Absence of Circular Dichroism in High-Temperature Superconductors

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We have repeated and improved upon the experiment of Lyons *et al.*, in which the observation of circular dichroism in high-T_c superconductors was reported. We found a simple extension to the previous technique which makes the apparatus *exclusively* sensitive to T-violating circular dichroism. We find no evidence for a temperature-dependent signal in all $YBa_2Cu_3O_{7-\delta}$ samples studied. We establish sensitivity of the apparatus to surface roughness and ascribe the previously observed signal to possible surface contamination.

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The microscopic origin of high-T_c superconductivity is a hotly debated issue. A view proposed by one of us [1] asserts that the condensation of particles obeying fractional statistics is responsible. Using this model, Wen and Zee [2] predicted that the optical properties of these materials should exhibit the underlying violation of the symmetries of parity (P) and time reversal (T). In a recent Letter, Lyons *et al.* [3] reported the observation of circular dichroism in the reflectivity of these materials, apparently confirming this prediction. However, as they discussed, their apparatus did not exclusively measure T-violating circular dichroism. Spielman *et al.* subsequently reported that they observed no signal, in transmission and reflection, with an apparatus sensitive only to T-violating circular *birefringence* [4]. Since these two experiments measured different quantities, the results of Spielman *et al.* did not conclusively refute or elucidate the nature of the results of Lyons *et al.* Hence, a number of recent Letters have been devoted to the theoretical implications assuming that Lyons' data are a clear signal of T violation [5]. The purpose of this Letter is to reconcile these experiments. We have carefully reproduced the apparatus of Lyons *et al.*, and discovered a very simple ratio technique that eliminates the source of non-T-violating signal. This improves the sensitivity, in some cases, by as much as a factor of 10, giving an absolute error of 2 μrad. We have discovered that the major source of error in such an apparatus is inhomogeneity in the detection optics. This causes the apparatus to be particularly sensitive to surface contamination, because roughness of the sample's surface induces nonuniformities in the reflected beam. We find no temperature-dependent signal from a variety of $YBa_2Cu_3O_{7-\delta}$ samples studied, provided the sample surface is kept clean. If not, we observe signals similar to those reported by Lyons, and these signals are greatly enhanced when the new ratio technique is *not* used.

The apparatus, illustrated in Fig. 1, ideally measures the imaginary part of the quantity γ, defined in terms of the sample's reflectivity matrix by

$$\mathbf{R}=\begin{pmatrix} R_{xx} & R_{xy} \\ R_{yx} & R_{yy} \end{pmatrix} = R \begin{pmatrix} 1+\alpha & \beta+\gamma \\ \beta-\gamma & 1-\alpha \end{pmatrix}. \tag{1}$$

γ is identically zero for any sample that does not violate time-reversal symmetry [6]. The basic idea is to deliver light of a known polarization to the sample surface and measure the light reflected in the orthogonal channel. The two polarizers in Fig. 1 are accurately crossed. If the Faraday modulator (FM) and half-wave plate (HWP) are removed, the light intensity I_d, detected by the photodiode, is proportional to the quantity $|\beta-\gamma|^2$. With the quarter-wave plate (QWP) in place and the FM driven with amplitude η at frequency Ω this becomes

$$I_d \propto |[1+\alpha]\eta\sin(\Omega t)+i[\beta-\gamma]|^2. \tag{2}$$

The QWP axis is accurately aligned with the input polarization axis, adding the i to the second term. Lyons *et al.* detected this signal with a lock-in amplifier, yielding

$$\phi_{CD}^{old}=\mathrm{Im}\{\gamma-\beta+\alpha\beta^*-\alpha\gamma^*\}. \tag{3}$$

