Ultrasonic Attenuation by the Vortex Lattice of High- T_c Superconductors

J. Pankert, G. Marbach, and A. Comberg

Philips GmbH Forschungslaboratorium Aachen, D-5100 Aachen, Federal Republic of Germany

P. Lemmens, P. Fröning, and S. Ewert

2. Physikalisches Institut, Rheinisch-Westfälische Technische Hochschule Aachen, D-5100 Aachen, Federal Republic of Germany

(Received 11 June 1990)

Measurements of the transverse sound attenuation and velocity in a ceramic $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ superconductor ($T_c = 110$ K) were performed with and without a magnetic field. Pronounced peaks in the field-induced attenuation and an increase of the velocity at low temperatures were observed. These effects can be attributed to the transition of a rigidly pinned flux-line lattice into a depinned state. A quantitative explanation is presented within the framework of thermally assisted flux flow and without any adjustable parameter.

PACS numbers: 74.30.Gn, 74.60.Ge, 74.70.Vy

The properties of the flux-line lattice (FLL) in the mixed state of high- T_c superconductors are among the most extensively studied aspects of high- T_c superconductivity. During the last two years, a great number of new phenomena have been observed including the most prominent effects such as "giant flux creep",¹ broadening of the resistive transition in a magnetic field,^{2,3} or frequency and field dependence of the ac susceptibility.^{4,5} While there is now a general agreement that flux motion is at the origin of all of these effects, the exact nature of the mixed state is still a matter of considerable debate. Many new ideas have been put forward in the literature, such as, for instance, FLL melting⁶⁻⁸ as a property of the FLL itself or vortex-glass phases as a consequence of pinning in conjunction with disorder.⁹⁻¹¹ Experimental evidence for both of these phenomena is discussed controversially¹²⁻¹⁷ and even the interesting crossover phenomena observed in the I-V characteristics of YBa₂Cu₃O₇ thin films¹⁷ cannot be considered as an ultimate proof for a nondiffusive motion of flux lines, as was recently pointed out by Esquinazi¹⁸ and Griessen.¹⁹

In spite of this intense research, the experimental access to these problems is rather limited. The most frequently used approach is the evaluation of the resistive transition in a magnetic field.^{3,17} Here, flux-line motion is studied in a stationary dissipative state where flux lines move from one side of the sample to the other. The resolution of these experiments requires current densities typically of the order of $10-10^5$ A/cm².¹⁷ A second approach consists of magnetization measurements either in the ac or dc mode.^{1,4} Here the interpretation is usually more difficult because of large inhomogeneities in the spatial distribution of flux lines. With the vibrating-reed measurements, a third approach has been introduced recently.^{14,15}

The main purpose of this Letter is to present ultrasonic attenuation as a novel means of access to the static and dynamic properties of the FLL. In ultrasound experiments, sound waves are coupled to the FLL via pinning. This coupling leads to a modified attenuation and dispersion of sound which allows us to extract detailed information about the mixed state. We shall demonstrate that this method has clear advantages over the above-mentioned approaches in several respects: First, ultrasonic measurements are real bulk measurements which implies that surface or grain-boundary effects are of minor importance. This is particularly interesting for granular samples in which resistive measurements cannot evaluate the flux-line dynamics because of grainboundary resistances. Second, once the flux lines have attained their equilibrium position, ultrasound waves will move them away from equilibrium by at most 10 nm which is the typical sound amplitude. With a magnetic field of 1 T and a wavelength of 200 μ m, this amounts to current densities of less then 0.1 A/cm^2 ; i.e., orders of magnitude smaller than in resistive measurements. Sound measurements are therefore best suited for the evaluation of the near-equilibrium dynamics of the FLL. Third, this method allows a direct measurement of the elastic constants of the FLL. This is important in anisotropic superconductors, where there is some controversy about the value of the tilt modulus C_{44} .^{6,8} Finally, we mention the possibility of varying the sound frequency over a broad range and of studying the different modes which could provide otherwise inaccessible information on anisotropies of the mixed state.

