

71. Title: Orbital fluctuations and orbital order below the Jahn-Teller transition in Sr₃Cr₂O₈

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Source: PHYSICAL REVIEW B

Volume:83

Issue:20

Pages: 201102

Publication year: 2011

Document type:Journal article (JA)

Abstract: We report on the magnetic and phononic excitation spectrum of Sr₃Cr₂O₈ determined by THz and infrared (IR) spectroscopy, and electron spin resonance (ESR) measurements across the Jahn-Teller transition, which is detected by specific-heat measurements to occur at $T_{JT} = 285$ K. We identify the singlet-triplet excitations in the dimerized ground state and estimate the exchange couplings in the system. ESR absorptions were observed up to $T^* = 120$ K with a linewidth. $\exp(-\Delta/k(B)T)$ and $\Delta/k(B) = 388$ K, indicating a phonon-mediated spin relaxation via the excited orbital state of the Cr e doublet in the orbitally ordered state. Upon entering the low-symmetry Jahn-Teller distorted phase below T_{JT} , we find an extended regime $T^* < T < T_{JT}$, where the IR active phonons change only gradually with decreasing temperature. This regime is associated with strong fluctuations in the orbital and lattice degrees of freedom, in agreement with the loss of the ESR signal above T^* . Using the measured magnetic and phononic excitation spectrum we model the orbital contribution to the specific heat and find the persistence of strong fluctuations far below T_{JT} .