

Orbital fluctuations and orbital order below the Jahn-Teller transition in Sr₃Cr₂O₈

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Zhe Wang, Joachim Deisenhofer, Michael Schmidt, Franz Mayr, Hans-Abrecht Krug von Nidda, and Alois Loidl

Experimental Physics V, Center for Electronic Correlations and Magnetism,
Institute of Physics, University of Augsburg, D-86135 Augsburg, Germany

Yuan Wan

Department of Physics and Astronomy, Johns Hopkins University, Baltimore, Maryland 21218, USA

Diana Lucia Quintero-Castro, A. T. M. Nazmul Islam, and Bella Lake

Helmholtz-Zentrum Berlin für Materialien und Energie, D-14109 Berlin, Germany

We report on the magnetic-, phononic-, and crystal-field-excitation spectrum of a spin-gapped system Sr₃Cr₂O₈ determined by Terahertz and infrared (IR) spectroscopy across the Jahn-Teller (JT) transition at $T_{JT} = 285$ K. We identify the spin singlet-triplet excitations in the dimerized ground state and reveal the corresponding selection rules, which rely on an inter-dimer Dzyaloshinskii-Moriya (DM) interaction with DM vector parallel with crystalline a -axis. The temperature-dependent feature of magnetic and phononic excitations supports the existence of strong orbital fluctuation in an extended temperature regime $T < T < T_{JT}$ with $T \sim 120$ K, in agreement with the results from Raman spectroscopy and electron spin resonance (ESR) measurements.[1,2] The strong fluctuation regime results from the competition between spin-orbital interaction and JT interaction, since, in contrary to the effect of JT distortion, spin-orbital interaction stabilizes the chromium orbital of $d_{x^2-y^2}$ with respect to d_{z^2} . The excitation corresponding to the split of the orbitals due to spin-orbital interaction is observed from $T > T_{JT}$ down to T , whose energy is around 3 meV consistent with the estimation based on the results of ESR and crystal-field-excitation spectrum. Below T , JT interaction is dominant that the ordering of lower-lying d_{z^2} orbital is achieved.

[1] Zhe Wang et al., Phys. Rev. B 83 201102 (2011)

[2] D. Wulferding et al., Phys. Rev. B 84, 064419 (2011)

Primary author: WANG, Zhe (EP 5, EKM, Institute for Physics, Augsburg University, Augsburg, Germany)

Presenter: WANG, Zhe (EP 5, EKM, Institute for Physics, Augsburg University, Augsburg, Germany)

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