

**Macroscopic Magnetic Properties of High Temperature Superconductors:
Effect of a Pinning Barrier Distribution**

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ABSTRACT:

The magnetic properties of YBaCuO crystals are strongly affected by thermally activated flux creep over pinning barriers. Here the effect of a power-law distribution of pinning barriers is explored. The critical current density is predicted to have a power-law dependence on the reduced temperature difference approaching the transition temperature T_c and a logarithmic dependence on the timescale of the measurement. The dependence of the critical current on the measurement time provides a possible explanation of the disparity between the critical current measured in thin film transport experiments and J_c obtained from magnetic data. Also, in contrast to some earlier theory, the normalized logarithmic time derivative of the remanent magnetization is shown to not diverge near T_c , irrespective of the pinning barrier distribution.

INTRODUCTION

In a series of recent papers, our group¹⁻⁶ and others⁷⁻¹³ have shown that flux creep effects are unusually great in the new high temperature superconductors. This is true even in non-granular material, that is, in crystals or epitaxial films, on which we concentrate exclusively here. These effects include the observation of very large time-logarithmic magnetic relaxation which shows a peak in relaxation rate $dM/d(\ln t)$ as a function of temperature^{1,3,7,8} and rises approximately as the cube of the magnetic field⁴ above the lower critical field H_{c1} . We have termed this a "giant flux creep." A related effect is the observation of an irreversibility line,^{1,2,5} defined by a peak in the ac susceptibility, exhibiting a dependence of the reduced temperature difference $\epsilon \equiv 1 - (T/T_c)$ on field H to a low power, typically $2/3$, with an amplitude depending logarithmically on the frequency of the measurement. Yet another effect is the rapid, almost exponential,⁹ dropoff in the magnetically measured critical current^{3,6,9} with temperature in the low temperature range. As will be discussed further below, there is even some evidence for a strong dependence of this dropoff on the measurement time.

Most of these effects have already received an initial explanation^{1-5,11-12} in terms of the classic flux-creep model,¹⁴⁻¹⁶ extended to take into account the giant size of the effects. Nevertheless puzzling features remain which do not appear to be easily explained by the phenomenology. This has led some authors to invoke glassy models⁸⁻¹⁰ as an alternative. In this paper we extend the standard flux-creep model to include a distribution of flux pinning barriers. In some sense this extension could be termed "glassy", although it does not involve the frustration which characterizes recent superconducting glass models.^{17,18} We show how including the pinning barrier distribution offers a new way to resolve some of the remaining problems in comparing to experiment, although so far in only a qualitative way. Relaxation time distributions have also recently been invoked by Foldeaki et al.¹⁰ to explain some of the same kinds of experiments, though in a superconducting glass context.

TEMPERATURE-DEPENDENT J_c AND $dM/d(\ln t)$: THE PROBLEMS

The first problem concerns the temperature dependence of the critical current density J_c , particularly when determined magnetically. At low temperatures, J_c appears to fall linearly with temperature,^{3,6,13,19-21} in some cases extrapolating to zero at temperatures far below T_c , even in crystals or epitaxial films. Furthermore, puzzling discrepancies have been reported in the literature where magnetic and transport determinations of J_c were compared.¹⁹⁻²¹ Typically, in such material, J_c determined from magnetic hysteresis measurements using the standard Bean model²²⁻²³ falls faster with temperature, by a factor of two or more, than the transport J_c , in the low temperature region.

A natural explanation of this effect emerges in the context of giant flux creep.^{1,3} In the standard theory,¹⁶ the critical current density can be expressed in terms of the temperature-dependent current density J_{cT} which would occur in the absence of flux creep, in terms of a temperature-dependent activation energy U_T for flux hopping in the absence of a driving force, and in terms of a characteristic time t for the measurement and an attempt time t_0 :

$$J_c = J_{cT} \left(1 - \frac{kT}{U_T} \ln \frac{t}{t_0} \right) . \quad (1)$$

Here the subscripts T indicate temperature-dependent quantities.

