BNL-113432-2017-JA

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Submitted to Applied Physics Letters

October 2016

Condensed Matter Physics and Materials Science Department

Brookhaven National Laboratory

**U.S. Department of Energy
USDOE Office of Science (SC),
Basic Energy Sciences (BES) (SC-22)**

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Angle-Resolved Photoemission with Circularly Polarized Light in the Nodal Mirror Plane of Underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ superconductor

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(Dated: 15 July 2016)

Angle-resolved photoemission spectroscopy (ARPES) experiments using polarized light have been proposed to detect possible symmetry breaking state in cuprates. The presence (absence) of an electronic order which breaks the mirror symmetry of the crystal, would in principle induce a finite (zero) circular dichroism in photoemission. Different orders breaking the reflection symmetry about different mirror planes can also be distinguished by the momentum dependence of the circular dichroism. Here, we report the ARPES experiments on an underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) superconductor in the Γ (0,0)-Y (π,π) nodal mirror plane using polarized light. No circular dichroism is observed on the level of $\sim 2\%$ at low temperature, which places clear constraints on the possible symmetry breaking orders in this sample. Meanwhile, we find that the geometric dichroism remains substantial very close to its perfect extinction such that a very small sample angle offset is sufficient to induce a pronounced dichroic signal. It highlights the importance to establish a perfect extinction of geometric dichroism as a prerequisite for the identification of any intrinsic circular dichroism in this material.

Pseudogap is one of the most striking phenomena associated with high temperature superconductivity. Unraveling the nature of the pseudogap phase is believed to be key to understanding the mechanism of superconductivity [1–5]. Numerous efforts have been made in both theories and experiments, among which is the proposal to detect the possible symmetry breaking state by ARPES measurements using polarized light [6, 7]. An electronic order that breaks the mirror symmetry of the system would contribute to the circular dichroism in photoemission. Various orders breaking the reflection symmetry about different mirror planes could also be differentiated by the momentum dependence of the circular dichroism. For example: an ordered pattern of circulating currents was predicted to break the reflection symmetry about the Γ (0,0)-M ($\pi,0$) antinodal mirror plane which gives a maximum circular dichroism at the antinode ($\pi,0$) [6]. On the other hand, the antiferromagnetic order at $Q=(\pi,\pi)$ and the “D-density wave” would produce a phase breaking reflection symmetry about the Γ (0,0)-Y (π,π) nodal mirror plane [7]. Another electronic order with increasing interest is the charge order observed in various cuprates [8–11] which might exhibit a chiral nature [12, 13]. Chiral symmetry breaking generally breaks all mirror symmetries of the system and would thus contribute to a circular dichroism in the photoemission performed in any mirror plane.

A prominent experimental realization of the proposal is the report of circular dichroism in the ARPES spectra at the antinode ($\pi,0$) of Bi2212 below the pseudogap temperature T^* [14–19]. The observed circular dichroism was first interpreted as direct evidence for the breaking of time-reversal symmetry [14], which seemed to be consistent with the theoretical proposal of a spontaneous ordered pattern of circulating currents [6, 7]. This interpretation was challenged by others considering the structural supermodulations in the Bi-O layer (superstructures) that breaks the reflection symmetry about the Γ (0,0)-M ($\pi,0$) antinodal mirror plane (Fig. 1a vs. Fig. 1b) and gives rise to the circular dichroism at the antinode ($\pi,0$) [15, 17, 19–21].

Compared to the Γ (0,0)-M ($\pi,0$) antinodal mirror plane (Fig. 1a,b), Γ (0,0)-Y (π,π) nodal mirror plane provides a cleaner platform to study possible intrinsic orders. As shown in Fig. 1c, this nodal plane remains a mirror plane even with the presence of superstructures [1, 21]. Therefore, the presence/absence of circular dichroism along the Γ (0,0)-Y (π,π) momentum cut in the nodal mirror plane would provide less entangled information on possible symmetry breaking orders in the sample.

