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Highly efficient third-order optical nonlinearities and their frequency dependence in donor-substituted cyanoethynylethene molecules. JOSHUA C. MAY, IVAN BIAGGIO, Department of Physics and Center for Optical Technologies, Lehigh University, TSUYOSHI MICHINOBU, FRANÇOIS DIEDERICH, Laboratorium für Organische Chemie, ETH-Hönggerberg — We report on a new class of organic molecules with record efficiency for application in third-order nonlinear optics (NLO). The third-order polarizability, γ, of several donor-substituted cyanoethynylethene molecules was determined at the off-resonant wavelength of 1.5 microns using four-wave mixing. The nonlinearities were found to be extraordinarily large relative to the small molecular masses and were found to be within 50 times Kuzyk's fundamental limit,[1] with 53×10^{-48} m^5V^{-2} as the highest γ value. Select molecules were further investigated at wavelengths on and surrounding their two-photon (TP) absorption peaks, revealing large TP cross sections and the resonant influence on the real and imaginary parts of γ. Several members of this molecular family can be vapor-deposited and are likely candidates for third-order NLO devices. When considering their small mass, the *specific* γ (γ per molecular mass) for this family (off resonance, at 1.5μm) is up to 6.5×10^{-23} $m^5V^{-2}Kg^{-1}$, approximately one order of magnitude larger than previously known large γ molecules.[2]

[1]M. G. Kuzyk, Opt. Lett. 25, 1218 (2000)
[2]J. C. May et al, Opt. Lett. 30, 3057 (2005)

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