Experimental report

15/02/2021

Proposal: **5-53-291** **Council:** 10/2019

Title: Possible octupolar phase in oxychlorine copper oxide

Research area: Physics

This proposal is a new proposal

Main proposer: **Dalila BOUNOUA**

Experimental team: Zachary ANDERSON
RICHARD SPIEKER
Martin GREVEN
Dalila BOUNOUA

Local contacts: Martin BOEHM
Paul STEFFENS

Samples: $Sr_2CuO_2Cl_2$

Instrument	Requested days	Allocated days	From	To
THALES	7	4	29/01/2021	02/02/2021
IN12	7	0		

Abstract:

Optical second harmonic generation (SHG) is a very powerful and sensitive technique to unveil novel and hidden phases in various compounds with strongly correlated electron systems. Nowadays, SHG studies encompass odd-parity order that breaks the inversion and rotational C4 symmetries in iridates as well as in the pseudogap state of high-Tc copper oxides superconductors, which can be described by a loop current phase originally, proposed for high-Tc cuprates. Recently, SHG measurements reported a new order parameter in $Sr_2CuO_2Cl_2$ that breaks both magnetic and structural point group symmetries, a potential new ferromagnetic octupolar order parameter. To search for this new phase, we apply for 7 days on Thales equipped with its polarized neutron set-up.

Possible octupolar phase in oxychlorine copper oxide

Optical second harmonic generation (SHG) is a very powerful and sensitive technique to reveal hidden phases in various compounds with strong electronic correlations. A well-known example is the magnetoelectric compound Cr_2O_3 which exhibits a Q=0 antiferromagnetic order [1]. Other recent SHG studies have uncovered odd-parity order that breaks the inversion and rotational C_4 symmetries in iridates as well as in the pseudogap state of high-Tc copper oxides superconductors [2]. Both cases can be described by a loop-current phase originally proposed for high-T_c cuprates [3,4] that breaks inversion and time-reversal symmetries, but not the product of both, and that preserves the lattice translation invariance. This phase can be described by a toroidal moment or an anapole along the diagonals of the CuO_2 plane with a ground state that contains four degenerate states. In the case of the iridates [2], it is remarkable that these four domains were identified by SHG through a rotational anisotropy with a domain size of about 10-50 µm. Microscopic currents running between Cu d-orbitals and oxygen p-orbitals produce orbital moments. Polarized neutron diffraction (PND) experiments performed in both cuprates [5-7] and iridates [8] have reported a Q=0 magnetic signal corresponding to the broken symmetries of the loop-current phases. However, these PND data can also be interpreted as evidence of spontaneous ordering of Dirac (or magnetoelectric) multipoles [9]. Within this quadrupole description, a microscopic model has been developed through a coupling between fluctuating spin magnetic dipole moments and polar optical phonons [10].

$Sr_2CuO_2Cl_2$ is a cuprate parent compound. It is an insulating layered perovskite with the tetragonal *(I4/mmm)* K_2NiF_4 structure which remains tetragonal down to at least 10 K [11]. In contrast to La_2CuO_4, the apical oxygen atoms are replaced by chlorine (Fig. 1a). Magnetic-susceptibility χ(T) and neutron-diffraction studies on single-crystal specimens have shown a three-dimensional antiferromagnetic structure similar to that in La_2CuO_4 below the Néel temperature T_N ~ 250 K by neutron diffraction [11]. Recently, it has been shown that, below T_N, SHG signal reveals a new order parameter, which breaks both magnetic and structural point group symmetries [13]. Contrary to the previous loop current and/or multipolar phases discussed above [3-11], the new order parameter in $Sr_2CuO_2Cl_2$ does not break inversion symmetry. Instead, this unexpected result suggests that a potential new ferromagnetic octupolar order parameter (called Φ phase) is at play in $Sr_2CuO_2Cl_2$ with *4/mm'm'* symmetry [12]. The symmetry 4/mm'm' is chiral and breaks time-reversal but not inversion. It is also consistent with a clockwise (or anti-clockwise) loop-current pattern from O-O-O-O in the part not enclosing Cu and the other way in O-O-O-O enclosing Cu [4,12].

