Influence of Hot-Carrier Extraction from a Photovoltaic Absorber: An Evaporative Approach

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The effect of energy-selective extraction on a hot carrier's population is addressed in this study. Using an evaporative cooling model inspired by the field of cold atoms, we derive an analytical expression supported by numerical calculations to account for the removal of particles from the distribution and subsequent energy redistribution among the remaining carriers. Depending on the filtering dimensionality and energy level of extraction, the distribution can be either heated up or cooled down, resulting in a modification of the current-voltage characteristic associated to the structure. The negative differential resistance peak indicating the selective extraction is shown to be markedly reduced when evaporation is considered, which may lead to an overestimation of the tunneling current in previous models. These results provide insights into the interpretation of experimental results on energy-selective contacts, as well as a straightforward method to unequivocally demonstrate the energy filtering of hot carriers in a structure operating under continuous illumination.

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I. INTRODUCTION

Within picoseconds after the absorption of a photon in a semiconductor, the promoted electron dissipates the imparted energy through carrier-carrier and carrier-phonon interactions leading the ensemble of photogenerated particles to relax towards a thermal distribution at equilibrium temperature with the surrounding lattice. This process implies a loss for the available work and gives rise to the celebrated Shockley-Queisser upper limit for photovoltaic conversion efficiency [1].

Hot-carrier solar cells (HCSCs) have been suggested as a way to circumvent this bound by harvesting carriers before they relax their excess energy and reach the bottom of the conduction band [2,3]. In its most simple form, a HCSC consists of two features: an absorber, in which photogenerated carriers are sufficiently decoupled from the surrounding lattice such that their equilibrium temperature is higher than the ambient one and an energy-selective contact (ESC), which allows for the discriminating extraction of hot carriers. In parallel to research carried out on both absorber [4–11] and contact [12–17] systems, some studies tackled the understanding of the complete structure theoretically [18,19] and experimentally [12,20,21], showing simultaneously the presence of hot carriers in the absorber and extraction of a current through the contact. Nevertheless, direct proof of the selective extraction of hot carriers for a structure under continuous illumination is still to be made.

A possible way to do so is to consider the feedback of current extraction on the hot carriers remaining in the absorber. In most semiconductor applications, carriers are buffered by their interaction with the lattice, which plays the role of a reservoir maintaining the temperature constant regardless of the extraction [22,23]. By contrast, in HCSC, carriers and leading-order (LO) phonons constitute a partially isolated system, allowing hot temperatures to be reached [24]. The energy loss induced by the carriers' extraction is, therefore, not fully compensated by the environment, and the ensemble subsequently relaxes towards a steady state determined by energy conservation rather than by the lattice temperature alone. However, this influence of extraction on the carriers' properties and, notably, their temperature, is omitted in some works or treated as a mere side product of the calculation. On the contrary, we show that this feedback should be explicitly taken into account, as it can constitute a smoking gun for hot carriers' selective extraction and lead to significant consequences for HCSC properties.

The influence of the particles' extraction on an isolated ensemble has been extensively studied in the field of cold atoms, where it is referenced as an *evaporative* process [25,26]. When cooling an atomic sample, particles with a given energy are actively removed, just like carriers are

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extracted through a resonant contact. In both cases, owing to energy conservation, the remaining particles redistribute the remaining energy, and the ensemble relaxes towards a thermal distribution, even without any contribution from the environment. If the extraction energy is high enough, the average energy per particle decreases, and the steady-state temperature decreases leading to the so-called *evaporative cooling*. Conversely, if the extraction is performed at low energy, the average energy per particle increases, and the remaining particles tend to form a hotter distribution, corresponding to an *antievaporative cooling* [27–29]. Cold atom experiments aim to reduce the temperature while extracting as little particles as possible, while HCSC aims to extract particles while reducing the temperature as little as possible. However, despite opposite optimization, the two systems can be depicted by an analog description.

In this article, we propose a minimalistic model inspired by evaporative cooling to account for the influence of energy-selective extraction on the remaining population. Our approach relies on a perturbative treatment, which consists of breaking the evaporation into discrete steps and treating the cooling induced on the initial state by a small extraction [30]. We extend this model by considering additional terms corresponding to the net energy and particle fluxes provided to the carriers' population through photoexcitation, thermal relaxation, and radiative recombination. We obtain a first-order analytic expression for the evaporative cooling, which compares with numerical calculations and allows for direct experimental verification. While the system under scrutiny is not a complete HCSC in working conditions, the results obtained here bring qualitative understanding of the feedback induced by selective extraction, suggest a method to prove the discriminating harvesting of hot carriers, and shed light on the influence of transmissivity on the output current.

