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Photoinduced EPR study of Sb²⁺ ions in photorefractive Sn₂P₂S₆ crystals

A. T. Brant, L. E. Halliburton, S. A. Basun, A. A. Grabar, S. G. Odoulov, A. Shumelyuk, N. C. Giles, and D. R. Evans Phys. Rev. B **86**, 134109 – Published 10 October 2012

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ABSTRACT

Single crystals of $\mathrm{Sn_2P_2S_6}$ are both ferroelectric and photorefractive. Antimony (Sb) ions are optically active in this material and play an important role in optimizing the photorefractive response. Electron paramagnetic resonance (EPR) is used to determine the site and charge states of the Sb ions in $\mathrm{Sn_2P_2S_6}$ and to illustrate the photocharging behavior of these ions. In as-grown crystals, $\mathrm{Sb^{3+}}$ ions substitute for $\mathrm{Sn^{2+}}$ ions. A multiline EPR spectrum from $\mathrm{Sb^{2+}}$ ions is observed after exposing a crystal at 30 K to either 633- or 442-nm laser light. These $\mathrm{Sb^{2+}}$ ions are thermally stable at low temperature after the light is removed. They revert back to $\mathrm{Sb^{3+}}$ ions when the crystal is warmed above 250 K for a few minutes. The EPR spectrum has S=1/2 and consists of well-resolved sets of hyperfine lines from $^{121}\mathrm{Sb}$ and $^{123}\mathrm{Sb}$ nuclei. Spin Hamiltonian parameters are obtained from the angular dependence of the spectrum (principal values are 1.810, 1.868, and 1.887 for the g matrix and 1404, 1687, and 1849 MHz for the $^{121}\mathrm{Sb}$ hyperfine matrix). These parameters provide evidence that the wave function for the unpaired spin has significant p-like character and has overlap with neighboring ions.

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Received 17 April 2012

DOI: https://doi.org/10.1103/PhysRevB.86.134109

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