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Thermionic-enhanced near-field thermophotovoltaics for medium-grade heat sources

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Abstract

Conversion of medium-grade heat (temperature from 500 to 1000 K) into electricity is important in applications such as waste heat recovery, or power generation in solar thermal and co-generation systems. At such temperatures, current solid-state devices lack of either high conversion efficiency (thermoelectrics) or high-power density capacity (thermophotovoltaics and thermionics). Near-field thermophotovoltaics (nTPV) theoretically enables high power density and conversion efficiency by exploiting the enhancement of thermal radiation between a hot emitter and a photovoltaic cell separated by nanometric vacuum gaps. However, significant improvements are possible only at very small gap distances (< 100 nm), and when ohmic losses in the photovoltaic cell are negligible. Both requirements are very challenging for current device designs. In this work, we present a thermionic-enhanced near-field thermophotovoltaic (nTiPV) converter consisting of a thermionic emitter (graphite) and a narrow bandgap photovoltaic cell (InAs) coated with low-workfunction nanodiamond films. Thermionic emission through the vacuum gap electrically interconnects the emitter with the front side of the photovoltaic cell and generates an additional thermionic voltage. This avoids the use of metal grids at the front of the cell, and virtually eliminates the ohmic losses, which are unavoidable in realistic nTPV devices. We show that nTiPV operating at 1000 K and with a realizable vacuum gap distance of 100 nm, enables a 10.7-fold enhancement in electrical power (6.73 W/cm²) and a 2.8-fold enhancement in conversion efficiency (18 %) in comparison with a realistic nTPV device having a series resistance of 10 m Ω ·cm².

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1. Introduction

Thermionics (TIC)^{1,2} and thermophotovoltaics (TPV)^{3,4} are highly efficient alternatives to thermoelectric generators (TEG)⁵. In TIC, electrons are thermally emitted from a hot emitter/cathode and collected in a cold anode/collector, subsequently producing an electrical current. In TPV, thermally radiated photons are absorbed in a low-bandgap semiconductor and excite electron-hole pairs, which are selectively collected to produce an electrical current. Both TPV and TIC have already demonstrated higher conversion efficiencies than TEG at temperatures beyond 1000 °C (~ 24 % for TPV^{6,7} and ~ 11% for TIC¹). However, the power density is comparatively very low (e.g. less than 1 W/cm² for TPV at 1039 °C⁶, while ~ 20 W/cm² for TEG at 595 °C⁸). The main reason is the lower energy flux of radiated particles compared with that carried by the electrons moved by a temperature gradient within a solid, as in TEG.

Boosting the power density of TIC and TPV is the motivation of current research efforts that aim at increasing the flux of radiated photons (for TPV) and electrons (for TIC). For TIC, most of the research focuses on reducing the workfunction of the emitter and collector, along with reducing the accumulated space-charge by applying magnetic fields or by reducing the vacuum gap that separates the cathode and the anode to micrometric scales^{1,2}. For TPV, at least three strategies were proposed for increasing the energy flux of radiated photons at moderate temperatures: light-pipe TPV (LTPV)⁹, thermophotonics (TPX)¹⁰, and near-field thermophotovoltaics (nTPV)¹¹. Recently, a combination of the last two has been also proposed¹². nTPV is the strategy with the highest theoretical potential. It consists of creating nanoscale vacuum gaps between the emitter and the photovoltaic (PV) cell, so that evanescent waves (photons) tunnel from the emitter to the cell and contribute to generating electrical power. Near-field thermal radiation transport was thoroughly investigated from both theoretical and experimental points of view^{13,14}, and its potential use for heat-to-electricity conversion widely analyzed¹⁵. Only very recently the proof-of concept of nTPV has been finally achieved by measuring a 40-fold enhancement of the electrical output power at gap distances of less than 100 nm¹⁶. However, nTPV has (at least) two main relevant issues that may impede its further deployment: first, the quite high ohmic losses due to the very high current densities that must flow laterally through thin semiconductor layers within the PV cell; second, the very small vacuum gaps that are needed to obtain a significant improvement in electrical power density.

In this work, we present a theoretical analysis of a thermionic-enhanced nTPV device (nTiPV) that eliminates the ohmic losses and enables higher power densities at larger gap distances than conventional nTPV. It is the aim of this work to illustrate the theoretical potential of a specific device with a medium-grade heat source operating at a temperature of 1000 K.

