

Supplemental material

Operating conditions and thermodynamic bounds of dual radiative heat engines

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I. ELECTRICAL CHARACTERISTIC AND ENERGY DIAGRAM OF INDIVIDUAL OPTOELECTRONIC COMPONENTS

We provide in Fig. S1 the current-voltage characteristic of the various optoelectronic components considered for dual radiative engines. Each component operates in a separate quadrant, two of them producing electrical power (TR and PV cells) while the two others consume power (LEDs and NEL diodes).

As an example, we also represent in Fig. S2 the energy diagrams of a TR cell and a NEL diode used in a TRNEL device, along with the various flows of energy exchanged. The negative bias applied to both components allows reducing the charge concentration in the respective bands, resulting in a decrease in emission. Because the TR cell is maintained at high temperature, it will nonetheless emit towards the cold NEL diode, generating electrical power. The low emission of the NEL diode further reduces the generation of electron-hole pairs in the TR cell that would otherwise repopulate the bands.

Additional information about the respective operating principle and energy diagram of the individual components employed in dual radiative heat engines can be found in [1].

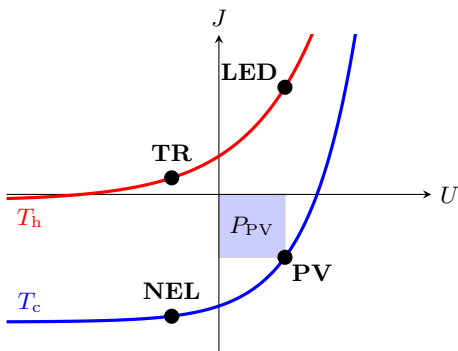


FIG. S1: Schematic of current-voltage characteristics of optoelectronic components, respectively when it is hotter (in red) or colder (in blue) than the surroundings. $U \cdot J > 0$ means that electrical power is consumed.

II. PERFORMANCE OF DUAL ENGINES FOR LOW BANDGAPS

We provide in Fig. S3 the variation of electrical power output and cooling power as a function of both chemical potentials, this time for $E_g = k_B T_h$. In comparison to the results obtained for larger bandgaps, the gap between the heat engine and heat pump operating regions is larger. The change is particularly visible in the TPX quadrant, since then $E_g - \mu_i$ becomes lower than $k_B T_i$. In this quadrant, the device is almost always capable of

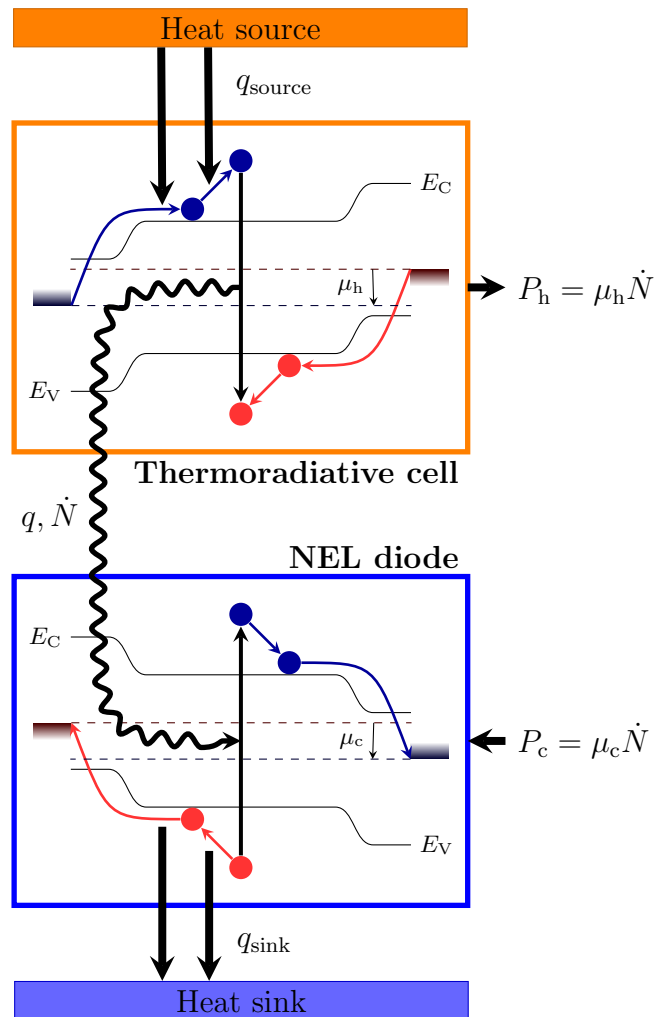


FIG. S2: Schematic of the energy flow through a thermoradiative-negative electro-luminescent (TRNEL) heat engine at the radiative limit, along with the energy diagram of the two components.

operating as a heat engine as long as $\mu_c \geq \mu_h$. Consequently, TPX devices are not able to operate as heat pumps for such low bandgaps.

