

Scaling of the hysteretic magnetic behavior in $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals

L. Civale, M. W. McElfresh,* A. D. Marwick, F. Holtzberg, and C. Feild
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598-0218

J. R. Thompson and D. K. Christen
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831
 (Received 26 February 1991)

We have studied the pinning force $F_p(B, T)$ in $\text{YBa}_2\text{Cu}_3\text{O}_7$ crystals as a function of fluence of 3-MeV protons. For a given ion-damage level, we could reduce all F_p data to a single function of reduced field $b = B/B^*$, where the scaling field $B^*(T) \ll H_{c2}(T)$ is related to the irreversibility line. This result rules out the possibility of matching effects between the vortex lattice and the distribution of pinning sites. We emphasize the influence of thermally activated relaxation in the determination of F_p and compare the data with simple pinning models.

Shortly after the discovery of cuprate superconductors, it became evident¹ that their magnetic and magnetotransport behavior was rather unusual, and the magnetic phase diagram of this material continues to be a problem of considerable interest and controversy.² The existence of an unpinned vortex regime at magnetic fields and temperatures above the "irreversibility line" $H_{\text{irr}}(T)$ is well established.¹⁻³ Several theoretical explanations for this unpinned regime have been proposed, including spin-glass-like behavior,^{1,4} giant flux creep,⁵ melting of the vortex lattice,^{6,7} and vortex-glass transitions.⁸

Due in part to the technological interest, an enormous number of studies of the critical current J_c for $\text{YBa}_2\text{Cu}_3\text{O}_7$ have been reported.⁹ Pinning in type-II superconductors results¹⁰ from interactions between flux lines and inhomogeneities in the material, which make the free energy of a vortex position dependent. The strength of the pinning center can be characterized by a pinning potential U . The pinning force per unit volume is $F_p = U/(aV)$, where a is the characteristic range of the potential well and V is an activation volume. Usually, all the quantities determining F_p can be written¹⁰ in terms of the internal field B and the temperature-dependent critical fields of the superconductor. With few exceptions, this results^{10,11} in an expression for F_p consisting of a product of a temperature-dependent factor and a function of the reduced field $b = B/H_{c2}$, where H_{c2} is the upper critical field. Thus, all the curves $F_p(B)$ measured at different temperatures can be superimposed in a single curve of F_p/F_p^{max} vs b , where $F_p^{\text{max}}(T)$ is the maximum pinning force at each temperature. This scaling property of the pinning forces has indeed been observed^{10,11} in a large number of traditional type-II superconductors.

In this paper we show that the pinning forces in $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals can also be scaled, provided that we replace H_{c2} by another temperature-dependent scaling field B^* , related to the irreversibility line $H_{\text{irr}}(T)$ of the material. This difference in scaling behavior is a consequence of the large relaxation rate in these samples. The scaling is observed both in unirradiated and proton-irradiated crystals. We discuss some implications in terms of possible pinning mechanisms. Several well character-

ized, twinned single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_7$ were used. Prepared using a flux growth method,¹² the crystals initially had superconducting transition temperatures of about 93.5 K, and transition widths $\Delta T_c < 0.5$ K. Typical dimensions were $1 \times 1 \times 0.03$ mm³ with the c axis parallel to the shorter dimension. Crystals were irradiated¹³ at room temperature using 3-MeV protons. At the highest dose used ($\Phi = 8 \times 10^{16}$ cm⁻²), T_c decreases by approximately 4 K. Measurements of the field- and temperature-dependent magnetization M were made in a Quantum Design superconducting-quantum-interference device (SQUID) magnetometer at applied fields $H \leq 5.5$ Tesla, for H parallel to the c axis. Samples were cooled in zero field and temperature was stabilized to within 0.05 K prior to field application and measurement. Scan lengths of 2 or 3 cm, providing a field uniformity of $\sim 0.05\%$, were used.

