Single and double-element doped GaSe for nonlinear applications

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In this work we present the last achievements on improvement of GaSe crystals. Data on Al, S, In, Te and Ag impurities in different concentrations were analyzed. It was shown that double doping by Al and S results in combined improvement of hardness and optical properties.

Searching for low optical loss, high nonlinear and optical damage threshold, suitable anisotropic materials is still one of the issues in the field of efficient phase matched THz generation. Among the most attractive materials GaSe shows poor mechanical properties and relatively low optical quality caused by the layered structure and technologically uncontrolled defects. These result in a very limited out-of-door application.

We have modified GaSe physical properties by modifying growth technology and by doping with chemical elements: isovalent Al, S, In, Te or Er, not isovalent Ag or the pair of elements (S and In or Al and S). The list of experimental samples consists of more than 50 crystals grown from the melt with different dopant concentrations.

It is possible to obtain high doping level if impurity is able to form solid solution on the base of GaSe. In case of sulfur its concentration can be as large as at least 11 weight %. On the other hand, In concentration can be up to 1.5 weight % and Te up to 1 weight %. For "nonisostructural" dopants the real solubility level was hard to measure. We estimate it to be around 10^{-3} . However, this small value has a large effect on optical properties in the THz range.

The modified technology of Bridgman growth with heat field rotation have decreased optical loss coefficient for 2-3 times. 2.5 weight % S-doped crystal also leaded to 2-3 times decrease optical losses and increased the damage threshold for 4-5 times. Most significant effect on the hardness (up to 300%) comes from Al doping at low doping but it results in small 20-30% increase in frequency conversion efficiency. Te and In resulted in about 20% increase of the hardness and close increase in the frequency conversion efficiency. For the first time we found cumulative effect on the modification of optical loss coefficient and hardness by two element doping. GaSe:S:Al demonstrates best result in the improvement of optical properties and hardness.



Fig. 1. GaSe crystal doped by S and Al.

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