MS19 Experimental and theoretical advances in quantum crystallography

MS19-03
X-ray diffraction - a suitable probe for superconductivity?
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Abstract
X-ray diffraction (XRD) techniques clearly have their merits in superconductor research: Over the past years, they have provided valuable information on the atomic structure of superconducting materials as a basis for further experimental and theoretical work. Temperature-dependent XRD experiments have also helped to elucidate subtle structural rearrangements linked to the onset of superconductivity in low-dimensional compounds like *1T*-$TiSe_2$ [1] or high-T_c superconductors like $YBa_2Cu_3O_{7-\delta}$ [2].
But may the sensitivity of XRD to the (one-)electron density distribution in a crystal be leveraged to directly track not only structural but also electronic changes connected to the onset of superconductivity? Previous powder XRD work on the BCS superconductor MgB_2 seems to imply this: Analysis of experimental data at 15 K and at room temperature by means of the maximum entropy method (MEM) pointed out an accumulation of additional electron density at the locations of bond-critical points between the boron atoms in MgB_2 below $T_c \approx 39$ K [3].
Yet, the large temperature difference between the experiments and the model-free nature of MEM leave open questions about the precise physical origin of these findings. We therefore tracked electron density changes across T_c by collecting temperature-dependent single-crystal XRD data with high resolution and refining flexible multipolar models [4]. In this way, detected electron density differences (see figures) could be decomposed into contributions from changes in lattice parameters, dynamic or static atomic displacements or bonding-induced density shifts. It turned out that the increase of electron density at the location of the boron-boron bond-critical point upon cooling from room temperature to 13.5 K is rather related to changes in thermal smearing than to changes in bonding density. We could furthermore show that previous reports on magnesium vacancy concentrations in the range of 5% in MgB_2 [5-8] may be related to the lacking flexibility of the independent atom model (IAM) to account for significant bonding-induced density shifts in the compound.

References
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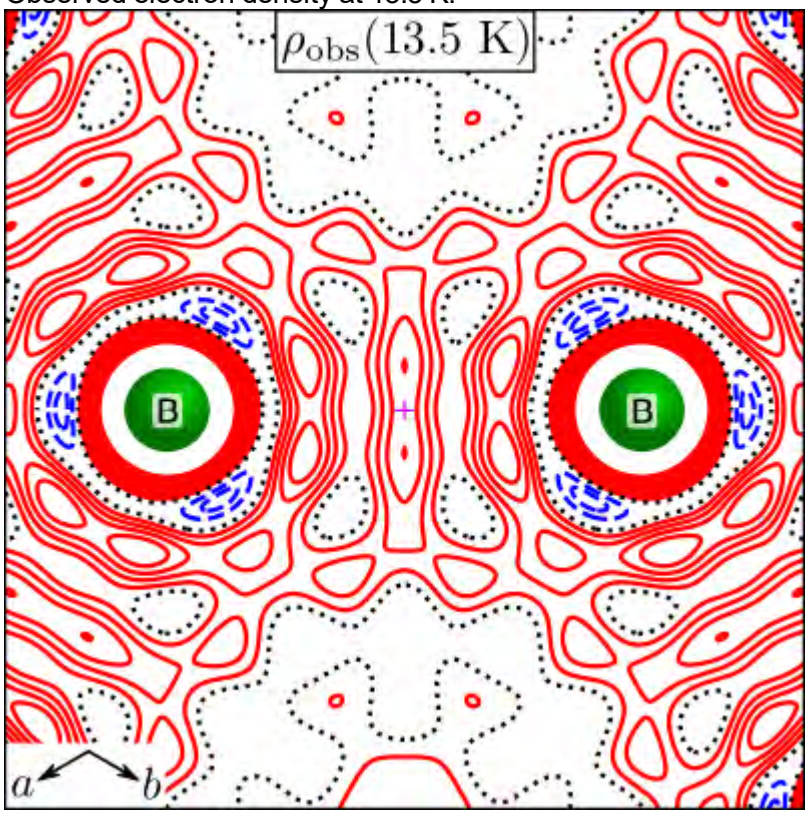
Observed electron density at 13.5 K.



Observed electron density at room temperature.