II. METHOD

In order to put emphasis on the evaporative mechanisms, we consider a minimalistic model composed of an illuminated absorber allowing for hot carriers and connected to a collector through an energy-selective contact such as a quantum-well resonant tunneling barrier (QWRTB) or a quantum-dot resonant tunneling barrier (QDRTB; see Fig. 1) [31]. Electrons and holes are treated in a fully symmetric way, and only electrons are mentioned in the following.

A. Open-circuit situation

In the open-circuit (OC) situation, the steady state results from the equilibrium between photoexcitation, carrier recombination, and thermalization with the lattice. This steady state can be characterized by a chemical potential $\mu_{\mathrm{OC}} = \mu_0 + \Delta\mu/2$ (where μ_0 is the intrinsic Fermi level, and $\Delta\mu$ the quasi-Fermi-level splitting resulting from the illumination) and an electronic temperature T_{OC}. In HCSC, inelastic processes are weak enough to allow the temperature T_{OC} to be partially decoupled from that of the lattice T_{latt}. Under the continuous-wave excitation considered in this work, the thermalization of hot carriers can be accounted for by the Q-factor model proposed by Le Bris *et al.* [7]

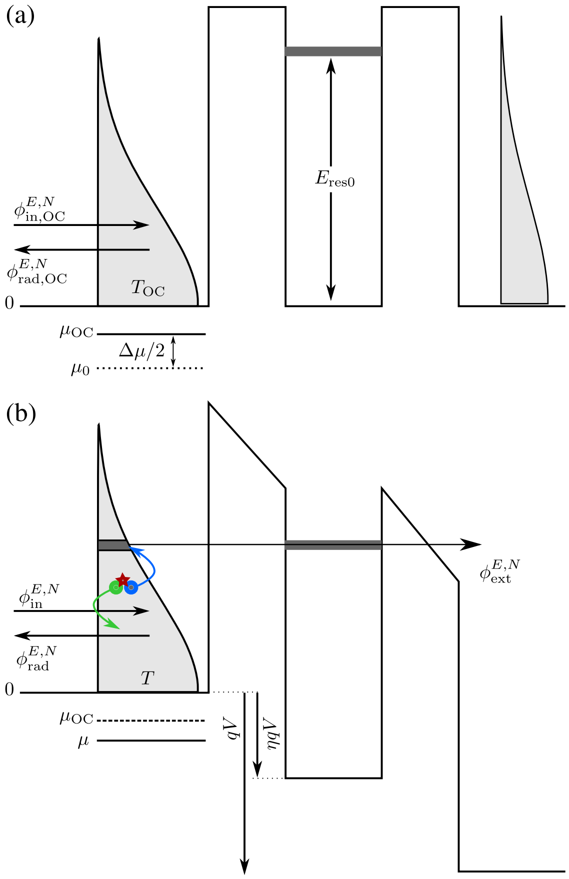

FIG. 1. (a) In OC conditions, the thermal population in the absorber is characterized by a temperature $T_{\mathrm{OC}} > T_{\mathrm{latt}}$ and a chemical potential μ_{OC} resulting from the photoinduced quasi-Fermi-level splitting $\Delta\mu$. (b) Under an external bias V, the resonant energy level is lowered inside the absorber population. A new equilibrium (T, μ) results from the balance between radiative recombination, net source terms, and extraction. From an evaporation perspective, elastic collisions (red star) redistribute the energy between particles. After the interaction, some particles (in green) might carry away most of the pair energy and reach the resonant level, while the remaining particles (in blue) are left with little energy and relax towards a colder distribution. Conversely, the process can lead to a heating of the population if the extraction is performed at the bottom of the population.

$$\phi^{E}_{\mathrm{rel}}(T) = Q(T - T_{\mathrm{latt}}) \exp\left(-\frac{\hbar\omega_{\mathrm{LO}}}{k_B T}\right), \tag{1}$$

where $\hbar\omega_{\mathrm{LO}}$ is the energy of the LO phonon responsible for energy loss, and Q is the so-called thermalization factor, including density effects and thermalization rate.