2. Device concept

68 *Figure 1* shows the band diagram of the proposed device, which is the near-field counterpart of
69 the hybrid thermionic-photovoltaic concept introduced previously¹⁷. The system consists of a
70 graphite emitter and an InAs (bandgap of 0.35 eV at 300 K) PV cell separated by a distance d .
71 The emitter and the PV cell are coated with very thin (~ 1 -2 nm) transparent H-terminated
72 diamond films, which have been proved experimentally to provide workfunctions in the range
73 of around 1.4 eV (N-doped films¹⁸) down to around 0.9 eV (P-doped films¹⁹). The emitter is
74 heated by an external heat source, and subsequently radiates photons and electrons towards the
75 PV cell. Due to the emission of electrons, the emitter surface is charged positively. Thus, the
76 radiated electrons are attracted back, and if the distance d is relatively large, they accumulate in
77 the vacuum gap. This regime of operation is named “space-charge-limited” mode, and it is
78 characterized by additional potential barriers ϕ_{EM} and ϕ_{CM} that oppose to the electrons’ flow. On
79 the contrary, if the distance d is small, the radiated electrons are effectively collected at the InAs
80 PV cell surface, without accumulating in the gap, subsequently eliminating any kind of potential
81 barrier, and leading to a drastic enhancement of the thermionic current. When the thermionically
82 emitted electrons reach the PV cell surface, they recombine with the holes photogenerated in the
83 PV cell. Ideally, no electrical potential is created in this process, as in ideal ohmic contacts.
84 Therefore, the full PV cell front side behaves as a transparent collector that ensures the wireless
85 electrical connection between the emitter and the PV cell. The output voltage is thus the
86 addition of the thermionic voltage (V_{TI} , generated between the emitter and the front side of the
87 PV cell) and the photovoltaic voltage (V_{PV} , generated between the front and rear sides of the PV
88 cell). Remarkably, this design avoids the use of front metal grids, eliminating the subsequent
89 shadowing losses, and mitigating the challenges of nano-gap implementation in space-
90 constrained near-field TPV devices. In conventional PV cells, either in front- or back-contacted
91 configurations, the main contributors to the ohmic losses are the currents that flow laterally
92 through the semiconductor and metal layers. These losses are fully eliminated in the nTiPV
93 device, where the current flow is nearly unidirectional and transversal to the device’s area.
94 Results will show that this is a particularly significant benefit for near-field operation, which
95 involves very large current densities.

96 The far-field counterpart of this device¹⁷ is being experimentally developed^{20,21}. The
97 experimental device operates in ultra-high vacuum (UHV) conditions and uses dielectric micro-
98 spacers to create a micrometer vacuum gap between the emitter and the PV cell. Modern
99 microfabrication techniques already enabled the development of thermally and electrically
100 insulated spacers that withstand large temperature gradients. These techniques eventually
101 enabled the experimental demonstration of micron-gap TIC^{22,23}. Sub-micron separation
102 distances were also experimentally realized using nano-spacers in the frame of near-field

103 thermal radiation experiments ²⁴⁻²⁷. Despite the recent experimental demonstration of nTPV ¹⁶
 104 was realized using a suspended emitter and precise alignment tools, current research efforts
 105 target the integration of spacers into stable nano-gap nTPV devices ²⁸. All these recent
 106 progresses should be directly transferrable to the experimental implementation of nTiPV
 107 devices.

108 3. Methods

109 Analysis of the nTiPV device described above requires the calculation of the total net flux of
 110 photons and electrons through nanoscale vacuum gaps, along with the generated current-voltage
 111 characteristics for both thermionic and photovoltaic converters in the near field.

112 For the thermionic part, the electrons' energy flux (Q_{el}), the generated current density (J_{TI}) and
 113 output voltage (V_{TI}), can be calculated by neglecting collector's back emission as ²⁹

$$114 \quad Q_{el} = J_{TI} \frac{(\phi_{max} + 2kT_E)}{e}$$

$$115 \quad J_{TI} = AT_E^2 e^{-\frac{q\phi_{max}}{kT_E}}$$

$$116 \quad qV_{TI} = \phi_E + \phi_{EM} - \phi_C - \phi_{CM}$$

117 where A is the Richardson-Dushman constant, k is the Boltzman constant, q is the electron's
 118 charge, and T_E is the emitter temperature. ϕ_{max} is the maximum of the electric potential created
 119 along the inter-electrode gap (*Figure 1*). In the space-charge-limited mode, $\phi_{max} = \phi_E + \phi_{EM}$
 120 or $\phi_{max} = qV_{TI} + \phi_C + \phi_{CM}$, ϕ_E (ϕ_C) being the emitter's (collector's) workfunction. The
 121 values of energy barriers ϕ_{EM} and ϕ_{CM} can be calculated using the Langmuir theory ²⁹. This
 122 theory assumes one-dimensional and collision-less electron flow with a half-Maxwellian
 123 distribution of velocities. In the so-called retarding mode, V_{TI} is large enough to locate the
 124 maximum of the electrostatic potential at the collector's surface, i.e. $\phi_{max} = qV_{TI} + \phi_C$. The
 125 latter will be the most typical case in the near field, where the very small inter-electrode
 126 distance will nearly eliminate the barriers ϕ_{EM} and ϕ_{CM} and the maximum power point (MPP)
 127 will happen at $\phi_{max} \approx \phi_E$ and $qV_{TI} \approx \phi_E - \phi_C$. In order to analyze the theoretical potential of
 128 the concept, a Richardson constant of 120 A/cm² is assumed. Significant deviations from this
 129 theoretical value are possible depending on the experimental conditions of the deposition of the
 130 emitter film, as well as on the interfacial layers that could be created during this process ¹⁸.

