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Optical, spintronics and electrical probes of Rashba-splitting in hybrid organic-inorganic lead trihalide perovskites*

Abstract

The electron dispersion relation, $E(k)$ near the extrema points in semiconductors are usually described by the effective-mass approximation, where the electrons and holes are treated as 'free carriers' having an effective mass, m*, that leads to spin-degenerate parabolic dispersion, $E(k)=\hbar^2k^2/2m^*$. However spin-orbit coupling (SOC) can split the spin-degenerate bands in non-centrosymmetric compounds, as firstly realized by Dresselhaus and Rashba (1, 2). In this case the spin-degenerate parabolic band splits into two spin-polarized bands, where the electron (or/and hole) dispersion relation may be described by $E_{\pm}(k)=(\hbar^2k^2/2m^*)\pm\alpha_R|k|$, where α_R is the Rashba-splitting parameter. This formulation yields new extrema at a momentum offset (k_0) and split energy (E_R) that are related to each other via the relation $\alpha_R = 2E_R/k_0$ (3). Importantly, the two Rashba-split branches have *opposite spin sense* that influences the photoexcitations optical and magnetic properties, a situation that may benefit spintronics since it enhances the spin-to-charge conversion efficiency (3, 4).

The hybrid organic-inorganic halide perovskite semiconductors possess strong SOC due to the heavy elements in their building block. These may lead to 'Rashba-splitting' close to the extrema in the electron bands. We have employed a plethora of ultrafast transient and nonlinear optical spectroscopies and photoconductivity for studying the primary (excitons) and long-lived (free-carriers) photoexcitations in thin films and crystals 2D and 3D hybrid perovskites (5-7). From our optical studies we found (5) a giant Rashba-splitting in 2D hybrid perovskites, with Rashba-splitting energy of ~40 meV and Rashba parameter of ~2.7 eV•A°; which are among the highest Rashba-splitting size parameters reported so far. These results have been confirmed by nonlinear optical studies (6) and transient spin-dependent photocurrent (7).

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