A coherent description of thermal radiative devices and its application on the near-field negative electroluminescent cooling

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A coherent description of thermal radiative devices and its application on the near-field negative 2 electroluminescent cooling 3 Chungwei Lin, Bingnan Wang, Koon Hoo Teo 4 Mitsubishi Electric Research Laboratories, 201 Broadway, Cambridge, Massachusetts 5 02139. USA 6 Zhuomin Zhang 7 George W. Woodruff School of Mechanical Engineering, Georgia Institute of Technology, 8 Atlanta, Georgia 30332, USA

10 Abstract

Using the transimissivity between two thermal reservoirs and the generalized Planck distributions, we describe the devices that use radiative energy transfer between thermal reservoirs in a unified formalism. Four types of devices are distinguished. For power generators that use the temperature difference between reservoirs, photovoltaic (PV) and thermoradiative (TR) devices respectively use the low-temperature photovoltaic cell and high-temperature thermoradiative cell to generate electricity. For active cooling, the electroluminescent (EL) cooling devices apply a forward bias voltage on the object we want to cool, whereas the negative EL cooling devices apply a reverse bias voltage to the heat sink. The relationship among these four devices is explicated. The performance of the negative EL cooling is analyzed, both in the Shockley-Queisser (blackbody spectrum and radiative recombination) framework and the near-field enhancement. The "impedance match" condition derived for PV systems is applied to the negative EL devices. One advantageous feature of the negative EL cooling is that it does not apply the voltage to the target object which we want to cool, and the near-field enhancement can apply to various target materials that support the surface resonant modes.

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20 1. Introduction

Usable work can be extracted from two reservoirs maintained at different 21 temperatures. Photovoltaic (PV) [1, 2, 3] and Thermoradiative (TR) [4, 5, 22 6, 7] devices are two power generators that use photons emitted at different 23 temperatures to generate electricity. PV devices use the low-temperature 24 (low-T) PV cell to generate charge current, whereas TR devices use the high-25 temperature (high-T) TR cell for power generation. As the energy transfer 26 is mediated by photons, these devices contains no moving parts, allowing the 27 possible stable and long-lived power generators. By reversing the light-to-28 electricity processes, work can be done to maintain the temperature difference 29 or to cool one of two reservoirs. The electroluminescent (EL) cooling devices 30 apply a forward bias voltage to the target object which we want to cool 31 [8, 9, 10], whereas the negative EL cooling devices [11, 12, 13, 14] apply a 32 reverse bias voltage to the heat sink that increases the thermal removal flux 33 from the target object. Recently proposed "thermophotonic heat pump" [15] 34 can be viewed as a combination of EL and negative EL cooling. As these 35 four devices share the same microscopic physics, any strategy that boosts 36 the performance of one of the devices, say the PV power generator, can be 37

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³⁸ used to enhance the performance of the other three types of devices.

For a general PV power generator [1], the incoming photons of high and 39 low energies are both wasted – photons of energies lower than the bandgap 40 of PV cell (E_q) cannot generate any electrons and holes, whereas photons of 41 energies higher than E_q can produce a voltage no larger than $E_q/|e|$ (e be-42 ing the electron charge). Therefore the ideal photon emission spectrum is a 43 δ -function peaked slightly above the PV bandgap, with the peak amplitude 44 as strong as possible [3]. In the far-field based devices, the radiative en-45 ergy transfer is limited by the blackbody spectrum. However, the δ -function 46 spectrum with a strong peak amplitude can be approximately achieved in the 47 near-field Thermophotovoltaic (TPV) [16, 17, 18, 19]. A basic TPV system 48 consists of an emitter and a PV cell [2, 20, 21, 22, 23, 24], with the emitter 40 placed between the heat source and the PV cell. The main role of the emitter 50 is to modify the photon emission spectrum that better fits the bandgap of the 51 PV cell. For the near-field based TPV system [25, 26, 27, 28, 29, 30, 31, 32], 52 the separation between the emitter and the PV cell is much shorter than 53 the characteristic wavelength of the emitted photons, and the resulting pho-54 ton emission spectrum approaches a δ -function of large amplitude. The 55 strong enhancement stems from the surface resonances supported by the 56 emitter/vacuum interface (surface plasmon polaritons) [16, 33]. Recently, us-57 ing the framework of Coupled Mode Theory (CMT) [33, 34, 35, 36, 37, 38], 58 Karalis and Joannopoulos show that the performance of the TPV system 59 can be strongly enhanced [32] when the emitter and PV cell are designed, as 60 a whole, to satisfy the "impedance matching" condition derived from CMT 61 [33]. The impedance matching condition for TPV can be stated as follows: if 62 both PV/vacuum and emitter/vacuum by themselves support their respec-63 tive surface resonances, the radiative energy transfer is maximized when the 64 (complex) resonant energies are identical; if there is only one resonant mode, 65 the radiative energy transfer is maximized when the resonant mode decays 66 to the PV cell and to the emitter at the same rate [33, 37, 39]. 67

In our previous work [40], we describe both PV and TR power generators in a unified formalism that involves the transmissivity between two reservoirs and the generalized Planck distributions [41, 42, 43]. We also showed how near-field TPV concept can enhance the TR performance. In

the present work, we further generalize the formalism to all four devices men-72 tioned above, and therefore the "impedance matching" condition derived for 73 TPV can be easily applied to all types of devices. In particular, we focus on 74 the negative EL cooling whose near-field enhancement has not extensively 75 studied in literatures [14]. Only the planar structure is considered, and the 76 transmissivity is computed using the dyadic Green function [44, 45, 46] (see 77 also Appendix) and the fluctuation-dissipation relation between the thermal 78 current and temperature [47]. The rest of the paper is organized as follows. 79 In section 2, we provide a general and unified formalism for thermal radia-80 tive devices including PV and TR power generators, as well as the EL and 81 negative EL active cooling devices. We show that the performance of these 82 devices, including the output power and the efficiency, can be expressed in 83 terms of transmissivity and the generalized Planck distribution. A coherent 84 description of these four devices are provided; the reverse saturation current 85 is found to be a good indicator of the near-field effect for all devices. In 86 section 3 we give a few model examples on the negative EL cooling devices, 87 emphasizing the near-field enhancement. The connection between all near-88 field devices are explicated. Some features and advantages specific to the 89 negative EL cooling are pointed out. Finally a brief conclusion is given in 90 Section 4. 91

