

Polar Kerr-Effect Measurements of the High-Temperature $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ Superconductor: Evidence for Broken Symmetry near the Pseudogap Temperature

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The polar Kerr effect in the high- T_c superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ was measured at zero magnetic field with high precision using a cyogenic Sagnac fiber interferometer. We observed nonzero Kerr rotations of order $\sim 1 \mu\text{rad}$ appearing near the pseudogap temperature T^* and marking what appears to be a true phase transition. Anomalous magnetic behavior in magnetic-field training of the effect suggests that time reversal symmetry is already broken above room temperature.

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One of the most challenging puzzles that has emerged within the phenomenology of the high-temperature superconductors (HTSC) is to understand the occurrence and role of the normal-state “pseudogap” phase in underdoped cuprates [1]. This phase exhibits anomalous behavior of many properties including magnetic [2], transport [3], thermodynamic [4], and optical properties [5] below a temperature, T^* , large compared to the superconducting (SC) transition temperature, T_c . Two major classes of theories have been introduced in an attempt to describe the pseudogap state: One in which the pseudogap temperature T^* represents a crossover into a state with preformed pairs with a d wave gap symmetry [6,7], and another in which T^* marks a true transition into a phase with broken symmetry that ends at a quantum critical point, typically inside the superconducting dome. While at low doping this phase may compete with superconductivity, it might provide fluctuations that are responsible for the enhanced transition temperature near its quantum critical point (e.g., as in Ref. [8]). Examples include competing phases of charge and spin density waves [9], or charge current loops that either do [10] or do not [8,11] break translational symmetry.

In this Letter, we report high resolution optical Kerr-effect measurements on $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ crystals with various hole concentrations p . (p is, in turn, a monotonic function of the oxygen concentration x , and it also depends on oxygen ordering in the chains [12].) We identify a sharp phase transition at a temperature $T_s(p)$, below which there is a nonzero Kerr angle, indicating the existence of a phase with broken time reversal symmetry (TRS). Both the magnitude and hole concentration dependence of T_s are in close correspondence with those of the pseudogap crossover temperature, T^* , which has been identified in other physical quantities. In particular, as shown in Fig. 1, T_s is

substantially larger than the superconducting T_c in underdoped materials, but drops rapidly with increasing hole concentration, so that it is smaller than T_c in a near optimally doped crystal and extrapolates to zero at a putative quantum critical point under the superconducting dome. The magnitude of the Kerr rotation in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (YBCO) is smaller by ~ 4 orders of magnitude than that observed in other itinerant ferromagnetic oxides [13,14], and the temperature dependence is “superlinear” near T_c ,

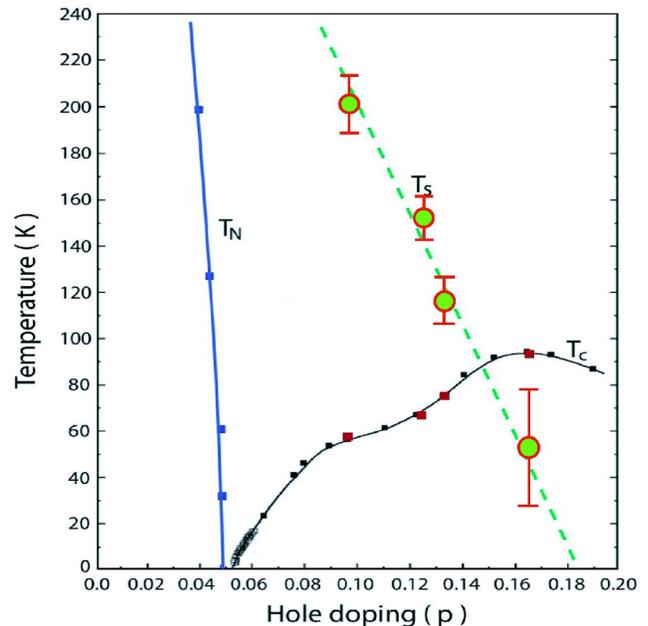


FIG. 1 (color online). The onset of the Kerr-effect signal, T_s (circles), and T_c (red squares) for the $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples reported in this Letter. Also shown are $T_c(p)$ (from [12]) and $T_N(p)$ (from [22]).