As temperature increases, the linear-T term in this equation increases in magnitude, and since U_T drops monotonically to zero at the superconducting transition temperature T_c , J_c in Eq. 1 must drop to zero at some temperature below T_c given by

$$T_0 = U_T/k \ln(t/t_0) . \quad (2)$$

Actually, near and above this temperature, Eq. 1 must be generalized, as has been emphasized recently by Dew-Hughes,¹² to take into account backward as well as forward hopping of the flux lines over the energy barriers.²⁴ This leads to the more general equation

$$J_c = J_{cT} \frac{kT}{U_T} \operatorname{arcsinh} \left(\frac{t_0}{2t} e^{U_T/kT} \right) , \quad (3)$$

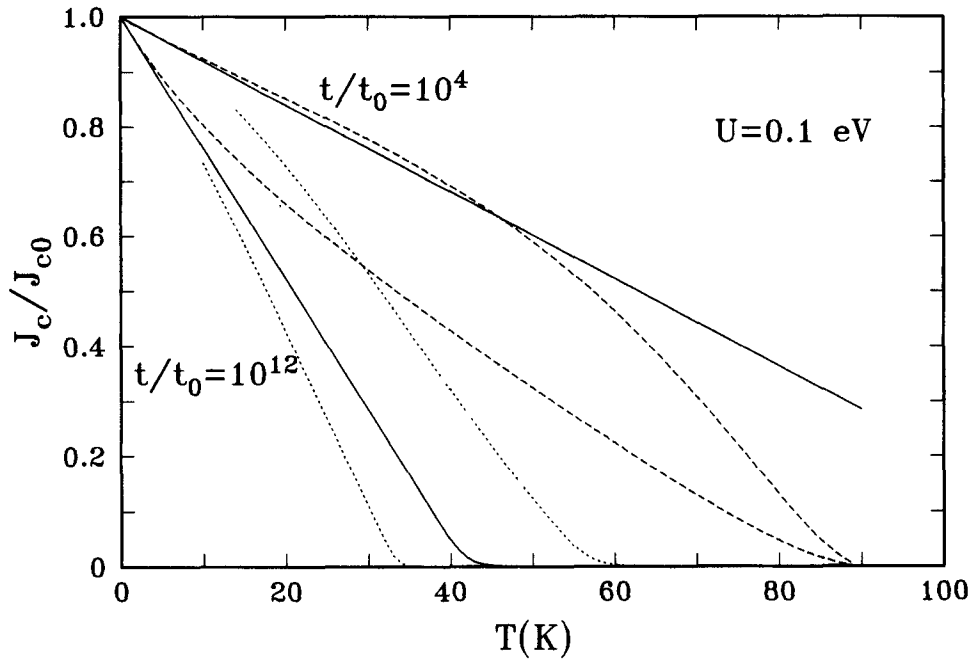
which, for temperatures well above T_0 , reduces to

$$J_c = J_{cT} \frac{kT}{U_T} \frac{t_0}{2t} e^{U_T/kT} . \quad (4)$$

A plot of the full Eq. 3 for typical values^{1,2,4} of $U_{T=0} = 0.1\text{eV}$ and $t_0 = 10^{-11}$ sec is shown in Fig. 1. The different lines represent two values of measurement time t , with the solid lines assuming temperature-independent U_T and the dotted lines assuming $U_T \propto (\epsilon')^{3/2}$, where $\epsilon' \equiv 1 - (T/T_c)^2$ rather than $1 - (T/T_c)$ because at low temperatures, all BCS superconducting parameters in the absence of flux creep are expected to have zero slope with temperature. However, the figure suggests that there can be very dramatic changes in the temperature dependence of the measured critical current density as a function of the measurement time.

In particular, a typical magnetic measurement time per point in a SQUID magnetometer is of order 100 sec or 0.01 Hz. In a typical transport measurement,² the effective time is determined by the minimum voltage criterion for observing resistance and is of order Ba_T/E_c , where B is the magnetic induction, a_T is the vortex jump distance and E_c is the minimum measurement voltage per meter. This works out typically to values of order 10^{-4} sec. Thus, Fig. 1 shows that factor of two or greater differences can

Fig. 1. Normalized critical current density versus temperature in different flux creep models. The two sets of lines represent different effective measurement times $t=10^2$ and 10^{-6} , with a characteristic time t_0 of 10^{-10} sec. The solid lines represent Eq. 3, the conventional theory (including backward flux hopping¹²) with a single temperature-independent activation energy U of 0.1 eV. Dotted lines represent Eq. 3 with a $(\epsilon')^{3/2}$ temperature dependence for U_T and a ϵ' temperature dependence for J_{cT}/U_T ($\epsilon' \equiv 1 - (T/T_c)^2$). The dashed lines represent the predictions of the barrier distribution model of Eq. 19, with $\bar{U} = 0.1$ eV, $n=3$, $m=1$ and $\ell = 1$.



