Abstract

The general objective of this Doctoral Dissertation, entitled *"Modification of quantum states in strongly correlated systems by applying uniaxial pressure"*, was to investigate how uniaxial stress, applied as a controlled symmetry-breaking perturbation of the crystal lattice, affects the structural, magnetic, and electronic properties of materials exhibiting strong electronic correlations. Uniaxial pressure has recently emerged as a powerful experimental tool that complements hydrostatic pressure, electric fields, and magnetic fields by enabling direction-selective tuning of lattice distortions without altering the chemical composition. While hydrostatic pressure uniformly shortens all interatomic distances, uniaxial pressure modifies only selected crystallographic directions, allowing one to probe and control the anisotropic balance between competing interactions such as electron–electron, electron–phonon, and magnetic couplings. This approach is particularly valuable in correlated systems where doping or substitution, such as carrier doping in oxides, inevitably introduces disorder, thereby obscuring intrinsic correlation effects.

A major technological challenge in studying (correlated) systems under uniaxial stress lies in achieving high mechanical precision and reliable quantitative calibration of the applied load. The few experiments performed so far have often lacked accurate pressure determination. Therefore, a key component of this Dissertation was the design, construction, and calibration of a compact uniaxial pressure cell (SOL), enabling quantitative, reproducible, and direction-selective compression. The developed cell operates as a screw-driven vise and is compatible with multiple measurement techniques, including X-ray diffraction (XRD), electrical resistivity, alternating current (AC) magnetic susceptibility, and X-ray absorption spectroscopy (XAS).

From the broad family of strongly correlated materials, stoichiometric magnetite (Fe_3O_4) was chosen as the model system because it combines long-range ferrimagnetic order with the Verwey transition at approximately $T_V \approx 123$ K, a coupled charge, orbital, and structural transition from cubic $Fd\bar{3}m$ to monoclinic Cc symmetry. Modifications of stoichiometry, substitutional doping, or hydrostatic pressure are known to suppress the Verwey temperature (T_V), whereas theoretical predictions and limited experimental data suggest that uniaxial compression can increase it. Thus, magnetite represents an ideal platform for studying how lattice symmetry breaking affects correlated electron states.

A series of synchrotron XRD experiments was performed at the ID11 beamline of the ESRF, with uniaxial compression applied along the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions. The diffraction data revealed an anisotropic elastic response: compression along $\langle 100 \rangle$ induced a tetragonal distortion, while compression along $\langle 110 \rangle$ resulted in an orthorhombic deformation. By comparing the evolution of lattice parameters measured in the SOL cell with reference data obtained from a calibrated ADMET mechanical press, a quantitative calibration was established between screw rotation and applied stress.

The microstructure of two types of high-quality magnetite single crystals, one grown by the traveling-solvent floating-zone (TSFZ) method and the other using the skull-melter technique, was first examined using Dark-Field X-ray Microscopy (DFXM) at the ID03 beamline of the ESRF. The synthesis method was found to leave a distinct structural fingerprint. Crystals produced by the skull-melter method exhibited low average strain but pronounced mosaic textures originating from rapid quenching, whereas TSFZ-grown crystals showed higher axial strain yet fewer extended defects. Since such microstructural characteristics can significantly influence the material's response to external stress, the TSFZ-grown crystals were selected for the subsequent uniaxial compression experiments.

Because uniaxial stress induces directional lattice distortions, the remanent strain fields persisting after decompression were also investigated using DFXM. The imaging revealed that stresses exceeding approximately 150 MPa generate stable, line-like dislocation networks on the order of 10 micrometers in size, preferentially aligned with the {111} slip planes. Despite the presence of these extended dislocations and residual strain fields, AC magnetic susceptibility measurements showed that the Verwey transition temperature remained unchanged even in samples subjected to stresses above 300 MPa. This observation indicates that extended dislocation networks do not affect the cooperative charge and orbital ordering associated with the Verwey transition.

The calibrated SOL cell was then used to study *in situ* the evolution of electrical resistivity and magnetic susceptibility under uniaxial compression along ⟨100⟩ and ⟨110⟩. In both orientations, the Verwey temperature increased linearly with applied stress, indicating the stabilization of the low-temperature monoclinic phase. The effect was strongly anisotropic: the enhancement of T_V under ⟨110⟩ compression was nearly an order of magnitude larger than under ⟨100⟩. This difference reflects the distinct strain symmetries induced by compression, orthorhombic for ⟨110⟩ and tetragonal for ⟨100⟩, and the stronger coupling of the former to the charge and orbital ordering mechanisms driving the Verwey transition. At room temperature, resistivity decreased linearly with compression, consistent with strain-enhanced carrier mobility, again showing a threefold stronger effect along ⟨110⟩ than along ⟨100⟩.

Complementary Fe K-edge X-ray absorption spectroscopy (XAS) measurements at room temperature were carried out to probe possible stress-induced modifications of the local electronic and structural environment. Analysis of the pre-edge region (7105–7120 eV), sensitive to $3d$–$2p$ hybridization and inversion-symmetry breaking, revealed no measurable variations in peak position, width, or intensity up to about 200 MPa. This invariance demonstrates that uniaxial stress along ⟨100⟩ and ⟨110⟩ induces tetragonal and orthorhombic distortions of the cubic lattice without breaking the local inversion symmetry of the FeO_6 octahedra. Consequently, the local Fe–O coordination remains unaffected, while macroscopic properties such as T_V are shifted due to collective, symmetry-preserving distortions.

Taken together, the results presented in this Dissertation establish a comprehensive and self-consistent picture of the coupling between lattice strain, electronic correlations, and phase transitions in magnetite. Moderate uniaxial stress modifies the global symmetry of the crystal and tunes the collective charge-orbital order without altering the local electronic configuration. The successful design and calibration of the SOL uniaxial cell provides a quantitative experimental platform for probing other correlated electron systems, such as high-temperature superconductors, charge-density-wave materials, or heavy-fermion compounds, where controlled symmetry breaking may serve as a route to tailor emergent quantum states.

Keywords: magnetite, uniaxial stress, correlated systems, synchrotron radiation, dark-field X-ray microscopy, electronic transport, magnetic susceptibility