The cost of solar energy is a primary factor that limits more widespread adoption, and improved conversion efficiency is the most effective strategy for lowering cost. Although commercial solar cells today generally provide less than 20% conversion efficiency, thermodynamic analyses indicate a maximum efficiency of 95% is possible. In this talk I will show how nanofabricated optoelectronic and plasmonic materials provide a route to achieve this maximum efficiency. Nanostructuring materials enables systematic control of the thermodynamic parameters governing optical power conversion, and optimization can shape, confine, and interconvert the energy and entropy of a radiation field, in order to maximize the excitation of an electronic or chemical storage pathway. Additionally, the remarkable nanoscale tailorability of a variety of structural properties, such as electrochemical potential, can further minimize entropic losses. I will describe two classes of nanoscale structuring that exemplify these opportunities: (1) Single electron transistors (SETs) comprised of individual semiconductor nanocrystals limit the flow of current to one electron at a time. (2) Plasmonic absorption in metal nanostructures provides an entirely new mechanism for generating electrochemical potential from photons. I have termed this behavior ‘the plasmoelectric effect’ (Science, 2014).