

ABSTRACT

PLD of heavy metal oxide thin film glasses: How metastable structures allow improving the nonlinear optical properties?

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Heavy metal oxide (HMO) glasses show physical properties that make them attractive candidates for the production of photonic devices. They are chemically stable, show a broad transparency range and have low phonon energies. Furthermore, they have high linear (n) and nonlinear (n_2) refractive indices, the latter being hundred times higher than that of silica.

In a previous work, HMO transparent thin film glasses of the system $\text{Nb}_2\text{O}_5\text{-PbO-GeO}_2$ were produced from glassy and crystalline targets with large Nb_2O_5 and PbO contents. The non linear response of the films ($\chi^{(3)}$, with n_2 proportional to $\text{Re}(\chi^{(3)})$) was determined by the DFWM technique at 800 nm using 100 fs laser pulses and low repetition rate (1 kHz). $\chi^{(3)}$ was found to be one order of magnitude higher than that of transparent targets and up to 10^3 times higher than that of SiO_2 . In the present work, the nonlinear optical response has been analyzed in the frame of semi-empirical models (modified Lines' equation and BGO model). These models fit for the bulk glasses but not for the film glasses. The strong differences in the optical response of bulk and film glasses are understood in terms of the structure of the glass network. Raman analysis of film glasses show that their network mainly comprises $[\text{NbO}_6]$ units, which ease the formation of transparent glasses in a wide range of compositions, but also $[\text{NbO}_4]$ units, which increase the nonlinear response of the films.