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Breaking the symmetry of a PbSe quantum dot by a hot electron-hole pair MINH-TUAN TRINH, XIAOYANG ZHU, Columbia University — Optical excitation of semiconductor nanocrystals or quantum dots (QDs) near the bandgap is now well-understood, but the same cannot be said about excitation significantly above the bandgap. Here, we report an observation of symmetry breaking imposed by a hot electron-hole pair from above-gap excitation in PbSe QDs using ultrafast pump-probe spectroscopy. The breaking of symmetry results in a modification of optical dipole selection rules, as well as the broadening and redshift of dipole-allowed transitions, during the picosecond lifetime of the hot carriers. The observations can be interpreted as a transient Stark effect resulting from the bulk-like behavior of the hot electron-hole pair. At a short time scale right after excitation, the hot electron and hole can be viewed as independent carriers that generate a net transient internal electric field which breaks the symmetry of the QD. By varying the excitation energy we show that the symmetry breaking effect increases with excitation energy and disappears at the bandgap excitation. Such a breaking of symmetry via transient Stark effect should be of general significance to the understanding of QD photophysics above the bandgap.

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