Shown in Fig. 1 are plots of M as a function of H at several temperatures, for the same $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystal both before and after proton irradiation of $\Phi = 6 \times 10^{15}$ cm⁻². A large enhancement of the irreversible magnetization with irradiation is evident, as reported previously.¹³ These $M(T, H)$ results can be used to determine values of the critical current density $J_c(T, H)$ by means of the critical state model. The simplest approximation (Bean model¹⁴) assumes that J_c is independent of H . In this case, for a rectangular sample,¹⁵ $J_c = 20\Delta M / L_1[1 - L_1/3L_2]$, where $L_1 \leq L_2$ are the sides of the rectangle, and ΔM is the difference between the upper and lower branches of the $M(H)$ loops.

Before pursuing this treatment we will determine the limits of applicability. Our crystals are thin plates and H is parallel to their shortest dimension t . In this situation, the currents flowing in the sample generate¹⁶ a radial component of magnetic field. If H is small compared to the self-field $H_s \sim J_c t$, the direction of the vortices will differ significantly¹⁶ from that of H , and will even be perpendicular to it in places. In that regime, the magnetic moment depends¹⁷ on two anisotropic components of J_c , corresponding to current flow in the (a, b) planes with vortices either parallel or perpendicular to the c axis ($J_c^{(a,b),c}$ and $J_c^{(a,b),(a,b)}$), respectively.⁹ Using the Bean

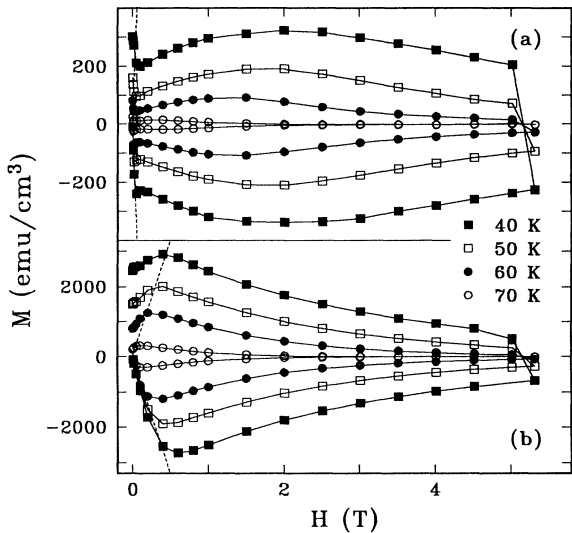


FIG. 1. Magnetization M vs applied field $H \parallel c$ axis, for $\text{YBa}_2\text{Cu}_3\text{O}_7$ crystal 6 at various temperatures. (a) Before irradiation. (b) After irradiation with 6×10^{15} ions/cm² at 3 MeV.

relation between ΔM and J_c , we estimate $H_s \sim 20t \times \Delta M / L_1 [1 - L_1/3L_2]$. In Fig. 1 the dotted lines represent the condition $H = H_s$, and approximately define the region of applicability of the Bean model. At lower fields, self-field effects dominate, the internal field has a component perpendicular to H , and ΔM is a complicated function of $J_c^{(a,b),c}$ and $J_c^{(a,b),(a,b)}$. At higher fields, the internal field is parallel to H , ΔM is a function of $J_c^{(a,b),c}$ only, and the Bean relation can be used. We will concentrate on the study of $J_c^{(a,b),c}(H)$, hereafter called J_c for simplicity. Also from inspection of Fig. 1 we find that, in the region $H > H_s$, the internal field B is very similar to H .

Figure 1(a) shows that $M(H)$ in the unirradiated crystal has a maximum at some intermediate field. These ‘‘bumps’’¹⁸ reflect the fact that J_c is not a monotonically decreasing function of H , but rather peaks at some intermediate field. We observed them in all the nonirradiated crystals investigated, although the field location differs. The bumps always disappear after proton irradiation, even at the lowest dose investigated ($\Phi = 3 \times 10^{15}$ cm⁻²). Figure 1(b) shows a monotonically decreasing $J_c(H)$ in the region of applicability of the critical state model.

