Test for nonreciprocal circular birefringence in Bi₂Sr₂CaCu₂O₈

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(Received 24 May 1991)

A fiber-optic Sagnac interferometer has been used to search for broken time-reversal symmetry in single crystals of $Bi_2Sr_2CaCu_2O_8$. The measurements reveal no nonreciprocal circular birefringence to a sensitivity of 10 μ rad. Data were taken in transmission at 1060 and 670 nm, from 20 to 300 K, in an applied field of up to 200 Oe.

In three recent papers¹⁻³ the proposal of the anyon mechanism^{4,5} for high- T_c superconductivity in the cuprates was tested optically. It has been predicted that the ground state of a system of anyons breaks the symmetries of two-dimensional parity (P) and time reversal (T) in a manner similar to a magnetic material.⁶ These three experiments were motivated by the prediction by Wen and Zee⁷ that an anyon superconductor should exhibit magneto-optic-like effects due to the broken time-reversal symmetry. In particular, one expects to observe the spontaneous Faraday effect in transmission and the spontaneous polar Kerr effect in reflection when a cuprate superconductor is cooled below some transition temperature.

Experimental results to date are inconclusive. Lyons et al.¹ have reported polar Kerr ellipticity on the order of 300 µrad in YBa₂Cu₃O₇ films and microtwinned crystals and in Bi₂Sr₂CaCu₂O₈ single crystals. To measure this small polarization effect in these birefringent materials, a special optical system was constructed to eliminate the effects of linear birefringence. The experiment was carried out at 514 nm wavelength with a spot size of 25 μ m. Upon scanning the laser beam across the sample, an effect of fluctuating sign and magnitude was recorded. The average over several points on the sample was zero, but the finite width in the distribution indicated that the signal at each point was the average of a finite number of domains. Their measurements on Bi₂Sr₂CaCu₂O₈ showed an effect just below room temperature that increased as the temperature was lowered.

Recently, Weber *et al.*³ have claimed another positive result in $Bi_2Sr_2CaCu_2O_8$ and $YBa_2Cu_3O_7$ untwinned single crystals at 633 nm wavelength. However, the data differ both qualitatively and quantitatively from those of Lyons *et al.* The observed effect was two orders of magnitude larger, and the onset for $Bi_2Sr_2CaCu_2O_8$ was at 140 K—about half that reported by Lyons *et al.* Most strikingly, Weber *et al.* found that the rotation was of the same sign over the entire sample, implying that only one domain existed. In their apparatus, linear birefringence was eliminated by aligning the polarization of the light with the principal axis of the sample. In transmission through a 1000-Å $Bi_2Sr_3CaCu_2O_8$ crystal, Weber *et al.* report 5 mrad of polarization rotation, implying a large circular birefringence. Polarization rotation is possible in materials that are T preserving; however, T breaking is suggested by their additional observation that the sign of the effect could be controlled by a small magnetic field, just as in a ferromagnet. It is interesting to note that the size of the rotation they found in both $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$ crystals is of the same order as is found in the best magneto-optic materials.

Measurements at 1060 nm wavelength at Stanford² showed less than 2 μ rad of nonreciprocal circular birefringence (Faraday effect) in transmission through $YBa_2Cu_3O_7$ films ranging in thickness from 50 to 800 Å. The apparatus consisted of a fiber-optic gyroscope (a type of Sagnac interferometer), modified to study Faraday rotation. The phase shift was measured between two beams of light which had traveled through the Sagnac loop and sample in opposite directions. Such an experiment with counterpropagating beams is inherently immune to reciprocal effects such as linear birefringence and the chirality associated with a "handed" material such as quartz. The null result was in apparent contradiction to the experiments of Lyons et al. and Weber et al. However, the difference in wavelength, and the polycrystalline morphology of the films made these results not directly comparable to those of Lyons and Weber.