Singling out the recombination process, a particle and energy balance for the thermalized carriers can be expressed within the radiative limit as

$$\phi_{\mathrm{in,OC}}^{E,N} = \phi_{\mathrm{rad}}^{E,N}(T_{\mathrm{OC}}, \mu_{\mathrm{OC}}), \tag{2}$$

where E (N) stands for the energy (particle) flux. At high carrier temperature and under maximal concentration, radiative fluxes $\phi_{\mathrm{rad}}^{E,N}$ for the electrons are given by [32]

$$\phi_{\mathrm{rad}}^{N}(T,\mu) = \frac{1}{4\pi^2\hbar^3c^2}\int_{E_g}^{+\infty} d\hbar\omega(\hbar\omega)^2 e^{[(\Delta\mu-\hbar\omega)/k_BT]}, \tag{3}$$

$$\phi_{\mathrm{rad}}^{E}(T,\mu) = \frac{1}{8\pi^2\hbar^3c^2}\int_{E_g}^{+\infty} d\hbar\omega(\hbar\omega)^2(\hbar\omega - E_g) \times e^{[(\Delta\mu-\hbar\omega)/k_BT]}, \tag{4}$$

and $\phi_{\mathrm{in,OC}}^{E,N}$ are *net source* fluxes accounting for the energy and particles reaching the thermalized population.

Note that $\phi_{\mathrm{in,OC}}^{E,N}$ are convoluted terms including both photoexcitation and thermalization processes. Their precise determination, which is required to estimate the steady-state properties $(T_{\mathrm{OC}}, \mu_{\mathrm{OC}})$ *ab initio*, is beyond the scope of this work. Instead, the strategy adopted here considers that the OC properties are accessible as in Ref. [33] and focuses on their perturbations induced by carrier extraction.

To simplify the picture, we assume in the following that the barrier is high and large enough to prevent all but resonant extraction and consider a unique resonant energy level initially much higher than the average thermal energy of the carriers, such that backward contribution to the current is negligible, and we take the open-circuit voltage as the voltage origin without loss of generality.

B. Under extraction

When an external bias V is applied, the resonant energy level is lowered inside the absorber's population [Fig. 1(b)], and a current is extracted. The description of the ESC then relies on the Tsu-Esaki model [34]

$$\phi_{\mathrm{ext}}^{N}(V,T,\mu) = \int d^3k\,\tau(\mathbf{k},V)\rho(\mathbf{k})e^{[(E-\mu)/k_BT]}\frac{\hbar k_z}{m}, \tag{5}$$

$$\phi_{\mathrm{ext}}^{E}(V,T,\mu) = \int d^3k\,\tau(\mathbf{k},V)\rho(\mathbf{k})e^{[(E-\mu)/k_BT]}\frac{\hbar k_z}{m}\times E, \tag{6}$$

where ρ is the electronic density of state and z the transport direction. The transmission τ takes the Breit-Wigner [35,36] form

$$\tau_{\mathrm{QWRTB}}(\mathbf{k},V) = \frac{A(V)}{1+\left(\frac{\hbar^2k_z^2/2m - E_{\mathrm{res}}(V)}{\Gamma}\right)^2}, \tag{7}$$

$$E_{\mathrm{res}}(V) = E_{\mathrm{res0}} - \eta \times qV, \tag{8}$$

where η is the voltage drop ratio between the two barriers and sharp resonance is assumed [37]. The transmittivity $A(V) = A_0$ is taken as a constant. Transmissivity variations with the bias [38] can be included without additional difficulty but do not add any physical content to the problem. Note that the transmission through a QDRTB can simply be accounted for by changing $k_z \to k$ in the expression of τ.

As in the case of evaporative cooling, this extraction results in the perturbation of the initial thermal state $(T_{\mathrm{OC}}, \mu_{\mathrm{OC}})$. Extracted particles take energy away with them, and the remaining population will share the remaining energy, relaxing towards (T, μ). As can be inferred from this simple picture, the temperature estimated as the average energy per particle can either increase or decrease depending on the position of the energy level. The main difference with a standard evaporation is the presence of source terms and additional loss fluxes due to radiative recombination and thermalization with the lattice. As photogeneration is unchanged with respect to open-circuit conditions, thermalization and recombination fluxes must decrease to balance the extracted quantities.