occur in the initial slope with temperature, as observed in experiment. This effect is not typical of low temperature superconductors and comes from the unusually low value of the activation energy U as expected in high temperature superconductors because of their small coherence lengths.^{1,4,11} Values of U as low as 0.02 eV have been deduced from magnetic relaxation measurements^{3,4} in YBaCuO crystals, while values of order 0.1 eV appear to be more appropriate to explain thin film J_c data, where different pinning mechanisms may come into play.

Nevertheless there is still a problem in that the high-temperature tail of J_c predicted by Eq. 3 and shown in Fig. 1 is far too small in the high temperature region, compared to experiment.^{3,6,9,21} Also, as shown in Eq. 4, J_c is no longer logarithmically dependent on time in the high temperature region. While the data^{1,7,8} show some early-time deviations, they still follow an approximately logarithmic form in the high temperature region. It is also not immediately obvious from Eq. 4 how an irreversibility line^{1,2} $\epsilon \propto H^{2/3}$ can be derived; this is discussed elsewhere.⁵

Another problem was first pointed out by Tuominen et al.,⁸ in measurements on ceramic material, but it remains an issue in epitaxial films and crystals.^{7,8} This concerns the temperature dependence of the magnetic relaxation $(1/M)(dM/d\ln t)$ as derived from a critical state model^{22,23} in which the magnetization is related to J_c . For example, in a slab of thickness D with field in the plane, the remanent magnetization is approximately²⁵

$$4\pi M_{rem} = f_b H_{c1} + (\pi D J_c / c) , \tag{5}$$

where f_b is some constant of order unity. At low temperatures, where Eq. 1 describes the logarithmic time decay and temperature dependence of the critical current, we find

$$\frac{1}{M} \frac{dM}{d\ln t} = - \frac{kT}{U_T} \frac{\pi D J_c T}{cf_b H_{c1} + \pi D J_c} . \tag{6}$$

Here M , $dM/d\ln t$ and J_c are all to be evaluated at the same time during the experiment. Clearly the normalized magnetic relaxation is predicted to rise monotonically with temperature. Such a model is not consistent with experimental results,^{1,7,8,10} which indicate a peak in the magnetic relaxation at some temperature $T < T_c$, and then a monotonic decay of the relaxation rate as T approaches T_c .

Initially it was argued⁸ that this observed decay of the magnetization could not be described by flux creep, since in Eq. 6 $(1/M)(dM/d\ln t)$ rises monotonically with temperature, and apparently diverges near T_c . (In the limit where we can ignore the H_{c1} term in Eq. 6, the normalized relaxation would go, to lowest order, as kT/U_T . Since U_T decreases monotonically to zero as temperature increases to T_c , the relaxation is expected to diverge.)

However, Eq. 6 is valid only in the low temperature limit. For temperatures $T > T_0$, the temperature dependence of J_c is described more accurately by Eq. 3 than by Eq. 1. In this high temperature limit, using the approximation given by Eq. 4 we find

$$\frac{1}{M} \frac{dM}{d \ln t} = - \frac{\pi D J_c}{c f_b H_{c1} + \pi D J_c} . \quad (7)$$

Now, in the limit as T approaches T_c , the magnetic relaxation tends to zero, rather than diverge, as long as the ϵ -dependence of H_{c1} is weaker than that of J_c . Coupled with the low temperature behavior, this implies a peak in the relaxation rate versus temperature, qualitatively as observed in experiment. It is interesting that this peak can be derived without any distribution of pinning barriers, while earlier work¹⁰ attempted to explain the peak in terms of a pinning barrier distribution. We shall see below that in our approach, rather similar results emerge whether or not one invokes such a distribution.

