

# Impact of slow carrier-carrier equilibration on spectroscopic carrier temperature determination and hot-carrier solar cell performance

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Spectroscopic characterization is a key tool for determining carrier temperature in optoelectronic materials and devices. Assigning and extracting carrier temperature requires the assumption of quasiequilibrium, where carriers follow a Fermi-Dirac distribution. However, under low illumination intensities, carrier-carrier equilibration may be too slow for this assumption to hold. This is particularly relevant for hot carrier solar cells (HCSCs), which are required to operate under solar photon fluxes. Using a nonequilibrium model with a relaxation time approximation, we show that slow carrier-carrier scattering may distort temperature extraction from photoluminescence spectroscopy. Furthermore, our work shows that inefficient carrier-carrier equilibration in HCSCs leads to depletion of carriers at the extraction energies, resulting in a reduction in the extracted current and therefore the efficiency of the device. Our findings highlight the significance of considering carrier transport and scattering properties when evaluating and engineering candidate HCSC absorber materials.

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## I. INTRODUCTION

Determining the temperature of charge carriers is key for a variety of optoelectronic systems; hot carriers can degrade the performance of metal-oxide-semiconductor (MOS) devices [1] and lasers [2,3], while they may enhance the performance of photovoltaic devices [4,5], photocatalysts [6,7], and photodetectors [8]. Photoluminescence (PL) spectroscopy is a useful tool in probing such systems, as PL spectra have been known to encode information about hot carriers in semiconductors [9]. Fitting the high-energy tail of PL spectra has become a common way to determine carrier temperatures in devices [10]. This fitting assumes the emission intensity  $I_{\text{PL}}$  as a function of emission energy  $E$  is given by the generalized Planck law as follows [11]:

$$I_{\text{PL}}(E) = \alpha(E) \frac{E^2}{4\pi^2 \hbar^3 c^2} \left[ \exp\left(\frac{E - \Delta\mu}{k_B T}\right) - 1 \right]^{-1}, \quad (1)$$

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where  $\Delta\mu$  is the quasi-Fermi-level separation (QFLS),  $T$  is the carrier temperature, and  $\alpha$  is the absorptivity. For energies  $E \gg \Delta\mu$ , the exponential term dominates and

$$I_{\text{PL}}(E) \propto E^2 \exp\left(-\frac{E}{k_B T}\right). \quad (2)$$

The  $E^2$  factor is often approximated to be constant; the high-energy tail (HET) of the PL spectrum can therefore be fit to an exponential decay, the decay constant of which depends on the carrier temperature. Such fitting techniques assume that carriers follow a Fermi-Dirac distribution, however, modeling has shown that there may be nonequilibrium steady-state carriers in semiconductors illuminated by low-power continuous-wave light [12]

In the case of hot-carrier solar cells (HCSCs), the impact of non-Fermi-Dirac carrier distributions can be significant. HCSCs harness the energy of hot charge carriers by extracting them via energy-selective contacts, forming an optoelectronic heat engine. While their detailed balance efficiency limit is 85% at full solar concentration [13,14], achieving such high efficiencies has been challenging due to fast carrier cooling in most materials. The nature of the carrier dynamics in a material is key in determining its suitability as a HCSC absorber. Experimentally, carrier dynamics are probed via time-resolved spectroscopy techniques, which examine carrier dynamics following an intense pulsed-laser excitation [15–17].

Such recent experiments have shown relatively slow carrier cooling in perovskites, attributed to their unusual phononic band structures [18,19]. Perovskites have complex dynamics and cooling mechanisms, which require careful interpretation of temperatures extracted via spectroscopy [20]. Fast and efficient extraction of long-lived hot carriers into carrier transport layers has been credited for performance boosts in perovskite solar cells [21–23], although how “hot” the carriers are remains debated [24].

