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Optically induced transient absorption related to formation of small polarons in $Sn_2P_2S_6$ in the sub-100-fs time domain[1] VOLKER DIECKMANN, HOLGER BADORRECK, MIRCO IMLAU, Department of Physics, University of Osnabrueck, Osnabrueck, Germany, ALEXANDR SHUMELYUK, Institute of Physics, National Academy of Science, Kyiv, Ukraine — The interaction of sub-100-fs light pulses with single crystals of nominally undoped $Sn_2P_2S_6$ is studied in the NIR spectral range ($590 - 1630$ nm). A predominant contribution of two-photon absorption (TPA) is verified. Scans over the photon energy show that the TPA coefficient increases in a superlinear way for photon energies exceeding $E_g/2$; for any photon energy it is nearly independent of propagation direction and polarization of the incident beam. The coefficient saturates at a maximum value of $\beta \approx 8$ cm GW^{-1} at $E_p \approx 1.8$ eV. It drops when reaching the bandgap E_g. The TPA coefficients are higher by a factor of two than the values reported for other wide bandgap ferroelectrics, such as $LiNbO_3$, while being lower in comparison to semiconductor crystals. Using fs-pump-probe measurements at 626 nm, a transient absorption is observed that persists for probe pulse delays much longer than the pump pulse duration, up to 2.5 ns. Such transients are typical for a variety of wide bandgap ferroelectrics, where they are described by optically generated polaronic states. We discuss our results in the frame of the microscopic structure of $Sn_2P_2S_6$ with emphasis on the optical generation of S^- small hole polarons.

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