Supplementary Information

An Asymmetric Aluminum Active Quantum Plasmonic Device

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Figure S1: The evolution of the absorption spectrum as a function of the applied external electric field U for d = 12 Å.



Figure S2: The absorption spectra of a model nanoantenna composed of a larger aluminum nanocube (43 atoms) suspended over an aluminum film simulated with TDDFT. The separation *d* was varied from 2 to 14 Å to observe the transition from charge transfer plasmon (CTP) coupling to conventional gap plasmon coupling. The spectra for the individual Al nanoparticle and Al film are included but their intensities were multiplied by a factor of 10 for clarity.



Figure S3: The absorption spectra evolution (as a function of positive- and negative-bias) for a system composed of an Al nanocube (13 atoms) suspended over an Al film.