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Higher-Order Nonlinearity of Refractive Index: the Case of **Argon¹** MARYAM TARAZKAR, (1,2), DMITRI ROMANOV, (1,3), ROBERT LEVIS, (1,2), (1) Center for Advanced Photonics Research, (2) Department of Chemistry, (3) Department of Physics, Temple University, Philadelphia, PA 19122 — Higher-order dynamic Kerr effect (HOKE) is currently at the center of a controversy regarding the mechanisms of laser filamentation. A strong HOKE with a crossover from positive to negative nonlinear refractive index at intensities well below the ionization threshold, would engender plasma-free filamentation and exotic new effects in light propagation. 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 atmospheric-pressure argon gas, calculated via developed coupled cluster cubic response approach implemented in Dalton program. All calculations are performed at the CCSD level of theory with t-Aug-cc-PV5Z basis set. The benchmark dispersion curve for n_2 reproduces correctly the available experimental data and agrees well with previously-reported theoretical calculations. The nonlinear refractive index n_4 is obtained using the relations between different hyperpolarizability coefficients, and the latter are calculated via the auxiliary static electric field approach on the basis of n_2 . We found that the higherorder nonlinear refraction index n_4 is positive over the wavelengths 300 nm-1500 nm. This result runs counter to the HOKE crossover hypothesis.

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