## Pair-breaking description of the vortex-depinning critical field in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> thin films

A. F. Hebard, P. L. Gammel, C. E. Rice, and A. F. J. Levi AT&T Bell Laboratories, Murray Hill, New Jersey 07974 (Received 28 April 1989)

The penetration length  $\lambda$  of *c*-axis-oriented thin films of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> in perpendicular magnetic fields *H* up to 14 T is determined from complex impedance measurements at 1.25 kHz. Vortex core pinning determines the functional form of  $\lambda(T,H)$ . The observed nonlinear temperature dependence of the depinning critical field, identified by  $\lambda \rightarrow \infty$ , can be described within the context of a pair-breaking formalism in which a type-II clean-limit pair-breaking rate ( $\alpha H^{1/2}$ ) replaces the dirty-limit rate ( $\alpha H$ ).

The penetration length  $\lambda$ , which defines the length scale over which magnetic fields and currents vary, is one of the more important parameters used to characterize a superconductor. Knowledge of  $\lambda$  and its temperature dependence also provides fundamental information about the strength and type of superconducting coupling. The detailed temperature dependence of  $\lambda$  can be used, for example, to distinguish between weak- and strong-coupled BCS behavior.<sup>1</sup> For the high- $T_c$  superconductor YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, experimental results on sintered ceramics,<sup>2</sup> crystals,<sup>3,4</sup> and films<sup>5</sup> are in general agreement that the temperature dependence of  $\lambda$  is consistent with BCS predictions. In the thin-film experiments, a complex impedance is extracted from ac screening measurements and  $\lambda$  is calculated directly from the inductive component.<sup>6</sup> The presence of pinned vortices, induced by an external magnetic field H, makes an additional contribution to this impedance<sup>7</sup> and hence to  $\lambda$ . At high enough fields this vortex-induced contribution can dominate the electrical response and hence, strongly affect such properties as the transition temperature, the critical current, and the screening. Thus, an understanding of these effects is crucially important to an understanding of the promise and potential of the new high- $T_c$  superconductors.

In this Rapid Communication, we present measurements of the temperature- and field-dependent penetration length  $\lambda(T,H)$  of c-axis-oriented YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> films with the magnetic field H applied parallel to the c axis. The functional form of  $\lambda(T,H)$  at high fields  $(H \ge 1 T)$  is shown to be a consequence of strong vortex pinning. At fixed H, extrapolation of  $\lambda(T)$  to a temperature where  $\lambda \rightarrow \infty$  enables a determination of the vortex-depinning critical field  $H_{cp}(T)$  which delineates the boundary between the superconducting and resistive flux-flow phases. The temperature dependence of  $H_{cp}$  near  $T_c$  is not linear as is  $H_{c2}$  in type-II superconductors, but rather nonlinear with strong positive upward curvature. Such curvature, also found from resistive transitions, appears to be ubiquitous in high- $T_c$  superconductors<sup>8,9</sup> and has been attributed to critical fluctuations,<sup>10</sup> a "quasi de Almeida-Thouless line,"<sup>11</sup> irreversible flux creep effects.<sup>8</sup> or inhomogeneities. Our own interpretation, supported by data on two high-quality YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> films with thicknesses of 700 and 2400 Å, invokes a pair-breaking description in which a clean-limit pair breaker  $(\alpha H^{1/2})$  replaces the conventional dirty-limit pair breaker ( $\alpha H$ ). This cleanlimit pair breaker dominates when H is high enough ( $\sim 1$ T for our films) so that the distance between vortices is smaller than the *ab*-plane quasiparticle scattering length. It is in this regime that the phase depairing between two quasiparticles defining a Cooper pair is dominated by ballistic rather than diffusive propagation between vortices.

The films were grown<sup>12</sup> with a strong c-axis orientation on (100)SrTiO<sub>3</sub> substrates. Samples, approximately 3-4 mm in size with leads attached in a van der Pauw configuration, were placed between two 1-mm-diam coils, so that data on the resistance and screening transitions, using a method previously described,<sup>6</sup> could be simultaneously acquired. The two samples chosen for study, 700 and 2400 Å thick, had narrow zero-field resistance transitions, similar temperature dependence in both van der Pauw components, and an absence of irregular features such as bumps or tails in the resistive and screening transitions. The 700- and 2400-Å samples had room-temperature resistivities of 360  $\mu\Omega$  cm and 270  $\mu\Omega$  cm, respectively, and the 2400-Å sample had a critical current density at 77 K of  $1.8 \times 10^6$  A/cm<sup>2</sup>. These values are indicative of extremely high-quality samples.