An equally important, yet technical issue regarding circular dichroism experiments arises from the experimental geometry (geometric dichroism) which might mask the intrinsic circular dichroism [15, 16]. Ideally, the geometric dichroism should vanish if the propagation vector of the light, the sample surface normal, and the final state momentum are all in a mirror plane of the sample [14, 22]. However, finite deviations from the desired perfect experimental geometry always exist and give rise to geometric dichroism with a magnitude that depends on the deviations and the material. This leads to a natural question: how accurate the experiment needs to be performed before the intrinsic circular dichroism can be discussed in this system?

In this paper, by using an ARPES setup equipped with

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FIG. 1: (Color online) Schematic of photoemission measurements in different mirror planes of Bi2212. (a) Incident photon $h\nu$, sample surface normal, and final state momentum p are all in the Γ (0,0)-M (π ,0) antinodal plane (blue plane). This plane is a mirror plane of the crystal without superstructures. The yellow line marks the slit of the electron analyzer. Antinodal M point (π ,0) locates in the mirror plane. (b) The reflection symmetry of the Γ (0,0)-M (π ,0) antinodal plane is broken with the appearance of superstructures. Superstructures are indicated by pink dashed lines and marked as MB+ q to be distinguished from the main bands (MB). (c) The Γ (0,0)-Y (π , π) nodal mirror plane remains a mirror plane even with the appearance of shadow bands (SB) and superstructure bands. MB+ q and MB+2 q represent the Fermi surface sheets from first order and second order superstructure bands respectively. The horizontal slit of the electron analyzer enables the measurement of the Γ (0,0)-Y (π , π) momentum cut in the mirror plane. (d) Schematic of the photoemission measurement and the definition of the θ rotation angle.

a horizontal slit in the electron analyzer and a sample holder with in-plane azimuthal rotation, we report the first study of the circular dichroism along the Γ (0,0)-Y (π , π) nodal cut of a Bi2212 underdoped $T_c=75$ K sample (as shown in Fig. 1c). No circular dichroism is observed along the nodal cut on the level of $\sim 2\%$ at low temperature, which places clear constraints on the possible symmetry breaking orders that might exist in this system. Meanwhile, the geometric dichroism in this material is found to be very sensitive to any slight deviation of the sample angle from its perfect experimental geometry. An angle offset as small as 0.3 - 0.5° is sufficient to induce a pronounced geometric circular dichroism.

The ARPES measurements were carried out at Beamline-9A of the Hiroshima Synchrotron Radiation Center with circularly polarized 21.2eV photons. The ARPES setup is equipped with a R4000 electron analyzer with a horizontal slit and a sample holder with in-plane azimuthal rotation. The experimental energy resolution was ~ 12 meV and the angular resolution was $\sim 0.3^\circ$. Only the center region of the detector ($\pm 10^\circ$) was selected to

FIG. 2: (Color online) Absence of circular dichroism along the Γ (0,0)-Y (π , π) nodal direction. (a) Underlying Fermi surface mapping obtained by integrating the spectral weight over a small energy window $[-10\text{meV}, 10\text{meV}]$ around the Fermi level. The theoretical Fermi surface sheets from the shadow band, first order and second order superstructure bands are appended. (b) Photoemission intensity plot for the momentum cut along Γ (0,0)-Y (π , π) direction. The momentum location of the cut is marked by the black line in (a). (c) Energy integrated $[-60\text{meV}, 30\text{meV}]$ spectral intensity as a function of momentum for circularly left-handed (I_{CL} , red curve) and right-handed (I_{CR} , black curve) polarized light. The energy window for the integration is marked by the red dashed rectangle in (b). (d) Relative difference $D=(I_{CL}-I_{CR})/(I_{CL}+I_{CR})$ of the energy integrated spectral intensity shown in (c). No difference is observed on the level of $\sim 2\%$.

eliminate any edge effect of the electron analyzer. The Fermi level was referenced to that of a polycrystalline Au piece in electrical contact with the sample. The measurements were performed at 30K with a base pressure better than 5×10^{-11} torr.