III. IMPACT OF NON-RADIATIVE LOSSES ON $\eta - P$ CHARACTERISTICS

The results presented in the main article are obtained at the radiative limit, and therefore provide an upper bound for dual radiative engines' performance. Obviously, if non-radiative losses are included, performance will be worsened: we aim at giving here some first insights about the impact of such losses. To do so, we manually set the quantum efficiency (QE), defined as the fraction of recombinations being radiative [2], to 0.9 for both components of the dual engine. This means that

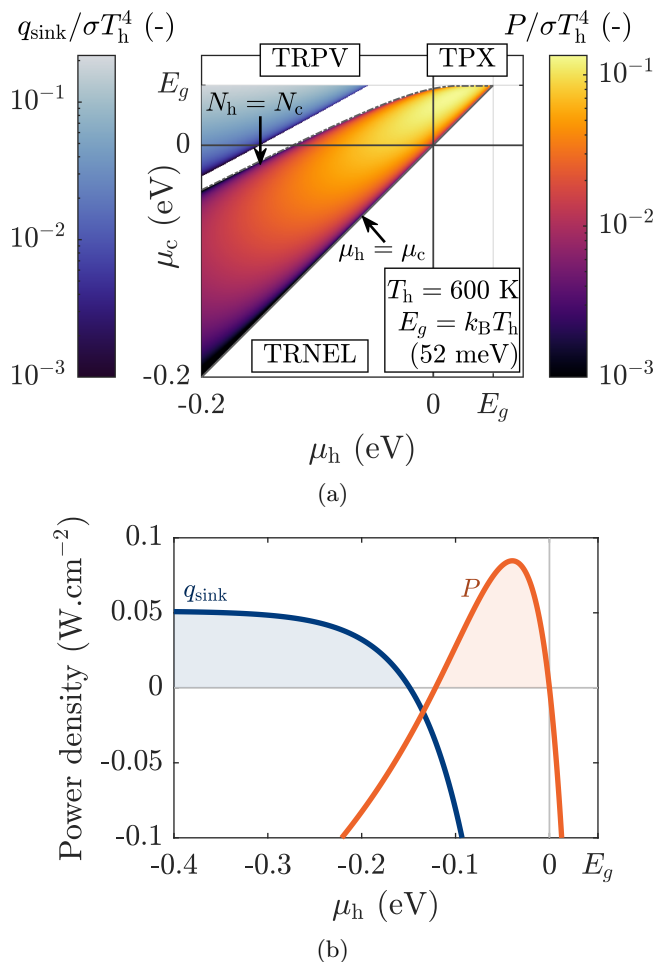


FIG. S3: (a) Performance of dual radiative systems operating as heat engines or heat pumps, at the radiative limit and for $E_g = k_B T_h = 52$ meV. (b) Zoom on the $\mu_c = 0$ scenario.

10% of the total recombination rate can be attributed to non-radiative losses. Since the non-radiative generation rate must balance the non-radiative recombination rate at equilibrium, we get a new expression of the power generated by an optoelectronic component [3]:

$$P_i = \mu_i \left(\dot{N}_j - \dot{N}_i - \frac{1 - \text{QE}}{\text{QE}} \left(\dot{N}_i - \dot{N}_i(\mu_i = 0) \right) \right). \quad (\text{S1})$$

Note that the definition of the QE used here corresponds to the one usually considered in the LED community, and should not be confused with its PV counterpart which corresponds to the conversion efficiency of photons into charges. For a 600 K heat source temperature, the addition of non-radiative events strongly modifies the performance of the engine, as shown in Fig. S4 where the variations of the electrical power output and the cooling power with the chemical potentials are depicted. The resulting $\eta - P$ plots are drawn in Fig. S5, and reveal several major

differences in comparison to the results obtained at the radiative limit. First, TRNEL devices have significantly lost interest, since they are no longer able to reach Carnot efficiency. Worse, their envelope is now within that of TRPV devices for both bandgaps considered. Therefore, TRNEL systems have interest only when operating close enough to the radiative limit. Second, the maximum efficiency is now highly dependent on the bandgap: being only 40% of Carnot efficiency for $E_g = k_B T_h$, it goes up to 80% of η_C for $E_g = 10 k_B T_h$. Last, the power output variation with the bandgap is way weaker; in fact, as long as $\text{QE} < 1$, there is an optimal bandgap that allows maximising the power output [4] (page 61).