Here, we present results for Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O_y ceramic. The sample was prepared by a sol-gel process.²⁰ X-ray-diffraction analysis showed a single superconducting phase with $T_c \approx 110$ K (determined from ac-susceptibility measurement linset of Fig. 1(b)]). The density was $\rho_0 = 4.5$ g/cm³, i.e., 72% of the theoretical density. Flat, parallel faces were prepared on two ends of the sample (5×5×5 mm³). Ultrasonic waves were generated by LiNbO₃ transducers²¹⁻²³ with resonance frequencies of 3 and 10 MHz. The pulse-echo technique



FIG. 1. (a) Attenuation and (b) velocity of a 3-MHz transverse sound mode with and without a magnetic field $(\mathbf{k} || \mathbf{B})$ in the $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ high- T_c superconductor. Inset: The ac susceptibility.

and an automatic phase-comparison method were used. The absolute transverse sound velocity was 1.944×10^5 cm/s at 4 K which implies wavelengths of $\sim 200 \ \mu m$ at 10 MHz, much larger than the typical grain size ($\sim 10 \ \mu m$). The magnetic field was applied in the field-cooled mode in order to ensure that the FLL had established its equilibrium position. Magnetic fields were always much larger than the lower critical field such that the internal field B_0 was almost identical with the applied field. In this Letter, we restrict ourselves to the discussion of the transverse sound mode with the propagation direction parallel to the magnetic field which implies that the displacement vector **u** of the crystal is perpendicular to the flux lines.

In Fig. 1, the results for the attenuation and velocity are shown for the 3-MHz transverse sound mode in zero field and in fields of 3 and 5 T. The zero-field attenuation is a structureless function with barely no change at T_c . Such a behavior seems to be a common feature of all high- T_c superconductors.²¹⁻²⁴ When applying a magnetic field, the curves split at T_c and a reach a maximum excess attenuation at T = 62.9 K (5 T) and T = 69.1 K (3 T) [Fig. 2(a)]. At the same temperatures the velocity



FIG. 2. Difference attenuation $\Delta \alpha = \alpha(B) - \alpha(0)$ and relative velocity shift $[c_t(B) - c_t(0)]/c_t(0)$ of the data of Fig. 1. Solid curves are theoretical predictions without any adjustable parameter.

increases by a certain fraction. The corresponding differences from the zero-field velocity are displayed in Fig. 2(b). We have performed the same measurements at different frequencies (2.855, 3, 9.044, 10, and 10.65 MHz) and at fields ranging from 1.5 up to 6.5 T. The positions of the corresponding attenuation peaks are shown in Fig. 3.

We shall demonstrate that all of these effects can be interpreted in a quantitative way within the framework of thermally assisted flux flow (TAFF).^{25,26} In the context of sound attenuation, this was discussed in detail by one of the authors.²⁷ The basic assumptions and results are the following: Any small distortion of the FLL will relax into equilibrium. Considering only tilt distortions, this can be expressed by a relaxation equation for the displacement vector $\mathbf{v} = (v_x, v_y)$ of the FLL (with $\mathbf{B} = B\mathbf{e}_z$),

$$\dot{\mathbf{v}} = \Gamma C_{44} \partial_z^2 \mathbf{v} \,, \tag{1}$$

where $C_{44} = \beta B_0^2/4\pi$ is the tilt modulus⁸ and β is a factor to correct for the empty space in a porous sample. In our experiment $\beta = \rho_0/\rho_0^{\text{theor}} = 0.72$. Equation (1) is valid for wavelengths larger than the London penetration



FIG. 3. Temperatures of the attenuation maxima of the transverse sound mode as a function of frequency and magnetic field (\bullet , 1.5 T; \Box , \bullet , 3 T; \circ , \bullet , 5 T; Δ , \bullet , 6.5 T). Solid curves are theoretical predictions, adjusted at the 10-MHz results (solid symbols).

depths⁸ and only in the immediate vicinity of the equilibrium state of the FLL. It is easy to show that (1) is identical to the diffusion equation of the magnetic field studied in TAFF.^{25,26} Hence the diffusion constant is nothing but the dc resistivity ρ ,²⁷

$$\rho = (4\pi/c^2) C_{44} \Gamma = \bar{\rho} \exp[-U(B)/T], \qquad (2)$$

where the last equation in (2) represents the empirical Arrhenius form of the resistivity as discussed in Ref. 3. U(B) is a temperature-independent activation energy and $\bar{\rho}$ a constant. In the presence of sound waves one has to study the combined motion of the crystal lattice and the FLL which for the transverse sound mode is described by²⁷

$$\ddot{\mathbf{u}} - c_t^2 \partial_z^2 \mathbf{u} + (1/\rho_0) \Gamma^{-1} (\dot{\mathbf{u}} - \dot{\mathbf{v}}) = 0, \qquad (3)$$

$$-C_{44}\partial_z^2 \mathbf{v} + \Gamma^{-1}(\dot{\mathbf{v}} - \dot{\mathbf{u}}) = 0, \qquad (4)$$

where c_t is the sound velocity and where, for simplicity, a background damping has been ignored in (3). The crystal lattice and the FLL are coupled by the viscous drag $\sim \Gamma^{-1}$ caused by the relative motion of **u** and **v**. The dispersion relation of (3) and (4) reveals for the excess attenuation $\Delta \alpha$, and for the relative velocity shift $\Delta c_t/c_t$.²⁷

