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Softening of the flux-line structure in $La_{1.8}Sr_{0.2}CuO_4$ measured by a vibrating reed

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Vibrating-reed measurements in $La_{1.8}Sr_{0.2}CuO_4$ in a magnetic field show that the flux-line structure (FLS) becomes uncoupled from the material at a temperature T_H of 23 K, much below the critical temperature of the sample, which is 32 K. The behavior is similar to that found in $Bi_2Sr_2CaCu_2O_8$ but shows some significant differences, the most important being that T_H changes little with magnetic field throughout the measured field range ($0.01\ T \leq H \leq 1\ T$). For low fields, T_H is below the reversibility line of the magnetization. This seems to imply that the reversibility line and the line at which the FLS soften have to be considered as distinct physical phenomena. It is argued that neither "flux-lattice melting" nor giant flux creep considered separately can account for the data in a wholly satisfactory manner.

The flux-line-structure (FLS) in high-T_c superconductors shows unusual behavior when compared to that of low-T_c materials. In particular, a change of regime in mechanical oscillator studies has been seen in $Bi_2Sr_2CaCu_2O_8$ and $YBa_2Cu_3O_7$ by different groups,[1-7] and several theoretical papers[8-11] have appeared, predicting amorphous, liquid, or hexatic phases of the flux-line structure, but in general the interpretation of the results is still controversial.

In this paper we report vibrating-reed measurements carried out in $La_{1.8}Sr_{0.2}CuO_4$ that also show a peculiar behavior of the FLS, similar in some ways to that observed in other high-T_c superconductors, but with some significant differences. Recent measurements of Higgins *et al.*[7] at ultrasound frequencies also show features related to our measurements at lower (700 Hz) frequencies.

The vibrating reed experiments were performed on ceramic $La_{1.8}Sr_{0.2}CuO_4$ prepared in air at 1200 °C for 24 h following the method reported previously.[12] A $15 \times 4 \times 0.17$ mm^3 slab was cut from the original pellet of the material using a diamond saw, and it was annealed afterwards in a pure atmosphere of oxygen at 600 °C for 12 h. X-ray analysis gave a single-phase structure, and from microstructural studies we found a medium grain size: $d \sim 90$ μm. The sample density was 6.5 g/cm^3.

The reed, clamped by one end, was excited and detected electrostatically, and the oscillation was maintained at resonance by means of a phase-locked loop. In the experiments the sample was cooled at 4.2 K in zero field; then the field was applied and the temperature increased. In each run, both the resonance frequency f and the vibration amplitude A were measured as a function of the temperature. The field was removed only when T_c was exceeded. By using a temperature controller, each experimental point could be taken at fixed temperature. Because of the relatively low values of the magnetic fields we have used, no corrections for magnetoresistence were made to our germanium thermometer.

By measuring resonance curves at different temperatures, absolute values of the internal friction Q^{-1} were obtained; however, in practice, we have calculated the dissipation using the relation $Q^{-1}(T) = A(4\ \mathrm{K})/A(T) \times Q^{-1}(4\ \mathrm{K})$.

In Fig. 1 we show the temperature dependence of the relative change in resonance frequency $\Delta f/f$ and the dissipation Q^{-1} of the reed for different magnetic fields. In both graphs, only the magnetic contribution is shown, while the intrinsic response of the material has been eliminated by substracting the zero-field background. As will be discussed later, the dynamical response of the FLS changes with the amplitude of vibration of the reed, so that to make a meaningful comparison all the measurements shown in Fig. 1 correspond to a constant excitation voltage of 10 V for the reed.

It can be seen that the resonance frequency decreases monotonically when the temperature is increased until a temperature T_H of around 23 K is reached. At temperatures higher than T_H no changes in resonance frequency can be detected, apart from those due to the background from the material. On the other hand, the dissipation also becomes equal to that of the material alone above a slightly higher temperature T_H^* as can be seen in the figure. Both these temperatures are well below the superconducting critical temperature T_c of 32 K as measured by the onset of flux expulsion in a superconducting quantum interference device (SQUID).

The points T_H, at which the hardening of the FLS starts, are plotted in Fig. 2. As can be seen, the slope of this curve is very steep and the line cannot be distinguished from a vertical line within our experimental resolution. However, it would be compatible with the measured slope[13,14] of H_{c2}, as can be seen by looking at the dashed line, which was drawn with a slope[14] of 2 T/K and whose meaning will be discussed later. This behavior is in contrast to that observed[2,3] in $Bi_2Sr_2CaCu_2O_8$, where there was a positive curvature in the field versus T_H line, and an easily observable variation of T_H with magnetic field. There is also a difference with $YBa_2Cu_3O_7$ be-

cause in this compound the "melting line" measured by Gammel *et al.*,[1] which is closely related to our T_H, has a slope close to that of H_{c2}, but it extrapolates near T_c in zero field. In our case, however, the extrapolation is to a reduced temperature $t_H = T_H/T_c$ of 0.7, well below T_c.

