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Delayed Higher-Order Optical Nonlinearities in Noble Gases[1]
MARYAM TARAZKAR, DMITRI ROMANOV, ROBERT LEVIS, Temple University, Philadelphia 19122 — The role of higher-order Kerr effect (HOKE) in femtosecond laser filamentation is currently at the center of a controversy, as alleged crossover from positive to negative nonlinear refractive index at higher intensities was proposed to cause filament stabilization. Experimental evidence of HOKE crossover or lack thereof is being hotly debated. Motivated by this debate, we report the frequency-dependent nonlinear refractive index coefficients n_2 and n_4 for a series of atmospheric-pressure noble gases: helium, neon, argon, krypton, and xenon. The corresponding atomic hyperpolarizability coefficients are obtained via auxiliary static electric field approach developed on the basis of *ab initio* calculations implemented in Dalton program and performed at the CCSD level of theory with t-Aug-cc-PV5Z basis set. The n_4 index is obtained using the relations between the degenerate six-wave mixing coefficient and some other frequency-dependent second hyperpolarizability coefficients, which can be calculated on the basis of n_2 via the auxiliary field approach. For all the investigated gases, the n_4 indices are found to be positive over the wavelength range 300 nm-1500 nm. This result runs counter to the HOKE crossover hypothesis. The calculated n_4 indices demonstrate considerable temporal dispersion, which progressively increases from helium to xenon. This feature implies delayed nonlinearity and calls for modifications in current theoretical models of filamentation process.

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