92 2. General Formalism

93 2.1. Overview

Four types of devices using radiation energy transfer are illustrated in 94 Fig. 1 – they are PV power generators, TR power generators, EL cooling de-95 vices, and negative EL cooling devices. All these devices involve at least two 96 different thermal reservoirs, with each reservoir characterized by a tempera-97 ture (T) and a chemical potential (μ , or equivalently a bias voltage $|e|V = \mu$). 98 Before providing the formalism that describes all these devices, in Section 99 2.2 we briefly review the basic description of non-equilibrium electron-hole 100 (e-h) concentrations, the generalized Planck distribution that introduces a 101 non-zero photon chemical potential to describe the e-h generation and re-102 combination, and fix the sign convention of charge current and bias voltage 103 used in this paper. Section 2.3 provides the general formalism for all four 104

¹⁰⁵ devices. Section 2.4 is devoted to the cooling performance, including the
¹⁰⁶ "thermophotonic heat pump" [15]. Section 2.5 gives a coherent description
¹⁰⁷ of all four devices.

2.2. Quasi-Fermi energies, generalized Planck distribution and sign conven tion

To describe the electron and hole concentrations away from their equilib-110 rium values, two (quasi) Fermi energies, one for electrons denoted as E_{FC} and 111 one for holes denoted as E_{FV} , are needed [3]. The difference $E_{FC} - E_{FV} = \mu$ 112 defines the photon chemical potential. At equilibrium, two Fermi energies are 113 identical, i.e. $E_{FC} = E_{FV} = E_F$. When the e-h concentration is larger than 114 that at equilibrium (under an illumination or a forward bias), the electron 115 Fermi energy increases $(E_{FC} > E_F)$ whereas the hole Fermi energy decreases 116 $(E_{FV} < E_F)$, and a positive μ is developed to account for the additional e-h 117 concentration. When the e-h concentration is smaller than that at equilib-118 rium (under a reverse bias), the electron Fermi energy decreases $E_{FC} < E_F$ 119 whereas the hole Fermi energy increases $E_{FV} > E_F$, and a negative μ is 120 developed to account for the reduced e-h concentration. Fig. 2 illustrates 121 the E_{FC} , E_{FV} , and μ under different conditions. We note that, reversely, 122 a photon chemical potential can be *defined* for the steady-state populations 123 that are different from those at thermal equilibrium. This allows analyz-124 ing the performance of the molecular light-to-current conversion in the same 125 framework of PV devices [48, 49]. 126

¹²⁷ The photon chemical potential is used in the generalized Planck distri-¹²⁸ bution [41, 42, 43] – for a thermal reservoir at a fixed temperature T and ¹²⁹ a photon chemical potential μ , the mean photon occupation number of the ¹³⁰ angular frequency ω is

$$\Theta(\omega; T, \mu) = \frac{1}{\exp\left[(\hbar\omega - \mu)/T\right] - 1},\tag{1}$$

with T the temperature measured in energy, i.e. the Boltzmann constant $k_B \equiv 1$. The photon chemical potential is used to describe $e+h \leftrightarrow \gamma$ processes in the cell (γ labels the photons). In this convention, a positive μ corresponds a larger e-h concentration, implying a larger e-h recombination rate and therefore a larger emitted photon number; a negative μ corresponds a smaller e-h concentration, implying a smaller e-h recombination rate and therefore a
smaller emitted photon number. From a fundamental point of view, the use of
quasi-Fermi energies assumes the electron and hole energy distributions still
possess the "Fermi-Dirac" form in the non-equilibrium steady state, based on
which the generalized Planck distribution for photons can be derived using
the principle of detailed balance [3].

The sign convention is defined with respect to a pn-junction [Fig. 3(a)]. Under a forward bias, the current flows from p-side to n-side, which defines the positive voltage and the positive current. The photo-generated current (photocurrent) is negative in this convention. For a pn-junction, the short-circuit current j_{sc} and the reverse saturation current j_S determine the characteristic of the current-voltage behavior

$$j = j_S \left[e^{|e|V/T} - 1 \right] + j_{sc}.$$
 (2)

 $j_{sc} = -|j_{sc}|$ is the photo-generated current, which is negative (see Eq. 6 and Eq. 21 for a derivation). A typical behavior of Eq. 2 is given in Fig. 3(b). Without providing the details, we note $|e|V = \mu$, and the photon chemical potential and the bias voltage can be used interchangeably [3].

Let us discuss the PV devices using Eq. 2. On the one hand, the illu-152 mination provides a short-circuit current j_{sc} that is along the reverse bias 153 direction. On the other hand, illumination increases the e-h concentration 154 that develops a forward bias voltage. This voltage-current relation means 155 that the photon current in the pn-junction, as a PV cell, is providing a "neg-156 ative" power [the red curve of Fig. 3 (b)]. In other words, the photocurrent 157 inside the PV cell flows against the voltage which the incident photons gen-158 erate – the holes flow to the +|V| side and electrons flow to the -|V| side. 159 Once an external load is attached, electrons and holes recombine through the 160 load to produce the electric power. 161

In the ideal PV cell where only the radiative recombination is considered, the photocurrent (I_c) is equal to the flux difference between the photon absorption (which generates current) and photon emission (which annihilates current). The power is given by

$$P_{cell} = (-I_c) \times V = \frac{-I_c}{|e|} \times \mu, \tag{3}$$

The minus sign comes from that the photocurrent is by definition negative. If P_{cell} is positive, the cell consumes energy; if P_{cell} is negative, the cell generates energy [Fig. 3 (b)].