Rotating the HWP averages over polarization directions, giving

$$\langle\phi_{CD}^{old}\rangle=\mathrm{Im}\{\gamma+\alpha\beta^*\}. \tag{4}$$

In our apparatus, however, the signal input to the lock-in detector is I_d/I_0, where I_0 is the intensity of the light rejected by the final polarizer, given by

$$I_0 \propto |1+\alpha-i[\beta-\gamma]\eta\sin(\Omega t)|^2. \tag{5}$$

Instead of Eq. (3), we then have [7]

$$\phi_{CD}^{new}=\mathrm{Im}\{\gamma-\beta+\alpha\beta-\alpha\gamma\}, \tag{6}$$

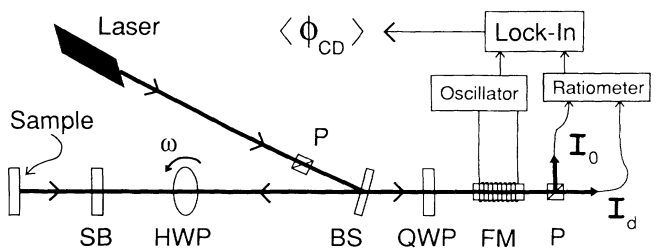

FIG. 1. Schematic of our experimental layout showing all major components: P, polarizer; BS, beam splitter; HWP, half-wave plate; SB, Soleil-Babinet compensator; QWP, quarter-wave plate; FM, Faraday modulator.

which leads to

$$\langle \phi_{CD}^{new} \rangle = \mathrm{Im}\{\gamma\} . \quad (7)$$

Thus, the addition of the ratiometer is crucial.

We used a diode-laser-pumped, single-frequency, single-mode, doubled Nd:YAG laser ($\lambda = 532$ nm). Our polarizers were Glan-Thompson calcite prisms with an extinction ratio of 2×10^{-7}. We used a multiorder HWP mounted on a rotational stage with a temperature controller. By stabilizing the HWP temperature, we obtained an extinction ratio of 10^{-4}, and improved this to 10^{-5} by including a stress-compensating element on the rotating assembly [7]. The QWP was zero order and had an extinction ratio of 10^{-5} and a phase delay error of 3×10^{-2} (which contributes only in third order). We used a $\lambda/50$, diffraction-limited lens. The FM was driven at a frequency of 1 kHz and had an extinction ratio of 3×10^{-6}. We calibrated our measurements by rotating the final polarizer by known amounts and by adding a dc current to the FM. We did *not* use the FM to null the measured signals, because we found that this introduced thermal birefringence.

Figure 2(a) demonstrates the insensitivity of the apparatus to linear birefringence. We substitute a polished silicon mirror for the sample and adjust the Soleil-Babinet (SB) compensator to introduce 10 mrad of linear birefringence. When the lock-in time constants are small, its output, ϕ_{CD}, oscillates with a 10-mrad amplitude at 4 times the 1-Hz rotation frequency of the HWP. When the time constants are increased, the amplitude becomes comparable with the systematic "drift" error of 5 μrad, as shown in the inset. In an actual experiment, the overall linear birefringence is compensated by the SB to less than 2–3 mrad.

Figure 2(b) demonstrates the effectiveness of the ratiometer. The experiment is the same as in Fig. 2(a), except that an optical wedge is inserted between the HWP and SB and tilted by an angle θ. This introduces an amount of linear dichroism, p, that can be calculated using Fresnel's formulas. With $\theta = 20°$, an index of refraction of 1.5, and wedge angle of 0.5°, we obtain $p = 13$ mrad. The $\alpha\beta^*$ term in Eq. (3) is then

$$\mathrm{Im}\{\alpha\beta^*\} = pg\sin[2\phi] = 12.6g(\mu\mathrm{rad}) , \quad (8)$$

where g is the linear birefringence introduced by the SB compensator (measured in mrad) and $\phi = 35°$ is the angle between the tilt axis of the wedge and the principal axis of the compensator. In Fig. 2(b) the linear birefringence setting of the SB is changed every 30 s. With the ratiometer off, the measured signal agrees with Eqs. (4) and (8). With the ratiometer on, the signal is zero, consistent with Eq. (7).