DISTRIBUTION OF PINNING BARRIERS

Although it is of course less elegant to introduce more parameters, nevertheless invoking a distribution of pinning barriers is a natural extension of the giant flux creep phenomenology. To see why this is of particular importance, we refer to Eq. 1 which reveals a complementarity between $\ln(t)$ and U_T : The curves of Fig. 1, labeled for two different measurement times, could equally well represent two different pinning strengths. Then at a temperature such as 50 K in the figure, J_c from the lower pinning barrier drops off strongly while the higher barrier still makes a substantial contribution. This makes it clear that the high pinning tail of any barrier distribution will dominate the high temperature behavior. As we show below, including a possible pinning distribution leads to some new properties which may be important in comparison to experiment.

Let us then consider such a distribution, taking as a simplest approximation the reduced temperature dependence of each pinning barrier to be the same normalized function u_T , which approaches 1 at $T=0$ and 0 at $T=T_c$. Let U be the amplitude of this temperature-dependent barrier

$$U_T = U u_T . \quad (8)$$

The temperature-independent U -values are assumed distributed according to a distribution function $P(U)$ with the normalization condition

$$\int_0^\infty dU P(U) = 1 , \quad (9)$$

and with an average U defined as

$$\int_0^\infty dU P(U) U = \bar{U} . \quad (10)$$

For specificity, we will use for $P(U)$ the simple mathematical form $(1 + x)^{-n}$, which, properly normalized to satisfy Eqs. 9 and 10, takes the form

$$P(U) = (n - 2)^{n-1}(n - 1)/\bar{U}(n - 2 + U\bar{U}^{-1})^n, \quad (11)$$

Here n cannot equal 1 or 2, and typically will be taken as 3 in the evaluations below. Eq. 11 implies a power-law fall off in $P(U)$ at large U .

Let us now evaluate the total critical current density using Eq. 3 and making the simple assumption that the current density is a weighted average integrated over all values of U . Whenever U is less than a critical value U_c defined by

$$U_c = kT \ln(t/t_0)/u_T, \quad (12)$$

J_c in Eq. 3 becomes very small. Thus, to a good approximation, we can re-write the integral as

$$J_c = \int_{U_c}^{\infty} dU P(U) J_{cT} [1 - (kT/Uu_T) \ln(t/t_0)]. \quad (13)$$

Now, using classical relations,^{16,24,26} we can write the pinning force density (in cgs units)

$$F_p = J_{cT} B/c \approx U_T/a_T V_T, \quad (14)$$

where B is the magnetic induction or flux density, a_T is a flux hopping distance and V_T is the activation volume. For simplicity we assume that the latter two quantities are independent of U . The T -subscripts suggest the possible temperature dependence of these quantities. We see then that J_c is in fact proportional to U_T , and so we can simplify Eq. 13 further:

$$J_c \approx \frac{cU_T}{Ba_T V_T} \int_{U_c}^{\infty} dU P(U) [U - kT/u_T \ln \frac{t}{t_0}]. \quad (15)$$

Performing the now-straightforward integrations, with the specific distribution function of Eq. 11, we find the simple result

$$J_c = \bar{J}_{cT} / (1 + \frac{T}{(n - 2)\bar{T}u_T})^{n-2}, \quad (16)$$

with

$$\bar{J}_{cT} = c\bar{U}u_T/Ba_T V_T, \quad (17)$$

and

$$\bar{T} = \bar{U}/k \ln(t/t_0). \quad (18)$$

Eq. 16 reduces to Eq. 1 in the limit $T \ll \bar{T}$, with J_{cT} and U being replaced by their averaged values \bar{J}_{cT} and \bar{U} . This makes contact with the earlier results, which were at least qualitatively successful in explaining the linear

falloff in J_c with temperature and its strong dependence on effective measurement time. On the other hand, in the opposite limit $T \gg \bar{T}$, Eq. 16 reduces to

$$J_c = \bar{J}_{cT} \left[\frac{(n-2)\bar{U}_{uT}}{k \ln(t/t_0)} \right]^{n-2} . \quad (19)$$

To discuss the resulting temperature dependences it is useful to parametrize the hopping distance a_T and the activation volume V_T as follows:

$$a_T \propto \epsilon^{-m/2} / B^{(1-m)/2} , \quad (20)$$

$$V_T \propto \epsilon^{-\ell/2} / B^{(3-\ell)/2} , \quad (21)$$

where, as before, $\epsilon \equiv 1 - (T/T_c)$ near T_c . Thus, for example, if the hopping distance goes as the coherence length $\xi \propto \epsilon^{-1/2}$, m would be 1, while if this distance were a flux lattice parameter $\sqrt{\Phi_0/B}$, m would be 0. Similarly, the various possibilities for V correspond to different choices of ℓ . Below we will take the choice $m = \ell = 1$ as an example. We also take $U_T \propto (H_c^2/8\pi)V_T$, with the thermodynamic critical field $H_c \propto \epsilon$.