suggesting that either we are not directly measuring the principal order parameter that characterizes the pseudogap phase in YBCO or we measure its very small “ferromagneticlike” component. In addition, we find a hysteretic memory effect that seemingly implies that TRS is broken in all cases at a still higher temperature (above room temperature), although no Kerr effect is detectable within our sensitivity at temperatures above T_s .

High quality $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ single crystals with $x = 0.5$ (ortho-II, $T_c = 59$ K), $x = 0.67$ (ortho-VIII, $T_c = 65$ K), $x = 0.75$ (ortho-III, $T_c = 75$ K), and $x = 0.92$ (ortho-I, $T_c = 92$ K) were grown by a flux method in BaZrO_3 crucibles [12]. The crystals, in the form of (ab plane) platelets several millimeters on a side and a fraction of a millimeter thick (c direction), were mechanically detwinned. X-ray diffraction measurements indicate long uninterrupted chain lengths (e.g., for ortho-II this length is $\sim 120 \times b$ [12]). The polar Kerr-effect measurements were performed using a zero-area-loop polarization-Sagnac interferometer at wavelength of $\lambda = 1550$ nm [15]. The same apparatus was previously used to detect (a noticeably weaker) TRS breaking below T_c in Sr_2RuO_4 [16]. The typical performance was a shot-noise limited $0.1 \mu\text{rad}/\sqrt{\text{Hz}}$ at $10 \mu\text{W}$ of incident optical power from room temperature down to 0.5 K.

Crystals were mounted on a copper plate using GE varnish. The system was aligned at room temperature, focusing the beam that emerges out of the quarter-wave plate to a $\sim 3 \mu\text{m}$ size spot [15]. A measurement cycle was then used in which the sample was first cooled in a field, the field was turned off at the lowest temperature (4.2 K), and the Kerr effect of the sample was measured while the sample was warmed to room temperature. Figure 2 shows the Kerr effect measured on $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ after cooling the sample in a field of 4 T. Three regimes are clearly observed. The low temperature Kerr effect is very large, indicating a large contribution from trapped vortices. This contribution, which follows the direction of the magnetic field, decays exponentially with increasing temperature and, at T_c , reaches a finite value that is of order $\sim 1 \mu\text{rad}$. This clearly indicates a new, unexpected state with a small but finite ferromagneticlike signal. As we continue to warm the sample above T_c , that remnant signal decreases until it disappears at a higher temperature denoted by T_s . Above T_s the Kerr signal is zero to within our sensitivity (± 30 nanorad). We emphasize that this is a true zero as all Kerr data shown in this Letter is raw, without any baseline subtraction. Error bars in all figures are one-sigma statistical error. For the sample in Fig. 2 we find $T_s = 155 \pm 5$ K.

One of the key features of a broken symmetry state is its sensitivity to small symmetry breaking fields. This leads to characteristic hysteresis loops. In particular, a ferromagnet cooled below its critical temperature in a small field will remain magnetized even when the field is turned off unless