A perfect sample alignment is the prerequisite for the experiment. We use large range Fermi surface mapping to align the orientation of the sample [1, 5, 23]. With the clearly resolved fine structures on the underlying Fermi surface (Fig. 2a) and three degrees of freedom in the angular rotation by the 6-axis manipulator, the sample was well aligned such that the propagation vector of the light, the sample surface normal, and the final state momentum are all located within the Γ (0,0)-Y (π , π) nodal mirror plane. If there is (no) reflection symmetry breaking induced by the electronic order, we should expect a finite (zero) circular dichroism along Γ (0,0)-Y (π , π) where the geometric dichroism also vanishes.

Energy-integrated photoemission intensities probed with circularly left-handed (I_{CL}) and right-handed (I_{CR}) polarized light are compared to show the circular dichro-

ism. As presented in Fig. 2c, when the momentum cut lies perfectly within the Γ (0,0)-Y (π,π) nodal mirror plane (Fig. 2a,b), the photoelectrons probed by light with different polarizations exhibit identical intensities along the entire cut, giving rise to zero circular dichroism within the error of $\sim 2\%$ (Fig. 2d). While one could argue that the intrinsic and geometric circular dichroism could happen to cancel each other and give zero dichroism at a certain momentum point when the sample is not perfectly aligned [15, 16], it is unlikely that they always have the same magnitude but with an opposite sign over the entire momentum cut. Therefore, our results suggest an absence of both intrinsic and geometric dichroism (on the level of $\sim 2\%$) along the nodal direction in our sample.

To understand the implications of our result, we consider various electronic orders. It has been suggested that the antiferromagnetic order at $Q=(\pi,\pi)$ and the existence of “D-density wave”, would in principle produce a phase breaking reflection symmetry about the Γ (0,0)-Y (π,π) nodal mirror plane and give rise to a finite circular dichroism in the ARPES spectra measured along the nodal momentum cut [7]. But for chiral symmetry breaking, one might ask whether it can in principle be detected along the nodal direction: The chiral order might be implicated with the pseudogap phenomena which are known to have a vanishing effect on the nodal electronic states near the Fermi level (E_F). Therefore, the nodal states, as the initial states of the photoemission process, may not be chiral. Nevertheless, we note the studies on chiral molecular systems have shown that the circular dichroism in photoemission is dominated by final-state (delocalized) interactions-scattering of the outgoing photoelectrons off the chiral molecular framework [24]. Such a mechanism should be at work independent of the nature of chirality, whether structural or electronic. Therefore, if a chiral order exists in the material, whether it involves the nodal states, a circular dichroism associated with the latter on the mirror plane along the nodal direction should be finite. Moreover, the circular dichroism in photoemission probes local chirality which is a pseudoscalar quantity and independent of chiral main axis orientation associated with possible chiral domains. Compared with circular dichroism in absorption which is associated with second-order interference terms of the light-matter interaction, circular dichroism in photoemission is associated with the dipole interaction term [24], which is of a much bigger magnitude. A dichroism asymmetry factor on the order of 20% was seen in chiral molecular systems [24]. Therefore, the absence of circular dichroism in our result does not seem to be consistent with the expected circular dichroism from the above electronic orders. On the other hand, we note more theoretical efforts are needed to quantitatively determine the magnitude of the expected circular dichroism from these orders, especially for the chiral order, of which limited knowledge is currently available. The expected value might be below our experimental sensitivity. In this regard, our study

FIG. 3: (Color online) Sensitivity of geometric dichroism to the angle offset. (a) Underlying Fermi surface mapping obtained by integrating the spectral weight over a small energy window $[-10\text{meV}, 10\text{meV}]$ around the Fermi level. (b-e) Energy integrated $[-60\text{meV}, 30\text{meV}]$ spectral intensity as a function of momentum for circularly left-handed (red curve) and right-handed (black curve) polarized light along 4 momentum cuts $\theta = 0^\circ \sim 0.5^\circ$ (shown in a).

places an upper limit, $\sim 2\%$, to the circular dichroism in photoemission due to the possible existence of the above electronic orders. Another possibility is that the symmetry breaking electronic orders only exist in a sample with lower doping level and/or at a particular temperature region between T_C and T^* , since our measurements were performed on an underdoped 75K sample below T_C . While more doping and temperature dependent measurements are needed to fully address this issue, we note the circular dichroism at the antinode ($\pi,0$) was observed on samples with similar doping levels and at various temperatures below T^* , regardless of T_C [14].