Because the maximum power no longer diverges with E_g , we can in fact trace the $\eta - P$ envelope of dual engines obtained over the whole bandgap space, indicating the best operating conditions achievable for a certain quantum efficiency. These are illustrated in Fig. S6 for three different quantum efficiencies, along with the TPV envelope in dashed line for comparison. The optimal bandgap appears to be $E_g = 5 k_B T_h$ for $\text{QE} = 0.9$, and decreases for lower QE. In all three scenarios, the performance of dual radiative engines at high efficiency are similar to those of TPV engines. In contrast, dual engines can reach higher power outputs than TPV even when non-radiative losses are included. However, quantum efficiencies of at least 0.75 are required to obtain a noticeable difference.

On a side note, including non-radiative losses causes a mismatch between the currents J_h and J_c of the two optoelectronic components. Since voltages are mismatched too, the two components cannot be directly bound electrically if both have the same area, and additional electronics is necessary to make the engine work. Otherwise, it is possible to design engines with components with mismatched areas [5] or bandgaps [6] to make them self-sustaining.

IV. IMPACT OF HEAT SOURCE TEMPERATURE ON $\eta - P$ CHARACTERISTICS

In Fig. S7 and S8 are provided $\eta - P$ plots obtained at the radiative limit, but respectively for a 400 K and 1200 K heat source temperature. The conclusion drawn for $T_h = 600$ K remains valid in these scenarios. We can still notice two slight differences. First, the normalised power output achieved becomes larger as T_h increases. Considering $E_g = 10 k_B T_h$ for instance, $P_{\text{max}} / \sigma T_h^4$ equals approximately 0.6 for $T_h = 400$ K, but goes up to 2.4 for $T_h = 600$ K and exceeds 5 for $T_h = 1200$ K. Second, the interest of TRPV devices rises with temperature, more and more of the total envelope corresponding to that of the TRPV device if the bandgap stays moderate. For $T_h = 1200$ K and $E_g = k_B T_h \approx 0.1$ eV, TRPV gives access to interesting trade-offs between power and effi-

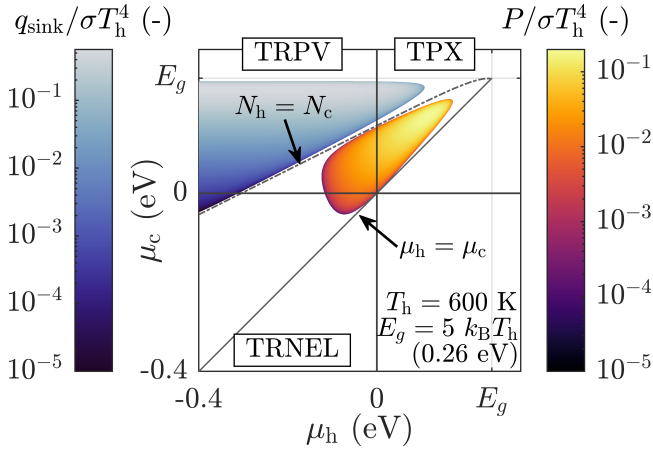


FIG. S4: Performance of dual radiative systems operating as heat engines or heat pumps, for $QE = 0.9$ and $E_g = 5k_B T_h = 0.26$ eV.