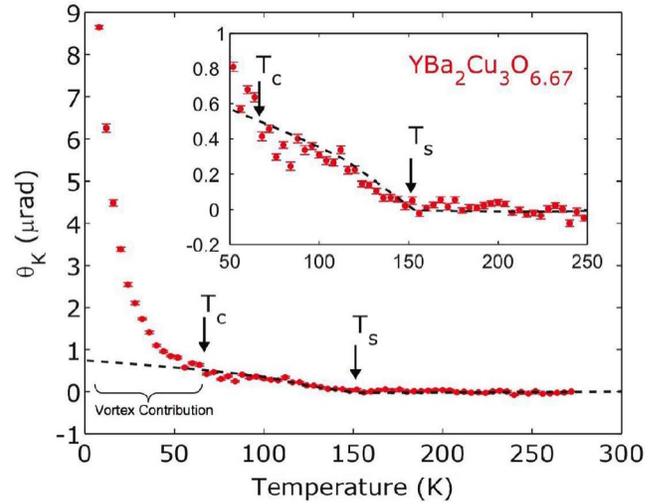


FIG. 2 (color online). The Kerr effect of $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ crystal. The sample was first cooled to 4.2 K in a +4 T field. The field was turned off at 4.2 K, and measurements were taken while warming the sample. Note the large vortex contribution that disappears just before $T_c = 65$ K. The inset shows the region above T_c with its zero baseline, indicating a finite Kerr signal that disappears at $T_s = 155$ K. Dashed lines are guides to the eye.

the temperature is subsequently raised above T_c , or an opposite field in excess of a certain temperature dependent “coercive field” is applied to reverse the magnetization. Therefore, to elucidate the character of the broken symmetry state detected in the present experiments, we have measured the history dependence of the Kerr rotation following several different protocols. The data in Fig. 3 were obtained on four different crystalline samples with $x = 0.92, 0.75, 0.67,$ and 0.5 as follows: First, the sample

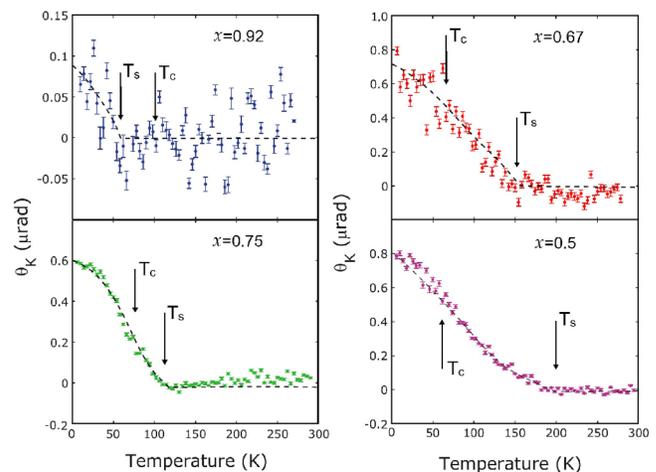


FIG. 3 (color online). The Kerr effect of zero-field warm-up for $x = 0.92, 0.75, 0.67,$ and 0.5 . All samples were cooled and measured in a magnetically shielded environment (field < 3 mOe). Dashed lines are guides to the eye.

was “trained” in a 4 T field in the “up” direction at room temperature. The field was then turned off, and the sample was cooled in zero field to 4.2 K [17]. All zero-field measurements were done in conditions where all magnets were open loops at room temperature, and the system was cooled in a double μ -metal-shield environment with a remnant field < 3 mOe, measured separately at the position of the sample.

The Kerr signal was measured upon warming, still in zero field (ZFC-ZFW). It is clear that for all four samples there is a temperature T_s at which a finite signal disappears when the temperature is raised. Moreover, it is evident that while for highly underdoped samples $T_s > T_c$, near optimal doping no signal is observed above and through T_c but rather appears at a temperature well below T_c [see Fig. 3(a)]. The crystal in Fig. 3(c) is the same as in Fig. 2. Note that the two traces look essentially identical above T_c , while the FC-ZFW trace shows a large vortex signal below T_c that is missing in the ZFC-ZFW trace. In all the underdoped samples, $T_s > T_c$. However, it is the lack of a vortex signal in the ZFC-ZFW traces that allows us to detect T_s in the near optimally doped sample, where $T_s < T_c$. Furthermore, inspecting the temperature dependence of the Kerr signal near T_s , we note a “superlinear” curvature, an observation that may point to the fact that we are probing a secondary order parameter.