Carrier dynamics at femtosecond and picosecond scales give insight into electron cooling effects; however, the pulse-width-limited resolution of many of these techniques may not be fine enough to detect carrier equilibration processes and characterize them. If detectable, carrier equilibration is found to be at least 10 times faster than other timescales [25]. Based on these observations in ultrafast spectroscopy, the typical assumption is that processes such as carrier scattering, momentum relaxation, and energy redistribution are fast enough for HCSC operation in general [4]. However, pulsed-laser excitations result in carrier concentrations orders of magnitude greater than that of steady-state solar illumination. Carrier scattering and equilibration is known to be dependent on carrier concentration [26]. A critical carrier density is required for efficient intercarrier energy exchange to dominate over carrier cooling, resulting in equilibrated carrier distributions [27]. Lower carrier densities under solar illumination may result in inefficient carrier-carrier scattering [28], which would inhibit energy redistribution within the carrier population.

For GaAs, modeling has shown that the effects of “spectral hole burning” become apparent for extraction rates greater than  $10^{-3}$  times the carrier scattering rate [29] under extraction of hot carriers via idealized energy-selective contacts. Spectral hole burning refers to a depletion of electrons in the range of energies at which they are being extracted and is most often encountered in semiconductor lasers [2]. However, unless fundamental electronic interactions are included, most HCSC models assume that carriers follow a Fermi-Dirac distribution, which assumes the fundamental requirement of efficient energy redistribution in the carrier population. A model that calculates carrier-carrier interactions using the exact quantum statistical equations is not computationally feasible, therefore a practical approach to include nonequilibrium distributions is to make a relaxation-time approximation, where processes such as carrier equilibration are assigned a time constant with which they contribute to the relaxation of the carrier distribution. Such models have shown that slow carrier-carrier scattering can deplete carriers and lower HCSC efficiency [30,31].

The impact of slow carrier-carrier scattering on HCSCs remains an underexplored area of research. In this work, we use a relaxation-time approximation model of an exemplary electron-hole symmetric system to show the impact of nonequilibrated carriers on photoluminescence

measurements of semiconductors, and the device operation and efficiency of HCSCs.

## II. MODEL

We consider an absorber material with a low band gap of 0.5 eV, which enables absorption of a wide range of the solar spectrum. The contacts are modeled as semi-infinite thermionic barriers, which have been shown to be a realistic route to achieving high HCSC efficiencies [32–34], bypassing the fabrication complexity of resonant tunneling quantum dot contacts [35,36].

We employ a relaxation-time approximation model in which we approximate scattering processes as a relaxation of electrons towards a Fermi-Dirac distribution with a certain time constant  $\tau_{cc}$ . This process is not constrained to conserve particle number, assuming that equilibration is due to a mixture of processes such as carrier-carrier scattering, impact ionization, and Auger recombination. Since we consider a low band-gap material, such intraband and interband processes are expected to occur on similar timescales [37,38]. When these processes are fast (low  $\tau_{cc}$ ), our model approaches the impact ionization model of HCSCs, in which there is no QFLS within the absorber and the voltage across the contacts is generated solely due to the temperature differential between the hot carriers in the absorber and the cold carriers in the contacts [14].

Using our model, we show how the interplay of excitation, scattering, and extraction rates affects the steady-state carrier distributions, device efficiencies and heat flows. To focus on these effects, we exclude carrier-phonon coupling and carrier cooling.

We consider the occupancy  $f$  of electrons at various energy levels  $E$ , with spacing  $\Delta E$ . The carriers relax towards a Fermi-Dirac distribution  $f_{FD}$  with time constant  $\tau_{cc}$ , assumed to be identical for electrons and holes. To ensure energy conservation,  $f_{FD}$  with temperature  $T_{FD}$  is defined as the Fermi-Dirac distribution that satisfies

$$\sum EDOS(E)f_{FD}(E, T_{FD}) = \sum EDOS(E)f(E), \quad (3)$$

where DOS is the density of states, assumed to be parabolic.

