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Phononic and electronic excitations in complex oxides studied with advanced Infrared and Raman spectroscopy techniques

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Promising candidates for next-generation quantum devices are the *transition metal-oxide perovskites* (also called *complex oxides*), which host a plethora of novel physics and are often very susceptible to external perturbation parameters, yielding complex phase diagrams. The macroscopic effects in complex oxides are governed, to a large extent, by the behavior of the phonons and electrons. A suitable approach to study these low-energy dynamics is optical spectroscopy, but its real potential for investigating and controlling solid state systems has not been fully exploited.

This PhD work presents advanced infrared and Raman spectroscopy techniques in combination with various external stimuli, which have been used to obtain valuable information on the low-energy electronic and phononic dynamics of several complex oxide systems in bulk form or as thin film heterostructures.

In particular, the magnetic field effect on an underdoped high-temperature superconductor $YBa_2Cu_3O_{6+x}$ has been studied by means of THz-infrared reflectometry and up to 30 Tesla. Besides a complete suppression of the macroscopic superconducting response, surprisingly weak changes in the optical spectra were observed, which is a weak reduction of the free carriers and a compensation by two electronic modes and the mid-infrared band. These optical signatures of the 3D charge density are readily seen at higher temperatures and even at zero magnetic field, which leads to the conclusion that the relationship between superconductivity and the charge ordering is not purely competitive.

Secondly, we have detected backfolded acoustic phonons for the first time in metal-oxide superlattices by means of confocal Raman spectromicroscopy, and have demonstrated the modes' immense capability as a reliable diagnostic for, in principle, any type of superstructures. These modes are not affected by the substrate and are highly sensitive to atomic-scale variations of the bilayer thickness.

Finally, the electric field effect on $SrTiO_3$- and $KaTiO_3$-based heterostructures was explored using a unique combination of infrared ellipsometry and confocal Raman spectroscopy. In the $SrTiO_3$ samples, we have observed a large electric field modulation which is non-collinear and asymmetric with respect to the applied electric field, that can be explained by the interplay of the oxygen vacancies and the tetragonal domain boundaries (absent in $KaTiO_3$). These results will be relevant for the growing research fields of oxide electronics and domain wall nanoelectronics, and have the potential to enhance the performance of oxide devices and give rise to new functionalities.

Jury:
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