We have made measurements at different excitation voltages, and the results are plotted in Fig. 3. Two regimes are distinguished by looking at the curves for $\Delta f/f$. At higher temperatures, between approximately 15 and 23 K (i.e., T_H), the shift in frequency is amplitude independent and the curves for different amplitudes superpose, while at lower temperatures the curves separate, and with higher excitation amplitude a smaller change in frequency is observed. The hardening temperature T_H, however, is independent of the excitation voltage, which again is in contrast to the behavior observed in $Bi_2Sr_2CaCu_2O_8$, where T_H shifted to lower temperatures when the excitation voltage was increased.[3] In both experiments the amplitudes of vibration of the reed are comparable, and we estimate the displacement at the end of the reed to be around 0.5 μm for the highest excitation voltages. In Ref. 3 it was suggested that the motion

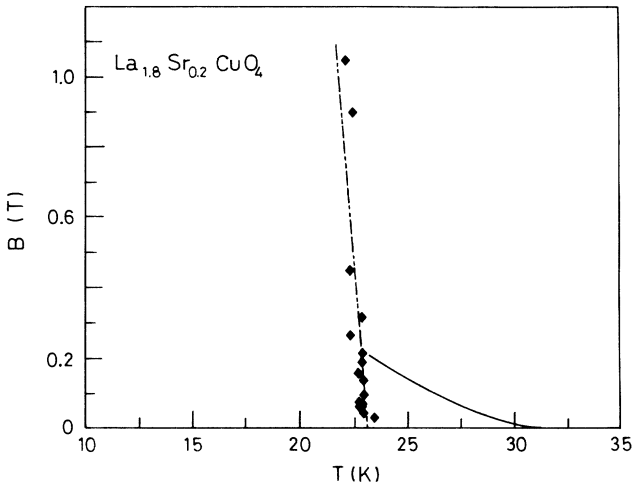

FIG. 2. Comparison of the reversibility line and the softening of the FLS measured with the vibrating reed. The temperature at which the SQUID signal becomes reversible for a given magnetic field defines the reversibility line, while the points corresponding to the softening of the FLS are defined at the temperature where the frequency response of the reed becomes equal to the response at zero applied field. Solid line: reversibility line measured by Civale (Ref. 18), dash-dotted line: fit to the melting line of Ref. 11, and diamonds: softening of the FLS (this work).

