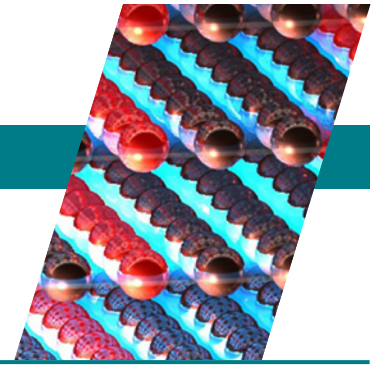


3D Charge Order Found in Superconductor

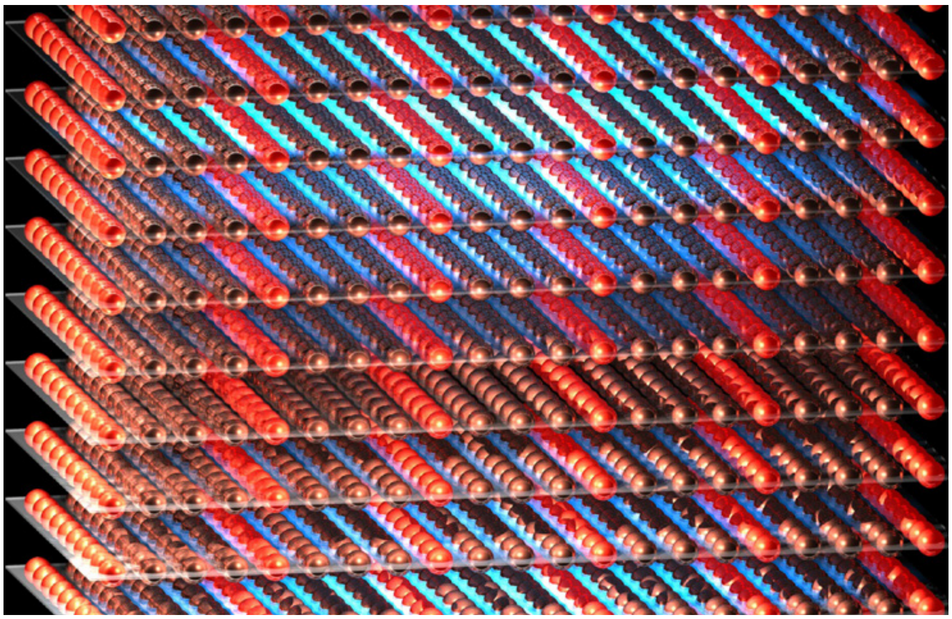


Illustration of the long-range electronic order discovered in YBCO thin films. Within each horizontal copper-oxide plane, the electronic charge modulation repeats every four copper atoms (indicated by red spheres); additionally, the charge modulation repeats vertically along the stack every eight copper-oxide planes (indicated by blue aura).

Despite 30 years of intense study, the explanation behind the zero-resistance current displayed by high-temperature superconductors (HTSCs) is still shrouded in complexity. HTSCs tend to be heterogeneous materials with multiple phases, and disentangling their various electronic behaviors for analysis can be difficult. At the ALS, researchers used resonant soft x-ray diffraction (RSXD), a technique sensitive to both structure and electronic state at the nanoscale, to study layered thin-film heterostructures containing the cuprate high-temperature superconductor, $YBa_2Cu_3O_{7-x}$ (YBCO). They found a surprising three-dimensional, long-range charge order—the first of its kind ever reported in a cuprate—that competes with superconductivity. A better understanding of such phenomena could help in the design of more robust superconductors with higher transition temperatures.

Many HTSCs, especially members of the cuprate family that includes YBCO, have been found to exhibit multiple ordering tendencies, such as periodic ripples in the electronic charge, which compete with the same electronic states involved in superconductivity. Such orderings of the electronic charge typically manifest within the cuprates' copper-oxide planes, which interact weakly (if at all) with each other. This two-dimensional charge order is typically a short-range effect, meaning that the order exists only weakly on a very local scale (a few atomic distances). So far, it is not yet known whether these various electronic orders ("supermodulations") are truly distinct or rather originate from the same underlying mechanism, nor why they manifest in so many forms.

