Glasses in the binary system (100-x)Pb2P2O7-xMoO3, with x = 10 – 80 mol %, were synthesized by conventional melt-quenching technique. Thermal analysis, linear optical absorption, refractive index measurements, Raman scattering and nonlinear (NL) optical experiments were performed to characterize the samples. The dependence of MoO3 content on thermal, structural and optical properties was investigated. Molybdenum oxide increases both the glass transitions temperature and thermal stability against devitrification up to 50 mol % due to formations of P-O-Mo linkages and the glass network connectivity increases. The nonlinear optical properties were studied at 1064 nm and 532 nm with pulses of ≈17 ps. The NL refractive index measured was +10-19 m2/W at both wavelengths for samples with different relative concentrations of the constituent compounds. At 1064 nm we determined that the two-photon absorption coefficient, , is smaller than the minimum that we can measure ( <0.01 cm/GW) while at 532 nm we measured 0.25 cm/GW. The nonlinear response of the samples is attributed to contributions from the lone electron pairs of Pb2+, MoO3 clusters, and to Mo5+ and Mo4+ ions.
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