Electrons at a given energy level can be generated by excitation from the solar flux or the ambient flux. Electron-hole symmetry and equal division of photon energy is assumed. This means that to generate an electron at energy  $E$ , a photon of energy  $E_\gamma = 2E$  is required. The electron flux resulting from solar excitation  $\phi_{el, Sun}$  at energy  $E$  is therefore

$$\phi_{el, Sun}(E) = C\phi_{BB}(E_\gamma = 2E, T_{Sun}), \quad (4)$$

where the Sun is assumed to be a Blackbody at  $T_{Sun} = 5800$  K. The factor  $C$  accounts for solar concentration,

with  $C = 1$  for full concentration and  $C = 1/46\,200$  for 1 Sun. Concentration of sunlight will likely be required to achieve practical HCSC operation [39], hence we choose 1000 Suns, which is achievable with practical optics.  $\phi_{BB}$  is given by the Planck radiation law as follows:

$$\phi_{BB}(E, T) = \frac{2\pi E^2}{h^3 c^2} \frac{1}{\exp\left(\frac{E}{k_B T}\right) - 1}. \quad (5)$$

Note that  $\phi_{\text{el,Sun}}(E)$  is the generated flux of electrons and is distinct from the incoming solar photon flux since a photon of energy  $E_\gamma$  generates an electron at energy  $E/2$ , resulting in a different flux distribution. Similarly, the generated flux due to the ambient is given by

$$\phi_{\text{el,amb}}(E) = (1 - C)\phi_{BB}(E_\gamma = 2E, T_A), \quad (6)$$

where the ambient temperature is assumed to be  $T_A = 300$  K. For laser illumination, assumed to have a Gaussian profile centred at the illumination wavelength  $\lambda_0 = hc/E_0$ , the generated electron distribution is given by

$$\phi_{\text{el,laser}}(E) = A \exp\left(\frac{1}{2} \left(\frac{E - (E_0/2)}{2\sigma}\right)^2\right), \quad (7)$$

where  $A$  is the factor representing the intensity of the illumination, and  $\sigma$  the spectral width.

Since the resultant steady state cannot necessarily be described by a thermal distribution and temperature, the radiative emission is also nonthermal and cannot be described in terms of blackbody radiation from a single temperature. Instead, an energy-dependent brightness temperature  $T_{\text{Br}}$  is ascribed, defined as

$$\begin{aligned} f(E) &= \frac{1}{e^{\frac{E}{k_B T_{\text{Br}}(E)}} + 1} \\ \rightarrow k_B T_{\text{Br}}(E) &= \frac{E}{\ln\left(\frac{1}{f(E)} - 1\right)}. \end{aligned} \quad (8)$$

The brightness temperature at the given energy  $E$  is the temperature of a blackbody that emits the same photon flux at that energy. This is a common way of ascribing a temperature to nonthermal light sources in fields such as radio astronomy [40]. Therefore, the radiation emitted at a given energy is described by

$$\phi_{\text{rad}}(E) = \phi_{BB}(E_\gamma = 2E, T_{\text{Br}}(E)), \quad (9)$$

resulting in the luminescence spectrum of the system. The balance of the occupancy is hence given by

$$\begin{aligned} \text{DOS}(E) \left( \frac{\partial f(E)}{\partial t} + \frac{f(E) - f_{\text{FD}}(E)}{\tau_{cc}} \right) t &= \phi_{\text{el,Sun}}(E) \\ &+ \phi_{\text{el,amb}}(E) - \phi_{\text{el,rad}}(E) - \frac{J_{\text{out}}(E)}{q} + \frac{J_{\text{in}}(E)}{q}, \end{aligned} \quad (10)$$

where  $t$  is the thickness of the absorber, set to 100 nm, and  $J_{\text{out}}$  and  $J_{\text{in}}$  are the outgoing and incoming components of the current density, respectively. Current extraction through a thermionic barrier of height  $E_{\text{ext}}$  depends on the momentum of the carriers orthogonal to the barrier,  $k_z$ . By integrating over the three-dimensional (3D) crystal momentum space and selecting for carriers with energy  $E$ , the energy dependent  $J_{\text{out}}$  is obtained, giving