169 2.3. The formalism

Now the radiative energy transfer between two reservoirs is considered. A reservoir is characterized by a temperature T and an voltage $|e|V = \mu$. Considering two reservoirs, labeled as 1 and 2, fixed respectively at (T_1, μ_1) and (T_2, μ_2) , the photon number flux and energy flux from 1 to 2 are given by

$$\dot{N}_{1\to 2} = \int_0^\infty \frac{d\omega}{2\pi} \varepsilon_{12}(\omega) \left[\Theta(\omega; T_1, \mu_1) - \Theta(\omega; T_2, \mu_2)\right], \tag{4}$$

$$P_{1\to 2} = \int_0^\infty \frac{d\omega}{2\pi} \hbar \omega \varepsilon_{12}(\omega) \left[\Theta(\omega; T_1, \mu_1) - \Theta(\omega; T_2, \mu_2)\right].$$
(5)

Here the transmissivity between the reservoirs 1 and 2 is given as $\varepsilon_{12}(\omega)$ 175 [33]. Eq. 4 and 5 imply that a non-zero chemical potential difference can 176 be generated via a temperature difference or vise versa. The transmissivity 177 $\varepsilon_{12}(\omega)$ is a dimensionless quantity for general geometries. In the planar con-178 figurations considered here, however, it is more convenient to compute the 179 transmissivity per unit area. In this case, Eq. 4 and 5 provide the photon 180 number flux density and energy flux density (i.e. flux per unit area). Since 181 systems composed of two reservoirs will be considered, the subscripts will be 182 neglected, i.e., $\varepsilon_{12}(\omega) \equiv \varepsilon(\omega)$, for the rest of the paper. 183

Four types of devices are now distinguished. The PV devices use the low-T reservoir (PV cell) to generate the power. The photocurrent and its generated power at PV cell is

$$I_{c} = |e|\dot{N}_{T_{h}\to T_{l}} = |e| \int_{0}^{\infty} \frac{d\omega}{2\pi} \varepsilon(\omega) \left[\Theta(\omega; T_{h}, 0) - \Theta(\omega; T_{l}, \mu)\right], \quad (6)$$
$$P_{cell} = -\mu \dot{N}_{T_{h}\to T_{l}}$$

Here T_h/T_l is the temperature of the high-T/low-T reservoir. When $\mu > 0$ and $\Theta(\omega; T_h, 0) - \Theta(\omega; T_l, \mu) > 0$ (so that $I_c > 0$), the power is "negative", meaning it generates power. Using Eq. 5, the power absorbed by the PV cell
is

$$P_{T_h \to T_l} = \int_0^\infty \frac{d\omega}{2\pi} \,\hbar\omega\varepsilon(\omega) \left[\Theta(\omega; T_h, 0) - \Theta(\omega; T_l, \mu)\right],\tag{7}$$

which is positive, meaning the PV cell gets energy from the other reservoir.
The TR devices use the high-T reservoir (TR cell) to generate the power.
The photocurrent and its generated power at TR cell is

$$I_{c} = |e|\dot{N}_{T_{l}\to T_{h}} = |e| \int_{0}^{\infty} \frac{d\omega}{2\pi} \varepsilon(\omega) \left[\Theta(\omega; T_{l}, 0) - \Theta(\omega; T_{h}, \mu)\right],$$

$$P_{cell} = -\mu \dot{N}_{T_{l}\to T_{h}}$$
(8)

When $\mu < 0$ and $\Theta(\omega; T_l, 0) - \Theta(\omega; T_h, \mu) > 0$ (so that $I_c < 0$), the power is "negative", meaning it generates power. Using Eq. 5, the power absorbed by the high-T TR cell is

$$P_{T_l \to T_h} = \int_0^\infty \frac{d\omega}{2\pi} \hbar \omega \varepsilon(\omega) \left[\Theta(\omega; T_l, 0) - \Theta(\omega; T_h, \mu)\right], \tag{9}$$

which is negative, meaning the TR cell losses energy to the other reservoir. For cooling devices, we assume reservoirs 1 and 2 are at temperatures T_1 and T_2 respectively, and reservoir 1 at T_1 is the target cell which we want to cool; reservoir 2 serves as the heat sink. The EL cooling devices apply a forward bias voltage to the target cell. The photocurrent and its generated power at the target cell is

$$I_{c} = |e|\dot{N}_{T_{2} \to T_{1}} = |e| \int_{0}^{\infty} \frac{d\omega}{2\pi} \varepsilon(\omega) \left[\Theta(\omega; T_{2}, 0) - \Theta(\omega; T_{1}, \mu)\right],$$

$$P_{cell} = -\mu \dot{N}_{T_{2} \to T_{1}}$$
(10)

When $\mu > 0$ and $\Theta(\omega; T_2, 0) - \Theta(\omega; T_1, \mu) < 0$ (so that $I_c < 0$), the power is "positive", meaning the target cell consumes the power. Using Eq. 5, the power absorbed by the target cell is

$$P_{T_2 \to T_1} = \int_0^\infty \frac{d\omega}{2\pi} \hbar \omega \varepsilon(\omega) \left[\Theta(\omega; T_2, 0) - \Theta(\omega; T_1, \mu)\right], \tag{11}$$

²⁰⁶ which is negative, meaning the target cell dissipates its heat to heat sink.