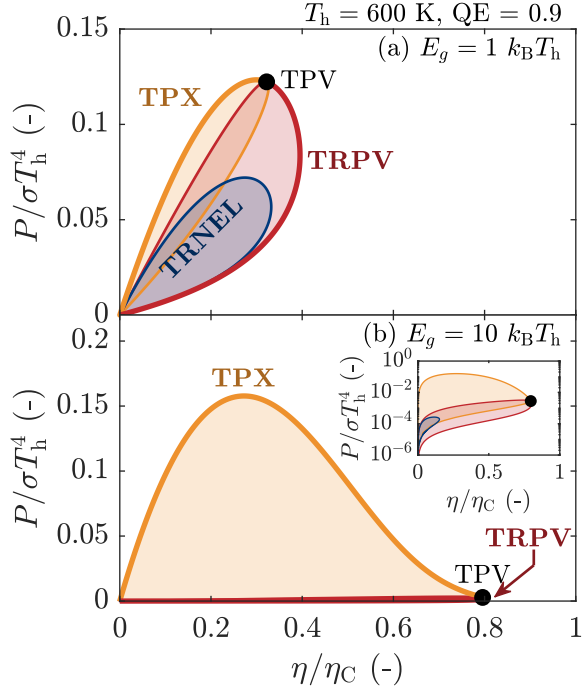


FIG. S5: $\eta - P$ plots obtained for dual radiative engines, for $T_h = 600$ K and for various bandgaps. A quantum efficiency of 0.9 is considered.

ciency. However, note that such high temperatures can hardly be withstood by optoelectronic components, and therefore limits TRPV interest.

V. VARIATION OF MAXIMUM POWER AND RELATED EFFICIENCY WITH BANDGAP

We provide in Fig. S9 the variations of the maximum power and related efficiency as a function of the

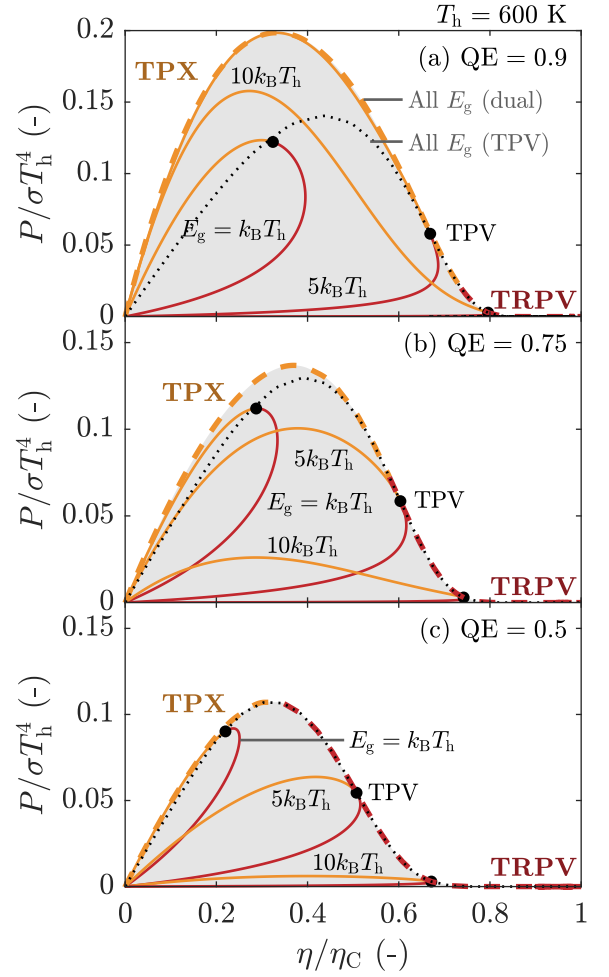


FIG. S6: Dual radiative engine $\eta - P$ envelopes obtained for varying bandgap energy E_g , depending on the quantum efficiency considered. The grey area corresponds to the operating region, while the dashed line represents the envelope obtained for TPV engines.

bandgap, respectively for the complete dual engine (thick black line) and for each individual engine. For a heat source temperature of 600 K, the dual engine MPP moves from the TRPV quadrant for low bandgaps to the TPX quadrant for higher bandgaps. In this case, the transition between the two quadrants occurs around $E_g/k_B T_h = 0.7$. Dual engines become especially attractive when $E_g > k_B T_h \approx 0.05$ eV, their power output increasing with E_g and being already 50% larger than that of single engines for $E_g = 1.9k_B T_h$. In practice, this condition is satisfied by any realistic bandgap at the temperature considered.

