

## Test for Nonreciprocal Circular Birefringence in $\text{YBa}_2\text{Cu}_3\text{O}_7$ Thin Films as Evidence for Broken Time-Reversal Symmetry

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We have measured the amount of nonreciprocal circular birefringence of 50 to 800 Å  $\text{YBa}_2\text{Cu}_3\text{O}_7$  films in transmission with a 15- $\mu\text{m}$  beam diameter. A novel instrument with a sensitivity of 2  $\mu\text{rad}$  for nonreciprocal phase shifts was developed by modifying a fiber-optic gyroscope. It is insensitive to reciprocal phase shifts. We observed no nonreciprocal phase shifts in any samples.

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One of the most exciting proposals for the theory of the cuprate high-temperature superconductors (HTSC) is the so-called "anyon superconductivity."<sup>1-3</sup> This theory is based on the concept that in two spatial dimensions, one can obtain not only quantum ideal Bose and Fermi gases, but also quantum ideal gases of new types of particles that interpolate between those two extremes, and hence obey fractional statistics. Those particles are known generically as *anyons*. It was first pointed out by Laughlin and co-workers<sup>4-6</sup> that these gases form superfluids, and become superconductors if the anyons are electrically charged. A striking property of such a superconductor is that its ground state exhibits violation of parity ( $P$ ) and time-reversal ( $T$ ) symmetries.<sup>3</sup> Similar to a magnetic material, an anyon superconductor will exhibit a spontaneous Hall effect and magneto-optical effects. If the cuprates are indeed anyon superconductors then in a transmission experiment through a thin HTSC film, one expects to observe a nonreciprocal rotation of polarization equivalent to the Faraday effect. In reflection, one expects to see an effect resembling the polar Kerr effect. Both effects are nonreciprocal as a consequence of the breakdown of time-reversal symmetry.<sup>1-3</sup>

In a recent experiment, Lyons *et al.*<sup>7</sup> reported such nonreciprocal optical effects in reflection from various HTSC. They constructed an optical system to measure the circular dichroism  $\phi$  which is related to the reflectivity for right and left circularly polarized light by  $R_{\pm} = Re^{\pm\phi}$ . Since one expects the anyon material to break into domains, the averaged nonreciprocal optical effect will be zero. The distribution width  $\sigma$  of the values of  $\phi$  is related to the circular dichroism of a single domain  $\phi_0$  by  $\sigma \sim \phi_0 d/s$ , where  $d$  is the domain size and  $s$  is the beam size. Lyon *et al.* reported a nonzero value for  $\sigma$  that increased from the noise level ( $\sim 25 \mu\text{rad}$ ) at 300 K to  $\sim 250 \mu\text{rad}$  as the temperature was lowered.

In this paper we present data taken on high-quality  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (YBCO) films which indicate no measurable nonreciprocal effect. Using a modified fiber gyroscope we have been able to measure nonreciprocal phase shifts with high sensitivity while simultaneously rejecting

all reciprocal effects. The phase shift we measure is  $\Delta\Phi = 2\pi l(n_+ - n_-)/\lambda$ , where  $n_+$  and  $n_-$  are the indices of refraction for circularly polarized light of the same handedness propagating in opposite directions through the sample. Equivalently  $n_+$  and  $n_-$  could refer to the two circular components of a single beam of linearly polarized light, in which case  $\Delta\Phi$  is related to the Faraday rotation angle  $\theta$  by  $\Delta\Phi = 2\theta$ .

Our experiment on YBCO did not show any nonreciprocal phase shift for any of the samples to a sensitivity of 2  $\mu\text{rad}$ . Depending on the way this value is interpreted, one can put bounds on the fundamental phase shift  $\Delta\Phi_0$  of a single Cu-O plane in the YBCO system. The interpretation of the data depends strongly on the size of the domains, the statistics of their distribution, and how the handedness of the individual planes order with respect to one another. A "ferromagnetic" ordering of the planes would produce a large signal that scales strongly with film thickness, whereas an "antiferromagnetic" ordering would tend to cancel the effect except for the case of an unpaired plane. Assuming that the domains distribute randomly, we can make the following statements for the single-domain shift at a wavelength of 1.06  $\mu\text{m}$ : If the layers order ferromagnetically (all have the same handedness) then  $\Delta\Phi_0 < 0.015s/d \mu\text{rad}/(\text{Cu-O plane})$ . If the layers order antiferromagnetically (alternating handedness) then  $\Delta\Phi_0 < 2s/d \mu\text{rad}/(\text{Cu-O plane})$ .