Quantitatively, the feedback of extraction on the thermalized population in the absorber can be accounted for by a particle and energy conservation

$$\phi_{\mathrm{in}}^{E,N} = \phi_{\mathrm{rad}}^{E,N}(T,\mu) + \phi_{\mathrm{ext}}^{E,N}(V,T,\mu). \tag{9}$$

To further simplify the picture, we assume that the extracted quantities are essentially compensated by an adjustment of the radiative fluxes, while the thermalization term is almost unaffected:

$$|\phi_{\mathrm{rel}}^{E}(T) - \phi_{\mathrm{rel,OC}}^{E}| \ll |\phi_{\mathrm{rad}}^{E}(T,\mu) - \phi_{\mathrm{rad,OC}}^{E}|. \tag{10}$$

This assumption is valid as $Q \lesssim 0.1$ W/K/cm^2, slightly below experimentally reported values [33], and we discuss possible extensions of this model is Sec. IV. Within this condition, the net source fluxes are almost unmodified from their open-circuit values:

$$\phi_{\mathrm{in}}^{E,N} \simeq \phi_{\mathrm{in,OC}}^{E,N}. \tag{11}$$

III. RESULTS AND DISCUSSION

A. First-order calculation

As long as the extracted fluxes remain small, an analytic expression for the temperature change can be obtained at first order of perturbation theory. In terms of particle density $n = [(mk_BT)/2\pi\hbar^2]^{3/2}e^{[\mu/(k_BT)]}$, the radiative recombination fluxes Eqs. (3) and (4) take the form

$$\phi_{\mathrm{rad}}^{N} = Bn^2, \qquad \phi_{\mathrm{rad}}^{E} = \frac{k_BT}{2}\times\phi_{\mathrm{rad}}^{N}, \tag{12}$$

where B is the radiative recombination coefficient [39], and a large gap limit is considered. The extracted fluxes Eqs. (5) and (6) can be reduced to

$$\begin{aligned}\phi^N_{\text{ext}} &= c(V)n,\\ \phi^E_{\text{ext}} &= \left(E_{\text{res}}(V) + d \times \frac{k_B T}{2}\right)\phi^N_{\text{ext}},\end{aligned} \tag{13}$$

where $c(V)$ is a tunneling speed, and d is the number of degrees of freedom (DOF) unconstrained by the contact ($d = 2$ for a QWRTB, $d = 0$ for a QDRTB). Each of these free DOF contribute to the extracted energy according to the equipartition theorem. The balance (9) then results in a temperature change

$$k_B \Delta T(V) = -2\left(E_{\text{res}}(V) + (d-1)\frac{k_B T_0}{2}\right)\frac{\phi^N_{\text{ext}}(V)}{\phi^N_{\text{rad,OC}}}. \tag{14}$$

This expression constitutes the main result of this work and holds several features. First, it contains only experimentally accessible quantities (see Sec. IV) and, thus, allows for direct assessment of the model. Second, it underlines a direct connection between hot-carrier extraction and temperature variation for the remaining population. Measuring a temperature variation under current extraction can, thus, provide direct proof of selective hot-carrier harvesting. Finally, Eq. (14) expresses several trade-offs which can lead to nonintuitive results. For instance, when increasing the bias, the balance between extracting more particles and lowering the extraction level results in a maximal cooling at a bias V_m different from the feed resonance $V_{\text{res}} = [E_{\text{res0}}/(q\eta)]$ at which the resonant level reaches the bottom of the band [38]. This bias can be estimated by considering $c(V) \propto \exp(-E/k_B T_{\text{OC}})$ for QWRTB and $c(V) \propto E \times \exp(-E/k_B T_{\text{OC}})$ for QDRTB, leading to

$$qV_m \simeq qV_{\text{res}} - \frac{1}{2\eta q} k_B T_{\text{OC}} \quad \text{for QWRTB}, \tag{15}$$

$$\simeq qV_{\text{res}} - \frac{(\sqrt{17}+5)}{4\eta q} k_B T_{\text{OC}} \quad \text{for QDRTB}. \tag{16}$$

As a second example, the formula predicts that a heating of the distribution can take place if the extraction level is below $(1-d)[(k_B T_0)/2]$. While this situation is not relevant for QWRTB, as the corresponding value is below the bottom of the band, it corresponds to an accessible situation for a QDRTB.

B. Numerical integration

The results of the numerical integration of Eq. (9) are shown in Fig. 2 and compared to first-order perturbation theory Eq. (14). The numerical values are taken as follows. The material has a gap of $E_g = 1.4$ eV, and the chemical potential in the dark is $\mu_0 = -E_g/2$ (intrinsic and symmetric situation). The illumination results in a carrier density of 10^{19} cm^{-3} at a temperature of 500 K. The contact has a width $\Gamma = 1$ meV small compared to the distribution spread [14]; the resonant level is initially located at $E_{\text{res0}} = 1$ eV, and the barrier is considered symmetric $k = 1/2$, leading to a current resonance at $2V$. Finally, the transmission amplitude A_0 is taken such that $A_0[(emk_B T_0 \Gamma)/4\pi^2\hbar^3] \simeq 10^2$ A cm^{-2} [37].