The negative EL cooling devices apply a reverse bias voltage to the heat sink. The photocurrent and its generated power at the heat sink is

$$I_c = |e|\dot{N}_{T_1 \to T_2} = |e| \int_0^\infty \frac{d\omega}{2\pi} \varepsilon(\omega) \left[\Theta(\omega; T_1, 0) - \Theta(\omega; T_2, \mu)\right],$$

$$P_{cell} = -\mu \dot{N}_{T_1 \to T_2}$$
(12)

When $\mu < 0$ and $\Theta(\omega; T_1, 0) - \Theta(\omega; T_2, \mu) > 0$ (so that $I_c > 0$), the power is "positive", meaning the heat sink consumes the power. Using Eq. 5, the power absorbed by the reservoir 2 is

$$P_{T_1 \to T_2} = \int_0^\infty \frac{d\omega}{2\pi} \hbar \omega \varepsilon(\omega) \left[\Theta(\omega; T_1, 0) - \Theta(\omega; T_2, \mu)\right], \tag{13}$$

which is positive, meaning the heat sink gets thermal radiation energy from the target cell.

214 2.4. Cooling performance and maximum heat removal flux

For a cooling device, the coefficient of performance or COP is defined by

$$\eta_{COP} = \frac{Q_c}{W} \tag{14}$$

with W being the work done to the device, and Q_c the heat removed from the reservoir of interest. Larger η_{COP} implies a larger heat removal flux for the same input work, or the same heat removal flux at a smaller input work. For EL cooling devices, using Eq. 10 and 11, one gets

$$\eta_{COP,EL} = \frac{-\int_0^\infty \frac{d\omega}{2\pi} \hbar \omega \varepsilon(\omega) \left[\Theta(\omega; T_2, 0) - \Theta(\omega; T_1, \mu)\right]}{-\mu \int_0^\infty \frac{d\omega}{2\pi} \varepsilon(\omega) \left[\Theta(\omega; T_2, 0) - \Theta(\omega; T_1, \mu)\right]}$$
(15)

²²⁰ μ being positive for the EL cooling ensures $\eta_{COP,EL} > 0$. For negative EL ²²¹ cooling devices, using Eq. 12 and 13, one gets

$$\eta_{COP,NEL} = \frac{\int_0^\infty \frac{d\omega}{2\pi} \hbar\omega\varepsilon(\omega) \left[\Theta(\omega;T_1,0) - \Theta(\omega;T_2,\mu)\right]}{-\mu \int_0^\infty \frac{d\omega}{2\pi}\varepsilon(\omega) \left[\Theta(\omega;T_1,0) - \Theta(\omega;T_2,\mu)\right]}$$
(16)

²²² μ being negative for the negative EL cooling ensures $\eta_{COP,NEL} > 0$.

In addition to COP, the net radiation flux *leaving* the cell (at T_1), given by the negative of Eq. 11 for the EL cooling and Eq. 13 for the negative EL cooling, is another important quantity of interest. For the negative EL cooling, there exist a maximum current and a maximum radiation flux

$$I_{c,max} = e \int_0^\infty \frac{d\omega}{2\pi} \varepsilon(\omega) \Theta(\omega; T_1, 0)$$
(17)

$$P_{TR,max} = \int_0^\infty \frac{d\omega}{2\pi} \hbar \omega \varepsilon(\omega) \Theta(\omega; T_1, 0)$$
(18)

because $\Theta(\omega; T_2, \mu) \to 0$ as $\mu \to -\infty$. To have a larger cooling power, Eq. 18 227 should be as large as possible, and increasing the transmissivity $\varepsilon(\omega)$ is the 228 key to enhance the cooling performance. The dimension of Eq. 13 is power 229 per unit area, and can be used as the boundary condition when we use the 230 Fourier law (thermal conductivity) to compute the temperature distribution 231 within the cell. We emphasize that the current description only concerns 232 the radiative processes, and thus represents the ideal condition. In the next 233 subsection we shall briefly describe how to take the non-radiative processes 234 into account. 235

Within our framework, the "thermophotonic heat pump" (THP) proposed by Oksanen and Tulkki [15] can be formulated as a combination of EL and negative EL cooling. It requires two semiconductors – one serves as the object one wants to cool with a forward bias $\mu_1 > 0$; the other as the heat sink with a reverse bias $\mu_2 < 0$. Assuming their respective temperatures are T_1 and T_2 , the COP is

$$\eta_{COP,THP} = \frac{\int_0^\infty \frac{d\omega}{2\pi} \hbar \omega \varepsilon(\omega) \left[\Theta(\omega; T_1, \mu_1) - \Theta(\omega; T_2, \mu_2)\right]}{(\mu_1 - \mu_2) \int_0^\infty \frac{d\omega}{2\pi} \varepsilon(\omega) \left[\Theta(\omega; T_1, \mu_1) - \Theta(\omega; T_2, \mu_2)\right]},$$
(19)

and the maximum heat removal flux is $\int_0^\infty \frac{d\omega}{2\pi} \hbar \omega \varepsilon(\omega) \Theta(\omega; T_1, \mu_1)$. Note that THP devices also work when $\mu_2 > 0$ (but $\mu_1 > \mu_2$ to ensure positive work), and in this sense THP is more general than the combination of EL and negative EL cooling.

246 2.5. A coherent description of all four types of devices

We conclude this section by discussing the performance using the voltagecurrent relations of all four devices. When the Planck distributions are approximated by the corresponding Boltzmann distributions, $-I_c$ in Eq. 6 and ²⁵⁰ Eq. 8 respectively reduce to

$$-I_{c} \equiv j_{PV}(V) = |e| \int_{0}^{\infty} \frac{d\omega}{2\pi} \varepsilon(\omega) e^{-\hbar\omega/T_{l}} (e^{|e|V/T_{l}} - 1) + |e| \int_{0}^{\infty} \frac{d\omega}{2\pi} \varepsilon(\omega) [e^{-\hbar\omega/T_{l}} - e^{-\hbar\omega/T_{h}}], \qquad (20)$$

$$-I_{c} \equiv j_{TR}(V) = |e| \int_{0}^{\infty} \frac{d\omega}{2\pi} \varepsilon(\omega) e^{-\hbar\omega/T_{h}} (e^{|e|V/T_{h}} - 1) - |e| \int_{0}^{\infty} \frac{d\omega}{2\pi} \varepsilon(\omega) [e^{-\hbar\omega/T_{l}} - e^{-\hbar\omega/T_{h}}].$$
(21)