We now analyze the pinning force density $F_p(B, T) = J_c B$. At a given temperature F_p is zero at $B = 0$ and $B = H_{c2}(T)$, and therefore reaches a maximum value $F_p^{\max}(T)$ at some intermediate field $B_{\max}(T)$. Shown in Fig. 2 are plots of the reduced pinning force $f_p = F_p/F_p^{\max}$ as a function of $b = B/B_{\max}$ for the sample in Fig. 1 at three different levels of proton irradiation ($\Phi = 0, 3$, and 6×10^{15} cm⁻²), and for two other $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals at higher levels of irradiation. Figure 3 contains similar data for another irradiated crystal. Each of the six data sets consists of 9 curves taken at evenly spaced temperatures between 40 and 80 K. Data from the region $H < H_s$ were excluded, as were data at low temperatures where B_{\max} was experimentally inaccessible. It is apparent that, at any level of irradiation, this scaling pro-

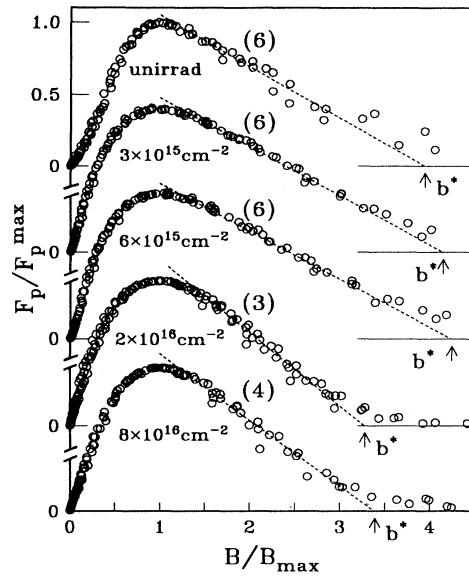


FIG. 2. Normalized pinning force density vs reduced field, for crystal 6 at three irradiation levels and for crystals 3 and 4. Doses are in units of ions/cm². Scaling variables F_p^{\max} and B_{\max} are peak values of the pinning force density and corresponding field, respectively, for each temperature.

cedure allows us to superimpose the data corresponding to a wide range of temperatures in a unique curve $f_p(b)$.

Scaling of the pinning forces at different temperatures has proved to be a useful tool^{10,11} in the study of pinning mechanisms in traditional type-II superconductors. The mere existence of a universal curve $f_p(b)$ that describes the data in a wide range of temperatures strongly suggests¹¹ that J_c is determined by a single type of pinning center. A lack of scaling, on the other hand, may also provide useful information. For instance, if the average separation of pins matches the vortex lattice spacing, scaling will not be observed,¹¹ because F_p will peak at the same field at all temperatures. Figures 2 and 3 show that good scaling is observed in these $\text{YBa}_2\text{Cu}_3\text{O}_7$ crystals at any

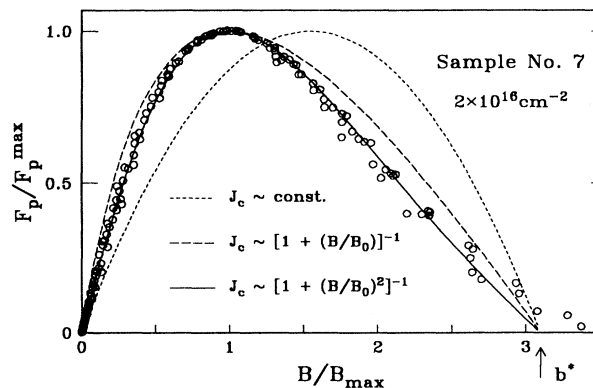


FIG. 3. Normalized pinning force density vs reduced field for crystal 7. Continuous lines are fits of the form $F_p = J_{c0}(B) \times B[1 - (B/B^*)]$.

level of proton irradiation, thus ruling out the possibility of matching between proton induced defects and the vortex lattice. The existence of the bumps in the magnetization of the unirradiated crystal is reflected in the $f_p(b)$ curve as an upward curvature at low b (top curve of Fig. 2). In this case J_c initially increases with B , and so f_p increases faster than linearly. In contrast, after irradiation J_c decreases monotonically with B and the upward curvature in f_p is absent. This observation implies that the relevant pinning centers differ in the two cases, but nevertheless both types of pinning sites satisfy a scaling rule.