Figure 1 shows a significantly broadened resistance transition at H = 14 T (×) when compared to the H = 0 T (•) transition. The reciprocal of  $\lambda$  for the 14-T data ( $\Delta$ ), derived from the screening measurements taken at 1.25 kHz, is seen to have a linear temperature dependence near  $T_c$  which extrapolates to a  $T_c(H)$  of 63.80 K. This functional dependence is different from the mean-field dependence [ $\lambda^{-2} \propto (T - T_c)$ ] found at low fields ( $H \lesssim 1$  T) and used to determine the H = 0 transition temperature  $T_c(0)$ of this film (dashed arrow). Mean-field BCS dependence at H = 0 has also been reported previously for a similar film 500 Å thick.<sup>5</sup>

Perhaps the most noteworthy feature of the data presented in Fig. 1 is the identification and location of  $T_c(H)$ below the tail of the resistive transition and well below the midpoint, an identification commonly used in  $H_{c2}$  determinations. We define  $T_c(H)$  as that temperature where  $\lambda \rightarrow \infty$ . This procedure is to be contrasted with the usual techniques of using field or temperature sweeps of the resistive transition in conjunction with an extrapolative procedure or an arbitrary percent-of-normal-state cri5244



FIG. 1. Temperature dependence for a 700-Å-thick YBa<sub>2</sub>-Cu<sub>3</sub>O<sub>7</sub> film of R at H=0 ( $\bullet$ ), R at H=14 T ( $\times$ ), and  $\lambda^{-1}$  at H=14 T ( $\Delta$ ). The solid-line fit extrapolates to the transition temperature  $T_c(H)$  at 14 T (dotted arrow). At H=0 a fit to the linear dependence of  $\lambda^{-2}$  vs T (not shown) extrapolates to  $T_c(0) = 87.78$  K indicated by the dashed arrow.

terion.<sup>13</sup> Such techniques are adequate when the resistive transition is narrow but become increasingly suspect for broad transitions, a situation which is typical for high- $T_c$  superconductors.

Experimental uncertainty is introduced, however, because extrapolation to a temperature defining a diverging quantity necessitates an assumption about the functional form of the extrapolation. In Fig. 1, a linear dependence of  $\lambda^{-1}$  gives a good fit to data which unfortunately cannot be extended to higher temperatures closer to  $T_c(H)$  due to uncertainty in the phase angle of the complex screening signal<sup>6</sup> in a region dominated by magnetic-field-induced dissipation. The extrapolation can be made over the entire temperature range of the high-field data by using the empirical functional form

$$\lambda(T,H) = \frac{\lambda(0,H)}{1-t^2}, \quad 1 \text{ T} \lesssim H \le 14 \text{ T},$$
 (1)

where  $t = T/T_c(H)$  is the normalized temperature and  $\lambda(0,H)$  is found experimentally to be proportional to  $H^{1/2}$ . Equation (1) has a  $(1-t)^{-1}$  dependence for  $t \sim 1$ , in agreement with the linear temperature dependence of  $\lambda^{-1}$  plotted in Fig. 1.

The functional form of Eq. (1) was used to model the temperature dependence of the screening data at each field using three fitting parameters,  $\lambda(0,H)$ ,  $T_c(H)$ , and an additive constant representing a small base-line mutual inductance in the unprocessed screening data. The linear dependence of the fitting parameter  $\lambda(0,H)$  on  $H^{1/2}$  is shown in Fig. 2 for the 700-Å ( $\Delta$ ) and 2400-Å-thick films ( $\bullet$ ), respectively. Near  $T_c(H)$ , Eq. (1) implies  $\lambda(0,H) = -2/T_c(H)(d\lambda^{-1}/dT)|_{T_c(H)}$ . This relationship and the values of  $T_c(H)$  and  $(d\lambda^{-1}/dT)|_{T_c(H)}$  calculated from the linear fits such as shown in Fig. 1 allow a separate calculation of  $\lambda(0,H)$  which is found to be in good agree-



FIG. 2. Magnetic field dependence of  $\lambda(0)$  for the 700-  $(\triangle)$  and 2400-Å-thick (•) films determined as described in the text by using a multiparameter fitting procedure at each field over the whole temperature range. The solid- and dashed-line fits should not be extended into the crossover regime below H = 1 T.

ment, especially for the 700-Å film ( $\triangle$ ), with the values of  $\lambda(0)$  determined by the fitting procedure. This good agreement is a self-consistency check for the functional form, both in temperature and field, of Eq. (1).