131 For the photovoltaic part, the photons' energy flux is calculated using fluctuational
 132 electrostatics ³⁰ and the S-matrix method for 1D-layered media ³¹. The current density (J_{PV}) –
 133 voltage (V_{PV}) characteristic is calculated by solving the minority carrier diffusion equation in

134 the frame of the low-injection approximation, using methods described elsewhere^{32,33}. The
 135 device consists of four layers sandwiched between two semi-infinite media, respectively made
 136 of graphite (emitter, semi-infinite), vacuum (gap with variable thickness d), p-doped InAs ($N_a =$
 137 10^{18} cm^{-3} , $0.75 \text{ }\mu\text{m}$ thick), n-doped InAs ($N_d = 10^{16} \text{ cm}^{-3}$, $6 \text{ }\mu\text{m}$ thick), gold (back surface
 138 reflector, 200 nm thick) and vacuum (semi-infinite). Radiative, Auger, and Shockley–Read–
 139 Hall (SRH) recombination mechanisms are considered with parameters from³⁴, along with
 140 finite doping and temperature-dependent mobilities for electrons and holes³⁵. The model
 141 assumes that the thermionic layer on the PV cell does not modify the PV cell band diagram in a
 142 way that holes could not diffuse towards the thermionic collector. This is a reasonable
 143 assumption given the presence of electrically active defects in the semiconductor-diamond
 144 interface, as well as the very high doping levels of both the p-doped InAs layer and the diamond
 145 thin film, both effects preventing the creation of Schottky barriers in the semiconductor-
 146 diamond interface^{18,36,37}. Optical properties of InAs corresponding to interband absorption and
 147 interactions with free carriers and phonons, are calculated using the method described in³⁸ and
 148 the Drude-Lorentz model³⁹, respectively, with the parameters of⁴⁰. The Drude model is used
 149 for gold⁴¹. For the sake of simplicity, diamond layers are omitted in the radiation transfer
 150 calculations. Their impact on emission by the graphite emitter and absorption by the InAs cell is
 151 assumed to be negligible, because layers are every thin ($\sim 1\text{-}2 \text{ nm}$), diamond's extinction
 152 coefficient is weak, and diamond's refractive index is of intermediate level between that of
 153 graphite and indium arsenide⁴².

154 Finally, the nTiPV conversion efficiency is given by

$$155 \quad \eta = \frac{[SJ(V_{TI} + V_{PV} - SJR_{lead})]_{\max}}{S(Q_{el} + Q_{ph}) + Q_{lead} - Q_d}$$

156 where $[SJ(V_{TI} + V_{PV} - SJR_{lead})]_{\max}$ is the maximum electrical power at a current density $J =$
 157 $J_{TI} = J_{PV}$ and voltage $V = V_{TI} + V_{PV} - SJR_{lead}$. $Q_{lead} = L(T_E^2 - T_C^2)/2R_{lead}$ is the minimum
 158 amount of heat lost through the emitter's leads having an electrical resistance R_{lead} , $L = \frac{\pi^2 k^2}{3e^2}$ is
 159 the Lorentz number of the metal²⁹, and $Q_d = S^2 J^2 R_{lead}/2$ represents the heat generated in the
 160 leads by Joule effect that is turned back to the emitter, S being the device area, equal to 1 cm^2 in
 161 the current study. The value of R_{lead} can be optimized to fulfil a trade-off between heat losses
 162 and power generation that ultimately maximizes conversion efficiency. For comparison
 163 purposes, the conversion efficiency of a standalone nTPV device is calculated by
 164 $\eta_{nTPV} = [J(V_{PV} - J_{PV}R_s)]_{\max}/Q_{ph}$, R_s being the PV cell series resistance in $\Omega \cdot \text{cm}^2$. In every
 165 calculation involving the search for maximum values, the Nelder-Mead algorithm is used⁴³.