²⁵¹ Eq. 2 is thus obtained by identifying

$$j_S = |e| \int_0^\infty \frac{d\omega}{2\pi} \varepsilon(\omega) e^{-\hbar\omega/T_l} > 0, \qquad (22)$$

$$j_{sc} = |e| \int_0^\infty \frac{d\omega}{2\pi} \varepsilon(\omega) [e^{-\hbar\omega/T_l} - e^{-\hbar\omega/T_h}] < 0.$$
(23)

Both $j_{PV}(-\infty)$ and $j_{TR}(-\infty)$ are negative. When using the same T_h and T_l , 252 $j_{PV}(0) = -j_{TR}(0) = -j_{sc} < 0$. The voltage-current (V-I) relations for PV 253 and TR devices are respectively illustrated in red and in blue in Fig. 3 (b). 254 When $T_h = T_l$, $j_{sc} = 0$ and $j_{PV}(V) = j_{TR}(V) = j_S(e^{|e|V/T_l} - 1)$, recovering 255 the voltage-current relation of a pn-junction in the dark [50]. Note that 256 the reverse saturation current j_s depends on $\varepsilon(\omega)$ and thus can be used 257 as a quantity to characterize the near-field effect without maintaining the 258 temperature difference and a vacuum gap between two reservoirs. 259

The PV power generators work in the $0 < V < V_{oc}^{PV}$ range of $j_{PV}(V)$ 260 [red curve in Fig. 3 (b)], whereas the TR power generators work in the 261 $V_{oc}^{TR} < V < 0$ range of $j_{TR}(V)$ [blue curve in Fig. 3 (b)]. The output power 262 of the PV and TR power generators are shown as the blue shaded areas in 263 Fig. 3 (b). Certainly one wants the output power as large as possible. The 264 EL cooling devices work in the $V > V_{oc}^{PV}$ range of $j_{PV}(V)$ [red curve in Fig. 3 265 (b)], whereas the negative EL cooling devices work in the $V < V_{oc}^{TR}$ range 266 of $j_{TR}(V)$ [blue curve in Fig. 3 (b)]. The work done to the EL and negative 267 EL devices are shown as the areas of dashed boxes in Fig. 3 (b). To have a 268 larger COP of cooling devices, for a given heat removal flux, the work done 269 to the cooling devices should be as small as possible. 270

It is worth emphasizing that both V-I curves in Fig. 3 (b) can be obtained 271 from measuring/computing the same physical device, which implies that the 272 same physical device can be used for different purposes, depending on the 273 applied voltage and temperature. Inclusions of non-radiative processes mod-274 ify the V-I relations, and its implications on PV and TR devices will be 275 presented elsewhere. Table 1 summarizes the function and the working pa-276 rameters of all four devices. Four types of devices work at four different 277 quadrants defined by the $(\mu, -I_c)$ plane. PV power generators and EL cool-278 ing devices work at $\mu > 0$, whereas TR power generators and negative EL 270 cooling devices work at $\mu < 0$. When only the radiative recombination is 280 considered, $\mu > 0$ devices generally have larger output powers (larger out-281 put power for power generators and larger heat removal flux for the cooling 282 devices) than $\mu < 0$ devices. 283

3. Near-field enhancement on the negative electroluminescent cool ing

286 3.1. Overview and material parametrization

In this section we apply the formalism to the negative EL cooling devices, 287 emphasizing the near-field enhancement. The same analysis on other three 288 devices are given in our previous work [39, 40]. The basic components of 289 a negative EL cooling device is illustrated in Fig. 4(a). The heat sink is 290 a semiconductor, characterized by a bandgap E_q . Applying a reverse bias 291 to the heat sink reduces the photon emission of the heat sink, effectively 292 enhancing the heat removal flux of the object one wants to cool. We choose 293 the heat sink to be a semiconductor of $E_g = 0.2$ eV, whose temperature 294 varies from $T_s = 320$ K to $T_s = 380$ K. 0.2 eV is roughly the bandgap of InSb 295 [51, 52]. The cell to be cooled, the target cell, is fixed at $T_c = 350$ K, and 296 its dielectric property will be specified shortly. We consider the semi-infinite 297 target cell and heat sink are separated by d = 20 nm. The goal is to see how 298 much heat flux can be removed from the cell when a reverse bias voltage is 299 applied to the semiconductor heat sink. 300

For the semiconductor heat sink, the dielectric function is governed by the direct valence-to-conduction interband transition [53, 54],

$$\epsilon_{pv}(\omega) = \epsilon_r(\omega) + i\epsilon_i(\omega) \tag{24}$$

$$\epsilon_i(\omega) = A\sqrt{x-1}/x^2, \ x > 1$$

= 0, x < 1
$$\epsilon_r(\omega) = B + A(2 - \sqrt{1+x})/x^2, \ x > 1$$

= B + A(2 - \sqrt{1+x} - \sqrt{1-x})/x^2, x < 1.

with $x = \hbar \omega / E_g$. As a model calculation, we use $(A, B, E_g) = (6, 10, 0.2 \text{ eV})$ [16]. We have varied A and B between 1 and 15 extracted from Refs. [53, 54], and found that they do not noticeably change the general behavior. The material of the dielectric functions of Eq. 25 will be referred to as "interband" material.