$$\begin{aligned} J_{\text{out}}(E) &= 2qf(E) \int d^3k \xi(k) v_z(k) \delta(E(k) - E) \\ &= \frac{qf(E)m_{\text{eff}}(E - E_{\text{ext}})}{2\hbar^3\pi^2}, \end{aligned} \quad (11)$$

where  $q$  is the elementary charge, and the effective mass  $m_{\text{eff}}$  is taken to be equal to the free electron mass for both electrons and holes. The transmission function  $\xi(k)$  is a step function

$$\begin{aligned} \xi(k) &= 1 \quad \forall k_z > \sqrt{\frac{2m(E - E_g)}{\hbar^2}} \\ \xi(k) &= 0 \quad \forall k_z \leq \sqrt{\frac{2m(E - E_g)}{\hbar^2}} \end{aligned} \quad (12)$$

$J_{\text{in}}$  is obtained by replacing  $f(E)$  with  $f_c(E)$ , the occupancy of the contact.  $f_c(E)$  is a Fermi-Dirac distribution with ambient temperature and quasi-Fermi-level set to  $V/2$ .  $J_{\text{in}}$  is therefore given by

$$J_{\text{in}}(E) = \frac{qf_c(E, T_A, V/2)m_{\text{eff}}(E - E_{\text{ext}})}{2\hbar^3\pi^2}. \quad (13)$$

Steady-state occupancies of the system were obtained by numerically solving Eq. (10) with  $\partial f(E)/\partial t = 0$ , which also included numerically solving for  $f_{\text{FD}}$ . By evaluating the net current at various values of  $V$ , current-voltage curves were also obtained, enabling assessment of device performance.

### III. RESULTS

#### A. Laser illumination

##### 1. No carrier extraction (open circuit)

We simulate the PL spectrum resulting from illumination by a Gaussian beam centred at  $E_0 = 2$  eV ( $\lambda_0 \approx 620$  nm) with a  $\sigma$  width of 0.01 eV.

We set the intensity parameter  $A$  such that the total excited carrier energy is similar to 1000-Sun illumination conditions. Figure 1(a) shows that for slow scattering, there is an accumulation of carriers at the excitation energy. This inefficient energy redistribution reshapes the entire PL spectrum, resulting in different slopes in the high-energy region. We choose a high-energy tail region of

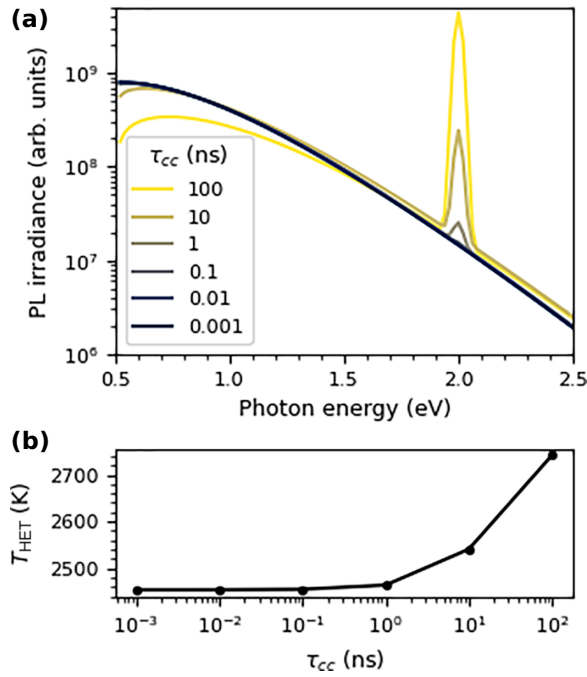


FIG. 1. (a) PL irradiance under 2-eV laser excitation for various  $\tau_{cc}$ , showing accumulation of carriers at the excitation energy for slow scattering. This distorts the temperature extracted via HET fitting, as seen in (b). The HET region chosen in this case was 1.4–1.6 eV.

