Contribution ID: **100** Type: **Student Poster Competition**

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

Monday, 23 July 2012 20:00 (2 hours)

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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 Dzyaloschinskii-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* ~120K, 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)

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Session Classification: Poster Session 1

Track Classification: Spin Phenomena