Three types of target cells are considered: the metal, the Lorentz material, 308 and the same interband material as the heat sink, with the blackbody as the 309 reference. Both metal and Lorentz material support the surface plasmon 310 polariton mode with a (surface) plasma frequency ω_0 . Our parameter choice 311 is guided by the impedance matching condition [39, 40]: a large transmissivity 312 can be obtained when the resonant energy $\hbar\omega_0$ is slightly larger than the 313 bandgap E_g . We therefore use $\hbar\omega_0 = 1.1 \cdot E_g$. The effect of decay rate will 314 be discussed in the next subsection. For the metal target cell, the dielectric 315 function can be approximated by the Drude model: 316

$$\epsilon_m = 1 - \frac{\omega_{pl}^2}{\omega^2 + i\gamma_m\omega}.$$
(25)

The surface resonant frequency is given by $\omega_0 = \omega_{pl}/\sqrt{2}$, so we choose $\omega_{pl} = \sqrt{2} \times 1.1 \cdot E_g$, and the decay is chosen to be $\gamma_m = 0.002\omega_{pl}$. For the Lorentz target cell, the dielectric function can be described by the Lorentz oscillator model:

$$\epsilon_L(\omega) = \epsilon_\infty \frac{\omega^2 - \omega_{LO}^2 + i\gamma\omega}{\omega^2 - \omega_{TO}^2 + i\gamma\omega}.$$
(26)

This is typical for many insulators. Here we choose $\epsilon_{\infty} = 4.46$. $\omega_{TO}/\omega_{LO} = 0.81$ and $\gamma/\omega_{LO} = 0.0041$. The resonant frequency is given by $\omega_0^2 = \frac{\omega_{LO}^2 + \omega_{TO}^2/\epsilon_{\infty}}{1+1/\epsilon_{\infty}}$ which is set to $(1.1 \cdot E_g/\hbar)^2$. The third choice is inspired by its symmetric configuration, which is shown to greatly enhance the transmissivity for both metal and Lorentz materials [16, 33, 39, 40]. For the blackbody reference, $_{326}$ the emissivity is given by [33, 40]

$$\varepsilon_{c,s}(\omega) = \frac{1}{2\pi} (\frac{\omega}{c})^2 \Theta(\omega - E_g/\hbar).$$
(27)

327 3.2. Simulation results

Fig. 4(b)-(d) show the input power (done to the heat sink) and the heat 328 removal flux (per unit area) for various target cells, with the blackbody 329 reference. The target cell is fixed at $T_c = 300$ K, whereas the heat sink varies 330 from $T_s = 250, 300, \text{ and } 350 \text{ K}$. Some general features are pointed out. We 331 first consider the case with zero bias voltage. When $T_s = T_c$, the target cell 332 and the heat sink are in equilibrium and there is no outgoing radiative flux 333 from the target cell; When $T_s > T_c$, the heat sink emits more photons than 334 the target cell such that the outgoing radiative flux from the target cell is 335 negative (heating); When $T_s < T_c$, the heat sink emits less photons than the 336 target cell such that the outgoing radiative flux from the target cell is positive 337 (cooling). When applying the reverse bias voltage, the heat removal flux 338 increases as the heat sink emits less photons, and reaches a saturation flux 339 given in Eq. 18. Note that when the heat sink is fixed at a higher temperature 340 $(T_s > T_c)$, a minimum applied voltage amplitude (V < 0) is needed for 341 cooling to happen. In our near-field arrangement (20 nm separation between 342 the planar target cell and the heat sink), the enhancement of the heat removal 343 flux is about 63 times (for the metal target cell), 22 times (for the Lorentz 344 target cell), and 11 times (for the same interband target cell) larger than the 345 blackbody reference. These results are comparable to the enhancement of 346 the TR power generator devices [40]. In the model simulations considered 347 here, the outgoing radiative flux saturates when |eV| is about 10-20% of the 348 bandgap E_q . The COP (η_{COP}) are computed using Eq. 16, and the results 349 for $T_s = 300 \text{ K}/250 \text{ K}$, $T_c = 300 \text{ K}$ are given in Fig. 5. Generally, the COP 350 for all target types of target cells are very close in value. The target cells 351 that support surface resonances, i.e., metal and Lorentz, have a COP about 352 5% lower than those that do not support surface resonances i.e., interband 353 and blackbody. This qualitative feature is found for other choices of $T_s \neq T_c$ 354 [see Fig. 5(a) and (b)]. 355

For metal and Lorentz target cells, the strong near-field enhancement originates from surface resonances introduced by the metal/vacuum and

Lorentz/vacuum interfaces. The energies of surface modes lie within a small 358 energy window [39]. In the far-field setup, the surface modes do not con-359 tribute to the radiative energy transfer due to the exponential decay in the 360 out-of-plane direction. In the near-field setup, the surface modes produce 361 a δ -function like peak with a strong peak amplitude in transmissivity and 362 greatly increase the radiative energy transfer [40]. An interesting and per-363 haps non-intuitive consequence derived from impedance matching condition 364 is that the damping of the resonance can sometimes help the radiative en-365 ergy transfer. In other words, there exists an optimal damping value for 366 the maximum radiative energy transfer. This is explicitly shown in PV [32] 367 and TR [40] devices. To illustrate this effect on negative EL cooling devices, 368 Fig. 6 shows the outgoing radiative fluxes per unit area for the metals with 369 plasma damping rate $\gamma_e/\omega_{pl} = 0.002, 0.004, 0.016, \text{ and } 0.030$ for two temper-370 ature differences. The radiative flux indeed shows a maximum value when 371 $\gamma_m/\omega_{pl} \sim 0.016$, above and below which the radiative flux decreases. 372