Figure 2(c) demonstrates that the apparatus detects circular dichroism. We configure the system as in Fig. 1, but without the lens and SB, and use a 1-cm-thick, polished steel disk as the sample. A bar magnet is placed

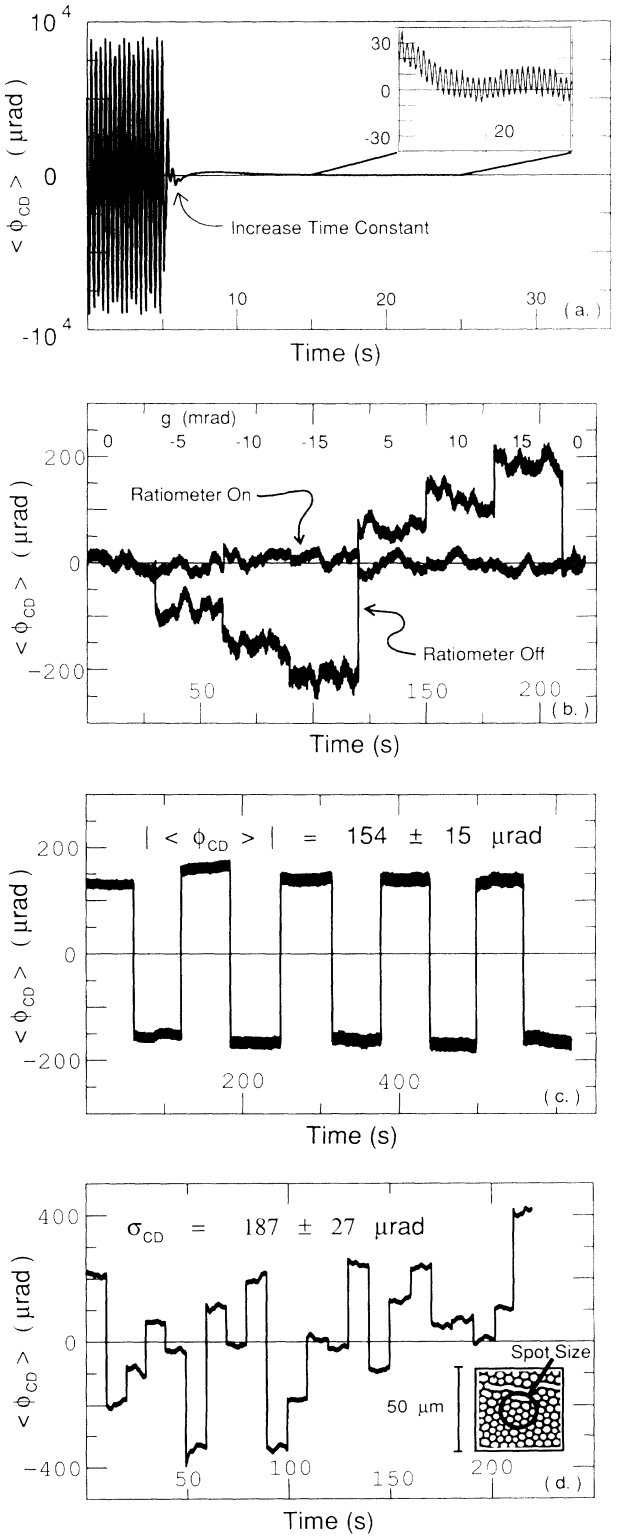

FIG. 2. Calibration measurements: (a) averaging of linear birefringence by increasing lock-in time constant; (b) discrimination of the $\mathrm{Im}\{\alpha\beta^*\}$ term with the ratiometer; (c) measurement of circular dichroism from an iron sample with an alternating 640 G applied field; (d) measurements on a YIG sample (inset—polarized light micrograph of the sample).

behind the sample and reversed every 60 s. The measured $\langle\phi_{CD}\rangle$ agrees well with the 150 μrad expected for the ± 640 G field strength measured at the sample surface [8].