In previous work<sup>7</sup> it has been shown that the complex sheet impedance  $Z_v$  due to field-induced vortices will, at sufficiently high fields, dominate over the inductive contribution from the background pair condensate. A useful insight into the functional form of Eq. (1) is gained by calculating  $Z_v$ , assuming uncorrelated vortex motion in harmonic pinning potentials. In this approximation, each vortex is independently pinned with a restoring force constant k and damping coefficient  $\eta$ . The equation of motion is  $\eta \dot{x} + kx = d_F J \Phi_0/c$ , where  $d_F$  is the film thickness, x the vortex displacement,  $\Phi_0$  the quantum of flux, and  $J \Phi_0/c$  the Lorentz force per unit vortex length due to an applied current density J. Combining this relation with the Maxwell relation  $\dot{x} = cE/H$ , we find the expression

$$Z_{v} = \frac{E}{Jd_{F}} = \frac{i\omega\tau}{1+i\omega\tau} \left(\frac{\Phi_{0}H}{c^{2}\eta}\right), \qquad (2)$$

where  $\tau = \eta/\kappa$  is a characteristic temperature-dependent response time. In the low-frequency strong-pinning limit  $(\omega\tau \ll 1)$  we see from Eq. (2) that  $Z_v = i\omega L_p$  where  $L_p = \Phi_0 H/c^2 k$  is the pinning inductance which dominates the response in this limit. To find the relation between  $\lambda$ and k we use the equation  $\lambda^2 = L_p c^2 d_F/4\pi$  to calculate

$$\lambda^2 = \frac{d_F \Phi_0 H}{4\pi k} \,. \tag{3}$$

This result is equivalent to the "pinning penetration length" calculated by Campbell<sup>14</sup> from a different point of view and found by him to be a measure of the small-signal screening response of pinned vortices.

It is important to point out that the ac fields used in our

measurements are typically on the order of  $10^{-2}$  Oe, a factor of  $10^{-6}$  less than the 1 T lower bound on the range of H for which Eq. (1) describes the data. Accordingly, the average vortex displacement resulting from the measuring field is, for  $H \gtrsim 1$  T, less than  $10^{-6}$  of the spacing between vortices. These small displacements of less than  $10^{-3}$  Å imply the small signal limit in which the vortices are undergoing *reversible* excursions about their equilibrium positions rather than hopping from site to site as would occur, for example, during flux creep.

For pinning which occurs on length scales comparable to the coherence length  $\xi$ , an upper bound on the pinning force can be found by equating the product of the energy density  $H_c^2/8\pi$  and the effective core volume  $\pi\xi^2 d_F$  to the stored energy  $k\xi^2/2$ , or  $k = H_c^2 d_F/4$ . Using Eq. (3) and the approximation  $H_c(t) \cong H_c(0)(1-t^2)$  for the critical field  $H_c(t)$ , it is straightforward to find

$$\lambda(t,H) = \frac{(\Phi_0 H)^{1/2}}{\pi^{1/2} H_c(0)(1-t^2)},$$
(4)

which has the same functional form as Eq. (1). Comparing the slope  $d\lambda(0,H)/dH^{1/2} = (\Phi_0/\pi)^{1/2}H_c(0)^{-1}$  with the experimental value of 0.14  $\mu$ m T<sup>-1/2</sup> for the 700-Åthick film (cf. Fig. 2), we estimate  $H_c(0) = 1.8$  kOe. This result for  $H_c(0)$  has a value near thermodynamic estimates<sup>15</sup> and thus implies strong vortex pinning. The increase of  $\lambda$  with increasing H, shown by Eqs. (3) and (4) to be a consequence of vortex motion in a pinning potential, is detrimental to small-signal ac screening and thus implies a heretofore unrecognized intrinsic limit to technological applications of these materials in high magnetic fields.