Note how, when TPX or TRPV engines do not maximise the power output, they operate as TPV devices. This is what causes the sudden change in slope observable in panel (b): around $E_g/k_B T_h = 0.7$, there is for both devices an abrupt change in the direction of displacement of the MPP in (μ_h, μ_c) coordinates, from that

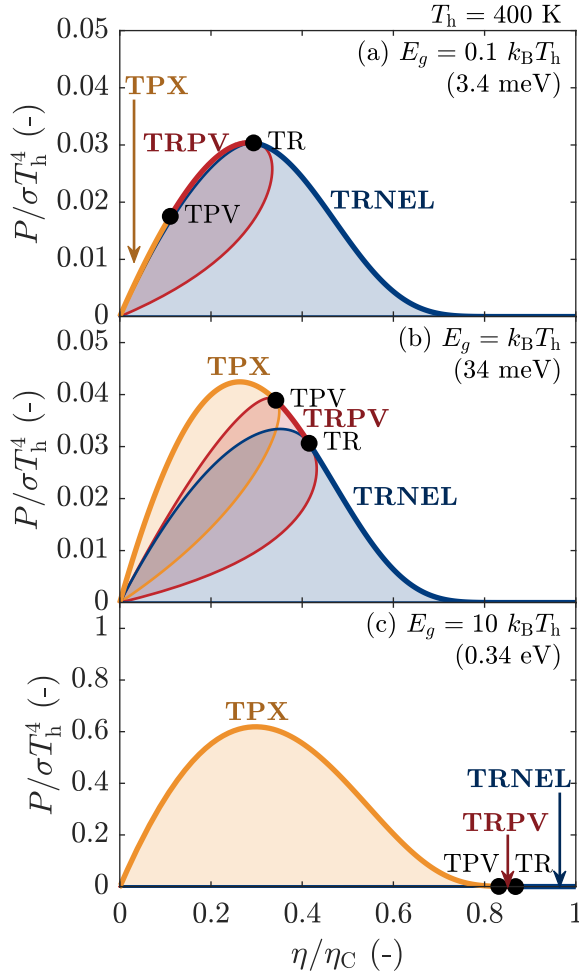


FIG. S7: $\eta - P$ plots obtained for dual radiative engines at the radiative limit, for $T_h = 400$ K and for various bandgaps.

of the complete dual engine to that of a TPV device (or vice-versa).

One can also observe that optimising TRNEL devices for electrical power production simply means operating as a TR device. Moreover, in the limit of zero bandgap, TR operation becomes optimal: this is because the TRNEL quadrant is the only one available since $\mu < E_g \rightarrow 0$.

VI. DETERMINATION OF THE MAXIMUM POWER AND RELATED EFFICIENCY FOR BROADBAND RADIATION

The goal of this section is to determine analytical expressions of the maximum power and related efficiency, in the limit of $E_g \rightarrow \infty$. Using the quantity $x_i = (E_g - \mu_i)/k_B T_i$, we obtain that $\mu_c - \mu_h = x_h k_B T_h - x_c k_B T_c$. Defining then X_i as $\exp(-x_i)$, the power output can be

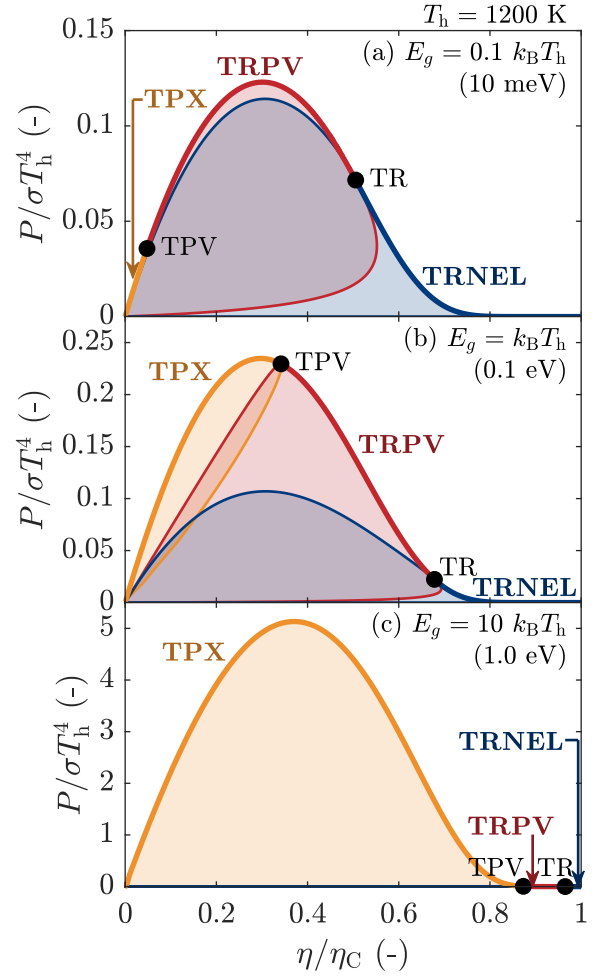


FIG. S8: $\eta - P$ plots obtained for dual radiative engines at the radiative limit, for $T_h = 1200$ K and for various bandgaps.