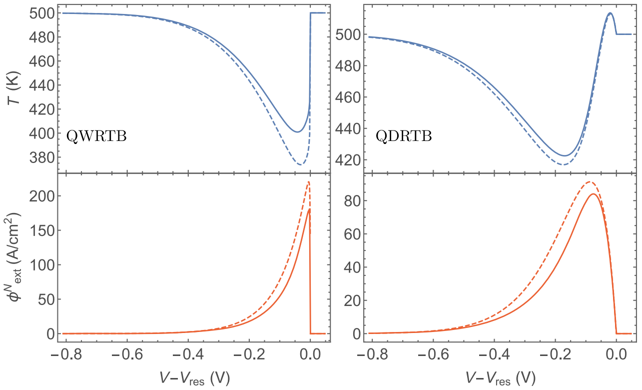

FIG. 2. Extracted current and temperature of the remaining carriers with an energy-selective contact filtering 1 DOF (QWRTB, left) and 3 DOF (QDRTB, right) in the radiative limit $f = 1$. Solid lines are given by the numerical integration of Eq. (9); dashed lines are first-order perturbation theory given by $\phi^N_{\text{ext}}(V, T_{\text{OC}}, \mu_{\text{OC}})$ and Eq. (14).

C. Evaporative cooling

The simulations show a clear decrease of the temperature up to several dozens of Kelvins. If this value is certainly overestimated by this simple model where several heating processes, such as the Joule effect, are neglected, the order of magnitude is such that we can reasonably expect to observe an experimental signature in real systems.

The numerical integration is well followed for the first-order result Eq. (14), which validates its relevance for experimental situations. We find indeed a good adequacy in the general behavior and notably in the position of the predicted minimal temperature. Furthermore, as expected, we observe an evaporative *heating* for QDRTB when the resonant level is lowered below $k_B T_0/2$.

We believe that the simultaneous measurement of the resonant current and this temperature change, which is opposite from the most parasitic effects such as Joule heating and can be explained only by evaporative processes, can provide a valuable tool to prove the selective extraction of hot carriers.

D. NDR peak

Another salient feature should be emphasized: The evaporation affecting both the temperature and the chemical potential also results in a reduction of the resonant current $\phi^N_{\text{ext}}(V, T, \mu)$, as compared to what can be expected from the open-circuit situation $\phi^N_{\text{ext}}(V, T_{\text{OC}}, \mu_{\text{OC}})$. The current peak-to-valley ratio is, thus, overestimated if one

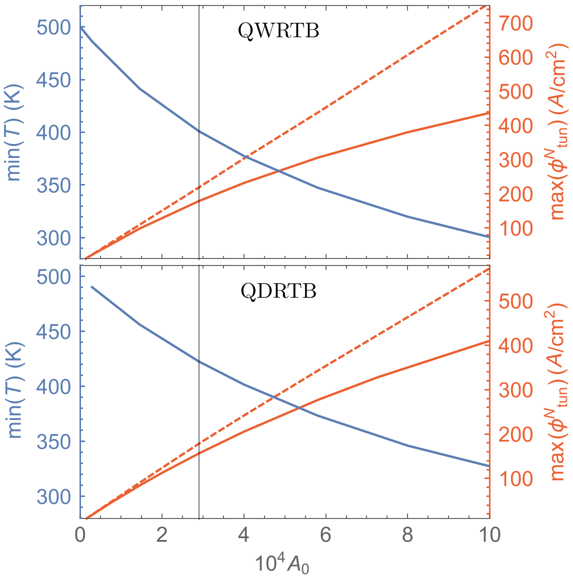

FIG. 3. Maximal extracted current ("peak value," in red) and minimal temperature (in blue) for increasing barrier transmittivity with a QWRTB contact (above) and a QDRTB contact (below). The dashed line is the linear behavior expected from Tsu-Esaki model Eq. (5) with open-circuit parameters [i.e., $\max[\phi^N_{\text{ext}}(V, T_{\text{OC}}, \mu_{\text{OC}})]$], and the solid line is the numerical integration Eq. (9) taking evaporation into account [i.e., $\max[\phi^N_{\text{ext}}(V, T, \mu)]$]. The vertical black line indicates the value used in Fig. 2.

considers the standard Tsu-Esaki formula Eq. (5) at constant temperature.