With these relationships and Eq. 19, we can work out the predicted temperature and field dependence of J_c in the high temperature regime. We find

$$J_c \propto \epsilon^{2+0.5m+(n-2)(2-0.5\ell)} / B^{0.5(1+m)+(n-2)(1.5-0.5\ell)} . \quad (22)$$

Thus, for $n=3$, $m=1$ and $\ell=1$, we find $J_c \propto \epsilon^4 / b^2$. A wide variety of dependences have been reported experimentally. Of particular relevance here is slow magnetic data analyzed carefully to include the H_{c1} contribution in Eq. 5. A recent such analysis²⁵ has indeed given evidence of such high powers of ϵ . In spite of this high power, the dependence is considerably more gradual than that predicted by Eq. 3, and so this treatment may help resolve the problem in comparison with experiment discussed earlier. A plot of the full dependence is shown by the dashed lines in Fig. 2 for two choices of measurement times (and using $\epsilon' \equiv 1 - (T/T_c)^2$ rather than $\epsilon \equiv 1 - (T/T_c)$). Clearly, the temperature dependences of J_c in this new model more accurately describe experimental data.

As discussed elsewhere,⁵ the approximate condition for the irreversibility line observed in ac susceptibility experiments is that J_c be a constant determined by the magnitude of the applied ac field. Combining this criterion with Eq. 19, we predict a line with ϵ proportional to

$$\{B^{m+1+(3-\ell)(n-2)} [\ln(t/t_0)]^{2(n-2)}\}^{1/[4+m+(4-\ell)(n-2)]} , \quad (23)$$

which for the same parameter choices as above yields

$$\epsilon \propto B^{1/2} [\ln(t/t_0)]^{1/4} . \quad (24)$$

This power of the dependence on B is not far from the frequently observed $2/3$ power, and the result of course depends on the specific values of n , m and ℓ .

The normalized time-logarithmic relaxation near T_c can be derived following the same steps as for Eq. 7, but now using J_c from Eq. 19

$$\frac{1}{M} \frac{dM}{d \ln t} = \frac{(n-2)\pi D J_c}{[\ln(t/t_0)](c f_b H_{c1} + \pi D J_c)} \quad (25)$$

Thus, except for logarithmic corrections, we recover the time-logarithmic dependence observed in experiment but missing from Eq. 4. In the limit $H_{c1} \rightarrow 0$, this reduces to the temperature-independent value $(n-2)/\ln(t/t_0)$. If we take H_{c1} into account, temperature dependence²⁵ is much weaker than the temperature dependence for J_c derived in Eq. 6. Thus H_{c1} will dominate in the denominator and the normalized relaxation rate will fall to zero at T_c . As before, this implies a peak in the normalized relaxation rate, in qualitative agreement with experiment.

CONCLUSION

In summary, a generalization of the giant flux creep phenomenology to include a distribution of barrier heights offers a new explanation of a number of previously puzzling aspects of the magnetic properties of YBCO, namely the temperature and time-scale dependence of the critical current density, the falloff in the normalized magnetic relaxation rate at high temperatures and its time-logarithmic behavior. The appearance of an irreversibility line in the ac susceptibility can also be derived in a consistent way.

Our goal here has been primarily the development of the phenomenology of flux creep with a barrier distribution, although we have indicated some qualitative comparisons to experiment. Further work on quantitative fits to experimental data must be reserved for a future article. Also many different barrier distributions and averaging schemes are possible and should be explored in comparison to experiment.