The films were deposited *in situ* on double-side-polished MgO substrates at 680°C. Our standard sputtering technique and the typical properties of the films have been discussed before.<sup>8</sup> The surfaces of these films are extremely smooth; no structure is observed under a scanning electron microscope with a lateral resolution of 100 Å.<sup>9</sup> The film thicknesses used for the experiment were 50, 200, 500, and 800 Å. (50 Å of amorphous YBCO was deposited *in situ* on the 50-Å film for protection.) The films are oriented with the  $c$  axis perpendicular to the substrate. The distance between twin boundaries is typically 100–500 Å and the grain size is 500–2000 Å. The transition temperatures of the 200-, 500-, and 800-Å films were 83.5 ( $\Delta T_c = 1.0$  K), 85 ( $\Delta T_c$

$=0.5$  K), and 85 K ( $\Delta T_c = 0.3$  K), respectively.

Since the expected magnitude of the signal is very small, special care must be taken to reject any undesired effects. To this end, we have developed a novel instrument consisting of a fiber-optic gyroscope modified to measure nonreciprocal circular birefringence. The fiber gyroscope has undergone much development since its first demonstration in 1976,<sup>10</sup> and is reviewed in several articles.<sup>11-13</sup> Briefly stated, the fiber-optic gyroscope consists of two counterpropagating optical beams in a fiber Sagnac interferometer. If the system undergoes no mechanical rotation, the beams that emerge from the Sagnac loop interfere constructively. If the system is rotating, the beams see different path lengths, resulting in a phase shift proportional to the rotation rate. When the fiber Sagnac interferometer is constructed in the minimum reciprocal configuration,<sup>14</sup> all reciprocal optical effects in the Sagnac loop will cancel out, leaving only nonreciprocal effects to affect the interference of the light at the output. The primary nonreciprocal effect of interest for the fiber gyroscope is obviously mechanical rotation; however, other nonreciprocal optical effects in the Sagnac loop, such as the Faraday effect,<sup>15</sup> can result in a phase shift in the counterpropagating beams of light.

The gyroscope used in this experiment is being developed independently, and will be described in much greater detail in a future publication. The layout of the apparatus is shown in Fig. 1(a). Light from the 1.06- $\mu\text{m}$

source passes through a directional fiber coupler, a linear polarizer, and then is split by a second directional coupler to propagate in both directions through the loop. 1 km of polarization-maintaining single-mode (at 1.06  $\mu\text{m}$ ) optical fiber is wound on a 20-cm-diam spool with a piezoelectric phase modulator<sup>11</sup> driven at 100 kHz at one end of the coil. The output of the fiber gyroscope was detected with a silicon *p-i-n* photodiode, and the 100- and 200-kHz components of the output corresponding to the first and second harmonics of the reference were measured with lock-in amplifiers with a time constant of 1 to 10 sec. In the phase-modulated Sagnac interferometer, the first harmonic is  $k_1 I_0 \sin(\Delta\Phi)$ , which for small nonreciprocal signals (or low mechanical rotation rate) is approximately  $k_1 I_0 \Delta\Phi$ . The second harmonic is  $k_2 I_0 \times \cos(\Delta\Phi)$ , which for small signals is approximately  $k_2 I_0$ .  $I_0$  is the optical power and  $k_1$  and  $k_2$  are determined by the system parameters. The first-harmonic scale factor  $k_1 I_0$  can easily be determined by rotating the gyroscope at a known rate. It is convenient to use the 15°/h Earth-rotation rate to generate a calibration signal for the first harmonic. Any changes in the optical power level, which may be more pronounced in this experiment than when the system is configured solely as a gyroscope, can be determined by monitoring the second-harmonic output. It is important to mention that the gyroscope, in its present configuration, has a constant dc offset of not more than 50  $\mu\text{rad}$  that is affected by internal losses in the system. This offset, which can vary from day to day (due to variations in the room temperature or the alignment of the fibers), can be zeroed by adding a constant phase shift to the system (e.g., by using the Earth rotation), or can serve as the reference value for the measurement. In all our measurements the offset was determined at the beginning of the experiment and stayed constant (to within our sensitivity) throughout the whole experiment including the temperature scan and the *x-y* translations of the sample.

