Dipole-Driven Photodetection in 0D/2D Heterostructures Enabled by Ligand Engineering and Insulating Interlayers

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Short-wave infrared (SWIR) photodetectors based on colloidal PbS quantum dots (QDs) have emerged as a cost-effective alternative to traditional semiconductor technologies. [1] However, the mechanisms responsible for photocurrent generation in QD/graphene hybrid systems are still not fully understood. In this work, PbS QDs capped with different ligands, including PbS-OA, PbS-$PbBr_2$, and PbS-EDT, are synthesized to systematically investigate the effect of ligand length. FTIR and GISAXS analyses reveal that the ligand type significantly influences the structural ordering of the QD films.To further probe the charge transfer process, a 10 nm Al_2O_3 insulating layer is inserted between the graphene and QD layers, effectively suppressing direct charge transfer and enabling the identification of a dipole-driven detection mechanism. For devices incorporating short-chain $PbBr_2$ ligands, the insertion of Al_2O_3 leads to a decrease in detectivity (from 5.5×10^{12} to 1.2×10^{12} Jones) along with an accelerated response speed, suggesting that photocurrent generation is dominated by carrier extraction. In contrast, PbS-EDT-based devices exhibit minimal changes after Al_2O_3 insertion, indicating that dipole-induced Fermi level modulation is the dominant mechanism.These findings demonstrate that ligand length plays a crucial role in determining the transition between extraction-driven gain and dipole-driven modulation, providing valuable insights for the design of high-performance, fast, and stable QD/graphene SWIR photodetectors.

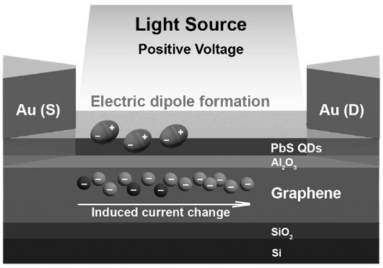

Figure 1. Schematic diagram of depicting the proposed operation model of PbS QDs/graphene photodetector.

[1] De Iacovo A., Venettacci C., Colace L., Scopa L. and Foglia S. (2016) Sci. Rep., 6, 37913.

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