373 3.3. Discussion

We begin the discussion by comparing the TR devices as a power gener-374 ator with the negative EL cooling (two $\mu < 0$ devices, see Table. 1). Both 375 power generators and cooling devices require attaching an external load to 376 one of the reservoirs, which is a semiconductor whose dielectric function is 377 approximated by the interband material in Eq. 25 – for TR power generators 378 it is the TR cell that generates electricity; for the negative EL cooling it 379 is the heat sink that uses the input power for cooling. For TR power gen-380 erators one chooses a heat sink to maximize the output power for a given 381 TR cell, whereas for the negative EL cooling devices one selects a target 382 cell for a given heat sink. In essence, we want to increase the radiative 383 power transfer from one reservoir to the other. Within the formalism given 384 in Eq. 4 and 5, the key is to design (via the material selection and their 385 geometry) two reservoirs (e.g. the target cell and heat sink for the negative 386 EL cooling) simultaneously that maximizes their mutual transmissivity, and 387 the impedance-matching condition derived from CMT provides a very good 388 guidance [33, 39]. These considerations are general for PV devices as well. 389 As the transmissivity determines the near-field performance, it could be a 390 good experimental quantity to measure. 391

Two advantages of negative EL cooling devices are worth noting. First it 392 does not apply a voltage to the target cell which we want to cool, allowing 393 more material choices of target cell. In contrast, the EL cooling requires 394 a p-doped region, an n-doped region, and a bias voltage to the target cell, 395 which has to be a semiconductor [8, 10]. Second, as the negative EL devices 396 aim to reduce the e-h concentrations in the heat sink, the Auger recombina-397 tion, which typically suppresses the PV and TR performances, becomes less 398 significant. Potential applications for the negative EL cooling include to cool 390 the wide-gap insulator (e.g. SiC, BN) that supports the surface polariton 400 (optical phonons) using a small-bandgap semiconductor as a heat sink, and 401 to cool a metal using a semiconductor heat sink whose bandgap is compara-402 ble to the metal/vacuum surface plasma energy. Finally we emphasize again 403 that any approaches that enhance the TPV performance, such as coating 404 the PV cell or coating the emitter [39], can be applied to the negative EL 405 cooling. 406

Finally, we recognize that the near-field devices, such as the setup in 407 Fig. 4(a), are experimentally very challenging. First, to maintain a small 408 vacuum gap between the target cell and the heat sink is difficult, and one 409 possible solution is to use a few short supports to separate the target cell 410 and the heat sink [55, 56, 57]. Second, to fabricate a P-I-N junction in the 411 heat sink is also difficult. Combining some thin film fabrication methods 412 [58, 59, 60] to diffusion driven charge transport structures [61, 62, 63, 64] 413 can potentially make the device. However, if not properly passivated, the 414 resulting large surface recombination can outweigh the near-field gain. 415

416 4. Conclusion

To conclude, we provide a coherent formalism that applies to all devices 417 that use radiative energy transfer for the power generation and the active 418 The formalism includes the generalized Planck distribution that cooling. 419 characterizes a thermal reservoir by a temperature and a chemical poten-420 tial, and the transmissivity between the cell (that generates power or is to 421 be cooled) and its corresponding heat source or heat sink. In this frame-422 work, four types of devices are distinguished: a PV device that uses the 423 low-temperature reservoir to generate electricity; a TR device that uses the 424

high-temperature reservoir to generate electricity; an EL device that applies 425 a voltage to the cell to be cooled; a negative EL device that applies a voltage 426 to the heat sink. In all these devices, the transmissivity is the material and 427 geometry specific quantity that plays the crucial role in the performance. 428 Once the "impedance match" condition, originally derived for optimizing 429 PV devices, is formulated using transmissivity, it can be straightforwardly 430 generalized to other types of devices. Using the formalism, we analyze the 431 performance of the negative EL cooling, in the Shockley-Queisser framework 432 as a reference and its near-field enhancement. For the near-field arrange-433 ment, a small bandgap semiconductor is chosen as the heat sink, which is 434 close to the target cell which we want to cool. The heat removal flux can be 435 more than ten times larger than the blackbody reference. As the negative 436 EL cooling applies the voltage on the heat sink instead of the target cell 437 (which we want to cool), it can be useful for cooling the wide-gap insulators 438 or metals. 439

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446 Appendix: Transmissivity for planar structure

For two semi-infinite materials (specified by its dielectric functions) separated by d, the emission power at a given frequency via a TM (transverse magnetic) mode [44, 46] is

$$S_{z}^{(TM)}(z,\omega) = \frac{1}{(2\pi)^{3}} \hbar \omega \Theta(\omega,T) \left(\frac{\omega}{c}\right)^{2} \times \int \frac{d^{2}\mathbf{K}}{(\omega/c)^{2}} \frac{1}{|\alpha_{TM}|^{2}} \frac{\operatorname{Re}[\tilde{\gamma}_{0}]\operatorname{Re}[\gamma_{2}]}{(\omega/c)^{2}} \left(1 + \frac{k^{2}}{|\gamma_{2}|^{2}}\right) e^{+2\operatorname{Im}[\gamma_{0}](z-d)}.$$
(28)

450 Here α_{TM} is defined as

$$\alpha_{TM} \equiv \frac{1}{4} \left[e^{i\gamma_1 d} \left(\frac{\tilde{\gamma}_0}{\tilde{\gamma}_2} + \frac{\tilde{\gamma}_0}{\tilde{\gamma}_1} + \frac{\tilde{\gamma}_1}{\tilde{\gamma}_2} + 1 \right) + e^{-i\gamma_1 d} \left(\frac{\tilde{\gamma}_0}{\tilde{\gamma}_2} - \frac{\tilde{\gamma}_0}{\tilde{\gamma}_1} - \frac{\tilde{\gamma}_1}{\tilde{\gamma}_2} + 1 \right) \right], \quad (29)$$

with $\tilde{\gamma}_i = \frac{\gamma_i}{\epsilon_i}$, $k^2 + \gamma_i^2 = \epsilon_i (\frac{\omega}{c})^2$. The total emitting power (integrating over 452 all frequency) is

$$S = \int d\omega S_{z}^{(TM)}(z = d, \omega)$$

=
$$\int \frac{d\omega}{2\pi} \hbar \omega \Theta(\omega, T) \times \left[\left(\frac{\omega}{c} \right)^{2} \frac{1}{(2\pi)^{2}} \int \frac{d^{2}\mathbf{K}}{(\omega/c)^{2}} \frac{1}{|\alpha_{TM}|^{2}} \frac{\operatorname{Re}[\tilde{\gamma}_{0}]\operatorname{Re}[\gamma_{2}]}{(\omega/c)^{2}} \left(1 + \frac{k^{2}}{|\gamma_{2}|^{2}} \right) \right]$$