1.4–1.6 eV and extract the carrier temperature by fitting to the Planck law.

Figure 1(b) shows that the altered slope due to slow scattering can result in deviations, in this case, an artificially high temperature. This shows that for a given illumination intensity, the PL spectrum and its HET fitting can yield significantly different temperatures depending on the  $\tau_{cc}$  of the system.

## 2. With carrier extraction

If a HCSC were to be used under monochromatic illumination, the accumulation of carriers due to inefficient scattering shown in Fig. 1(a) may result in an enhanced performance if all or the majority of the carriers are generated at the extraction energies. For laser excitation below the barrier height, however, scattering is needed to promote photogenerated carriers to higher energies and enable extraction.

To illustrate this effect, we use a 2-eV barrier and consider carriers generated below and above the barrier energy. By electron-hole symmetry, generating carriers at energy 2 eV requires a 4-eV laser excitation. We consider laser excitations detuned to 4.2 and 3.8 eV, which respectively generate carriers above and below the barrier height. As shown in Fig. 2(a), for 4.2-eV laser excitation, fast scattering results in a reduction in the extracted current. Furthermore, redistribution also increases the excess heat

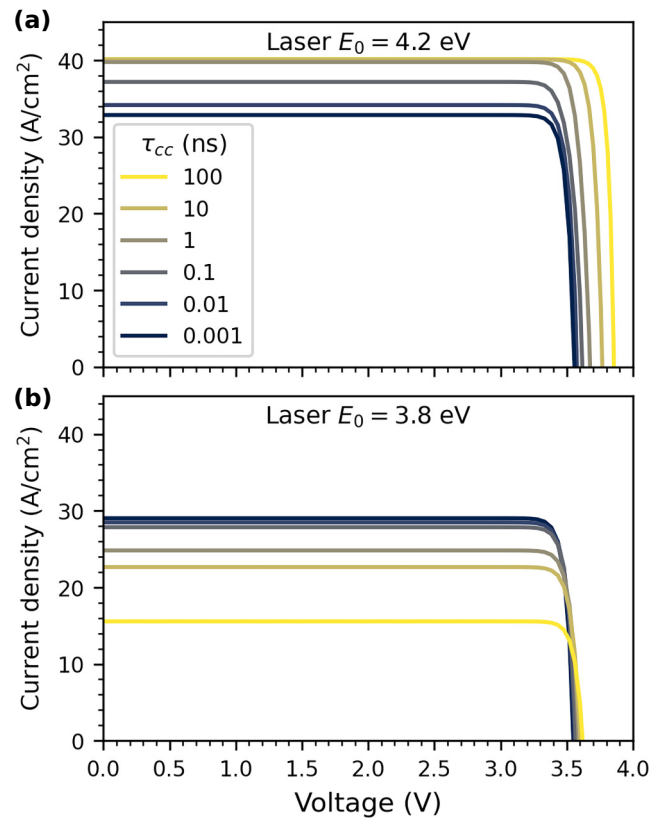


FIG. 2. Current-voltage curves with a 2-eV thermionic barrier under (a) 4.2 eV and (b) 3.8 eV laser illumination for various  $\tau_{cc}$ .

flow lost to the contacts, resulting in a reduction of the  $V_{OC}$ . The opposite trend is seen for 3.8 eV, where faster carrier equilibration dramatically improves current extraction, as shown in Fig. 2(b).

## B. Solar illumination

### 1. No carrier extraction (open circuit)

As mentioned before, we study the absorber and device operation under 1000-Sun illumination, since HCSC operation is expected to require solar concentration. To isolate the effect of a finite scattering rate, we first consider carrier populations in an absorber with no carrier extraction. The steady-state distributions simulated for various  $\tau_{cc}$  values are shown in Fig. 3.

