Netiesiniai optiniai reiškiniai polikristaluose

Nonlinear optical phenomena in polycrystalline media

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Random quasi phase matching in disordered materials with second-order nonlinearity opens interesting perspectives in broadband frequency conversion of ultrashort light pulses [1]. A possibility to phase match virtually any wavelength that falls into the transparency range of these materials is provided by a natural disorder of orientations and grain-size distribution of individual crystallites [2]. To this end, polycrystalline zinc-blende semiconductors, such as ZnSe and ZnS, which are optically isotropic but possess second-order nonlinearity due to $\bar{4}3m$ symmetry, emerge as very attractive nonlinear materials. The frequency conversion processes in these materials become particularly efficient in the femtosecond filamentation regime due to high achieved intensity and localization of the pump [3-5]. Polycrystalline strontium barium niobate (SBN) is another interesting nonlinear material, which produces various nonlinear optical processes at multiple wavelengths and directions without angle tuning [6]. Unlike polycrystalline zinc-blende semiconductors, polycrystalline SBN consists of birefringent needle-like randomly distributed ferroelectric domains. Owing to that, harmonics generation produces completely different contributions to the output radiation.

In this Contribution, the results of nonlinear propagation of intense femtosecond mid-infrared laser pulses in polycrystalline ZnS, ZnSe and SBN are presented. It is demonstrated that efficient generation of even and odd harmonics in zinc-blende semiconductors (ZnS and ZnSe) stems from the polycrystalline structure of these materials, which provides broadband random quasi-phase matching for multiple simultaneous three-wave mixing processes, considerably extending the spectral range of the output radiation. More specifically, using sub-μJ, 60 fs, 3.6 μm input pulses, 3.6 octave-wide harmonics-enhanced supercontinuum spanning the 0.4 – 5 μm wavelength range was generated in ZnS, and 3.3 octave-wide supercontinuum spanning the 0.5 – 5 μm wavelength range was generated in ZnSe, whose maximum blue shifts were limited by the short-wave absorption of the crystals. A full set of even and odd harmonics up to the 10th order in ZnS and up to the 8th order in ZnSe were recorded with 100 fs input pulses at 4.6 μm (Fig. 1).

Another set of experiments demonstrate that polycrystalline SBN serves as an excellent nonlinear material for supercontinuum generation in the near- and mid-infrared, being pumped by 100 fs pulses in the regions of its normal (1.2 μm), zero (1.96 μm) and anomalous (2.4 μm) group velocity dispersion. Moreover, it is shown that broadband out-off axis second harmonic generation usefully serves to visualize and monitor precisely filamentation dynamics, allowing to accurately establish position of the nonlinear focus and to determine the nonlinear index of refraction applying the Marburger`s law. It is found that n_2 surprisingly increases toward longer wavelength, which is in stark contrast with what is expected from the general dispersion law of n_2. It is suggested that the measured increase of n_2 is resulted by the contribution of cascaded second-order nonlinearity, which increases with increasing the incident wavelength.

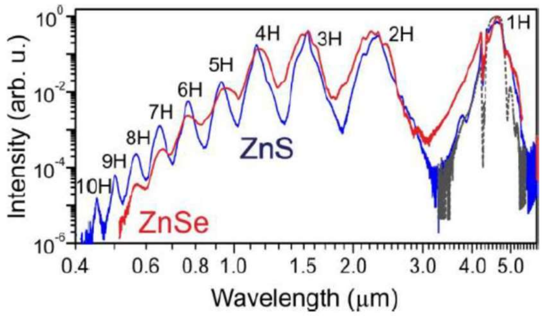

Fig. 1. Output spectra of 2 mm thick ZnS (blue) and 3 mm thick ZnSe (red) polycrystalline samples, produced by filamentation of 100 fs, 1.5 μJ input pulses at 4.6 μm. The input pulse spectrum is shown by grey dashed curve. Harmonic orders are labelled on the tops of the peaks

These results demonstrate that simultaneous nonlinear effects in polycrystalline materials: harmonics generation due to quadratic nonlinearity, and self-focusing and filamentation due to cubic nonlinearity, eventually produce ultrabroadband, multioctave supercontinuum radiation that covers a remarkably broad spectral range from the visible to the mid-infrared.

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