Here we report measurements of the nonreciprocal circular birefringence at 670 nm in three cleaved crystals of $Bi_2Sr_2CaCu_2O_8$ of thicknesses 600, 1000, and 1500 Å. The $Bi_2Sr_2CaCu_2O_8$ crystals used in this study were grown from a near-stoichiometric melt in a strong thermal gradient using a magnesia crucible. Electron probe microanalysis indicates a composition of $Bi_{2.12}Sr_{1.96}Ca_{0.86}Cu_{2.05}O_{8+\delta}$. Details of the crystal growth method will be reported elsewhere.⁸ Each thin crystal was prepared as follows: A thick crystal was attached to a microscope slide with wax and cleaved repeatedly until a transparent region showed. The wax was dissolved in acetone and the sample floated onto a 200- μ m-spaced copper grid. After the *T*-symmetry measurements, the samples were examined under a polarizing microscope and found to be birefringent and of single crystallographic domain. Cleaving the crystals did not significantly affect their superconducting properties. Magnetometry of the 1000 Å sample revealed a Meissner signal comparable to the best Bi₂Sr₂CaCu₂O₈ samples,

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with $T_c = 85$ K.

The details of the fiber-optic Sagnac interferometer technique have been described previously in our work with the 1060 nm fiber-optic gyroscope.² The counterpropagating beams of a Sagnac interferometer are passed through the sample in opposite directions. The phase shift difference detected when these beams interfere is a measure of the nonreciprocal (T breaking) circular birefringence in the sample. All reciprocal effects (linear birefringence, chirality) cancel out. A new interferometer has been constructed for operation at 670 nm wavelength, in an attempt to duplicate more closely the conditions of the other two experiments, in particular that of Weber et al. at 633 nm. This system operates on the same principle as the 1060 nm gyroscope but is more suited for the study of magneto-optic materials. The loop length is 20 m rather than 1000 m reducing the effects of thermal and acoustic fluctuations. The proportionally higher phase modulation frequency requires an electrooptic modulator, rather than the simpler piezoelectric fiber stretcher described in our earlier work.² This system, which is still under development, has about 5 μ rad of noise in a 1-Hz bandwidth. In addition, an offset of $0-200 \ \mu rad$ exists, which can be nulled out optically,⁹ but drifts on a 20-min time scale by about 20 μ rad.

Measurements were carried out on the 1000 Å sample with 1060 nm radiation and on all three samples with 670 nm radiation. Two techniques were used to search for the effect. With the 1060 nm system, we placed the sample in the beam and monitored the signal as the sample was cooled. For the 670 nm interferometer we first stabilized the temperature, nulled the offset, and then moved the sample into the beam by translating the cryostat. Any change in signal would have indicated broken Tsymmetry. Since the samples are so thin, their insertion does not disturb the path of the beam, and, of course, the linear birefringence cannot disrupt this measurement. With the sample in the beam, a calibration signal of $100-400 \ \mu rad$ was produced by applying a field to a piece of terbium borosilicate glass (FR-5), a Faraday rotator, also in the beam. The crystals were supported in the cryostat vacuum by the copper grids, and cooled in a field of 200 Oe at a typical rate of 5 K per minute. The temperature was measured on the sample holder $\sim 5 \text{ mm}$ from where the copper grid was glued, and data were taken down to 20 K. These conditions are similar to those in the experiment of Weber et al. We saw no nonreciprocal phase shift in $Bi_2Sr_2CaCu_2O_8$ to a sensitivity of 10 μ rad.

The results for the nonreciprocal optical effects reported in Refs. 1-3 and this work are difficult to reconcile with one another. To resolve the discrepancy, one must consider what difference in the experimental techniques might have affected the results. Dispersion seemed to be a possible explanation in reconciling Refs. 1-3, but in light of the work reported here at 670 nm, wavelength difference no longer seems to be a reasonable source of discrepancy. Here, we will consider briefly possible differences between the samples and implications of the differences between reflection and transmission experiments. It is possible that the domain structure of the anyon state varies moderately from sample to sample. However, Weber reports a Faraday rotation 10^3 times larger than the upper limit imposed by this work. To explain the discrepancy, the domains would have to be 10^{-3} of our probe size, i.e., $10^{-3} \times 15 \ \mu m = 150$ Å. Since Weber observed a constant rotation over his entire 1 mm sample, the ratio of the domain size in his sample to that in ours would be approximately 10^5 . It is unclear why such a large difference in domain size should exist between samples of high quality.