For slow scattering rates, due to a low efficiency of carrier equilibration and energy redistribution, the carrier distribution corresponds to the initial generation profile of the carriers that results upon excitation from the Sun. Note that this generation profile is not a Fermi-Dirac distribution at temperature  $T_{\text{Sun}}$ , since each photon of energy  $E$  gives an electron of energy  $E/2$ . The total energy of the profile corresponds to a Fermi-Dirac distribution of temperature 2400 K, which is approached for fast scattering rates ( $\tau_{cc} < 1$  ns).

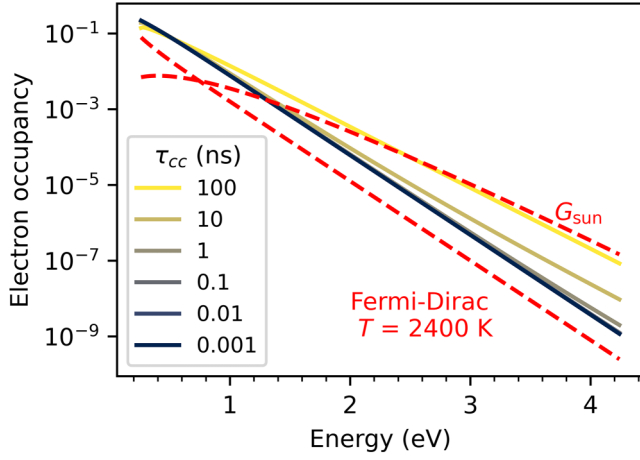


FIG. 3. Electron distribution functions under 1000-Sun illumination for various  $\tau_{cc}$  values. While a steady-state distribution of temperature 2400 K (shown with a dashed line, scaled) is seen for fast equilibration, for slow equilibration the distribution begins to approach the shape of the initial generation profile (shown with dashed line labelled  $G_{\text{sun}}$ , scaled).

## 2. With carrier extraction

To illustrate the effect of slow carrier equilibration on energy conversion, the HCSC device characteristics under solar illumination for a range of  $\tau_{cc}$  values were studied. We set the thermionic barrier height to 2 eV and focus primarily on the maximum power point (MPP).

Figure 4(a) shows that carriers above the barrier energy get depleted if  $\tau_{cc}$  is large, resulting in a distribution that deviates from Fermi-Dirac. The low-energy carriers cannot transfer their energy effectively, and their concentration builds up in the absorber. This high density of carriers leads to a larger recombination rate and energy loss via radiation. This ineffective energy redistribution and carrier extraction results in a drop in current density apparent in the  $J$ - $V$  curves in Fig. 4(b). It is worth comparing this result to Fig. 2, which showed that slow carrier equilibration may be advantageous for above-barrier carriers, but presents a significant decrease in extraction of below-barrier carriers. Since the Sun is a broadband source, generating a high density of low-energy carriers, fast equilibration is overall necessary for efficient energy conversion. The above points become clearer upon examination of energy flows for various  $\tau_{cc}$  values [see Fig. 5(a)]. Part of the energy associated with the current extracted from the device is converted to electrical power (blue), while the rest is lost as excess heat flow (green). Carriers that are not extracted lose their energy via radiative recombination (red). The energy flows show that carrier extraction drops and most of the carrier energy is lost via radiation as equilibration becomes inefficient. For very high  $\tau_{cc}$ , the efficiency drops below the 1000-Sun single junction limit for an absorber with the same band gap (shown in white). Figure 5(b)

