

On the detection of time-reversal symmetry breaking by photoemission with circularly polarized light in the high- T_c superconductor Bi2212

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ABSTRACT

We argue that that in their recent experiment in which they claim to have found evidence for a time-reversal symmetry broken state, Kaminski *et al.* (2002) (*Nature*, **416**, p. 610) overlooked small temperature-dependent changes in the superstructure of Bi2212. These subtle changes may manifest themselves by changing the final state configurations of the photoemission process and thus invalidate their ultimate conclusions.

Kaminski *et al.* (2002) recently reported the results of an experiment in which they found a small but significant asymmetry ($\approx 3\%$) in the angle-resolved photoemission intensity of the high- T_c superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) below its approximate pseudogap temperature T^* when using light of different helicities. This was interpreted as indicative of a hidden time-reversal-symmetry (TRS)-breaking phase in the underdoped regime. However, as there is ample evidence for low-temperature structural changes in the underdoped regime of Bi2212, we feel that Kaminski *et al.* (2002) are incorrect to infer the existence of a TRS breaking state from their experiment.

The Kaminski *et al.* (2002) experiment is not directly sensitive to TRS and is, in fact, a probe of reflection symmetry (RS). Although this point is made in the theoretical proposal (Simon and Varma 2002) and implicit in the experiment, it is worth making explicit. It can be easily seen by considering the photoemission optical matrix element

$$M^r(\mathbf{k}) = \sum_{fi} | \langle \psi_f | \hat{\Delta}^r | \psi_i \rangle |^2,$$

where $\hat{\Delta}^{r(l)}$ is the dipole operator for right (left) circularly polarized light and $|\psi_i\rangle$ and $|\psi_f\rangle$ are, for illustration purposes, one-electron states. With a reflection symmetry operation \hat{R} , the terms of the above matrix element equal $| \langle \psi_f | \hat{R}^{-1} \hat{\Delta}^r \hat{R}^{-1} \hat{R} | \psi_i \rangle |^2$. If states $|\psi_i\rangle$ and $|\psi_f\rangle$ are eigenstates of reflection, this quantity is proportional to $| \langle \psi_f | \hat{\Delta}^r | \psi_i \rangle |$. The associated matrix element can be defined as M^l and therefore the dichroism quantity probed by Kaminski *et al.* (2002), $D = M^r - M^l$, is sensitive to reflection. However, because a crystal's point

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group and reflection symmetries can only be preserved if all magnetic moments from orbital currents or spin are confined to the irreducible unit cell, all proposed TRS-breaking states for the cuprate superconductors break RS across at least one structural mirror plane. In this regard, RS breaking is a necessary, but not sufficient, condition for TRS breaking. A system that has a dichroism photoemission signal across a certain direction and no structural aspects which break RS across that direction may be a candidate for a TRS-breaking state, but a state that breaks RS alone can also give such an effect.

Although the $\Gamma \leftrightarrow (\pi, 0)$ direction that Kaminski *et al.* (2002) use as a mirror plane in their experiment is superficially a mirror plane of an idealized CuO_2 square lattice, RS is broken across this direction in Bi2212 by the well-known \hat{b} direction [$\Gamma \leftrightarrow (\pi, \pi)$] incommensurate superstructure modulation, which then ceases to be a mirror plane (Gao *et al.* 1988, Kirk *et al.* 1988). In principle, a supermodulation at 45° to the $\Gamma \leftrightarrow (\pi, 0)$ plane would give a maximum dichroism signal at $(\pi, 0)$. As evidenced by the experiment, however, whatever are the effects of the superstructure, they apparently do not cause an appreciable dichroism at high temperatures. The conclusion that the low-temperature effect is not caused by the superstructure and instead by a TRS-breaking state is only reasonable if there are no changes in the modulation as a function of temperature and doping. If there are such changes, it would seriously undermine the claim that TRS breaking could be inferred from RS breaking. Note that, monitoring only a single main Bragg peak via X-rays, as done by Kaminski *et al.* (2002), is likely to be a very incomplete measure of temperature-dependent changes for a complicated oxygen-rich incommensurate crystal such as Bi2212.

We should also note that, although Kaminski *et al.* (2002) argued, from their X-ray diffraction data and the fact that their observed dispersion is symmetric around $(\pi, 0)$, that any temperature-dependent changes in the crystal structure were much smaller than that necessary to cause a macroscopic shift of the mirror plane of the near- E_F electronic states, this point is irrelevant. Because $(\pi, 0)$ is not in a true mirror plane of the Bi2212 crystal, any changes in the structure do not have to be such as to influence the spectral function $A(\mathbf{k}, \omega)$ of the near- E_F states (of which the dispersion is indicative of) greatly; the purported measurement is of the photoemission optical matrix element $M^{r,l}$, which will be sensitive to subtle changes in the hybridization of the final-state configuration $|\psi_f\rangle$.

There are a number of studies that do, in fact, suggest low-temperature structural changes of the incommensurate modulation in underdoped samples. For instance, Anderson *et al.* (1992, 1997) in a series of careful ultrasonic measurements, found a sharp internal-friction loss peak, indicative of a bulk structural change, at 167 K only in oxygen-deficient (underdoped) Bi2212 samples. Miles *et al.* (1997), in a detailed Rietveld refinement of neutron diffraction data, found a sharp discontinuity in the \hat{b} axis lattice parameter as well as the superstructure period for underdoped Bi2212 crystals around 160 K. Although the only comprehensive temperature-dependent X-ray study of the incommensurate structure found no such strong anomalies (perhaps due to the weak oxygen sensitivity of X-rays, which makes them a poor probe for this type of system), it did find a suppression of the super structure satellite intensities around ~ 130 K (Johnson and Hatton 1995). The doping and temperature dependence of these structural changes mimic that of T^* .

Generally speaking, strong temperature-dependent interlayer stresses arise in such low-dimensional incommensurate structures from the differences in expan-

sivities of intralayer bonds (Bak 1982). Such stresses may be relieved by the vacancies in oxygen-deficient Bi–O layers through subtle bulk reconstructions. We should mention, however, that such stresses can also be relieved at the surface. As photoemission is an eminently surface-sensitive probe, the surface structural symmetry needs to be considered separately. Although low energy electron diffraction studies have claimed that the surface structure is commensurate with the bulk, analysis was not performed at the requisite level of detail to allow strong statements about subtle differences (Lindberg *et al.* 1988, Claessen *et al.* 1989). In fact, scanning tunneling microscopy routinely shows (Kirk *et al.* 1988, Inoue *et al.* 1996, Pan *et al.* 1998) a strong additional surface modulation of the bismuth atoms at approximately the superstructure periodicity of which no corresponding feature has been observed in bulk-sensitive X-ray diffraction. The differences between surface and bulk structure mean that this issue needs to be investigated in more detail before one can rule out doping-dependent low-temperature surface reconstructions.

As judged from the experiments detailed above, which find structural changes in the 130–160 K temperature range in underdoped samples, there is evidence for a subtle structural reconstruction with a doping dependence that mimics T^* . Because the changes found in Bi2212 violate the structural symmetry condition necessary to infer a TRS-breaking state from RS breaking, we feel that Kaminski *et al.* (2002) cannot reasonably deduce the existence of an exotic TRS-violating state from their experiment.

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