a phase transition between the vortex-liquid state and the vortex-solid state, no matter whether first or second order. As previously proposed by Gammel *et al.*, the mechanical dissipation peak measured by the high- Q oscillator is the evidence for flux melting; our measurements in the audio-frequency range by using the vibrating-reed technique also give a demonstration for the phase transition of the vortex as further proof of their proposition. Furthermore, how to determine whether a first-order lattice-liquid melting or a second-order glass-liquid melting occurs, needs further investigation

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APPENDIX: DERIVATION OF EQS. (1) AND (2):

The derivation of Eqs. (1) and (2) is similar to the procedure used in Ref. 12. The derivation is given for the slab geometry $0 < x < l_h$. It is shown that the superconductor is glued at the free end of the host reed and its typical dimension is far smaller than that of the host reed. The composite reed is a nonuniform system and we give its density ρ , cross section area s , and the radius of gyration of cross section κ as a function of x along the length of the reed:

$$\rho(x) = \begin{cases} \rho_h(1 + g(x)), & l_h - l_s < x < l_h \\ \rho_h, & \text{others,} \end{cases} \quad (\text{A1})$$

where

$$g(x) = \frac{d_s w_s}{d_h w_h + d_s w_s} \frac{\rho_s - \rho_h}{\rho_h}$$

in which w is the width, d is the thickness, and ρ is the density, the subscript h and s indicate the host and superconductor, respectively. Also,

$$s(x) = \begin{cases} d_h w_h (1 + \alpha(x)), & l_h - l_s < x < l_h, \\ d_h w_h & \text{others,} \end{cases} \quad (\text{A2})$$

where

$$\alpha(x) = \frac{d_s w_s}{d_h w_h}$$

and

$$\kappa(x) = \begin{cases} \kappa_s(1 + \sigma(x)), & l_h - l_s < x < l_h \\ \kappa_s, & \text{others,} \end{cases} \quad (\text{A3})$$

where $\sigma(x) = (M_s/M_h)\alpha + \beta$ in which M_s and M_h indicate the modulus of the host reed and superconductor, respectively. $\alpha = (1/s) \int_{\xi}^{d_s + \xi} z^2 dz$ and $\beta = (1/s) \int_{-d_h + \xi}^{\xi} z^2 dz$ in which

$$\xi = \frac{M_s d_h^2 - M_h d_s^2}{2(d_s M_s + d_h M_h)}.$$

Due to the equation given in Ref. 12, the resonance frequency ω is

$$\omega = \frac{2\pi}{\sqrt{\rho_h/M_h \kappa_h^2}} \left(\frac{\beta_1}{2l_h} \right)^2 \sqrt{1 + \eta}, \quad (\text{A4})$$

in which $\beta_1 = 1.057$ and η is given as the following form:

$$\eta = \frac{2}{l_h} \int_0^{l_h} \Phi_1 \left\{ (\sigma - g)\Phi_1 + \left(\frac{l_h}{\pi\beta_1} \right)^4 \left[2 \frac{d(\alpha + \sigma)}{dx} \cdot \frac{d^3\Phi_1}{dx^3} + \frac{d^2(\alpha + \sigma)}{dx^2} \frac{d^2\Phi_1}{dx^2} \right] \right\} dx. \quad (\text{A5})$$

Inserting Eqs. (A1)–(A3) into Eq. (A5), it can be obtained that

$$\eta = 1 + \eta_0 + \alpha \left(\frac{4l_s}{l_h} + \frac{4\sqrt{2}\Phi_{(l_h)}''''l_s}{l_h} \right) \frac{M_s}{M_h}, \quad (\text{A6})$$

where η_0 is a constant independent of M_s and M_h , and $\Phi_{(l_h)}''''$ is the fourth-order derivation of the wave function of the reed at the free end.

Then we obtain the resonance frequency:

$$\omega = \frac{2\pi}{\sqrt{\rho_h/M_h\kappa_h^2}} \left(\frac{\beta_1}{2l_h} \right)^2 \times \sqrt{1 + \eta_0 + \alpha \left(\frac{4l_s}{l_h} + \frac{4\sqrt{2}\Phi_{(l_h)}''''l_s}{l_h} \right) \frac{M_s}{M_h}}. \quad (\text{A7})$$

Following the same procedure as above, we obtain the IF in the following form:

$$Q^{-1} = Q_h^{-1} + \xi \frac{l_s}{l_h} \cdot \frac{M_s}{M_h} Q_s^{-1}. \quad (\text{A8})$$

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