Although the observed scaling of F_p resembles that frequently found in traditional type-II superconductors, there is an important difference. While in the low- T_c superconductors B was scaled¹⁰ by the experimental value of H_{c2} , in our case H_{c2} is too large to be measured^{19,20} except in a narrow temperature range close to T_c . Consequently, our scaling field $B_{\max}(T)$ is not determined independently. The values of $B_{\max}(T)$ used to scale the six curves in Figs. 2 and 3 are plotted in Fig. 4. A more direct comparison with traditional scaling can be established if we plot the values of the field at which F_p drops to zero. Inspection of Figs. 2 and 3 shows that such a determination is difficult due to the long tails in the high-field region. An alternative approach consists of taking the linear portion of $f_p(b)$ and extrapolating to $f_p=0$, as indicated in Fig. 2. This procedure determines a reduced field b^* and another characteristic field $B^*=b^*B_{\max}$, which is not the field where $F_p=0$, but is a good approximation to it. The fact that b^* is temperature independent is a consequence of the scaling law obeyed by F_p , and implies that the scaling field can be either $B^*(T)$ or $B_{\max}(T)$; the latter is simpler to determine experimentally. Values of $B^*(T)$ for all the crystals are shown in Fig. 4, along with $H_{c2}(T)$ of a similar crystal.²⁰ It is seen that the lines of $B^*(T)$ lay far below $H_{c2}(T)$, reflecting the fact that high- T_c superconductors present an unpinning vortex regime between $H_{c2}(T)$ and $H_{\text{irr}}(T)$. As a consequence, the scaling field B^* should be associated with H_{irr} rather than with H_{c2} .

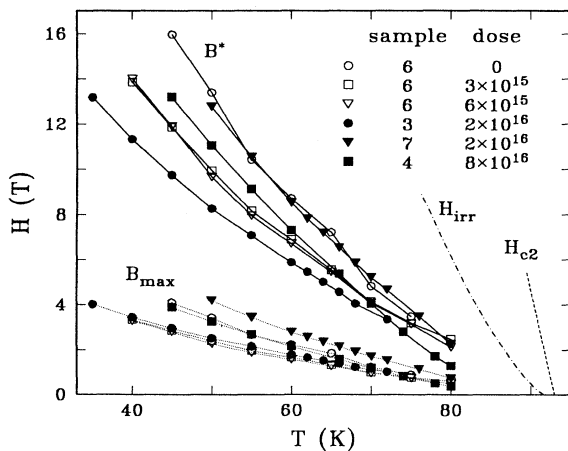


FIG. 4. Characteristic fields vs T for various crystals, including upper critical field H_{c2} , the ac irreversibility line H_{irr} , field B^* at which F_p extrapolates to zero, and field of maximum pinning force B_{\max} .

The position of H_{irr} as determined¹³ by ac susceptibility at $f=1$ MHz is also shown in Fig. 4. The location of this line is very similar in all the crystals measured. A comparison between the dc and ac results is difficult because the data obtained by both techniques only overlap in a small temperature window, between 76 and 80 K. In that region, H_{irr} is about three times higher than B^* . Two factors contribute to this difference. First, it is seen in Fig. 2 that B^* underestimates the field where $F_p=0$ by no less than 20%. Second, H_{irr} in $\text{YBa}_2\text{Cu}_3\text{O}_7$ crystals shows³ a frequency dependence $H_{\text{irr}} \approx C[\ln(f_0/f)]^{-2/3}(1-t)^{4/3}$, where $t=T_{\text{irr}}/T_c$, $f_0 \approx 10^9$ Hz, and C is independent of frequency. Estimating that our dc data have an associated “frequency” of $\sim 10^{-2}$ Hz, corresponding to the measuring time for each point, the ac H_{irr} at the same frequency would thus be a factor of ~ 2.4 lower than the 1 MHz result. The combination of both effects produces a factor of ~ 2.9 , which is almost exactly the experimental value, thus giving further support to the assertion that H_{irr} and B^* are measurements of basically the same phenomenon. Although there is some dispersion in the B^* curves corresponding to different crystals, no correlation is observed between the location of the line and the dose of irradiation. This result reinforces our previous conclusion¹³ that the location of H_{irr} is almost unaffected by proton irradiation, in spite of the large enhancement of J_c induced below that line.