=
$$\int \frac{d\omega}{2\pi} \hbar \omega \Theta(\omega, T) \left[\frac{d^{2}\mathbf{K}}{(2\pi)^{2}} \varepsilon_{12}(\omega, \mathbf{K}) \right]$$
(30)

⁴⁵³ Note that in Eq. 30, the term in the square bracket has the dimension $1/L^2$, ⁴⁵⁴ and the dimensionless transmissivity at a given in-plane **K** and frequency ω ⁴⁵⁵ is give by

$$\varepsilon_{12}(\omega, \mathbf{K}) = \frac{1}{|\alpha_{TM}|^2} \frac{\operatorname{Re}[\tilde{\gamma}_0] \operatorname{Re}[\gamma_2]}{(\omega/c)^2} \left(1 + \frac{k^2}{|\gamma_2|^2}\right).$$
(31)

Eq. 31 is the transmissivity for two planar reservoirs. Let us consider the vacuum case, where $\epsilon_0 = \epsilon_2 = 1$ so $\tilde{\gamma} = \gamma$. In this case, $|\alpha_{TM}|^2 = 1$, and $1 + \frac{k^2}{|\gamma_2|^2} = \frac{(\omega/c)^2}{|\gamma_2|^2}$. As Eq. 31 vanishes when $\operatorname{Re}[\gamma] = 0$, we obtain $\varepsilon_{12}(\omega, \mathbf{K}) = \Theta(\omega/c - |\mathbf{K}|)$, which is the formula for the blackbody radiation.

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Function	Type	$(\mu, -I_c)$	description
PG	PV	(+, -)	low-T cell provides usable work
PG	TR	(-,+)	high-T cell provides usable work
Cooler	EL	(+, +)	work done to the to-be-cooled object
Cooler	Negative EL	(-, -)	work done to the heat sink

Table 1: Summary of four devices. $(\mu, -I_c)$ gives the signs of the photon chemical potential and the "negative" of the photocurrent for the device working region. PG stands for power generator. The sign of product $-I_c\mu$ determines its function: negative corresponds to power generators; positive corresponds to cooling devices.



Figure 1: (a) The PV cell as a power generator: the power is generated via the low-T PV cell. (b) The TR cell as a power generator: the power is generated via the high-T TR cell. (c) The EL cooling device. A forward bias is applied to the object 1 (at T_1) we want to cool. (d) The negative EL cooling device. A reverse bias is applied to the heat sink (at T_2). The arrows indicate the net photon flux.



Figure 2: Illustration of the relationship between the photon chemical potential μ , the (quasi) electron/hole Fermi energies E_{FC}/E_{FV} , and the equilibrium Fermi energy E_F . The box indicates the number of electron-hole pairs at thermal equilibrium. (a) At thermal equilibrium, $E_F = E_{FC} = E_{FV}$, and $\mu = E_{FC} - E_{FV} = 0$. (b) In the non-equilibrium situation where e-h concentration is larger than that at equilibrium, $E_{FC} > E_F$, $E_{FV} < E_F$ and $\mu = E_{FC} - E_{FV} > 0$. (c) In the non-equilibrium situation where e-h concentration is smaller than that at equilibrium, $E_{FC} - E_{FV} < 0$.



Figure 3: (a) The sign convention of a pn-junction. Forward-bias voltage and current directions are chosen to be '+'; reverse-bias voltage and current directions '-'. (b) Current-voltage relation for PV (red) and TR (blue) cells. The blue shaded areas represent the power delivered by the PV and TR devices: the larger area gives the larger output power. The areas of dashed boxes represent the work done to the EL and negative EL devices: the smaller area gives the better cooling performance. The rectangular areas are randomly chosen and for illustration only.



Figure 4: (a) The illustration of negative EL device. The heat sink is a semiconductor of a bandgap E_g . When a reverse bias voltage is applied, the heat sink emits much less photons (the arrows with a red cross), effectively enhancing the heat removal flux of the object one wants to cool. (b)-(d) The input power (dashed curves) and the outgoing radiative flux (solid curves) per unit area of the metal (black), Lorentz (red), interband (green) target cells, with blackbody reference (blue). The heat sink temperature is T_s , whereas the cell temperature is T_c . (b) $T_c = 300$ K, $T_s = 300$ K. (c) $T_c = 300$ K, $T_s = 350$ K (beware of the linear scale in power density) (d) $T_c = 300$ K, $T_s = 250$ K. When the applied voltage is large in amplitude, the outgoing heat flux saturates.



Figure 5: COP for the metal (black), Lorentz (red), interband (green) target cells, with blackbody reference (blue). (a) The cell and sink are both fixed at 300 K; (b) The cell is fixed at 300 K whereas and heat sink temperatures 250 K. The target cells that support surface resonances, i.e., metal and Lorentz, have a COP about 5% lower than those that do not support surface resonances, i.e., interband and blackbody. The dashed horizontal line represents $\eta_{COP} = 1$.



Figure 6: The outgoing radiative flux (in linear scale) per unit area for the metals of plasma damping rate $\gamma_e/\omega_{pl} = 0.002$ (black), 0.004 (red), 0.016 (green), and 0.030 (blue). (a) The cell and sink are both fixed at 300 K; (b) The cell is fixed at 300 K whereas and heat sink temperatures 250 K. The radiative flux shows a maximum value when $\gamma_m/\omega_{pl} \sim 0.016$ for both temperature differences.