expressed as:

$$P = \frac{E_g^2 k_B^2 T_h^2}{4\pi^2 c^2 \hbar^3} \left(\ln(X_h) - \frac{T_c}{T_h} \ln(X_c) \right) \times \left(\ln(1 - X_h) - \frac{T_c}{T_h} \ln(1 - X_c) \right), \quad (\text{S2})$$

since $\text{Li}_1(x) = \ln(1 - x)$. First, we must verify whether the maximum power is reached inside the (X_h, X_c) domain (i.e. for $0 < X_i < 1$) or at the boundary. For instance, if X_h goes to 0, it gives

$$P \sim -\frac{E_g^2 k_B^2 T_h^2}{4\pi^2 c^2 \hbar^3} \ln(X_h) \ln(1 - X_c) \frac{T_c}{T_h} \rightarrow -\infty, \quad (\text{S3})$$

and the maximum power is therefore not reached at this boundary (the power being negative). A similar treatment can be done at the three other boundaries to ensure that the maximum power is indeed located inside the domain. We should then find the couple (X_h, X_c) which makes both partial derivatives equal to zero. If the

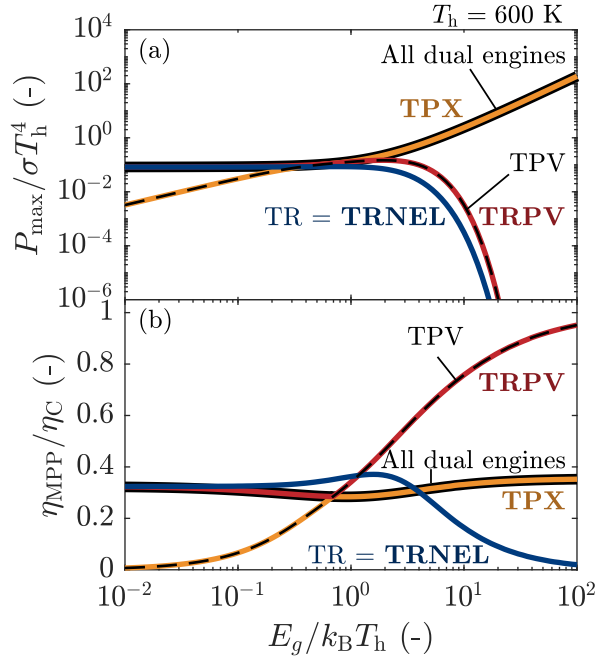


FIG. S9: Variation of (a) the maximum power, (b) the efficiency at maximum power, for the different radiative engines considering $T_h = 600$ K.

couple found is unique, it is the maximum power point since P goes to $-\infty$ at the boundaries (which means that a maximum power point must exist).

Setting both partial derivatives of P with respect to X_i to zero, we get

$$\frac{\ln(1 - X_h) - \ln(1 - X_c) \frac{T_c}{T_h}}{\ln(X_h) - \ln(X_c) \frac{T_c}{T_h}} = \frac{X_h}{1 - X_h}, \quad (\text{S4a})$$

$$= \frac{X_c}{1 - X_c}. \quad (\text{S4b})$$

Note that this holds only if $\ln(X_h) - \ln(X_c) \frac{T_c}{T_h}$ is non-zero at the maximum power point. If the former term was equal to zero, then $\ln(1 - X_h) - \ln(1 - X_c) \frac{T_c}{T_h}$ should also be zero to satisfy that partial derivatives are equal to zero, which would lead to $1 - X_c^{T_c/T_h} = (1 - X_c)^{T_c/T_h}$. This equation being satisfied only for X_c equal to 0 or 1 (which are not inside the domain), $\ln(X_h) - \ln(X_c) \frac{T_c}{T_h}$ is therefore non-zero at the maximum power point.