$$\Delta \alpha = \frac{\omega^2}{2\rho_0 c_l^3} \frac{C_{44}^2 \Gamma k^2}{\omega^2 + (C_{44} \Gamma k^2)^2},$$
 (5)

$$\frac{\Delta c_t}{c_t} = \frac{1}{2\rho_0 c_t^2} \frac{C_{44}\omega^2}{\omega^2 + (C_{44}\Gamma k^2)^2},$$
 (6)

with $\omega/2\pi$ being the frequency and k the wave vector. Inserting the expression (2) for Γ into (5) and (6), we find an attenuation peak at $T = U(B)/\ln(c^2\bar{\rho}k^2/4\pi\omega)$ and a step in the velocity at the same temperature. This is qualitatively in perfect agreement with our experimental results in Fig. 1. For a quantitative comparison we need to know the resistivity ρ , i.e., the values of $\bar{\rho}$ and U(B). Since we are not aware of any such measurement in the Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O_y phase, we have to adopt a somewhat different strategy. For $\bar{\rho}$ we choose as a reasonable guess the value of the Bi_{2.2}Sr₂Ca_{0.8}Cu₂O_{8+ δ} phase, i.e., $\bar{\rho}$ =0.1 Ω cm.³ Since this quantity enters our results only logarithmically, this choice is certainly not very critical. Next, we determine the activation energy U(B) from the position of the attenuation maxima of the 10-MHz measurements (see Fig. 3). The resulting values of U(B) are 643 K (1.5 T), 548 K (3 T), 510 K (5 T), and 495 K (6.5 T). They are surprisingly close to the corresponding values of the Bi_{2.2}Sr₂Ca_{0.8}Cu₂O_{8+ δ} phase.³

With $\bar{\rho}$ and U(B) fixed, we are now left with no adjustable parameter for the explanation of the 3-MHz data in Fig. 2. The resulting theoretical prediction is shown by the solid curves in Fig. 2. The agreement between theory and experiment is remarkable both in height, shape, and position of the attenuation peak as well as the velocity shift. The only significant deviation occurs in the low-temperature regime, where the attenuation stays at a temperature-independent level. This is probably due to hysteretic losses which also occur in vibrating-reed experiments.^{14,16} In Fig. 3, we have plotted the positions of the attenuation peaks of the other measurements not shown in Fig. 2, and find almost perfect agreement with the predicted positions (solid curves).

This excellent agreement between theory and experiment excludes other possible explanations (such as, for instance, FLL melting¹²) of the attenuation peak. We therefore conclude from these results that for sufficiently small driving forces, flux-line motion is mainly diffusive in character. In order to substantiate this point, we have reduced the power of the ultrasound by a factor of 25 and find almost no difference in our results. This implies that nonlinear contributions to the attenuation are negligible. As a further result we find that the tilt modulus C_{44} is not reduced by the anisotropy or the granularity of the superconductor. Otherwise, the step height in Δc_t and the height of $\Delta \alpha$ would have to be reduced as well. We consider this as a direct proof of Brandt's conjecture that the long-wavelength limit of C_{44} is solely given by the Maxwell energy density of the magnetic field.⁸ In fact, finite-wavelength corrections to C_{44} of anisotropic superconductors are given by⁷

$$C_{44}(k)/C_{44}(0) = 1/(1+k_{\perp}^2\lambda_c^2+k_{\parallel}^2\lambda_{ab}^2)$$

where λ_c and λ_{ab} are the London penetration depths in the c and ab directions and k_{\perp} , k_{\parallel} are the corresponding components of the wave vector. For $\lambda_{ab} \approx 1500$ Å, ²⁸ and an anisotropy $\lambda_c/\lambda_{ab} \approx 55$, ²⁹ we find $\lambda_c \approx 8 \ \mu m$ which is indeed very large, but nevertheless much smaller than all wavelengths used in our experiments.

In summary, we have shown that the ultrasound

method provides a new approach to the study of the mixed state of high- T_c superconductors. It can be used as a measure of the elastic and the pinning properties and as a sensitive probe of the activation energies in bulk samples.³⁰ An interesting extension of our work would be to go to much higher frequencies in single-crystalline samples (e.g., by Brillouin scattering). Then one can expect to observe directly the crossover to the shortwavelength limit, where the "pancake" structure of the vortices becomes relevant and where the superconductor is two dimensional in character.³¹ Lastly, it also opens the possibility to measure the large shear moduli predicted by Kogan and Campbell.^{32,33}

This work was supported by the Bundesministerium für Forschung und Technologie Grant No. FKZ: 13 N5487/5 and by the Deutsche Forschungsgemeinschaft, Sonderforschungsbereich No. 341.