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References

1. Y. Yeshurun and A. P. Malozemoff, *Phys. Rev. Lett.* **60**, 2202 (1988).
2. A. P. Malozemoff, T. K. Worthington, Y. Yeshurun, F. Holtzberg and P. H. Kes, *Phys. Rev. B*, to be published.
3. Y. Yeshurun, A. P. Malozemoff and F. Holtzberg, *J. Appl. Phys.*, to be published (Proceedings of the Joint Intermag/3M Conference, Vancouver, Canada, July 12-15, 1988).
4. Y. Yeshurun, A. P. Malozemoff, F. Holtzberg and T. Dinger, *Phys. Rev. B*, submitted.
5. T. K. Worthington, Y. Yeshurun, A. P. Malozemoff, W. J. Gallagher, R. M. Yandroski, F. Holtzberg, T. R. Dinger and D. L. Kaiser, submitted to the International Conference on Magnetism, Paris, France, July 25-29, 1988.
6. T. R. Dinger, T. K. Worthington, W. J. Gallagher and R. L. Sandstrom, *Phys. Rev. Lett.* **58**, 2687 (1987); T. K. Worthington, W. J. Gallagher and T. R. Dinger, *Phys. Rev. Lett.* **59**, 1160 (1987); W. J. Gallagher, T. K. Worthington, T. R. Dinger, F. Holtzberg, D. L. Kaiser and R. L. Sandstrom, *Physica* **148B**, 228 (1987); T. K. Worthington, W. J. Gallagher, T. R. Dinger and R. L. Sandstrom, in *Novel Superconductivity*, ed. S. A. Wolf and W. Z. Kresin (Plenum Press, New York, 1987), p. 781; T. K. Worthington, W. J. Gallagher, D. L. Kaiser, F. H. Holtzberg and T. R. Dinger, *Physica*, to be published (Proceedings of the Interlaken Conference, Switzerland, Feb. 29-Mar. 4, 1988).
7. C. Rossel and P. Chaudhari, *Physica*, to be published.
8. M. Tuominen, A. M. Goldman and M. L. Mecartney, *Physica*, to be published; *Phys. Rev. B* **37**, 548 (1988).
9. S. Senoussi, M. Oussena and G. Collin, *Phys. Rev. Lett.*, submitted; L. Fruchter, M. Oussena, C. Giovannella and I. A. Campbell, preprint.
10. M. Foldeaki, M. E. McHenry and R. C. O'Handley, *Phys. Rev. Lett.*, submitted.
11. M. Tinkham, *Helv. Phys. Acta*, to be published.
12. D. Dew-Hughes, *Cryogenics*, to be published.
13. P. H. Kes, *Physica*, to be published; J. van den Berg, C. J. van der Beek, P. H. Kes, J. A. Mydosh, A. A. Menovsky and M. J. V. Menken, *Physica*, to be published ; R. J. Wijngaarden, K. Heeck, R. Griessen, A. A. Menovsky and M. J. V. Menken, *Physica*, to be published. (Proceedings of the Interlaken Conference, Switzerland, Feb. 29-Mar. 4, 1988).
14. P. W. Anderson, *Phys. Rev. Lett.* **9**, 309 (1962); Y. B. Kim, *Rev. Mod. Phys.* **36**, 39 (1964).
15. M. R. Beasley, R. Labusch and W. W. Webb, *Phys. Rev.* **181**, 682 (1969).
16. A. M. Campbell and J. E. Evetts, *Adv. Phys.* **21**, 199 (1972).

17. C. Ebner and D. Stroud, *Phys. Rev. B* **31**, 165 (1985).
18. I. Morgenstern, K. A. Müller and J. G. Bednorz, *Z. Phys. B* **69**, 33 (1987).
19. R. B. van Dover, L. F. Schneemeyer, E. M. Gyorgy and J. V. Waszczak, *Appl. Phys. Lett.* **52**, 1910 (1988).
20. B. Oh, K. Char, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond and A. Kapitulnik, *Phys. Rev. B* **37**, 7861 (1988).
21. J. Mannhart, P. Chaudhari, C. C. Tsuei, D. Dimos and T. R. McGuire, to be published.
22. C. P. Bean, *Phys. Rev. Lett.* **8**, 250 (1962).
23. H. Ullmaier, *Irreversible Properties of Type II Superconductors* (Springer Verlag, Berlin 1975).
24. M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York 1975).
25. L. Krusin-Elbaum, A. P. Malozemoff, Y. Yeshurun, D. C. Cronmeyer and F. Holtzberg, *Phys. Rev. Lett.*, submitted.