The divergence of  $\lambda$  at  $T_c(H)$  shown by the data of Fig. 1 is thus associated with vortex core depinning where the pinning force, proportional to k, becomes negligible. The temperature dependence of the vortex-depinning critical field  $H_{cp}$ , shown for the 700-Å-thick film in the inset of Fig. 3, is seen to be nonlinear with positive curvature. Our model for this nonlinearity is based on simple pairbreaking theory<sup>16,17</sup> in which the ratio  $T_c(H)/T_c(0)$  is related to a pair-breaking energy  $\alpha \ll k_B T_c(H)$  by the equation

$$\ln\left(\frac{T_c(H)}{T_c(0)}\right) = \frac{-\pi\alpha}{4k_B T_c(H)}.$$
 (5)

We see in Fig. 3 that a pair-breaking energy  $\alpha \propto H^{1/2}$ used in this equation gives an excellent fit to the  $H \gtrsim 1$  T data for both films. To account for this field dependence, we recall that the pair-breaking rate  $\tau_p^{-1} = 2\alpha/\hbar$  is a measure of the time rate of change of the phase of the wave function describing the pair.<sup>18</sup> When the quasiparticles move a distance  $l_H = (\Phi_0/H)^{1/2}$  between vortices, the phase change is on the order of unity and the pair is broken. For a type-II dirty superconductor, the transport scattering length  $l_{\rm tr} < l_H$  is small and the pair-breaking rate  $\tau_{\rm dirty}^{-1} = D/l_H^2 = DH/\Phi_0$  with diffusion constant D has the dirty-limit form. For ballistic propagation,  $l_{\rm tr} > l_H$ and  $\tau_{\rm clean}^{-1} = v_F/l_H = v_F (H/\Phi_0)^{1/2}$ , which is the form used in Eq. (5) to characterize the data plotted in Fig. 3. Thus, there is a crossover from diffusive ( $\alpha \propto DH$ ) to ballistic



 $(\alpha \propto v_F H^{1/2})$  pair breaking near a length  $l_H \sim l_{tr} \sim 450$  Å corresponding to the distance between vortices at H = 1 T. From resistivity saturation arguments we would expect the in-plane scattering length of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> to be on the order of 100 Å, <sup>19</sup> almost a factor of 5 less than the 450 Å at 1 T where we begin to observe ballistic behavior.

A physically intuitive understanding is gained <sup>18</sup> by considering the magnetic vector potential A as the source of an energy perturbation  $\alpha = ev_F \cdot A/c$  on the paired quasiparticles. Using the expression  $A = Hl_H/2$  for the in-plane vector potential and  $v_F = 8.2 \times 10^6$  cm/sec, <sup>15</sup> we calculate  $\alpha$  and hence a "critical field" slope which is larger by a factor of 3 than the slopes for the two films shown in Fig. 3. This estimate does not take into account the unknown angular average of the quantity  $v_F \cdot A/|v_FA|$ . Finally, taking the average slope of 5.4 T<sup>-1/2</sup> K for the two films of Fig. 3 and solving for the pair-breaking energy  $\alpha_c = 0.88k_BT_c(0)$  necessary to suppress superconductivity at zero temperature, <sup>16-18</sup> we find  $H_{cp}(0) = 133$  T.

Although the vortex contribution to  $\lambda$  clearly vanishes at  $T_c(H)$ , it is difficult to ascertain, because of *H*-field dissipation, if the smaller background condensate term does likewise. Recent dc magnetization measurements on YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> crystals<sup>20</sup> indicate a persistence of a diamagnetic response to temperatures near the normal-state part of the resistance transition, a response which is consistent with a finite pair condensate part of the penetration depth. We note, however, that the zero-field transition temperature  $T_c(0)$ , indicated by the dashed arrow in Fig. 1, has been determined by extrapolation<sup>5</sup> of a BCS mean-field temperature-dependent  $\lambda$  and found to be located in the low-resistance part of the transition. Thus, for this particular film, the temperature  $T_c(0) = 87.78$  K serves as an upper bound for the existence of a finite pair condensate extending uniformly over the entire film.

In conclusion, we have presented experimental results



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on the magnetic-field-dependent penetration length of c-axis-oriented YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> films. The functional form of  $\lambda(T,H)$  for  $H \ge 1$  T has been shown to be a consequence of strong vortex core pinning which vanishes at  $T_c(H)$ . Extrapolation to a temperature  $T_c(H)$  where  $\lambda \rightarrow \infty$  determines the vortex-depinning critical field  $H_{cp}$ . The phase boundary determined by  $H_{cp}$  is well described by simple pair-breaking theory and may be related to the "melting" line found in mechanical measurements.<sup>21</sup> The upward curvature of  $H_{cp}(T)$  is intrinsic and is described by a different set of parameters than those which have been extracted from  $H_{c2}$  measurements which are assumed to reflect type-II dirty theory and which have been

<sup>1</sup>J. Rammer, Europhys. Lett. 5, 77 (1988).