E. Influence of transmittivity

This result also sheds light on the influence of the contact transmissivity A_0. While increasing the transmissivity improves the extracted current, it also enhances the evaporation, which tends to reduce the extracted current. This effect is most important close to the peak current. As shown in Fig. 3, the trade-off results in a departure from the linear increase of the maximal extracted current with the transmissivity, which can be expected from Eq. (5) if the distribution parameters are kept at their open-circuit values. This result supports and brings physical insight into the previous analysis that predicted a detrimental effect of a too large conductance [18,19].

IV. PERSPECTIVES

As shown on Fig. 2, the first-order expression (14) is in satisfying agreement with the numerical simulations and should, therefore, also be relevant to account for experimental measurements. It is worth noting that all parameters included in this expression are accessible experimentally, for instance, through a photoluminscence (PL) measurement performed simultaneously with I-V characterization. At zero bias, the open-circuit temperature of the carriers T_0 and recombination flux $\phi^N_{\text{rad,OC}}$ can be estimated from the PL signal [33]. Such a measurement would require an absolute calibration of the luminescence signal in photon per second [40]. As an external bias V is applied, the temperature change ΔT can be recovered from the PL signal, while the output current $\phi^N_{\text{ext}}(V)$ is readily accessible through the electrical contact. The resonant energy level E_{res} position can either be taken as a fitting parameter or be inferred from the position of the NDR peak and the asymmetry of the contact, which determines the voltage drop ratio η.

While the approach developed in this work is kept minimalist in order to underline the physical picture, it can readily be applied to a more complex description of the problem. For instance, for absorbers with a higher thermalization parameter $Q \gg 0.1\ \text{W/K/cm}^{-2}$, the extraction-induced modification of the thermalization rate can be accounted for in the energy balance by including the term (1), which is found negligible in this study. Furthermore, nonradiative recombinations can also be considered by enhancing radiative fluxes by a factor f^{-1}, f being the radiative efficiency [1,41]. Up to the first order, this correction will appear as a scaling prefactor in Eq. (14). Both of these considerations will result in a damping of the evaporation influence without changing its qualitative behavior. By contrast, the Joule effect will lead to a monotone heating of the distribution with increasing extraction and constitute a parasitic contribution that might hinder the cooling signal. In the situation considered here, a sheet resistivity of approximately $150 \times 10^{-3}\ \Omega\,\text{cm}^2$ is required to compensate for the evaporation influence.

Finally, the model can be adapted to include or focus on a semiselective extraction, where all particles above a certain energy threshold are harvested. This extraction strategy has been considered for HCSC [14] and underlines a connection with thermionic cooling applied to hot carriers [42,43]. From this perspective, the evaporative scheme considered here resembles a light-induced Nottingham effect in the absence of a lattice buffer [44–47]. Similarly, one can consider nonselective extraction, such as nonresonant tunneling through the potential barriers [48], which relates to the nonresonant Nottingham effect [49–51]. All three components (selective, semiselective, and nonselective) can also be treated together to account for more realistic systems [38].

V. CONCLUSION

We develop a simple model inspired by atomic evaporative cooling to describe the feedback of energy-selective extraction on the remaining hot-carrier population. The two key assumptions of this model are to take as a starting point open-circuit conditions, which can be obtained experimentally and to treat perturbatively the influence of extraction. Within this theory, we show that extraction results in the cooling or heating of the steady temperature depending on the extraction level and constraints. This signal readily accessible through photoluminescence measurements under *I*-*V* characterization can constitute a smoking gun of hot-carrier extraction and is well accounted for by an analytic expression Eq. (14). Furthermore, the evaporation induces a reduction of the extracted current and, hence, of the resonant peak-to-valley ratio. This effect leads to a reduced increase of the extracted current with the contact transmissivity as compared to the linear behavior expected from the Tsu-Esaki expression. Our work, thus, brings insights into the required properties of energy-selective contacts in terms of extraction level and conductivity, adding qualitative understanding of the previous analysis. Several variations of this model are considered, showing the broad versatility of this approach.

While this study underlines the influence of evaporative cooling on the extracted current, it should be noted that the voltage provided by a HCSC also depends on the carriers' temperature [52]. The present model can, therefore, be used to account for a hot-carrier solar cell under working conditions and estimate the impact of extraction feedback on the energy conversion efficiency.

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