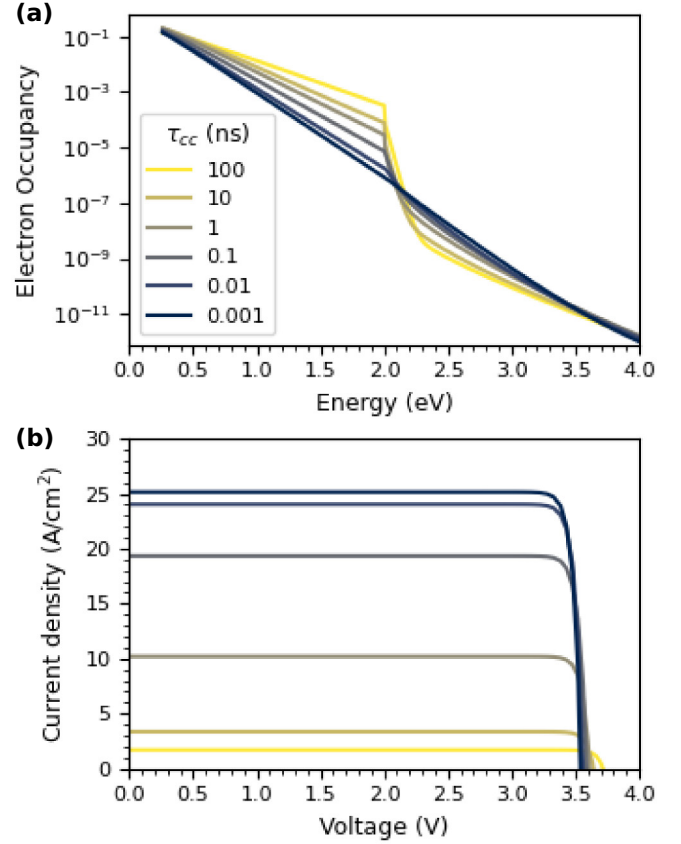


FIG. 4. (a) Carrier distributions at MPP under 1000-Sun illumination for various  $\tau_{cc}$ . (b) Current-voltage curves for the  $\tau_{cc}$  simulated, showing degradation in performance primarily through a lower current output.

shows that the Fermi-Dirac equivalent temperature  $T_{\text{FD}}$  of the carriers is also higher, indicating that there is increased energy within the distribution. Ineffective scattering therefore is shown to be a barrier to extraction, resulting in smaller current, analogous to what occurs when the barrier height is excessively high [33].

Inefficient equilibration therefore also favours lower extraction energies; the optimal barrier height reduces with increasing  $\tau_{cc}$ , as shown in Fig. 6. When carrier equilibration is slow, lower extraction energies are optimal as they can dramatically increase the current density of the device, compensating for the lower achievable  $V_{\text{OC}}$ . For fast equilibration, the optimal barrier approaches 1.6 eV, with an efficiency of approximately 60%—the known quasiequilibrium limit for such systems [33].

## IV. CONCLUSION

Our relaxation-time approximation model shows that steady-state nonthermal carrier distributions can result from inefficient carrier equilibration. Without carrier extraction, the carrier distributions were found to result

