Supporting Information

Solar Energy Conversion with Tunable Plasmonic Nanostructures for Thermoelectric Devices

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Experimental Procedure:

Synthesis of silver nanocubes: In a typical synthesis (refer to Q. Zhang, W. Li, L.-P. Wen, J. Chen and Y. Xia, *Chem. Eur. J.*, 2010, **16**, 10234), ethylene glycol (50 mL, EG, Aladdin, 1095698-500mL) was added into a 250-mL round bottom flask and heated under magnetic stirring in an oil bath preset to 150 ºC. NaSH (0.6 mL, 3 mM in EG, Sigma-Aldrich, 02326AH) was quickly injected into the heated solution after its temperature had reached 150 ºC. After 2 min, a HCl solution (5 mL, 3 mM in EG) was injected into the reaction solution, followed by the addition of poly(vinyl pyrrolidone) (PVP, 12.5 mL, 20 mg/mL in EG, M.W.=55,000, Sigma-Aldrich, 856568-100g). After another 2 min, silver trifluoroacetate (4 mL, 282 mM in EG, Sigma-Aldrich, 04514TH) was added into the mixture. The reaction mixture was heated at 150 °C in air for 30 min. The samples were washed with acetone and then with ethanol and water several times to remove most of the EG and PVP by centrifugation.

Synthesis of silver nanoplates: In a typical synthesis (refer to Q. Zhang, N. Li, J. Goebl, Z. Lu and Y. Yin, *J. Am. Chem. Soc.*, 2011, **133**, 18931), a 24.75 mL aqueous solution containing silver nitrate (50 μL, 0.05 M, Sigma-Aldrich, 209139-100g), trisodium citrate (0.5 mL, 75 mM) and H_2O_2 (30 wt %, 100 μL) was vigorously stirred at room temperature in air. Sodium borohydride ($NaBH_4$, 250 μL, 100 mM) was rapidly injected into this mixture to initiate the reduction. The reaction mixture was maintained in air for 5 min. The samples were washed with acetone and then with ethanol and water several times by centrifugation.

Characterizations: A drop of the aqueous suspension of particles was placed on a piece of carbon-coated copper grid or silicon wafer and dried under ambient conditions. TEM images were taken on a JEOL JEM-2010 LaB6 high-resolution transmission electron microscope operated at 200 kV. SEM images were taken on a FEI Sirion 200 field emission scanning electron microscopy operated at 5 kV.

UV-vis extinction spectra were taken using an Agilent Varien Cary 60 spectrophotometer. The concentration of silver nanoparticles was measured with a Thermo Scientific PlasmaQuad 3 inductively-coupled plasma mass spectrometry (ICP-MS) after dissolving them with a mixture of HCl and HNO_3 (3:1, volume ratio). I-V measurements of devices were carried out at room temperature using a DC source meter (model 2420, Keithley) operated by LabVIEW5, and a 1,000-W full-spectrum solar simulator (model 94063A, 6×6 inch source diameter, ±4° collimation, Newport Oriel) equipped with an AM 1.5 direct filter.

DDA calculation: The calculation used in this work is based on DDA method. DDA is a computational method developed for studying both the scattering and absorption of electromagnetic radiation by particles with sizes on the order of or smaller than the wavelength of incident light. In the calculation, the particle is divided into an array of *N* polarizable point dipoles, each of which is characterized by a polarizability of $\boldsymbol{\alpha_i}$. When the system is excited by a monochromatic incident plane wave $\mathbf{E_{inc}}$, each dipole of the system will be subjected to an electric field that can be split in two contributions: *i*) the incident radiation field $\mathbf{E}_{i,\mathbf{inc}}$; and *ii*) the field radiated by all of the other induced dipoles $\mathbf{E}_{i,\mathbf{dip}}$. The sum of both fields is the so-called local field at each dipole ($\mathbf{E}_{i,\mathbf{loc}} = \mathbf{E}_{i,\mathbf{inc}} + \mathbf{E}_{i,\mathbf{dip}}$). Each dipole can be expressed as an oscillating polarization with the dipole moment being $\mathbf{P}_i = \boldsymbol{\alpha}_i \cdot \mathbf{E}_{i,\mathbf{loc}}$. Both absorption and scattering cross-sections (C_{abs} and C_{sca}) can be directly obtained from $\mathbf{P}_i$.

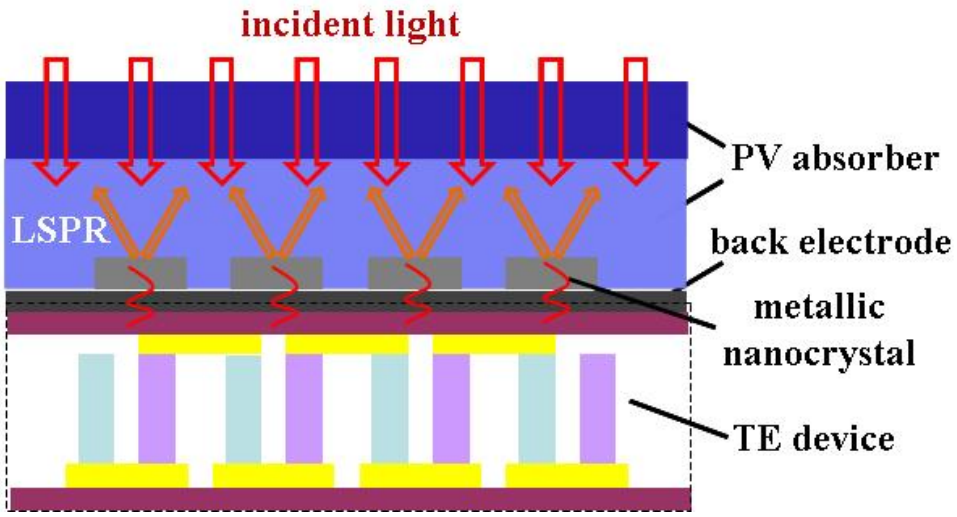

Fig. S1. Schematics illustrating the concept of PV-TE tandem cell structure with plasmonics as the mediation for light harvesting.