Electronic Symmetry Breaking Probed via Local In-Gap Excitations in NiO and CoO: Application of First-Principles Wannier Functions to Linear Response of Strongly Correlated Systems

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Recent inelastic X-ray scattering measurement discovered striking sharp peaks well below the Mott-Hubbard gap in NiO and CoO at large q-vectors. More puzzling is the strong angular dependence of these excitations that shows different trend in NiO and CoO, regardless their similar electronic structure.

Within local Wannier picture, these in-gap modes are shown to be dipole forbidden, on-site d-d excitations, which become dominant at large q. Employing first-principles energy-resolved Wannier functions, obtained with LDA+U functional and our symmetry-respecting construction, a new set of "selection rules" along special nodal directions are identified, from the symmetry of the underlying Wannier functions of the particle-hole pairs. Our results explain very well the experimentally observed strong angular dependence of the excitations. Moreover, the different trend in CoO is shown to result from breaking of local cubic symmetry into rhombohedral ones upon magnetic order, thus demonstrating the sensitivity of the response function to the electronic symmetry breaking.

This work illustrates the usefulness of first-principles Wannier functions in understanding strongly correlated systems, and highlights one potential approach of linear response of time-dependent density functional theory beyond Kohn-Sham scheme.

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