Both reflection and transmission measurements have been made in the search for magneto-optic-like effects in the cuprates. The transmission experiments reported here and by Weber³ are sensitive to Δn , the real part of the difference between the index of refraction for left- and right-circularly polarized light. The reflection measurements^{1,3} are sensitive to Δn and the imaginary part Δk as well. In a magneto-optic medium where the real and imaginary parts of the index, n and k, are comparable, Δn and Δk , which both arise from the off-diagonal term in the conductivity tensor, will generally be of the same order. So, barring some accidental degeneracy at both 1060 and 670 nm, the null results reported here and in Ref. 2 cannot be explained by such reasoning.

The possibility of observing a finite effect in reflection but not in transmission was considered by Dzyaloshinskii.¹⁰ If the ordering of the conducting planes is antiferromagnetic, their T-breaking effects would cancel in transmission. However, in Bi₂Sr₂CaCu₂O₈ and YBa₂Cu₃O₇ where there are two planes per unit cell, switching the handedness of all the planes does produce a distinguishable state. Dzyaloshinskii argues that Tbreaking in a two-layer system would result in a signal in a reflection experiment comparable to that of one layer. This effect could account for the difference between our results and those of Lyons et al. In transmission, the P breaking could still be observable as reciprocal circular birefringence; possibly, this is what Weber et al. see.

In conclusion, the only reasonable reconciliation of the various experimental results that is consistent with an anyon mechanism is the possibility that the ordering of the planes is antiferromagnetic. The hypothesis that broken T is responsible for the ellipticity in the Weber and Lyons reflection experiments can be tested by a reflection measurement using a Sagnac interferometer. A null result would be in direct contradiction with these reflection measurements, whereas a positive result would help to resolve the discrepancies in these works.

Useful discussions with B. Halperin, K. Lyons, and R. Laughlin, are gratefully acknowledged. This work was supported by the National Science Foundation through the PYI program (A.K.), by AFOSR Grant No. 91-0145 (S.S.), and by Stanford University through the Office of Technology Licensing. $Bi_2Sr_2CaCu_2O_8$ crystals were made with support from the Stanford Center for Materials Research through the MRL/NSF program.

- ¹K. B. Lyons, J. Kwo, J. F. Dillon, Jr., G. P. Espinosa, M. A. P. Ramirez, and L. F. Schneemeyer, Phys. Rev. Lett. **64**, 2949 (1990).
- ²S. Spielman, K. Fesler, C. B. Eom, T. H. Geballe, M. M. Fejer, and A. Kapitulnik, Phys. Rev. Lett. 65, 123 (1990).
- ³W. W. Weber, D. Weitbrecht, D. Brach, H. Keiter, A. L. Schelankov, W. Weber, Th. Wolf, J. Geerk, G. Linker, G. Roth, P. C. Splittgerber-Hünnekes, and G. Güntherodt, Solid State Commun. **76**, 511 (1990).
- ⁴V. Kalmeyer and R. B. Laughlin, Phys. Rev. Lett. **59**, 2095 (1987).
- ⁵R. B. Laughlin, Phys. Rev. Lett. **60**, 2677 (1988).

- ⁶B. I. Halperin, J. March-Russel, and F. Wilczek, Phys. Rev. B 40, 8726 (1989).
- ⁷X. G. Wen and A. Zee, Phys. Rev. Lett. **62**, 2873 (1989).
- ⁸L. W. Lombardo and A. Kapitulnik, J. Cryst. Growth (to be published).
- ⁹The offset is apparently due to imperfections in the phase modulator. It can be nulled by slight adjustments in the optics which guide the light through the modulator. Alignment of the sample, or its associated optics does not affect the offset.
- ¹⁰I. E. Dzyaloshinskii, TCSUH Workshop on the Physics and Mathematics of Anyons (World Scientific, Singapore, 1991).