Combining the two expressions given in Eq. (S4), we easily get that $X_h = X_c$. Writing this common value X , we obtain

$$P = \frac{1}{\hbar} \left(\frac{E_g k_B (T_h - T_c)}{2\pi c \hbar} \right)^2 \ln(X) \ln(1 - X). \quad (\text{S5})$$

A quick study of the function $\ln(X) \ln(1 - X)$ immediately reveals that it reaches a maximum at $(X_h = 1/2, X_c = 1/2)$, which therefore corresponds to the maximum power point.

To derive a closed-form expression of the efficiency at maximum power, we use that $x_{h,\text{MPP}} = x_{c,\text{MPP}} = \ln(2)$ to obtain $(\mu_{c,\text{MPP}} - \mu_{h,\text{MPP}})/(E_g - \mu_{h,\text{MPP}}) = \eta_C$. Dividing both the numerator and the denominator of Eq. 6 by $(\dot{N}_h - \dot{N}_c)(E_g - \mu_h)$, one gets

$$\eta_{\text{MPP}} = \frac{\eta_C}{1 + \left(\frac{1}{\rho} - 1\right) \frac{1}{\ln(2)} \frac{E_g}{k_B T_h}}, \quad (\text{S6})$$

where $\rho = E_g(\dot{N}_h - \dot{N}_c)/(q_h - q_c)$ corresponds to the fraction of radiative energy being useful to optoelectronic conversion. To express it, both Li_1 and Li_2 terms are necessary. One obtains

$$\left(\frac{1}{\rho} - 1\right) \frac{1}{\ln(2)} \frac{E_g}{k_B T_h} \underset{E_g \rightarrow \infty}{\sim} (2 - \eta_C) \frac{1}{\ln(2)} \frac{\text{Li}_2(1/2)}{\text{Li}_1(1/2)}. \quad (\text{S7})$$

The polylogarithmic terms having closed-form expressions for $x = 1/2$, the efficiency at maximum power obtained as $E_g \rightarrow \infty$ is

$$\eta_{\text{MPP}}^{E_g \gg k_B T_h} = \frac{\eta_C}{1 + (2 - \eta_C)\chi}, \quad (\text{S8})$$

χ being a constant defined in the main paper.

VII. DETERMINATION OF THE EFFICIENCY AT MAXIMUM POWER FOR QUASI-MONOCROMATIC RADIATION

To derive an expression of the efficiency at maximum power in the case of quasi-monochromatic radiation, we use that $E_g - \mu_i \ll k_B T_i$. To verify that this is indeed correct, we show in Fig. S10 the variation of power output with μ_h for $E_g = 1$ eV, considering only the TPX quadrant. The power output associated to each μ_h corresponds to the maximum achievable with the whole range of μ_c available. We observe that $E_g - \mu_{h,\text{MPP}} \ll k_B T_c$: since $T_h > T_c$, therefore $E_g - \mu_{h,\text{MPP}} \ll k_B T_h$. Moreover, $\mu_c \geq \mu_h$, and thus $E_g - \mu_{c,\text{MPP}} \ll k_B T_c$.

Because $E_g - \mu_i \ll k_B T_i$, the Bose-Einstein distributions can be simplified using that $[\exp(x) - 1]^{-1} \sim x^{-1}$ around 0. Setting to zero any of the two partial derivatives of P with respect to μ_i then leads to

$$\eta_{\text{MPP}}^{\delta E \ll k_B T_c} = 1 - \sqrt{\frac{T_c}{T_h}}. \quad (\text{S9})$$

VIII. MODIFICATION OF THE ACHIEVABLE OPERATING CONDITIONS UNDER SPECTRAL FILTERING

To study the impact of spectral filtering (i.e. of reducing the radiation bandwidth δE , see Fig. S11), we show in Fig. S12 how the $\eta - P$ envelope of dual engines vary with δE . In addition, the complete set of operating conditions achievable by varying δE is represented by

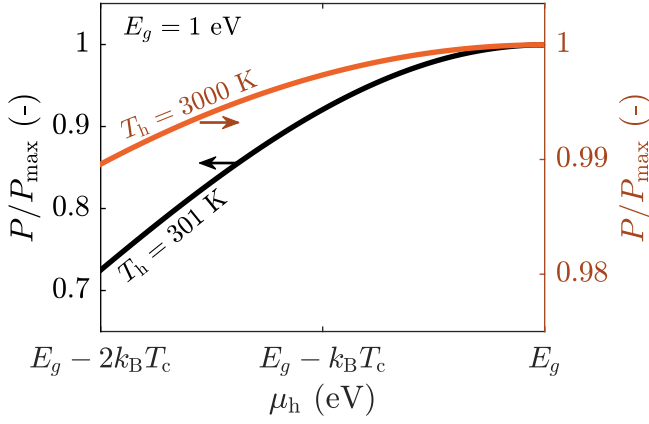


FIG. S10: Variation of the power output with the LED chemical potential considering quasi-monochromatic radiation exchanged, for $E_g = 1$ eV and for two different heat source temperatures. In this case, only the TPX quadrant is considered.

the grey area. Three different scenarios are considered to highlight the variability of the influence of spectral filtering on the achievable operating conditions.