It is important in identifying the Kerr effect with a state with spontaneous symmetry breaking to demonstrate that it can be reversed in a sufficiently high magnetic field. However, one very unexpected feature of our data (which is implicit already in the above) is that the coercive field does not vanish at T_s , nor indeed up to room temperature. (We have not gone above room temperature in order not to destroy the oxygen ordering in the crystals.) Both aspects of this are demonstrated by the traces shown in Fig. 4 taken on the same crystal with $x = 0.67$ as in Figs. 2 and 3(c). (Similar, although not quite as extensive, results have been obtained for the other crystals.) First, the crystal is trained at room temperature in a +4 T field, which is then removed. In Fig. 4(a), this crystal is field cooled in a “small” field of +60 Oe, and then measured in a zero-field warm-up. Not unexpectedly, the trace looks very similar to that in Fig. 2, except with a substantially smaller vortex contribution below T_c . In Fig. 4(b), the crystal is cooled in a small field of -60 Oe, and then measured in a zero-field warm-up. Clearly, the vortex signal below T_c has the opposite sign, as expected, but the signal above T_c is unchanged by the application of the -60 Oe field, proving that the coercive field at room temperature is in excess of this value. To obtain the traces in Figs. 4(c) and 4(d), the crystal was first retrained by the application of a -4 T field at room temperature, then field cooled in a field of ± 60 Oe, respectively, and measured in a zero-field warm-up. Clearly, these two curves are just the reverse of the curves in Figs. 4(a) and 4(b). A detailed study of the training

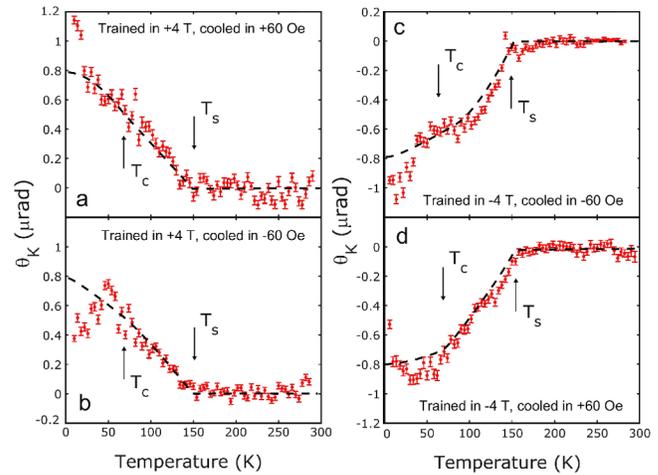


FIG. 4 (color online). The Kerr-effect measurement of $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ crystal taken while warming the sample after cooling it in fields of -60 Oe and +60 Oe, and switching the field off at 4.2 K. These measurements were taken after the sample was trained in a field of -4 T (left) and +4 T (right), as shown in Fig. 2 (see text). Note the much smaller vortex contribution and the fact that it tracks the sign of the field in which it was cooled in.

hysteresis showed that a field of ~ 3 T applied at room temperature fully flips the orientation of the effect below T_s , hence the choice of 4 T for our training schedule. This study will be the subject of a future publication.

So far we have discussed a series of single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. However, we note that all the effects we observed above were also observed on a series of thin films of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. Such films are known to be inhomogeneous with transitions that are broader than in single crystals. This may be due to grain boundaries, twin boundaries, disorder in the chains, or lattice distortion induced by the substrate, among other reasons. Thus, exploring the Kerr signal in thin films will be an excellent test of the genuine origin of the effects and their sensitivity to disorder. While a detailed study of the films will be given in a different publication, we introduce Fig. 5 here as an example of a c -axis film with onset temperature $T_c \sim 60$ K. Compared to the crystals, this is a sample that should be in between $x = 0.5$ and $x = 0.67$. Indeed, the signal we observe is similar to the one observed in Fig. 3(c), with a broad onset at $T_s \sim 160$ K, similar to that of the crystal with $x = 0.5$ [Fig. 3(d)].

