Optical second harmonic generation reveals hidden odd-parity order in Sr_2IrO_4

Evidence of an odd-parity hidden order in a spin-orbit coupled correlated iridate, L. Zhao, D. H. Torchinsky, H. Chu, V. Ivanov, R. Lifshitz, R. Flint, T. Qi, G. Cao and D. Hsieh, *Nature Phys.* **12**, 32 (2016).

Recommended with commentary by Joseph Orenstein, Department of Physics, UC Berkeley

The subject of the study reported by Zhao et al. is the single-layer perovskite iridate Sr_2IrO_4. Interest in this compound spiked soon after the discovery of cuprate superconductivity because Sr_2IrO_4 has the same K_2NiF_4 crystal structure as La_2CuO_4 and the unconventional (possibly p-wave) superconductor Sr_2RuO_4. However failure to make Sr_2IrO_4 go superconducting caused research efforts to quickly taper.

Following the topological insulator boom of the late 2000s, interest in Sr_2IrO_4 (and iridates in general) was reignited because of its strong spin-orbit coupling (SOC), which reaches an energy scale on par with short-range Coulomb repulsion (U). Early on it was recognized that the interplay between SOC and U causes Sr_2IrO_4 to be an antiferromagnetic insulator[1], despite the large spatial extent of the Ir 5*d* orbitals. SOC splits the Ir t_{2g} band into fully occupied $J_{\text{eff}} = 3/2$ bands and a half filled $J_{\text{eff}} = 1/2$ band. The latter has a bandwidth smaller than U which favors the opening of a Mott gap. For this reason Sr_2IrO_4 is often referred to as a spin-orbital entangled Mott insulator.

While there is nothing topological about Sr_2IrO_4, the fact that its electronic structure is well approximated by a single ($J_{\text{eff}} = 1/2$) band Hubbard model further strengthens its similarity to the cuprates and led to theoretical predictions of high-temperature superconductivity[2] with electron doping. Recent results reveal a deeper connection to cuprates than previously known. Most notably, the opening of a pseudogap in the electronic density of states has now been observed by ARPES in both hole and electron doped Sr_2IrO_4 in the form of Fermi arcs[3–5]. Both ARPES[6] and STM[7] have shown that in samples doped by deposition of a fractional monolayer of K, the pseudogap evolves to a *d*-wave gap upon further cooling, suggesting possible superconductivity with a critical temperature near 30 K.

The work of Zhao et al. addresses the question of whether additional electronic orders exist in the vicinity of the antiferromagnetic parent phase. Cuprates have a tendency towards various forms of charge order at low doping levels whose relationship to the pseudogap and to superconductivity has been a source of extended debate[8]. With an analogous system now available in the form of Sr_2IrO_4, one can perhaps separate those electronic orders that are universally associated with the pseudogap and high-T_c superconductivity from those that are material specific.

Zhao et al. searched for signs of symmetry-breaking induced by electronic order in both parent and hole-doped Sr_2IrO_4 through the use of optical second harmonic generation (SHG). In SHG optical excitation at frequency ω induces a current at 2ω that can be detected as frequency doubled radiation in the far-field. This mechanism for SHG is forbidden in materials that possesses a center of inversion, thus its appearance is a sensitive probe of inversion symmetry

breaking. To this basic experiment Zhao et al. added a unique technical feature – the ability to measure SHG from a local region of the sample at low temperatures while rotating the optical scattering plane with respect to the principal axes of the crystal. This extra information – the dependence of SHG intensity on angle – allowed Zhao et al. to pin down rotational and mirror symmetries that may remain despite the loss of inversion symmetry. The bottom line is that Zhao et al. can use SHG to probe point group symmetry as a function of spatial position and temperature.

Unexpected results already arose in the parent compound. SHG showed unambiguously that inversion symmetry and all rotational symmetries about the *c*-axis are broken at a temperature T_Ω that is very close to the Neel temperature. Such a lowering of symmetry is not consistent with the antiferromagnetic structure of Sr_2IrO_4 reported by neutron and resonant x-ray diffraction measurements, which retains inversion and two-fold rotational symmetry. Local distortion of the antiferromagnetic structure also appears to be an unlikely explanation because the same SHG angular dependence appears throughout the sample, albeit with different domain orientations.

As Sr_2IrO_4 is hole-doped away from half-filling, T_Ω is monotonically suppressed, but not nearly as rapidly as the Neel temperature. Thus there exist swaths in the temperature-doping plane where inversion-breaking order is unaccompanied by long-range antiferromagnetism. The obvious question is: what is going on microscopically?

Using their SHG data, Zhao et al. narrow the possible point groups in the ordered phase to a very short list. This list includes candidates from the so-called magnetic point groups, which contain symmetry elements in which conventional operations are accompanied by a reversal of time. For example, 2′ denotes 2-fold rotation accompanied by time reversal. They find that the only non-magnetic point group that fits their data has very low symmetry (m), whereas the groups 2′/m and m1′ are in play as well. Although m symmetry cannot be ruled out, the authors believe that the very low symmetry of this group, as well as the proximity of the system to magnetic ordering, favor an interpretation in terms of magnetism. Both 2′/m and m1′ groups are consistent with ordering of intra-unit cell current loops like those first proposed by Varma in the context of cuprates[9]. Of course other magnetic phases sharing the same point groups cannot be ruled out. Borrowing from ideas spawned from the cuprates, candidates include some long-range ordered configuration of oxygen moments[10], an inversion broken variant of the Kalmeyer-Laughlin chiral spin liquid[11], or long-range ordering of local parity-odd multipole moments[12].

This work naturally begs for follow up experiments: 1) The closeness of T_Ω to the Neel temperature in the parent compound raises the possibility that they are actually one and the same. It is conceivable that the magnetic structure refined from diffraction measurements is simply incorrect and actually has 2′/m or m1′ symmetry. After all, even the crystallographic structure of Sr_2IrO_4 is being continually revised ($I4/mmm$[13] $\rightarrow$ $I4_1/acd$[14] $\rightarrow$ $I4_1/a$[15,16]) with each successive and more careful experiment; 2) What is the relationship of T_Ω to T^*, the onset temperature of the pseudogap? A pseudogap has been observed at low temperature by ARPES[4] in the same compounds as those used in the study by Zhao et al., but so far no temperature dependent ARPES data exists; 3) According to theory, Sr_2IrO_4 and La_2CuO_4 are electron-hole conjugates[2],

meaning that the compounds studied by Zhao et al. are analogues of electron-doped cuprates. But no evidence of loop-current order has been observed in the pseudogap region of the electron doped cuprates[17]. Therefore it would be instructive to repeat the SHG experiments on electron doped iridates to examine whether they exhibit electron-hole asymmetries similar to the cuprates.

Clearly, the findings reported by Zhao et al. stir up as many questions as answers, but the introduction of a new symmetry sensitive probe to the world of correlated electron materials is an important step forward – one that may eventually help solve the puzzle of high-T_c superconductivity in correlated oxides.

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