Femtosekundinių šviesos gijų indukuojamas kelių oktavų pločio superkontinuumo formavimasis bei harmonikų generacija polikristaliniame ZnSe

Multi-octave supercontinuum and harmonic generation induced by femtosecond filamentation in polycrystalline ZnSe

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Nonlinear photonic crystals are the structures with spatially modulated quadratic nonlinearity [1] offering new possibilities to manipulate the nonlinear three wave interactions in a desired way. Naturally grown disordered polycrystalline materials consisting of a large number of single-crystal domains with random orientations, random shapes and random sizes, represent a particular class of nonlinear photonic crystals, often termed as random, or short-range order nonlinear photonic crystals [2]. The so called "random quasi phase matching", which stems from the disorder of nonlinear domains [3], greatly extends the fundamental limits of frequency conversion that are imposed by the phase mismatch between the interacting waves, without any additional adjustments [4]. Due to greatly relaxed phase matching conditions, random quasi phase matching allows phase matching conditions for any second-order (three wave interaction) process to be fulfilled, therefore enabling a broadband frequency conversion within a wide spectral range, with the limitations of the bandwidth being imposed just by the transparency window of the material.

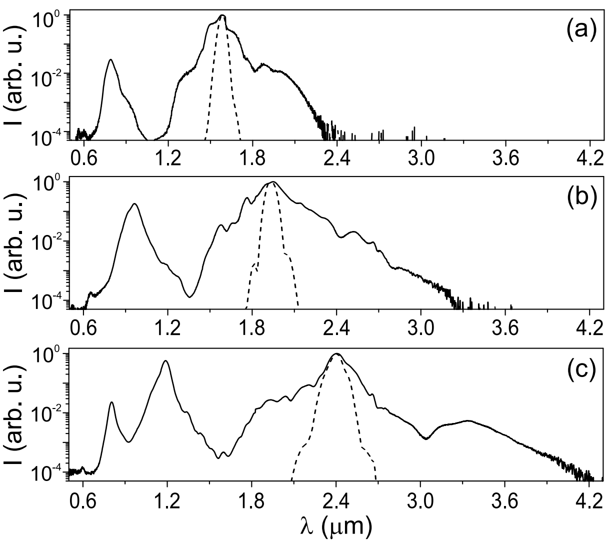

Fig. 1. The output spectra of 5 mm-long polycrystalline ZnSe sample, as generated with incident wavelengths: (a) $\lambda_i = 1.55$ μm, (b) $\lambda_i = 1.9$ μm, (c) $\lambda_i = 2.4$ μm. The input pulse energies are 1.25 μJ, 1.9 μJ and 3 μJ, respectively. The input spectra are shown by dashed curves.

Polycrystalline materials, such as zinc-blende semiconductors, appear as attractive nonlinear media in the mid-infrared spectral range. These materials are optically isotropic, but do not have a center of inversion as they belong to the point group $\bar{4}3$m, thereby possessing a nonzero second-order nonlinearity. In particular, polycrystalline zinc selenide (ZnSe), owning a promising set of optical properties, is a long-known nonlinear crystal for frequency conversion in the mid-infrared spectral range [5].

In this work we studied self-focusing and filamentation of $1.5-2.4$ μm tunable 100 fs pulses in polycrystalline ZnSe. We have shown efficient generation of infrared SC accompanied by the broadband emissions at second, third and fourth harmonics (see Fig. 1) with the input energy chosen so as to keep a fairly constant ratio of the input power to the critical power for self-focusing (≈ 50). In particular, with 2.4 μm incident wavelength, the total spectral coverage corresponded to 2.8 optical octaves spanning from 600 nm to 4.2 μm. We also demonstrate that the spectral features, polarization properties and linear energy trends prove that harmonics are generated via simultaneous randomly quasi phase matched three-wave mixing processes due to nonvanishing quadratic nonlinearity of the crystal. Extremely high conversion to broadband second and third harmonic (19% and 0.5%, respectively) is achieved due to filamentary propagation.

Summarizing the above, polycrystalline ZnSe shares the properties of both, isotropic nonlinear material and random nonlinear photonic crystal, opening intriguing perspectives in the rapidly developing field of ultrafast mid-infrared nonlinear optics.

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