

MAR07-2006-002703

Abstract for an Invited Paper
for the MAR07 Meeting of
the American Physical Society

Nonlinear optical signatures of the tensor order in $\text{Cd}_2\text{Re}_2\text{O}_7$ ¹

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The pyrochlore oxide $\text{Cd}_2\text{Re}_2\text{O}_7$ undergoes a structural phase transition at $T = 200$ K with an unusual tensor character.² The order parameter for this state is two-dimensional, and associated with a phonon mode with E_u symmetry. In magnetically frustrated pyrochlores, magnetoelastic coupling to similar modes can induce fascinating magnetically ordered states;³ $\text{Cd}_2\text{Re}_2\text{O}_7$ is nonmagnetic, so it is possible to study the structural instabilities of the pyrochlore lattice in isolation. We have used optical second harmonic generation with polarization sensitivity to resolve an ambiguity in the low-temperature crystal structure, and verify an auxiliary condition on the structure that is implied by the order parameter symmetry. We also show that the temperature-dependence of the order parameter is consistent with thermal occupation of a Goldstone mode that results from the E_u order parameter symmetry. The methodology that we have developed may be applied more widely in characterizing ordered states in matter.

¹Work supported by NSERC; the Canadian Institute for Advanced Research; the Sloan Foundation; the Research Corporation; and the Division of Materials Sciences and Engineering, Office of Basic Energy Sciences, US Department of Energy.

²J. C. Petersen *et al.*, *Nature Phys.* **2**, 605 (2006); C. A. Kendziora *et al.*, *PRL* **95**, 125503 (2005); I. A. Sergienko *et al.*, *PRL* **92**, 065501 (2004).

³Y. Yamashita and K. Ueda, *PRL* **85**, 4960 (2000); O. Tchernyshyov *et al.*, *PRL* **88**, 067203 (2002) and *PRB* **66**, 064403 (2002).