We previously obtained four days of beam time on THALES (proposal 5-53-291) (experiment done in January/february 2021) to study this potential new order phase in the AFM insulator parent cuprate using PND technique. We obtained very interesting results consistent with the SHG result [12], using single crystals grown at the University of Minnesota as in ref. 12. Figures 1 and 2, show the thermal dependence of the inverse of the measured flipping ratio (1/FR) for two Bragg peaks: as discussed in refs. [13], a positive upturn on 1/FR shows a novel magnetic contribution whereas a smooth decrease corresponds to a parasitic change of the polarization. As in the other cuprates [5-7], the magnetic scattering is observed at Q=(1,0,1) Bragg reflections below an ordering temperature of roughly 400 K (Fig. 2) whereas nothing is observed at (0,0,4) on fig. 1b (where the expected structure factor is zero for the expected magnetic patterns discussed above [13]). Attempts to measure magnetic scattering at forbidden Bragg positions ((1,0,L) for L even) were unsuccessful, indicating that the magnetic scattering preserves lattice-translational symmetry. We need now to perform a polarization analysis on both Bragg peaks to control the polarization on (0,0,4) and determine the moment direction on (1,0,1). Measurements, on the (1,0,L) (L=3,5..) Bragg positions, are also needed to address the issue of the magnetic structure factor.

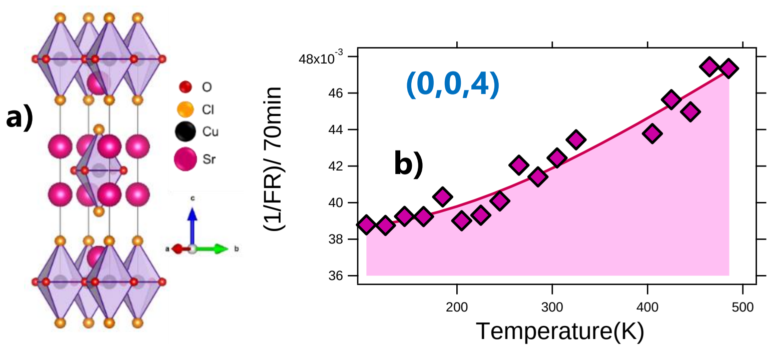

Figure 1: a) crystal structure of $Sr_2CuO_2Cl_2$. *b) Inverse of the flipping ratio of on the Bragg peak (0,0,4) where no magnetic signal is expected.*

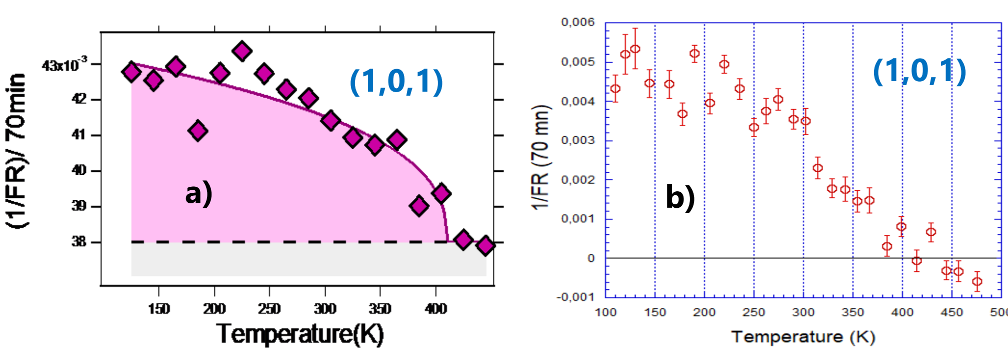

Figure 1 a) Inverse of the flipping ratio of the Bragg peak (1,0,1) versus temperature. b) Inverse of the flipping ratio of the Bragg peak (1,0,1) versus temperature. Here data, obtained from two different temperature ramps, have been averaged. Both figures indicate a magnetic signal arising around 400 K. There is a good agreement between both data.

References

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