Promising materials for THz and second harmonic generation by femtosecond laser pulses

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Nanocomposites (NC) are promising media for the goal of creation of compact intense THz- source [1] as well as for frequency convertor to higher harmonics (HH) due to their large nonlinearities bringing wide set of the opportunities. Large values of permanent dipole moment (PDM) are revealed in nanostructures (~10^2 - 10^3 Debye), such as semiconductor quantum dots (QDs) of ZnO, ZnS, CdSe what is comparable with values of transition dipole moments between the exciton states. As well, the additional transitions between exciton states allowed due to PDM may play a significant role in the nanoparticle response, in particular for the generation of new frequencies in the THz range.

In the work presented we optimize the conditions of THz and second harmonic (SH) generation in NC consisted of QD inclusions (ZnO) in transparent dielectric matrix (SiO_2 or KDP host). Pumping pulse carrier frequency is resonant to the frequency transition between lower excitonic states. Theoretical models allowing study coupled resonant and non-resonant mechanisms of frequency down and up conversion in nanocomposite have been developed. Simulations of the pulse propagation were performed on the base of self-consisted system for the density matrix (Bloch) equations describing multilevel excitonic transitions and the unidirectional propagation equation, accounting for chromatic dispersion, second- and third-order optical nonlinearities of both host and inclusions, photoionization of inclusions, plasma dynamics (in case of higher pulse input power) and its influence on the dielectric function of the inclusions. Numerical simulations were performed using a SOLPIC software developed in the frame of RISE-ATLANTIC project. As an analysis shows, for the case of PDM smaller than 10 Debay and moderate input intensity (less than 0.07 TW/cm^2) the output efficiency of THz may reach 0.23% after 50 mkm of propagation of two 15-fs pulses at central circular frequencies (FWHM) of 2.26 fs^{-1} and 2.40 fs^{-1}, what may be explained by significant exciton resonance contribution. With increase of the input intensity up to 1 TW/cm^2 and PDM up to 100 Debay for pulse propagating over distances 50 mkm in NC it is established that the THz part of the spectrum is increased with the propagation distance. The THz efficiency at the same propagation length is by 2-3 order of magnitude more for the case with lagrer PDM. It has been found for SH possible conversion efficiency for such materials may reach up to 65-70%. The few approaches to the real level structure computation have been proposed.

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References

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