Contribution ID: **144** Type: **Invited**

Nano-optical imaging and spectroscopy of mesoscopic phase behavior in quantum matter

Friday, 27 July 2012 10:45 (25 minutes)

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The rich phase behavior of correlated matter including colossal magnetoresistance, superconductivity, or multiferroicity has made them candidates for a wide range of technological applications. However, the underlying microscopic electronic, lattice, and spin interactions that give rise to these complex materials properties are yet poorly understood. The investigation of these materials is further complicated by frustration and degenerate ground states that can lead to phase competition and coexistence of multiple phases.This often gives rise to structural and electronic inhomogeneities and mesoscopic spatial phase separation on atomic to microscale dimensions.

We will discuss the combination of the new nano-optical scanning probe techniques of scattering scanning near-field optical microscopy (s-SNOM) with different linear, inelastic, nonlinear, and ultrafast spectroscopies. By selective optical near-field coupling to electronic, lattice, and spin degrees of freedom and their symmetries these different spectroscopic implementations allow for probing the associated domain architecture with nanometer spatial resolution. By selecting one or more suitable optical interactions, multiple order parameter can be imaged simultaneously under the influence of sample strain, high magnetic or electric fields, or cryogenic and variable temperature.

Specific examples include the investigation of the complex nanoscale phase separation between metallic and different insulating phases in the metal-to-insulator transition in VO_2 microcrystals by combining near-field Raman and IR (Drude response) spectroscopy. The resulting nanoscale strain-temperature phase diagram provides insight into the competition between the intrinsic phase behavior and external strain, temperature, and photo doping. The extension of tip-enhanced Raman spectroscopy allows for phonon Raman nano-crystallography, e.g., for the study of finite-size effects on the ferroelectric order in BaTiO_3 nanocrystals or BiFeO_3. The spatial and time-reversal symmetry selectivity of optical second-harmonic generation can access the coupled ferroelectric and antiferromagnetic nanoscale domain topology and order in intrinsic multiferroics, e.g., YMnO_3, and provide insight into the mechanisms of the magnetoelectric coupling. In conclusion I will provide an outlook based on our recent developments on femtosecond optical control for nanoscale ultrafast spatio-temporal imaging.

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Session Classification: Nanoscale Spectroscopies

Track Classification: Nanoscale Spectroscopies