For the purpose of searching for nonreciprocal effects in a thin film, the bulk-optic section shown in Fig. 1(b) was inserted into the Sagnac loop. The linearly polarized light from the fiber was collimated with a microscopic objective, converted into right circularly polarized light by a  $\frac{1}{4}$ -wave plate, and focused onto the sample. The transmitted light was then converted back into the original linear polarization by another  $\frac{1}{4}$ -wave plate before being coupled into the other fiber end. Note that light propagating in the opposite direction along the same path will also be right circular in the central portion of the bulk-optics setup. A minor misalignment of the  $\frac{1}{4}$ -wave plates, while losing signal by coupling light into the wrong polarization mode of the fiber [which is eventually rejected by the polarizer in Fig. 1(a)], cannot create a nonreciprocal effect. The  $1/e^2$  beam diameter was determined to be 15  $\mu\text{m}$ , with a depth of focus of  $\sim 0.5$  mm. The cryostat was mounted on an *x-y-z* stage

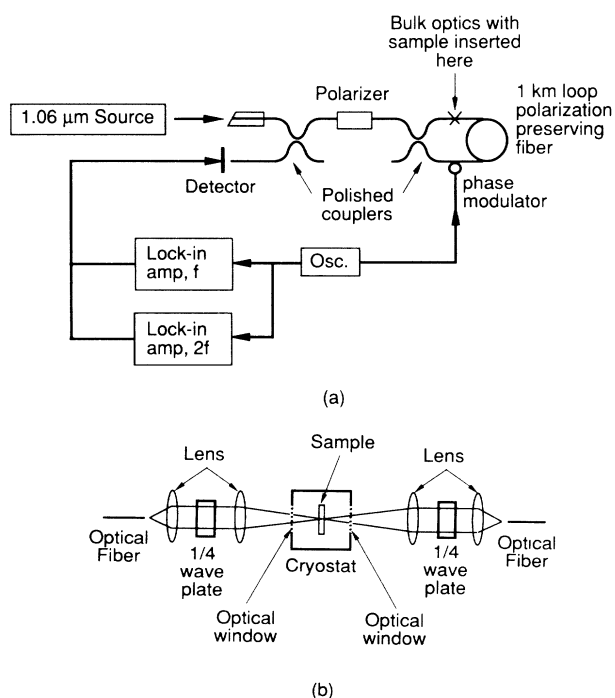


FIG. 1. The optical apparatus used for the experiment. (a) The fiber-gyroscope circuit and (b) the part that is inserted into the gyroscope loop as described in the text.

such that several samples and many points on the same sample could be scanned.

Since both beams have the same handedness, the apparatus is insensitive to an optically active material (i.e., a chiral molecule like sucrose) whose index depends on the handedness of the light, but not the direction of propagation. Furthermore, a linearly birefringent material, while coupling some light into the other polarization mode, would still behave identically in both propagation directions, and the beams would again emerge with no relative phase shift. However, in the case of the Faraday effect, the refractive index of the material depends not only on the handedness of the light, but also on the direction of propagation. The apparatus is inherently sensitive only to nonreciprocal processes like the Faraday effect.

We have performed several tests to ensure that our instrument rejects any signals due to reciprocal effects and at the same time is capable of measuring nonreciprocal phase shifts of the type proposed by the anyon theory. It was shown recently by Schlesinger and Collins<sup>16</sup> that in the  $a$ - $b$  plane of  $\text{YBa}_2\text{Cu}_3\text{O}_7$ , the dielectric constant, even for visible light, is highly anisotropic. Thus a strong linear birefringence is expected. The following materials served as tests for rejection of spurious effects: (1) a polypropylene sheet of thickness  $\sim 0.2$  mm exhibiting strong linear birefringence; (2) an aqueous solution of sucrose (10 g/100 ml), in a cuvette of  $\sim 1.5$  mm thickness ( $\sim 1$  mrad of reciprocal optical rotation was expected); (3) freshly polished MgO substrates; and (4) MgO substrates on which YBCO was grown and then etched away, to simulate the effects from the high-temperature cycle of the film deposition or any surface effect due to the interface with YBCO. All the above tests showed null results.

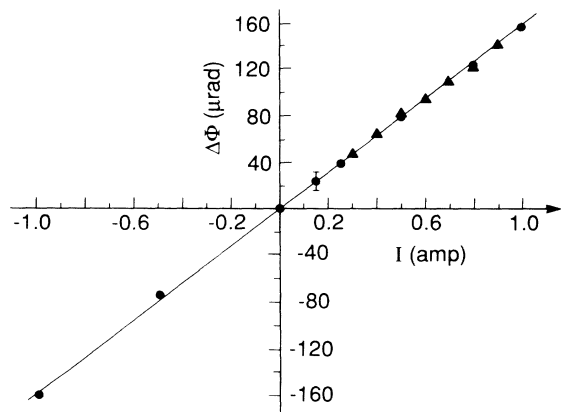


FIG. 2. Calibration signal from FR-5 glass as a function of the electromagnet coil current. The conversion is 30 G/A. The error bar denotes the peak-to-peak variation of the signal. Note that the data represent twice the Faraday rotation angle, as discussed in the text.

