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Effect of post-growth annealing on the optical properties of LiGaS2 nonlinear crystals

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Crystals of the LiBC2 family with B=In, Ga and C=S, Se, Te are considered promising for nonlinear optics in the mid-IR. Among them LiGaS2 (LGS) exhibits the largest band-gap and lowest two-photon absorption and group velocity mismatch. As in the case of other multi-component chalcogenides, deviations from the stoichiometric composition occur in the growth process. To develop a way to correct the composition and improve the optical quality of LGS, we studied changes in transmission, photoluminescence (PL) and photo-induced absorption (PIA) produced by annealing LGS in vacuum, in the presence of Li2S, Ga2S3, and S. The S-containing inclusions were found responsible for light scattering and absorption near 8 μm in the mid-IR. Anion vacancies of two structural types, VS1 and VS2, produce absorption in the 370 nm band and intense PL in pairs of bands at 426/510 and 450/532 nm. The VS concentration grows after vacuum annealing and drops after annealing in Li2S, Ga2S3, and S. The concentration of cation antisite defects (GaLi) is determined mainly by the growth conditions. These defects are responsible for intense yellow PL which is excited through band-to-band transitions and for the intense 330 nm band near the fundamental absorption edge. PIA in the $\leq$850 nm range, responsible for the grey track effect, was found reversible: It appears after UV/blue light illumination and can be removed by IR light. PIA is a result of the capture of free charge carriers by the traps and parameters of the latter were estimated using thermo-activation spectroscopy.