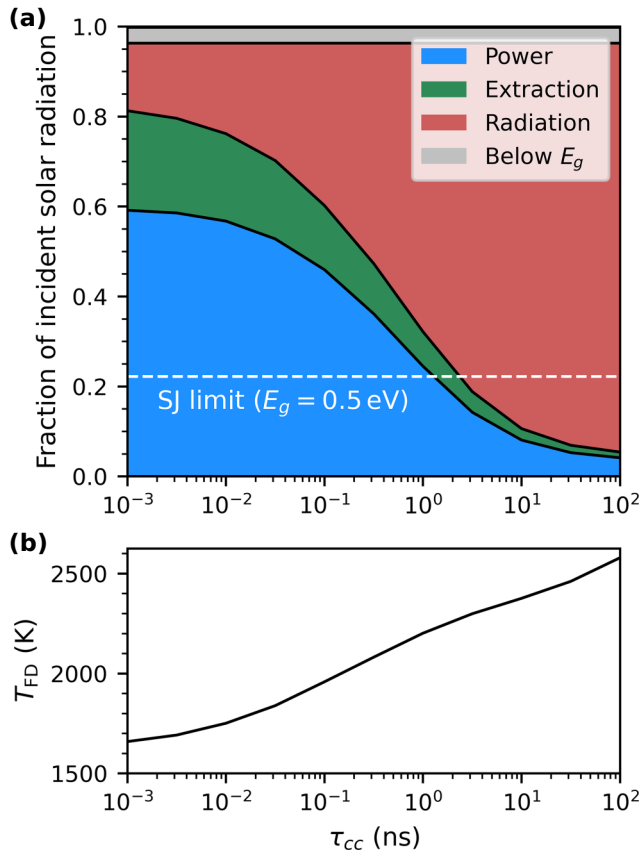


FIG. 5. (a) Energy-flow breakdown at MPP for various  $\tau_{cc}$  under 1000-Sun illumination, with barrier height fixed to 2 eV. The single junction detailed balance limit for the same band gap is shown as a dashed white line. A decrease in power output is seen with slower carrier equilibration, with increased energy being lost as radiation. (b) This corresponds to a higher  $T_{FD}$  as there is higher energy within the carrier population.

from a competition between the generation rate and carrier equilibration rates that reshape the carrier distribution. In the regime where these reshaping processes dominate, the quasiequilibrium assumption holds and the carriers follow a Fermi-Dirac distribution, but a deviation occurs otherwise. This may lead to errors in carrier temperatures extracted through photoluminescence spectra, since they typically assume an equilibrated carrier distribution. In the case of hot-carrier solar cells, carrier equilibration plays a key role by refilling carriers at energies where they are extracted via the energy-selective contacts. Under slow equilibration, carriers are depleted by extraction through the contacts and cannot be effectively replaced by scattering. This reduces the extracted current and therefore the efficiency of the device.

It must be noted that assigning a single timescale to a complex process is a simplification, and is not necessarily an accurate reflection of dynamics in real devices. For instance, carrier scattering is strongly dependent on carrier

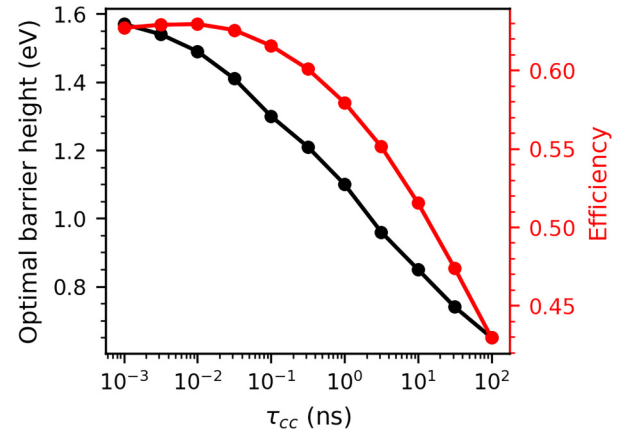


FIG. 6. Optimal barrier height (black) and the corresponding efficiency (red) for various  $\tau_{cc}$ . Efficient redistribution of energy within the carrier populations enables efficient extraction at high barrier heights.

concentration, and hence on the illumination intensity. In this work, it was modeled with a constant, concentration-independent rate and hence this effect was not taken into account. Electron-hole symmetry was also assumed for simplicity. The goal of this work was to explore the implications of slow intercarrier scattering rates relative to other timescales, and we expect our broad conclusions to apply to real, asymmetric systems. Our results highlight the general significance of considering carrier transport and scattering properties when evaluating and engineering candidate HCSC absorber materials, the investigation of which is limited in the literature [41,42]. Alongside slow carrier cooling, efficient carrier-carrier scattering under practical solar fluxes is a key material property required for efficient HCSC operation.

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## DATA AVAILABILITY

The data that support the findings of this article are available from the authors upon reasonable request.

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- Correction:* The previously published Fig. 4 contained an incorrect legend and has been replaced.