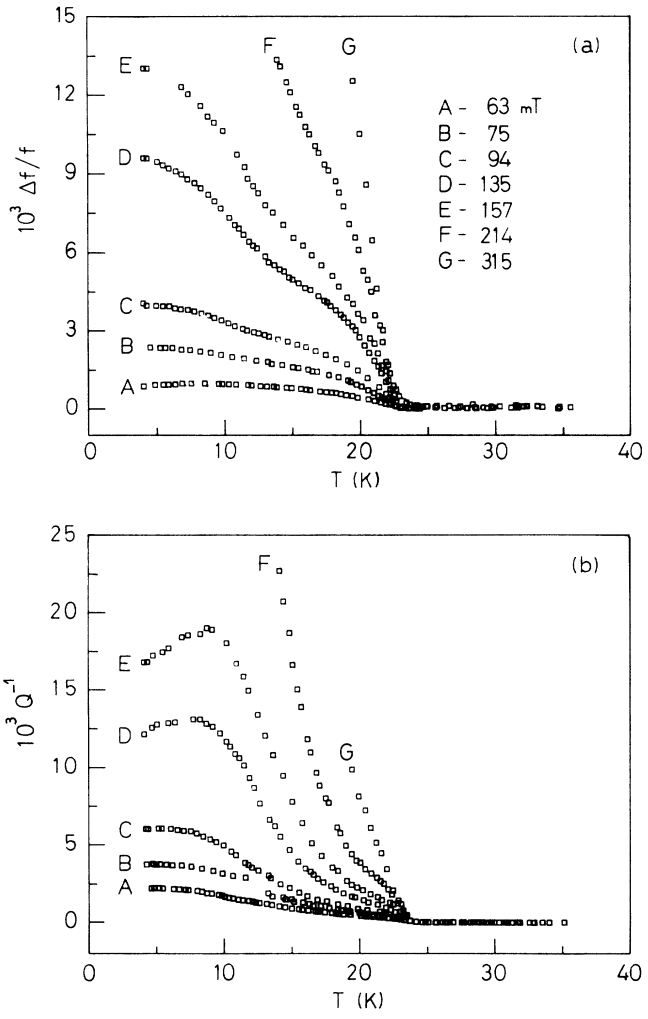

FIG. 1. (a) Relative change in resonance frequency of the vibrating reed as a function of temperature for different applied magnetic fields. (b) The attenuation Q^{-1} measured for the same applied fields. In both graphs the background due to the material has been substracted so that only the extra stiffness and dissipation due to the flux lines are represented. It can be seen that above 23 K there is no signal from the FLS.

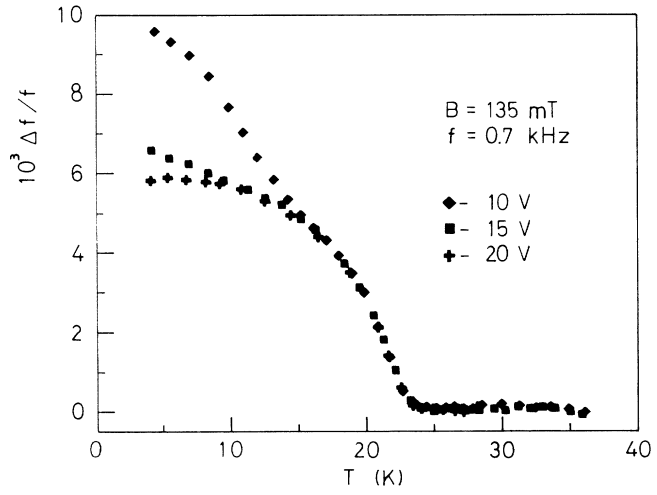

FIG. 3. The relative change of resonance frequency as a function of temperature for different driving voltages and at a constant magnetic field of 135 mT. It can be seen that the curves superpose between 15 and 23 K, and the point where the FLS softens does not shift with the amplitude of vibration of the reed. The background due to the material has also been substracted from these curves.

of the reed could favor melting or entanglement of the vortices in $Bi_2Sr_2CaCu_2O_8$, but this does not seem to be the case in $La_{1.8}Sr_{0.2}CuO_4$. The dependence of the amplitude of vibration with applied voltage, which was observed[3] to increase linearly in $Bi_2Sr_2CaCu_2O_8$ at low excitation and to increase faster at higher excitation, is difficult to fit to one simple behavior in the present experiments, and no linear region can be defined.

Higgins *et al.*[7] have observed that there is a change in their ultrasound velocity measurements in $La_{1.8}Sr_{0.2}CuO_4$, at a temperature $T_{CR}(H)$, which they attribute to a change in pinning regime. We also observe an anomaly when measuring at different amplitudes, and the point at which the frequency curves for different amplitudes separate could be related to $T_{CR}(H)$. A quantitative comparison is difficult, however, because the conditions in the experiments are different and the measured field ranges scarcely overlap.

In the paper of Higgins *et al.*[7] at significantly higher frequencies a superposition of the sound velocity with and without a field was reported at temperatures above 20 K, very close to our T_H of 23 K. The published curve taken at 6.5 T, a field much higher than those measured here, indeed shows a superposition up to 20 K, and this would be compatible with our results and a slope of around 2 T/K (close to that of H_{c2}), for the change of T_H with magnetic field. If the superposition is not fortuitous, this would imply that T_H is frequency independent to a very high degree.

It has been assumed, when the anomalous response of the FLS was first found in mechanical oscillator studies,[1] that a melting of the vortex lattice would imply zero critical current because the vortices would be free to move above the melting temperature $T_M(H)$. This would in turn imply that the sample magnetization should be reversible above $T_M(H)$, since the irreversibility is the consequence of the existence of a finite critical current. A "reversibility line" $T_R(H)$ was first reported by Müller *et al.*[15] in high-T_c materials, and one of the interpretations of this line was given by Yeshurun and Malozemoff[16] and Tinkham,[17] in terms of "giant flux creep." In the giant flux creep picture, the vortices above $T_R(H)$ are moving over the pinning barriers because of thermal activation, and the resultant critical current is zero. One then expects the melting line and the reversibility line to coincide, apart from a possible shift due to the different frequencies involved, since in a thermally activated process the temperature at which changes in response are seen depend on the measurement frequency. In $Bi_2Sr_2CaCu_2O_8$ it was observed,[2,3] that the reversibility line measured with a SQUID occurs at higher temperatures than $T_H(H)$ for low magnetic fields. This relative shift of $T_H(H)$ and $T_R(H)$ is in the opposite direction than one would expect in a thermally activated process, since the vibrating-reed measurements are at a higher frequency than those of the SQUID by about four orders of magnitude.