The absence of circular dichroism along the nodal direction also provides a unique chance to study the geometric dichroism as a function of the sample angle deviation. Fig. 3 shows the results for the momentum cuts in the vicinity of the Γ (0,0)-Y (π,π) nodal direction. The angle difference (tilt angle θ , see Fig. 1d for its definition in the experimental setup) between these cuts is within 0.5° . Energy-integrated photoemission intensities for circularly left-handed (I_{CL}) and right-handed (I_{CR}) polarized light along these cuts (Fig. 3a) are shown in Fig. 3(b-e) respectively. A moderate geometric dichroism starts to show up at $\theta=0.3^\circ$ and becomes pronounced when θ reaches 0.5° . Our results suggest that the geometric dichroism in Bi2212 is very sensitive to small angle deviation from the perfect alignment.

To understand the origin of the geometric dichroism, we consider the photoemission under three-step model and sudden approximation. The photoemission intensity I is proportional to $|M_{f,i}|^2$, where $M_{f,i} \equiv \langle \phi_f | H | \phi_i \rangle$ is the one-electron dipole matrix element describing the ejection of an electron from an initial state $|\phi_i\rangle$ to a final state $|\phi_f\rangle$ [1]. When the incident light is circularly polarized, the dipole operator containing the vector potential of photons with different polarizations can be written as H_{CL} (circular left) and H_{CR} (circular right). Then the circular dichroism is denoted as

$D = | \langle \phi_f | H_{CL} | \phi_i \rangle |^2 - | \langle \phi_f | H_{CR} | \phi_i \rangle |^2$ [22]. If we define an operator R as reflection of the blue plane of Fig. 1a (which contains the propagation vector of the light, the sample surface normal, and the final state momentum), then $R^{-1}H_{CL}R = H_{CR}$ and $\langle \phi_f | H_{CR} | \phi_i \rangle = \langle \phi_f | R^{-1}H_{CL}R | \phi_i \rangle$. When the blue plane is a mirror plane, $R|\phi_i\rangle = \pm|\phi_i\rangle$, $R|\phi_f\rangle = \pm|\phi_f\rangle$, and thus D is zero [22]. However, if there is a small angular offset and the blue plane is no longer a mirror plane, then the dichroism given by $D = | \langle \phi_f | H_{CL} | \phi_i \rangle |^2 - | \langle \phi_f | H_{CR} | \phi_i \rangle |^2$ has a nonzero value. In this regard, materials with different electronic structure would generally have different response in the photoemission circular dichroism to the angle deviation in experimental geometry. Theoretical calculations considering the band structure of Bi2212 are needed to address the high sensitivity of the geometric dichroism in this system.

In summary, by measuring the photoemission spectra along Γ (0,0)-Y (π,π) in the nodal mirror plane of a Bi2212 underdoped 75K sample with polarized light, we report the absence of any detectable circular dichro-

ism on the level of $\sim 2\%$ at low temperature. This result is not affected by the structural supermodulations in the sample, thus places clear constraints on the possible symmetry breaking electronic orders. Our result also reveals the high sensitivity of geometric dichroism to the slight angle deviation from the perfect experimental geometry. It highlights the importance to establish a perfect extinction of geometric dichroism before identifying any intrinsic circular dichroism in this material.

Acknowledgments

The work at Boston College was supported by a BC startup fund (J.H.), the US NSF CAREER Award DMR-1454926 (R.-H.H.) and Graduate Research Fellowship DGE-1258923 (T.R.M.). ARPES experiments were performed at Hiroshima Synchrotron Radiation Center under proposal No.14-A-1. The work at BNL was supported by the DOE under contract No. DE-AC02-98CH10886.

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