- <sup>2</sup>D. R. Harshman, G. Aeppli, E. J. Ansaldo, B. Batlogg, J. H. Brewer, J. F. Carolan, R. J. Cava, M. Celio, A. C. D. Chaklader, W. N. Hardy, S. R. Kreitzman, G. M. Luke, D. R. Noakes, and M. Senba, Phys. Rev. B 36, 2386 (1987).
- <sup>3</sup>D. R. Harshman, L. F. Schneemeyer, J. V. Waszczak, G. Aeppli, R. J. Cava, B. Batlogg, L. W. Rupp, E. J. Ansaldo, and D. L. Williams, Phys. Rev. B **39**, 851 (1989).
- <sup>4</sup>L. Krusin-Elbaum, R. L. Greene, F. Holtzberg, A. P. Malozemoff, and Y. Yeshurun, Phys. Rev. Lett. 62, 217 (1989).
- <sup>5</sup>A. T. Fiory, A. F. Hebard, P. M. Mankiewich, and R. E. Howard, Phys. Rev. Lett. **61**, 1419 (1988).
- <sup>6</sup>A. T. Fiory, A. F. Hebard, P. M. Mankiewich, and R. E. Howard, Appl. Phys. Lett. **52**, 2165 (1988).
- <sup>7</sup>A. F. Hebard and A. T. Fiory, in Ordering in Two Dimensions, edited by S. K. Sinha (North-Holland, Amsterdam, 1980), p. 181; A. T. Fiory and A. F. Hebard, in Inhomogeneous Superconductors—1979, edited by D. U. Gubser, T. L. Francavilla, S. A. Wolf, and J. R. Leibowitz, AIP Conference Proceedings No. 58 (American Institute of Physics, New York, 1979), p. 293.
- <sup>8</sup>A. P. Malozemoff, T. K. Worthington, Y. Yeshurun, and F. Holtzberg, Phys. Rev. B 38, 7203 (1988).
- <sup>9</sup>G. W. Crabtree, W. K. Kwok, U. Welp, R. Burriel, H. Claus, K. G. Vandervoort, and J. Z. Liu, J. Magn. Magn. Mater.

used almost universally to characterize the high- $T_c$  oxides. Finally, use of accepted values for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> of the thermodynamic critical field, transport scattering length, and Fermi velocity in our models for  $\lambda(T, H)$  and  $H_{cp}$  gives results in good qualitative agreement with the data.

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**76 & 77**, 547 (1988).

- <sup>10</sup>B. Oh, K. Char, A. D. Kent, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond, and A. Kapitulnik, Phys. Rev. B 37, 7861 (1988).
- <sup>11</sup>K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).
- <sup>12</sup>A. F. J. Levi, J. M. Vandenberg, C. E. Rice, A. P. Ramirez, K. W. Baldwin, M. Anzlowar, A. E. White, and K. Short, J. Cryst. Growth **91**, 386 (1988).
- <sup>13</sup>For a discussion, see M. A. Paalanen and A. F. Hebard, Appl. Phys. Lett. 45, 794 (1984).
- <sup>14</sup>A. M. Campbell, J. Phys. C 2, 1492 (1969).
- <sup>15</sup>M. B. Salamon and J. Bardeen, Phys. Rev. Lett. **59**, 2615 (1987).
- <sup>16</sup>P. G. de Gennes, Superconductivity of Metals and Alloys (Benjamin, New York, 1966).
- <sup>17</sup>K. Maki, in *Superconductivity*, edited by R. D. Parks (Dekker, New York, 1969), p. 1035.
- <sup>18</sup>M. Tinkham, Introduction to Superconductivity (McGraw-Hill, New York, 1975), p. 264.
- <sup>19</sup>M. Gurvitch and A. T. Fiory, Phys. Rev. Lett. **59**, 1337 (1987).
- <sup>20</sup>U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, Phys. Rev. Lett. **62**, 1908 (1989).
- <sup>21</sup>P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D. J. Bishop, Phys. Rev. Lett. **61**, 1666 (1988).