166

4. Results and Discussion

168 *Figure 2 (a) (Figure 2 (b))* shows the generated current density (voltage) of the nTiPV device as
169 a function of gap distance d . Results are shown for two values of the emitter workfunction
170 ($\phi_E = 1.3$ and 1.4 eV), a fixed collector's workfunction ($\phi_C = 1$ eV), and an emitter temperature
171 of $T_E = 1000$ K. The lead resistance (R_{lead}) is optimized at every distance to maximize the
172 nTiPV conversion efficiency. The voltage generated in the photovoltaic (V_{PV}) and thermionic
173 (V_{TI}) stages is also shown in *Figure 2 (b)*, along with the voltage drop in the leads ($-J \cdot R_{lead}$).
174 As explained in the previous sections, both thermionic and photovoltaic currents must be
175 identical within the nTiPV device due to the series interconnection, i.e. $J = J_{TI} = J_{PV}$. This
176 means that their respective internal voltages, V_{TI} and V_{PV} (see *Figure 2 (b)*), must be adapted to
177 meet this condition. However, the maximum power density attainable for each sub-device
178 would be higher if they were biased independently. This is illustrated in *Figure 2 (a)*, which also
179 shows the current densities at the MPP for the independently-biased thermionic (J_{TI}^*) and
180 photovoltaic (J_{PV}^*) devices. This information is valuable for the following discussion.

181 There are three main different regions in *Figure 2 (a)*. For large distances, thermionic emission
182 is strongly reduced by the space-charge effect, also illustrated by the larger thermionic voltage
183 contribution in *Figure 2 (b)*, and limits the total current of the nTiPV device. This causes the PV
184 cell to be biased near open circuit. For intermediate distances, the space charge is mitigated and
185 the flux of thermionically emitted electrons exceeds that of photogenerated charges in the PV
186 cell. This causes an increase (decrease) of the thermionic (photovoltaic) voltage that reduces
187 (increases) the thermionic (photovoltaic) current until both thermionic and photovoltaic currents
188 are identical. In this region, the thermionic device undergoes the transition from space-charge-
189 limited to saturation mode, and the photovoltaic device undergoes the transition from far field to
190 near field. The third region corresponds to the smallest distances at which photovoltaic
191 photogeneration exceeds the thermionic electrons' flux due to the strong near-field enhancement
192 of photons' flux. In this region, the thermionic current is already saturated, with no space-charge
193 effect, and limits the total current of the nTiPV device. Saturation of the thermionic sub-device
194 is characterized by voltages approaching $V_{TI} = \phi_E - \phi_C$ at very small distances (0.3 and 0.4 eV
195 in *Figure 2 (b)*). The transitions through these three regimes take place at two specific distances
196 for which both thermionic and photovoltaic currents are equal. There is one in the near field and
197 another in the far field. Only at these specific distances, both thermionic and photovoltaic sub-
198 devices are biased simultaneously at their respective MPP. In all other situations, one of the
199 devices produces a lower current and it is consequently biased at larger voltages than that of its
200 MPP.

201 *Figure 3* shows the maximum electrical power density (*Figure 3* (a)) and conversion efficiency
202 (*Figure 3* (b)) of nTiPV as a function of gap distance. *Figure 4* rearranges the results from these
203 figures to show conversion efficiency as a function of electrical power density. Results for two
204 kinds of “conventional” nTPV devices are also included: “ideal” nTPV assumes negligible
205 ohmic losses, and “real” nTPV assumes a PV cell with a series resistance of $10 \text{ m}\Omega\cdot\text{cm}^2$.

206 nTiPV generally outperforms nTPV, especially when considering a “real” nTPV device with
207 non-negligible ohmic losses. Even in the case of an “ideal” nTPV (with negligible ohmic losses)
208 nTiPV outperforms nTPV provided that the inter-electrode distance is larger than 100 nm and
209 the emitter workfunction is lower than 1.5 eV. Impact of the emitter workfunction is evident for
210 small distances, where a low emitter workfunction (e.g. $\phi_E = 1.3 \text{ eV}$) is needed to produce a
211 high enough thermionic current and fully exploit the enhancement of the photovoltaic power
212 generation in the near field. In the case of larger emitter workfunctions, the low thermionic
213 current limits the total current of the device and near-field effects are not fully exploited. At
214 larger distances, impact of the emitter’s workfunction is negligible because the nTiPV device is
215 limited either by the photovoltaic current or by the space charge. Quite importantly, nTiPV
216 produces a significantly higher power at larger (more feasible) gap distances (*Figure 3* (a)). For
217 instance, a nTiPV device with an emitter (PV cell) surface workfunction of 1.3 eV (1 eV)
218 produces 6.73 W/cm^2 for a gap distance of 100 nm. This is 3.7 times more electrical power than
219 an idealized nTPV device with negligible ohmic losses (1.82 W/cm^2), and 10.7 times more
220 electrical power than a realistic nTPV device having a series resistance of $10 \text{ m}\Omega\cdot\text{cm}^2$ (0.63
221 W/cm^2). Besides, the conversion efficiency is similar to that of an idealized nTPV device (~ 18
222 %), but significantly higher than that of a realistic nTPV device with non-negligible ohmic
223 losses (6.4 %). Generally speaking, we can state that nTiPV operating at 1000 K theoretically
224 enables reaching power densities and conversion efficiencies greater than 10 W/cm^2 and 15 %,
225 respectively, while realistic nTPV is limited to $\sim 1 \text{ W/cm}^2$ and $\sim 7 \%$ (*Figure 4*).