The abrupt decay of f_p at fields much smaller than H_{c2} is due to the fast time relaxation induced by thermal activation. In a similar analysis²¹ performed in $\text{YBa}_2\text{Cu}_3\text{O}_7$ thin films, the high-field portion of the $f_p(b)$ was fitted using standard flux creep. Invoking flux-creep models, we outline a more general analysis that fits $f_p(b)$ over the whole range, similar to phenomenological treatments.²² A more complete study will be presented elsewhere.

We start with the usual expression for J_c in the presence of flux creep, $J_c = J_{c0}[1 - (kT/U)\ln(t/t_0)]$, and multiply by B to obtain the pinning force density

$$F_p = J_{c0}B[1 - (kT/U)\ln(t/t_0)]. \quad (1)$$

If, as is frequently postulated,⁵ and observed in epitaxial films,²³ $U = \alpha(T)/B$, the expression inside the square brackets is linear in B and drops to zero at $B^* = kT\alpha(T)\ln(t/t_0)$. It is important to note that Eq. (1) only applies when $kT \ll U$. When the thermal activation becomes large, this approximation fails and F_p decreases more slowly, thus originating a tail as experimentally observed in Fig. 2. We can associate the linear decreasing portion of $f_p(b)$ with the region of applicability of the logarithmic decay, and rewrite Eq. (1) as

$$F_p = F_{p0}(1 - B/B^*) = F_{p0}(1 - b/b^*), \quad (2)$$

where $F_{p0} = J_{c0}B$ is the pinning force in absence of thermal effects. In the Bean model¹⁴ $F_{p0} = J_{c0}(T)B$ where $J_{c0}(T)$ is independent of B . In this approximation the expression for $f_p(b)$ results in a parabola. The comparison with experimental data for an irradiated crystal is shown in Fig. 3. The temperature dependence of J_{c0} disappears in the scaling procedure. A better agreement with experiment can be achieved by using the Kim formula²⁴ $J_{c0} \sim 1/[1 + B/B_0(T)]$, which implies $F_{p0} \sim 1/$

$[1 + (b_0/b)]$. The parameter $b_0 = B_0/B_{\max}$ can, in principle, depend on temperature. Assuming that it is temperature independent within our experimental range, it can be determined by the condition that f_p peaks at $b = 1$. The result is $b_0 = 1/(b^* - 2)$. This fitting is also shown in Fig. 3. An almost perfect agreement with the experiment is obtained if we assume that $J_{c0} \sim 1/[1 + (B/B_0)^2]$, although we recognize that there is no theoretical justification for this expression. In this case $F_{p0} \sim 1/[(b_0/b) + (b/b_0)]$ and $b_0 = [b^*/(b^* - 2)]^{1/2}$. The result is also shown in Fig. 3. This analysis shows that the maximum effective pinning force that can be obtained in these materials is strongly limited by thermally activated relaxation. There is increasing evidence^{25,26} that Eq. (1) is modified in the intermediate temperature regime to a form based on the vortex-glass theory,⁸ wherein $J_c = J_{c0}/[1 + (kT/U)\ln(t/t_0)]^{1/\mu}$. This model can, in principle, produce a similar scaling with F_p vanishing for fields below H_{c2} , provided that $\mu < 1$. However, the comparison of experiment with this theory is complicated by the fact that, according to recent studies,²⁶ μ is both temperature

and field dependent. An analysis of our results in the context of the vortex-glass model will be presented in a future report.

In summary, we have shown that the pinning forces of $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals, both unirradiated and proton irradiated, obey a scaling law over the whole range of experimentally accessible temperature and field. The scaling field is closely related to the irreversibility line. We have analyzed the implications in terms of pinning mechanisms and discussed the differences between unirradiated and irradiated samples.

The authors thank A. P. Malozemoff and T. K. Worthington for useful discussions and T. Zabel for his cooperation in the irradiations. Technology development was jointly funded by IBM and the Oak Ridge National Laboratory High Temperature Superconductivity Program, Office of Energy Storage and Distribution, Conservation and Renewable Energy, under Contract No. DE-AC05-84OR21400, with Martin Marietta Energy Systems, Inc.

*Present address: Department of Physics, Purdue University, West Lafayette, IN 47907.

¹K. A. Müller, M. Takashige, and J. G. Bednorz, *Phys. Rev. Lett.* **58**, 408 (1987).

²A. P. Malozemoff, *Mater. Res. Bull.* **15** (6), 50 (1990).