Figure 2(d) demonstrates that our focus on the sample is near the diffraction limit. We focus the light onto a thin film of yttrium iron garnet (YIG). We move the spot to a new location every 10 s. The measured standard deviation agrees roughly with the Gaussian expression [7]

$$\sigma_{CD}=(\overline{\langle\phi_{CD}\rangle^2})^{1/2}\sim\frac{2}{3}\frac{dD}{\lambda f}\langle\phi_{CD}^{sat}\rangle, \quad (9)$$

where $\langle\phi_{CD}^{sat}\rangle=1100$ μrad is the measured value of the saturated single-domain circular dichroism signal, $d=1.8$ μm is the effective domain size, $D=4.5$ mm is our beam diameter, and $f=5.3$ cm is the focal length of the lens.

The exact nature and size of the focused beam on the superconducting samples was determined accurately by two other measurements. First, we replaced the sample with a telescope and projected the spot onto the wall. We found an Airy profile with a 15-μm spot size. Second, we monitored the autocollimation of the reflected beam during measurements. This constrained the lens position to ± 0.1 mm, corresponding to 1.4 Rayleigh depths, so that the beam diameter at the sample was ≤ 20 μm.

We now consider measurements of σ_{CD} for high-T_c superconductors. The setup is that of Fig. 1, with a superconducting sample placed at the lens focus and cooled with a cold-finger cryostat. Vacuum is maintained at 10^{-6} torr. For each temperature we measure $\langle\phi_{CD}\rangle$ at $N\approx 25$ points, compute the standard deviation, σ_{CD}, and assign an error bar of $\sigma_{CD}/\sqrt{2N}$. Figure 3 shows typical results. Figure 3(a) is for an unetched, untwinned, single crystal of $YBa_2Cu_3O_{7-\delta}$ (YBCO). Figure 3(b) is for an 800-Å film of YBCO on an MgO substrate. For comparison, data reported by Lyons *et al.* on similar samples are also shown. Our data consistently show a complete lack of temperature dependence within the error of the measurement. There is, however, a "background" signal due to inhomogeneities in the system, as discussed below. Lyons *et al.* reported the presence of such a background, but subtracted it from their data.

Table I shows the "background" σ_{CD} for all of the samples we measured, including a polished silicon reference. For the superconducting samples, which were grown by standard methods [9], the transition temperatures and widths are shown as obtained with resistance, mutual inductance, and magnetometry measurements. Samples 2–5 were characterized both before and after our experiments and showed no significant changes in superconducting properties.

The background σ_{CD} has the same physical origin as the drift seen in the inset of Fig. 2(a). Table I shows that σ_{drift}, defined as the standard deviation of the time-dependent signal observed when the spot is left in a single position on the sample, is equal to σ_{CD} in all cases studied. In addition, the size of both signals correlates with the amount of scattered light observed at large angles. We conclude that both are caused by surface roughness.

