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Large Optical Nonlinearity Induced by Singlet Fission in Pentacene Films¹ YUNLONG LIU, CHUNFENG ZHANG, Nanjing Univ, MIN XIAO, Nanjing Univ: University of Arkansas — By creating two triplet excitons from one photo-excited singlet exciton, singlet fission in organic semiconductors has drawn tremendous attention for its potential application in boosting the efficiency of solar conversion. Here, we show that this carrier-multiplication effect can be used to dramatically improve the nonlinear optical response in organic materials. With the technique of dual-wavelength optical Kerr effect (OKE), we have observed large optical nonlinearity with a magnitude of $\chi^{(3)}$ up to 10^{-9} esu in pentacene films, which is further shown to be a result of singlet fission as demonstrated by the detailed temporal dynamics and wavelength dependence experiment. Through the use of optical heterodyne detected OKE experiment, we have determined both the sign and value of $\text{Re}\chi^{(3)}$ of the pentacene film. Such efficient third order nonlinear optical response has been successfully applied to demonstrate the all-optical switching. The results observed in this work indicate that the singlet fission could be served as an effective strategy to promote the optical nonlinearity in organic molecule systems.

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