The envelopes obtained at the radiative limit for a bandgap of $k_B T_h$ are shown in panel (a), the results being mostly similar to those obtained at $E_g = 10k_B T_h$. In this scenario, filtering gives access to new operating conditions in the high-efficiency region. For $P/\sigma T_h^4 < 0.03$, this can allow increasing the efficiency by up to 10 percent points. The effect is even stronger when decreasing the quantum efficiency (QE) to 0.9 (see Section III for more details on QE), as depicted in panel (b): an efficiency increase of the order of 25 percent points can be reached for $P/\sigma T_h^4 = 0.03$, and goes up to 45 percent points for $P/\sigma T_h^4 = 0.01$. In this case, the MPP obtained with filtered radiation can even move beyond the envelope obtained in the broadband scenario. In other words, spectral filtering can limit the power loss undergone in high-efficiency operation.

There are however cases where filtering has no significant benefit. We provide one such example in panel (c), obtained for $\text{QE} = 0.9$ and $E_g = 10k_B T_h$. Under such conditions, reducing the bandwidth mostly make the envelope shrink, the only benefit being a small increase of the efficiency achieved for powers close to zero. In such scenarios, it is thus better to prevent filtering radiation.

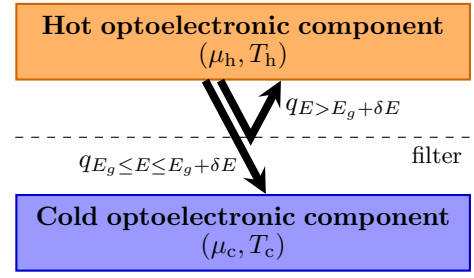


FIG. S11: Representation of spectral filtering in dual radiative engines.

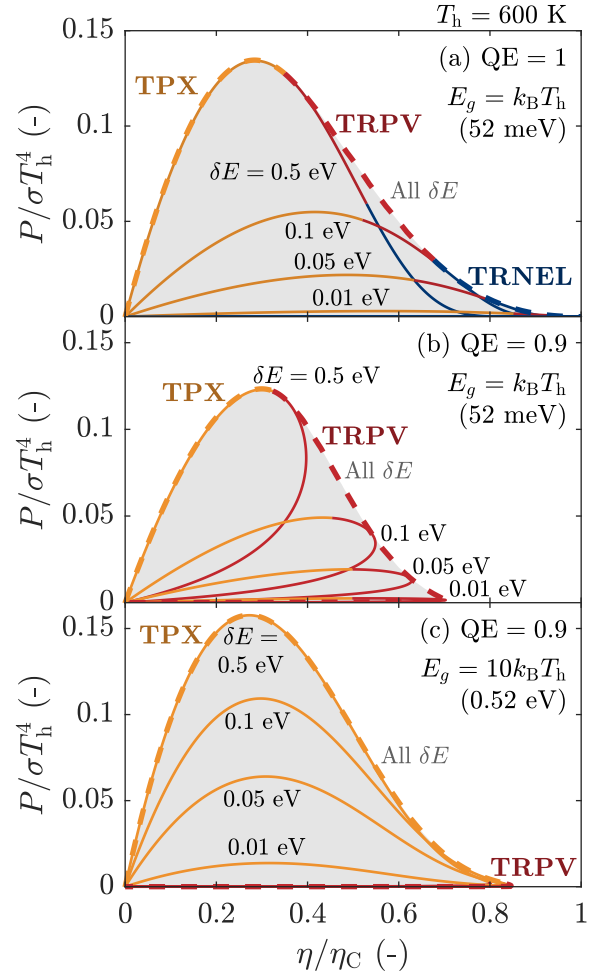


FIG. S12: Dual radiative engine $\eta - P$ envelopes obtained for varying spectral bandwidth δE .

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