226 5. Conclusions

227 We have established a conceptual thermionic-enhanced near-field thermophotovoltaic (nTiPV)
228 device for the conversion of medium-grade heat into electricity. The converter comprises an
229 InAs photovoltaic cell and a graphite emitter separated by a nanometric vacuum gap, both
230 elements having engineered low workfunctions. Based on an analytical theoretical model that
231 combines fluctuational electrodynamics and the Langmuir theory, we have shown that nTiPV
232 produces significantly higher electrical power (6.73 W/cm^2) and conversion efficiency (18 %)
233 than conventional near-field thermophotovoltaics (nTPV) using moderately large gap distances
234 (100 nm). The major advantages are the elimination of the ohmic losses and the enhancement of

235 the output voltage. According to these results, nTiPV could significantly outperform current
236 thermoelectric devices for the conversion of medium-grade heat sources into electricity.

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249 emitter and the PV cell.

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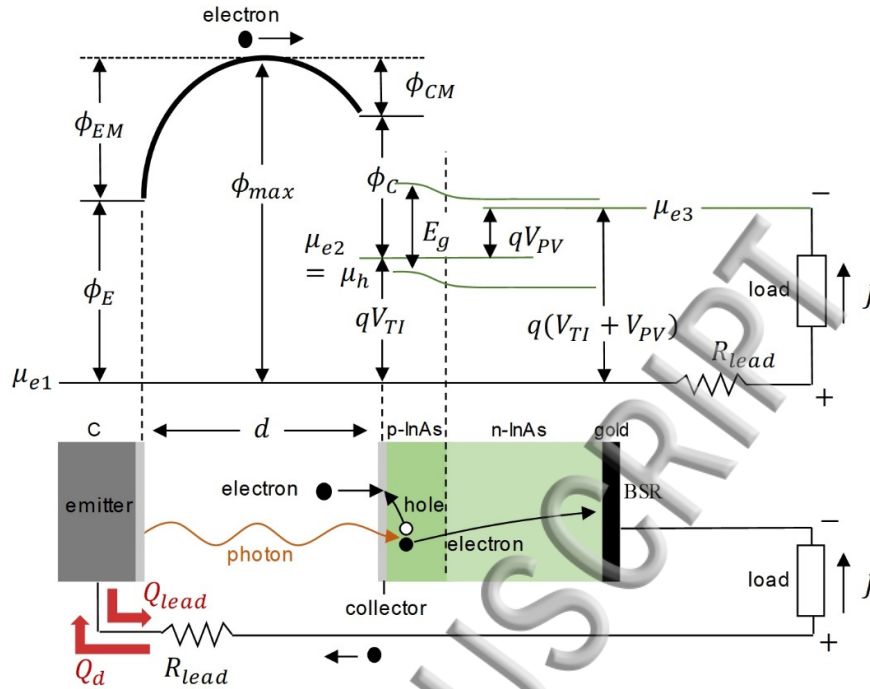
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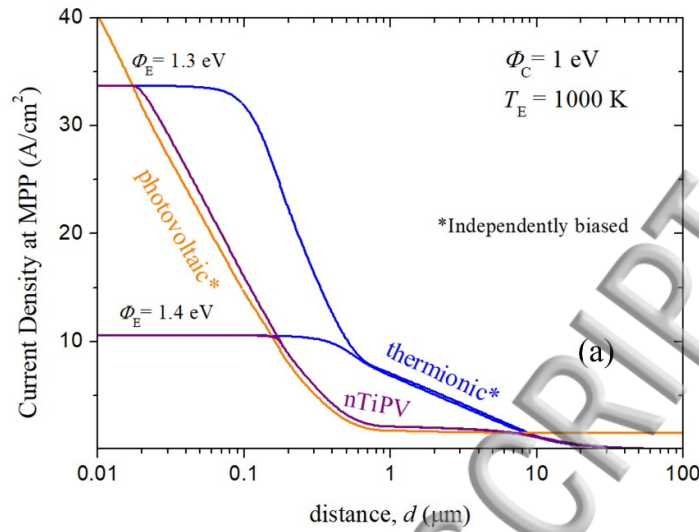
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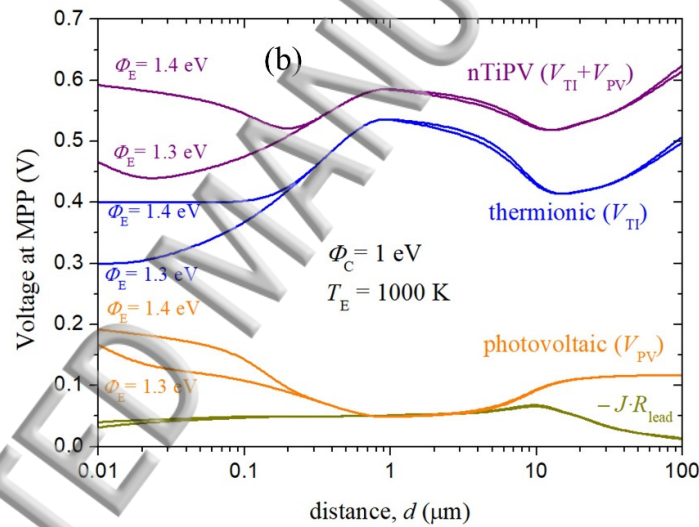
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345 *Figure 1. Band diagram of the proposed nTiPV device comprising a C thermal emitter and an*
 346 *InAs PV cell with engineered low workfunction materials and/or coatings. The thermally excited*
 347 *electrons having enough energy to overcome the emitter workfunction ϕ_E and space-charge*
 348 *barrier ϕ_{EM} are radiated towards the InAs PV cell, which is separated by distance d from the*
 349 *emitter. The electrons are collected at the PV cell surface, also named collector, which is biased*
 350 *at voltage V_{TI} . The photons are absorbed within the PV cell and generate an electron-hole pair.*
 351 *The photogenerated holes recombine with the thermionically collected electrons coming from*
 352 *the emitter. The photogenerated electrons are collected in the rear contact, which also*
 353 *comprises a gold back surface reflector (BSR). The electrochemical potential of electrons*
 354 *gradually increases from μ_{e1} (when injected in the emitter from the lead), to μ_{e2} (after being*
 355 *collected in the PV cell surface) and finally to μ_{e3} (when collected in the rear terminal of the PV*
 356 *cell).*