³A. P. Malozemoff, T. K. Worthington, Y. Yeshurun, F. Holtzberg, and P. H. Kes, *Phys. Rev. B* **38**, 7203 (1988).

⁴I. Morgenstern, K. A. Müller, and J. G. Bednorz, *Z. Phys. B* **69**, 33 (1987).

⁵Y. Yeshurun and A. P. Malozemoff, *Phys. Rev. Lett.* **60**, 2202 (1988).

⁶P. L. Gammel, D. J. Bishop, G. J. Dolan, J. R. Kwo, C. A. Murray, L. F. Schneemeyer, and J. V. Waszczak, *Phys. Rev. Lett.* **59**, 2592 (1987).

⁷A. Houghton, R. A. Pelcovitz, and A. Sudbo, *Phys. Rev. B* **40**, 6763 (1989).

⁸M. P. A. Fisher, *Phys. Rev. Lett.* **62**, 1415 (1989).

⁹A. P. Malozemoff, in *High Temperature Superconductivity II*, edited by S. H. Whang, A. Das Gupta, and R. B. Laibowitz (TMS Publications, Warrendale, PA, in press).

¹⁰W. A. Fietz and W. W. Webb, *Phys. Rev.* **178**, 657 (1969).

¹¹H. Ullmaier, *Irreversible Properties of Type II Superconductors* (Springer-Verlag, Berlin, 1975), Chap. 3.

¹²D. L. Kaiser, F. Holtzberg, M. F. Chisholm, and T. K. Worthington, *J. Cryst. Growth* **85**, 593 (1987).

¹³L. Civale, A. Marwick, M. W. McElfresh, T. K. Worthington, A. P. Malozemoff, F. H. Holtzberg, J. R. Thompson, and M. A. Kirk, *Phys. Rev. Lett.* **65**, 1164 (1990).

¹⁴C. P. Bean, *Phys. Rev. Lett.* **8**, 250 (1962).

¹⁵A. M. Campbell and J. E. Evetts, *Ad. Phys.* **21**, 199 (1972).

¹⁶M. Däumling and D. C. Larbalestier, *Phys. Rev. B* **40**, 9350 (1989).

¹⁷L. W. Conner, A. P. Malozemoff, and I. A. Campbell, *Phys. Rev. B* (to be published).

¹⁸M. Däumling, J. M. Seuntjens, and D. C. Larbalestier, *Nature* (London) **346**, 332 (1990).

¹⁹U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, *Phys. Rev. Lett.* **62**, 1908 (1989).

²⁰Z. Hao, J. R. Clem, M. W. McElfresh, L. Civale, A. P. Malozemoff, and F. H. Holtzberg, *Phys. Rev. B* **43**, 2844 (1991).

²¹J. D. Hettinger, A. G. Swanson, W. J. Skocpol, J. S. Brooks, J. M. Graybeal, P. M. Mankiewich, R. E. Howard, B. L. Straughn, and E. G. Burkhardt, *Phys. Rev. Lett.* **62**, 2044 (1989).

²²H. R. Kerchner, R. Feenstra, J. O. Thomson, J. R. Thompson, D. K. Christen, S. T. Sekula, and L. A. Boatner, in *High Temperature Superconductors, Fundamental Properties and Novel Materials Processing*, edited by D. K. Christen, J. Narayan, and L. F. Schneemeyer, MRS Symposia Proceedings No. 169 (Materials Research Society, Pittsburgh, 1990), p. 883.

²³D. K. Christen, R. Feenstra, D. H. Lowndes, D. Norton, H. R. Kerchner, J. R. Thompson, S. T. Sekula, J. D. Budai, L. A. Boatner, and R. Singh, in *High Temperature Superconductors, Fundamental Properties and Novel Materials Processing* (Ref. 22), p. 903.

²⁴Y. B. Kim, C. F. Hempstead, and A. R. Strnad, *Phys. Rev. Lett.* **9**, 306 (1962).

²⁵A. P. Malozemoff and M. P. A. Fisher, *Phys. Rev. B* **42**, 6784 (1990).

²⁶J. R. Thompson, Y. Sun, and F. H. Holtzberg, *Phys. Rev. B* (to be published).