We have therefore compared our results in $La_{1.8}Sr_{0.2}CuO_4$ with the data of Civale[18] and Civale *et al.*,[19] which have measured the reversibility line in a sample very similar to ours using a SQUID magnetometer. They find that $T_R(H)$ obeys the equation

$$T_R(H) = H^{2/3}/\alpha - T^* \ , \tag{1}$$

where $\alpha = 21\ \mathrm{Oe}^{2/3}\mathrm{K}^{-1}$ and T^*=30.9 K. The fit is good up to the measured field of 0.2 T and measurements of H. Pastoriza[20] confirmed that Eq. (1) holds for the sample reported here. We have plotted this equation in Fig. 2, and it can be seen that for fields below 0.2 T $T_R(H)$ is indeed above $T_H(H)$, but that above this field the opposite seems to be true, although we do not have data for $T_R(H)$ above 0.2 T and one should be careful in extending the validity of Eq. (1) outside the measured range [in $Bi_2Sr_2CaCu_2O_8$ for example, it has been found that for high fields the dependence of $T_R(H)$ can change from an $H^{2/3}$ to an exponential field dependence[21]]. The difference in the melting and reversibility curves is hard to explain in terms of critical currents or pinning forces, and we do not have a plausible explanation at the moment, although it seems to indicate that the melting and the irreversibility are distinct physical phenomena. The conclusion seems to be that a definitive explanation has to take into account both the thermal activation over the pinning barriers (and that pinning is of fundamental importance) and the possible changes in the rigidity of the flux-line structure. This is a complicated problem, and much depends on the density and strength of the pinning centers involved. For example, if the FLS looses its rigidity in the presence of very dense pinning centers each vortex could find a defect in which to pin, and the result would be a strongly pinned amorphous FLS that would *enhance* the response of a vibrating reed. Sparse pinning centers, even if they were strong, would pin only a few vortices and leave the rest free to move, therefore weakening the response of the reed when the FLS becomes fluid.

We have compared our measurements with a theory of the melting of the FLS proposed by Markiewicz[11] based on earlier work on two-dimensional systems.[22–24] The formula obtained in Ref. 11 for the melting field $b_M = H/H_{c2}$ is

$$b_M = 1 - \gamma_M[t/(1-t)^4]^{1/2} \tag{2}$$

with $\gamma_M^2 = 2(4\pi)^3\sqrt{3}\lambda_o^2 k_B T_c/(d\alpha\Phi^2)$, where λ_o is the London penetration depth at zero temperature, d is the thickness of the film, α is a constant of order unity, and Φ is the flux quantum. We have plotted this equation in Fig. 2 as a dashed line taking $H_{c2} \simeq 2(T_c - T)$ T/K. There is a difficulty in defining the meaning of d, since this is the thickness of the film where two-dimensional melting occurs, and in our case the field is parallel to the long dimension of the sample ($\sim$1 cm). In the paper by Markiewicz,[11] when the thickness of the film was used in the formulas, the resultant λ_0 was an order of magnitude greater than the measured value, so there are grounds to believe that the value of d may differ from

that of the thickness. We have accordingly left d as a free parameter, to be found in the following way: extrapolating the experimental data, t_M at zero field was obtained, γ_M was then calculated by Eq. (2) and from the definition of γ_M and known superconducting parameters of the material, a value of d was obtained. If we take[18,19] λ_0=3000 Å and T_c=32 K, we obtain $d \simeq$65 Å. The value of d is of the order of the coherence length of the material (~40 Å), and this could perhaps mean that the vortex structure behaves as almost uncoupled layers of thickness comparable to the coherence length. This is a highly speculative hypothesis, however, and further work is needed to corroborate or deny it. The theory[11] does not include the effect of pinning or flux creep, and so may provide an incomplete description of the problem, as was discussed in the preceding paragraph. If we use the above-mentioned procedure for calculating d in $YBa_2Cu_3O_7$ or $Bi_2Sr_2CaCu_2O_8$ with the available data, we obtain results that are not easy to interpret and will be reported in the future.

In conclusion, we have shown that the unusual behavior of the FLS is not exclusive of "1-2-3" or bismuth-based superconductors but can be found in $La_{1.8}Sr_{0.2}CuO_4$ as well. We also observe a difference between the anomalies observed and the reversibility line, which seems to imply that they arise from different physical phenomena.

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