The observed large training fields suggest that TRS is already broken above room temperature, but the nature of this state is unclear. If it has a ferromagnetic component, then its moment is less than $\sim 10^{-5} \mu_B/\text{Cu}$, which is our estimated sensitivity. The signal that we do measure at T_s cannot be a consequence of a gradual increase of the high-temperature signal because its sharp onset resembles a true broken symmetry at T_s . We therefore suggest that another order parameter orders at T_s , which is coupled to the high-

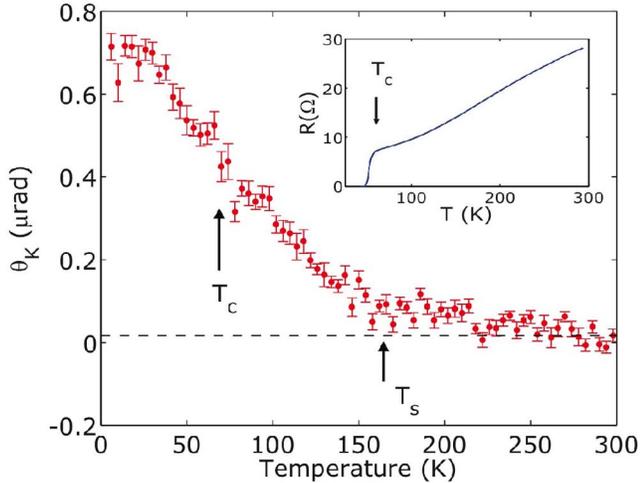


FIG. 5 (color online). The Kerr effect of a c -axis film with estimated average oxygen stoichiometry of $0.5 < x < 0.67$. The inset shows the resistive transition of the sample. We also note the position of T_c and the range of T_s (see text) for this sample.

temperature TRS-breaking order parameter. Either this order parameter may itself break TRS or it becomes “visible” through a ferromagneticlike component that is induced in the high-temperature order through some distortion below T_s .

The fact that T_s marks the onset of a true symmetry breaking effect gains support from the recent elastic neutron scattering measurements [18,19], and earlier μ SR measurements [20]. Using polarized elastic neutron diffraction, Fauqué *et al.* identified a magnetic order in the $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ system that does not break translational symmetry and is consistent with either opposite moments on oxygens of adjacent bonds or two countercirculating charge current loops within the unit cell. The circulating current state was proposed by Varma [8] to account for a symmetry breaking effect at T^* . We note that the first possibility necessarily produces a small ferromagnetic moment due to the orthorhombicity of YBCO. The current loop state, by itself, is incompatible with ferromagnetism, but a ferromagnetic component can be induced by any additional effect (e.g., impurities), which further reduces the spatial (point-group) symmetry [21] of the crystal. Comparing the neutron data to our T_s reveals that the onset of the effect for similar dopings is ~ 30 K higher for the neutron experiments. The μ SR measurements were done for $x = 0.67$ and for $x = 0.95$, and both give onset temperatures for increased muon relaxation that are identical to our T_s . Both the neutron and μ SR experiments also note the existence of magnetic scattering above the onset temperature that persists all the way to room temperature. However, no proposal has been put forward to explain this effect.

In conclusion, we have reported in this Letter the discovery of a novel magnetic order in a wide range of doping of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. The new effect is ferromagneticlike and onsets at a temperature that matches the pseudogap behavior in underdoped cuprates. We further find evidence that the line defined by the onset of this effect crosses the superconducting dome to appear below T_c for a near optimally doped sample. Finally, we find that this effect couples to another time reversal symmetry effect that occurs at high temperatures and dictates the sign of the Kerr effect that appears at the pseudogap temperature.

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