Of course, equally important is to calibrate the apparatus for nonreciprocal rotation by using optical materials, such as magnetic materials, exhibiting the Faraday effect to verify the quoted sensitivity. To calibrate our apparatus, we have used two procedures. We aligned the fiber gyroscope along and perpendicular to the axis of rotation of the Earth as described earlier. Also, a 1-mm-thick plate of terbium borosilicate (FR-5) glass with a Verdet constant at  $1.06 \mu\text{m}$  of  $2.1 \mu\text{rad}/\text{Gmm}$  (corresponding to a relative phase shift of  $4.2 \mu\text{rad}/\text{Gmm}$ ) was inserted into the apparatus. Figure 2 shows typical data for the nonreciprocal phase shift when a magnetic field (using a Cu solenoid and placing the FR-5 in its center) is applied along the beam direction. Note that the direction of the effect reverses appropriately when we reverse the magnetic-field direction. The phase shift measured agreed with the tabulated value to within 10%.

The YBCO samples were measured as a function of temperature in the range 11 to 300 K. The temperature was controlled to better than  $\pm 1\%$  and at each temperature six to twelve points selected from a  $2 \times 2\text{-mm}^2$  section of the sample were measured. Data were also recorded during the transitions between temperatures. Thus, the thermal expansion of the sample holder (typically  $\sim 5 \mu\text{m}/\text{K}$ ) served as another scanning procedure on the sample. A typical set of data on an  $800\text{-\AA}$  film at 12.3 K is shown in Fig. 3. An Earth-rotation calibration is also shown for reference. The rms value of  $\Delta\Phi$  is  $\sim 0.2$  times the peak-to-peak value of the data, which implies an rms noise of  $2 \mu\text{rad}$  for our 10-sec averaging time. As is evident from the data in Fig. 3, which are similar to all our measurements on the YBCO films, no nonreciprocal effect is observed.

The results presented here contradict those of Lyons *et al.*<sup>7</sup> There are a few major differences between the two experiments. The first is the wavelength of the light used

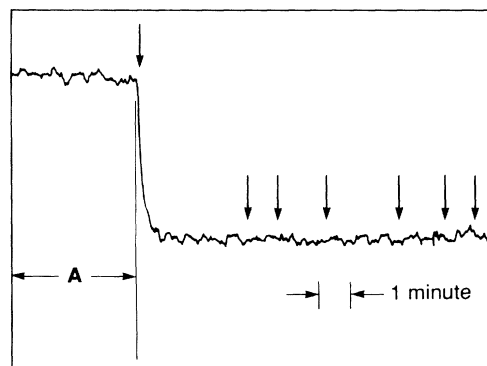


FIG. 3. Region A is an Earth-rotation calibration of  $100 \mu\text{rad}$  ( $5^\circ/\text{h}$  mechanical rotation) followed by a typical scan of six different positions of the sample. Arrows denote the instant when a new beam location was obtained. Typical distance between the spots is  $100 \mu\text{m}$ .

in our experiment (Lyons *et al.* have used a 514-nm laser). We do not believe this should make a difference unless a resonance effect occurs,<sup>16</sup> given the large detuning of both wavelengths from the gap frequency. A second difference is the transmission mode that we have been using as compared to the reflection mode of Ref. 7. The magnitude of the reflectivity  $R_{\pm}$  depends on both the real and imaginary parts of the index of refraction  $n_{\pm}$  and  $k_{\pm}$ , while the phase shifts reported in this paper depend only on  $n_{\pm}$ . However, since  $n_{\pm}$  depend on both the real and imaginary parts of the dielectric tensor, only an accidental degeneracy of  $n_{\pm}$  could reconcile the results of the two experiments on the grounds that a different quantity was measured. As described above, our results restrict the phase shift  $\Delta\Phi_0$  of a single Cu-O plane in YBCO to an upper bound that depends on the domain size and the ordering of the planes. More theoretical work addressing these unknowns is needed before the consistency of our results with an anyon description of HTSC can be determined.

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