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Spectroscopy of the Primary Photoexcitation and the Origin of the Photocurrent in Rubrene Single Crystals HIKMAT NAJAFOV, IVAN BIAGGIO, Lehigh University, VITALY PODZOROV, MATT CALHOUN, MICHAEL GERSHENSON, Rutgers University — By studying and correlating the photoexcitation spectra of several observables connected with excitons and charge transport we are able to gain new insights into the nature of the primary photoexcitation in organic molecular crystals. By simultaneously measuring the excitation spectra of the transient luminescence and of the transient photoconductivity after picosecond pulsed excitation in rubrene single crystals we show that free excitons are photoexcited starting at photon energies larger than 2.0 eV. We observed a competition between photoexcitation of free excitons and photoexcitation into vibronic excited states that subsequently decay into free carriers, while self-trapped molecular excitons are instead formed predominantly through the free exciton. At photon energies smaller than 2.25 eV free charge carriers are only created through a long-lived intermediate state with a lifetime of up to 0.1 ms and no free carriers appear during the exciton lifetime.

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