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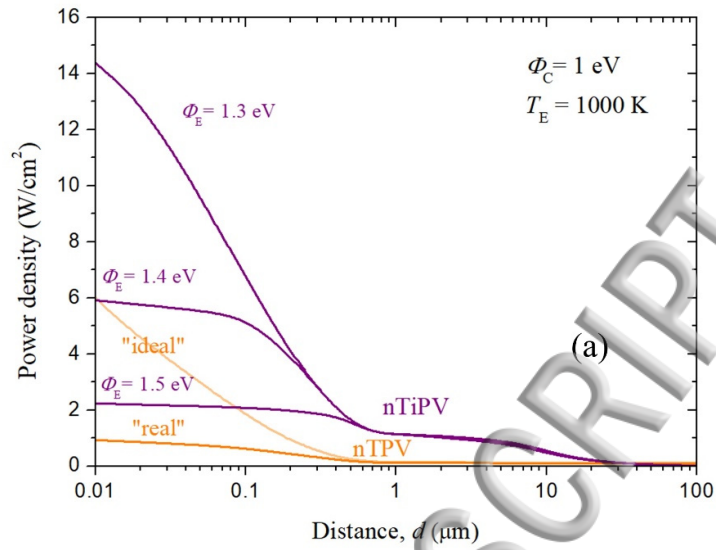
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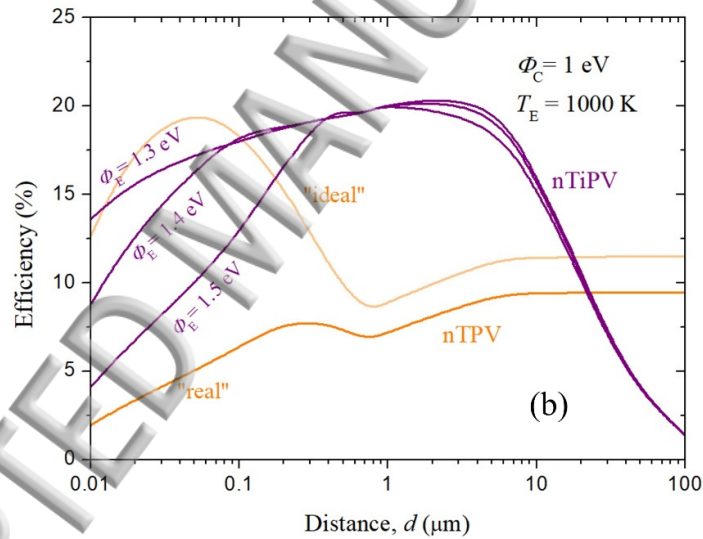
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360 *Figure 2. Current densities (a) and voltages (b) of nTiPV as a function of gap distance between*
 361 *the emitter and the PV cell. The current densities for the photovoltaic and thermionic sub-*
 362 *devices are identical in the nTiPV device, but independently-biased photovoltaic and thermionic*
 363 *current densities are shown in (a) to illustrate which of them is limiting the total device current.*
 364 *Different workfunctions of the emitter (ϕ_E) are considered. R_{lead} is optimized at every distance*
 365 *to maximize the nTiPV conversion efficiency. The device area is 1 cm^2 .*