¹Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).

²M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).

³T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **61**, 1662 (1988).

⁴A. P. Malozemoff, T. K. Worthington, Y. Yeshurun, F. Holtzberg, and P. Kes, Phys. Rev. B **38**, 7203 (1988).

⁵Th. M. Nieuwenhuizen and J. Pankert, Europhys. Lett. 11, 457 (1990).

⁶D. R. Nelson and H. S. Seung, Phys. Rev. B **39**, 9153 (1989).

⁷A. Houghton, R. A. Pelcovits, and A. Sudbø, Phys. Rev. B 40, 6763 (1989).

⁸E. H. Brandt, Phys. Rev. Lett. **63**, 1106 (1989).

⁹M. P. A. Fisher, Phys. Rev. Lett. 62, 1415 (1989).

¹⁰M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin, and V.

M. Vinokur, Phys. Rev. Lett. 63, 2303 (1989).

¹¹T. Nattermann, Phys. Rev. Lett. **64**, 2454 (1990).

¹²P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D.

J. Bishop, Phys. Rev. Lett. 61, 1666 (1988); R. N. Kleinman,

P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D. J. Bishop, Phys. Rev. Lett. **62**, 2331 (1989).

¹³E. H. Brandt, P. Esquinazi, and G. Weiss, Phys. Rev. Lett. **62**, 2330 (1989).

¹⁴A. Gupta, P. Esquinazi, H. F. Braun, and H.-W. Neumüller, Phys. Rev. Lett. **63**, 1869 (1989).

¹⁵A. Gupta, P. Esquinazi, H. F. Braun, W. Gerhäuser, H.-W. Neumüller, K. Heine, and J. Tenbrink, Europhys. Lett. **10**, 663 (1989).

¹⁶S. Gregory, C. T. Rogers, T. Venkatesan, X. D. Wu, A. Inam, and B. Dutta, Phys. Rev. Lett. **62**, 1548 (1989); **63**, 695 (1989); P. H. Kes, Phys. Rev. Lett. **63**, 694 (1989).

¹⁷R. H. Koch, V. Foglietti, W. J. Gallagher, G. Koren, A. Gupta, and M. P. A. Fisher, Phys. Rev. Lett. **63**, 1511 (1989). ¹⁸P. Esquinazi, Solid State Commun. **74**, 75 (1990).

¹⁹R. Griessen, Phys. Rev. Lett. **64**, 1674 (1990).

²⁰L. M. Fatter, D. A. Payne, T. A. Friedmann, W. H.

Wright, and D. M. Ginsberg, Br. Ceram. Proc. 41, 261 (1989). ²¹S. Ewert, S. Guo, P. Lemmens, F. Stellmach, J. Wynants,

G. Arlt, D. Bonnenberg, H. Kliem, A. Comberg, and H. Passing, Solid State Commun. 64, 1153 (1987).

²²P. Lemmens, F. Stellmach, J. Wynants, S. Ewert, A. Comberg, H. Passing, and G. Marbach, J. Less-Common Met. **151**, 153 (1989).

²³P. Lemmens, Ch. Hünnekes, M. Brakmann, S. Ewert, A. Comberg, and H. Passing, Physica (Amsterdam) **162-164C**, 452 (1989).

²⁴S. Bhattacharya, M. J. Higgins, D. C. Johnston, A. J. Jacobsen, J. P. Stokes, D. P. Goshorn, and J. T. Lewandowski, Phys. Rev. Lett. **60**, 1181 (1988).

²⁵P. H. Kes, J. Aarts, J. van den Berg, J. C. van der Beek, and J. A. Mydosh, Supercond. Sci. Technol. 1, 242 (1989).

²⁶E. H. Brandt, Z. Phys. B 80, 167 (1990).

²⁷J. Pankert, Physica (Amsterdam) 168C, 335 (1990).

²⁸Y. J. Uemura *et al.*, Physica (Amsterdam) **162-164C**, 857 (1989).

²⁹D. E. Farrell et al., Phys. Rev. Lett. 61, 2805 (1988).

³⁰P. Lemmens, P. Fröning, S. Ewert, J. Pankert, H. Passing, and A. Comberg, Physica (Amsterdam) **165-166B**, 1275 (1990).

³¹P. H. Kes, J. Aarts, V. M. Vinokur, and C. J. van der Beek, Phys. Rev. Lett. **64**, 1063 (1990).

³²V. G. Kogan and L. J. Campbell, Phys. Rev. Lett. **62**, 1552 (1989).

³³J. Pankert, Physica (Amsterdam) 165-166B, 1273 (1990).