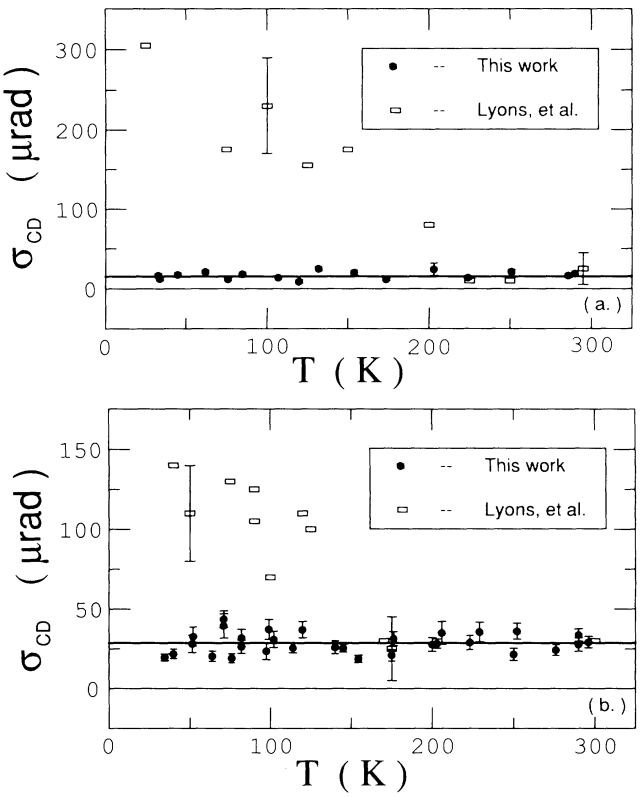

FIG. 3. (a) Our data on an untwinned, single crystal of YBCO, compared with data of Lyons *et al.* [3], on a twinned, etched, single crystal of YBCO. (b) Our data on an 800-Å film of YBCO on MgO, compared with data of Lyons *et al.* [3] on 500-Å films of YBCO on MgO.

The time dependence of the drift comes from modal and pointing fluctuations in the laser. We verified this by inserting a 1-m length of single-mode optical fiber be-

TABLE I. Samples which we measured along with their superconducting transition temperatures and widths, and the value of the temperature-independent σ_{CD} and σ_{drift}.

Sample	T_c (K)	ΔT (K)	σ_{CD} (μrad)	σ_{drift} (μrad)
1. Polished silicon	···	···	6	5
2. YBCO (crystal)	90.5	<1.5	15	15
3. 3000-Å YBCO/MgO	82	1.5	18	···
4. 800-Å YBCO/MgO	87	0.7	29	22
5. 500-Å YBCO/MgO	87	0.7	59	···
6. 2000-Å YBCO/YSZ [a]	89	1.1	145 [b]	···

[a]Yttrium-stabilized zirconia.

[b]No temperature scan was done for this sample.

tween the laser and the input polarizer and observing a reduction in σ_{drift} by a factor of 3. In addition, we eliminated coherent interference effects between optical surfaces as a possible source of this time dependence by mechanically tapping and heating each optical element and observing no changes in the measured signal.

Surface roughness and laser drift communicate to the detector via inhomogeneities in the detection optics. Four measurements support this conclusion: (1) We observed a large (≈ 200 μrad) drift signal when another SB compensator was used as the QWP; (2) we observed a "signal" of ≈ 50 μrad in $\langle\phi_{\text{CD}}\rangle$ when the reflected beam was translated with respect to the detection optics by roughly a quarter beam diameter; (3) we observed a 100-μrad change in $\langle\phi_{\text{CD}}\rangle$ when we intentionally induced model changes in the laser, which was eliminated by installing the single-mode fiber; and (4) we observed no significant change in σ_{drift}, with a mirror as the sample, when the HWP and lens were removed, showing that "steering" of the beam by the HWP was not the source of σ_{drift}. Thus, changes in the position of the beam with respect to the detection optics result in different averaging over inhomogeneities in the latter, giving a new baseline ϕ_{CD}, and this effect is enhanced by surface-roughness-induced intensity nonuniformities.

Temperature-dependent changes in σ_{CD} are only seen in our apparatus when the cryostat vacuum is bad. In light of the sensitivity of the apparatus to surface roughness, these can be understood as changes in the surface topography resulting from condensation. In a poor vacuum of 10^{-3} torr, *without* using the ratiometer technique, σ_{CD} on sample 4 was seen to increase by 50 μrad at 200 K and by 25 μrad at 80 K. These temperatures correspond to the condensation of ice and air, respectively, at this pressure. Use of the ratiometer suppressed these effects by a factor of ≈ 3. The particular shape of the data observed depended on the history and environment of the sample and the increases were not seen with the smooth reference (sample 1). We observe that the characteristic temperature at which the signals reported by Lyons *et al.* appear is 200 K and that they have also reported an additional feature at ≈ 80 K [10]. In addition, recent measurements by Lyons *et al.* made with a new, more complex method of obtaining Eq. (7) are inconclusive and include a variety of null results [11].

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