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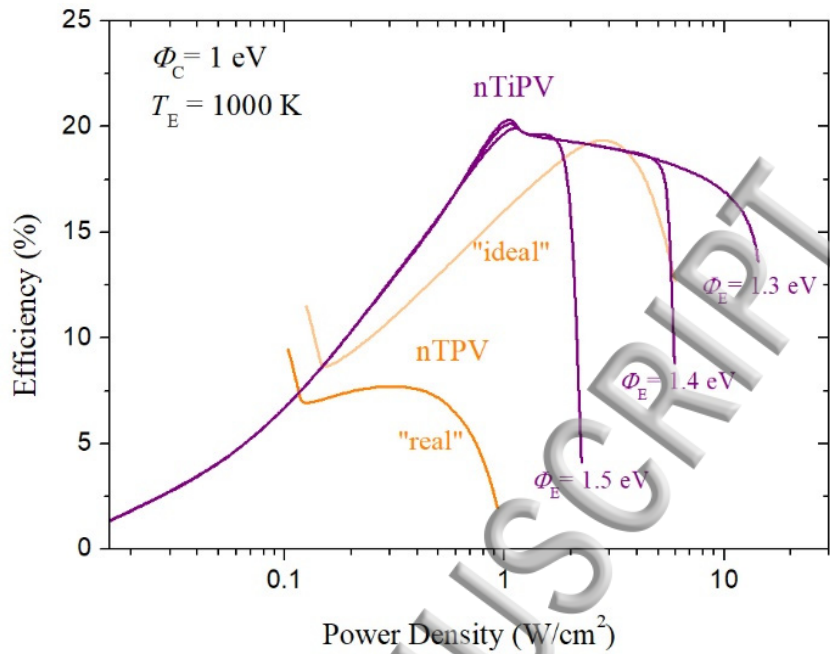


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369 Figure 3. Electrical power density (a) and conversion efficiency (b) of nTiPV and nTPV
 370 converters as a function of gap distance between the emitter and the PV cell. Different
 371 workfunctions of the emitter (ϕ_E) are considered for nTiPV. "ideal" and "real" nTPV refer to
 372 the case with negligible ohmic losses and the more realistic case with a series resistance of 10
 373 $\text{m}\Omega\cdot\text{cm}^2$, respectively. R_{load} is optimized at each distance to maximize the nTiPV conversion
 374 efficiency. The device area is 1 cm^2 .



375

376 *Figure 4. Conversion efficiency as a function of electrical power density for nTiPV and nTPV*
 377 *converters, rearranged from the results shown in Figure 3. Different workfunctions of the*
 378 *emitter (ϕ_E) are considered for nTiPV. “ideal” and “real” nTPV refer to the case with*
 379 *negligible ohmic losses and the more realistic case with a series resistance of $10 \text{ m}\Omega\cdot\text{cm}^2$,*
 380 *respectively. R_{lead} is optimized at each distance to maximize the nTiPV conversion efficiency.*
 381 *The device area is 1 cm^2 .*