In this work, the researchers studied 30-nm thin films of YBCO grown epitaxially on a sublayer of the ferromagnetic material, $La_{0.7}Ca_{0.3}MnO_3$ (LCMO). Interestingly, such

A "Twofer" Technique

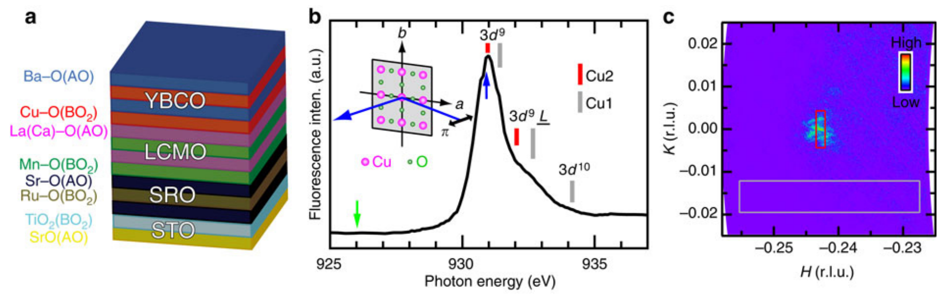
Resonant soft x-ray diffraction (RSXD) combines two techniques, diffraction and spectroscopy, in one experiment—two for the price of one, as the saying goes. X-ray diffraction is a technique that identifies periodic order in materials because x-rays diffracted from a periodic structure interfere constructively at well-defined angles that directly correspond to both the period and the direction of the ordering. Spectroscopy is a technique that can probe the electronic states of specific elements in a heterogeneous material. Each element in the material has its own set of core-level transitions that occur at characteristic energies, making the photon-energy tunability of synchrotron radiation essential.

In high-temperature superconductors, scientists have found that the electrons self-organize into periodic structures (i.e., they display "charge order"). RSXD uses soft x-ray wavelengths that reveal or resonantly enhance the diffraction pattern when the x-ray wavelength matches resonant transitions into periodically ordered electronic states. In this way, RSXD can identify the electronic states that are involved in the ordered structure. The RSXD endstation at ALS Beamline 4.0.2 was designed specifically for detecting and measuring such phenomena and is one of very few places in the world where this type of research can be performed.

heterostructures have previously been reported to exhibit "long-range proximity effects," in which the two major competing orders in the system—the superconductivity in the YBCO and the ferromagnetism in the LCMO—tend to supress each other across the interface. RSXD, by combining diffraction (information about structure) with spectroscopy (information about electronic state), is ideally suited to explore spatial charge modulations in thin films and at buried interfaces in nanoscale heterostructures. The experiments were performed at ALS Beamline 4.0.2 at an x-ray scattering endstation specifically built for studies of electronically ordered phases.

The researchers expected their diffraction data to show a rod-shaped peak, indicating the presence of a short-range, two-dimensional supermodulation in the YBCO/LCMO film. Instead they discovered that the diffraction peaks were concentrated into a single pair of sharp points, indicating that the electronic supermodulation found in YBCO/LCMO bilayers exists across much larger distances than previously thought and extends in all directions, in sharp contrast to the traditional two-dimensional order. The correlation length (a measure of the extent of the supermodulation), estimated from the sharpness of the diffraction peaks, is about 42 unit cells in-plane (double the length reported for bulk YBCO) and about 19 unit cells out-of-plane (compared to 1 unit cell in bulk cuprates). Moreover, the periodicity of the supermodulation has both an in-plane component and an out-of-plane component, with the in-plane component repeating every four copper atoms and the out-of-plane component repeating every eight copper-oxide planes.

Discovery of this three-dimensional electronic supermodulation in YBCO/LCMO is a significant advance in our understanding of the mechanisms that underlie superconductivity and competing orders in cuprates. Furthermore, it may also help explain the seemingly paradoxical "long-range proximity effect" observed across several tens of nanometers near YBCO/LCMO interfaces, whereas mutual suppression of superconductivity in YBCO and ferromagnetism in LCMO is expected to extend only about one nanometer. The existence of competing orders in YBCO, stabilized by the LCMO, may help explain the long-range suppression of superconductivity even when ferromagnetism is limited to the interfacial region. Improving our understanding of the phases that compete with and degrade superconductivity helps theorists and experimentalists recognize and focus on the important physical interactions in condensed matter physics that are dominated by "electron correlation" effects.



(a) Schematic of a sample thin-film heterostructure. The thin films consist of ABO_3-type alternating layers (perovskite structures), with the individual atomic layers within each (AO and BO_2) indicated by different colors. (b) X-ray absorption spectrum measured near the copper L_3 absorption edge. Inset: Schematic of the scattering geometry with linear (π) incident polarization in the horizontal scattering plane. (c) Diffraction data at the 930.85 eV resonance peak from (b), showing a sharp spot near (-0.245, 0, 1.38) in reciprocal lattice units (r.l.u.). A second diffraction peak (not shown) appears near (+0.245, 0, 1.38).

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