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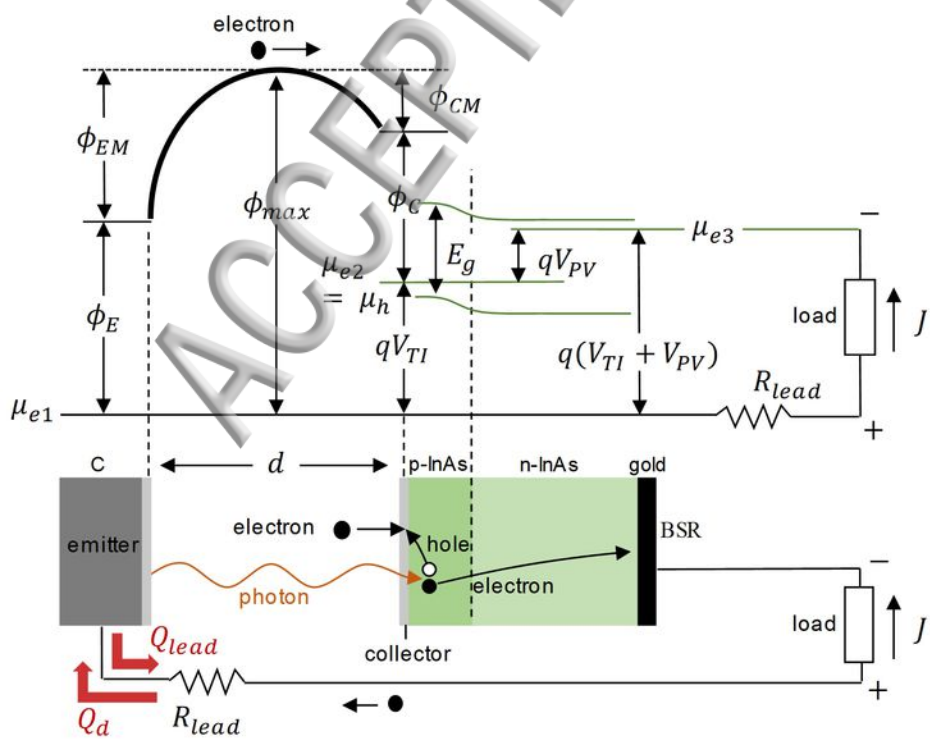
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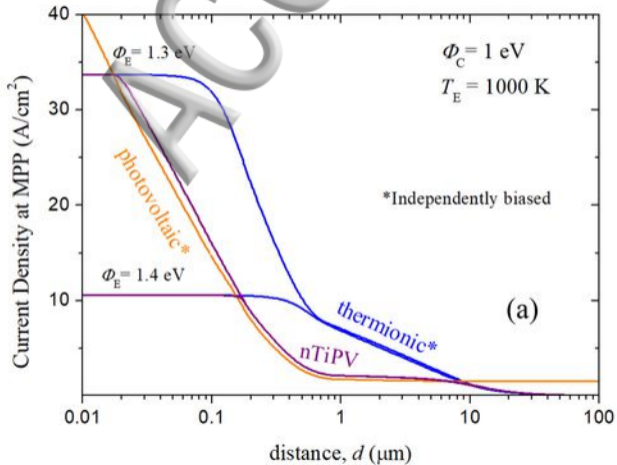
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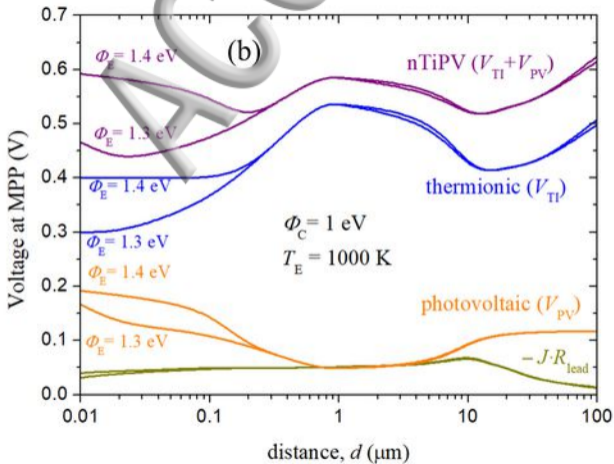
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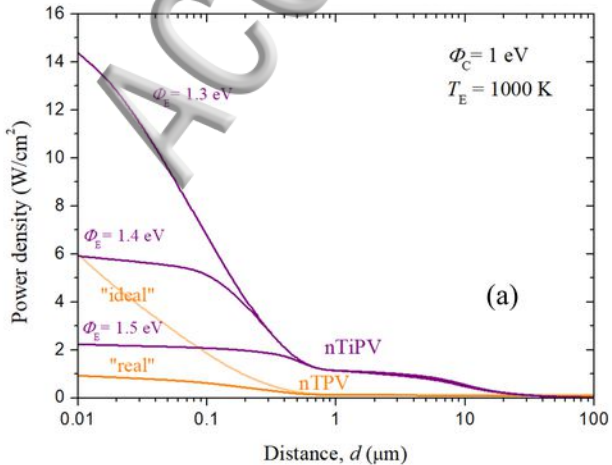
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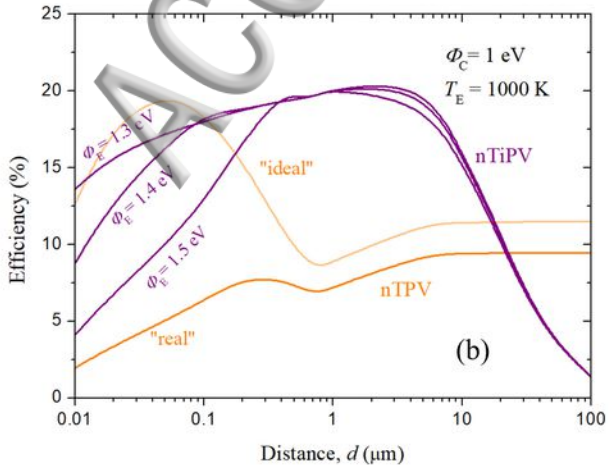
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Efficiency (%)

