Photodetection by Hot Electrons or Hot Holes: A Comparable Study on Physics and Performances

Qingxin Sun,†‡§ Cheng Zhang,†‡§ Weijia Shao,†‡ and Xiaofeng Li*†‡§

Article

ABSTRACT: Hot-carrier photodetectors are drawing significant attention; nevertheless, current researches focus mostly on the hot-electron devices, which normally show low quantum efficiencies. In contrast, hot-hole photodetectors usually have lower barriers and can provide a wide spectral range of photodetection and an improved photoconversion efficiency. Here, we report a comparable study of the hot-electron and hot-hole photodetectors from both underlying physics and optoelectronic performance perspectives. Taking the typical Au/Si Schottky contact as an example, we find obvious differences in the energy band diagram and the consequent hot-carrier generation/transport/emission processes, leading to very distinguished photodetection performances. Compared with hot electrons, hot holes show higher density below the Fermi level, the longer mean free path arising under the lower electron–electron and electron–phonon scatterings, a lower barrier height, and a lighter effective mass in Si, all of which lead to larger number of high-energy hot holes, larger transport probability, higher emission efficiency, and higher photoresponsivity. However, the low barrier height can cause poor performances of hot-hole device in dark current density and detectivity. The study elucidates the intrinsic physical differences and compares the key performance parameters of the hot-hole and hot-electron photodetections, with the objective of providing complete information for designing hot-carrier devices.

INTRODUCTION

Recently, hot carriers generated from the nonradiative decay of surface plasmons have attracted increasing attention for wide applications in photodetecion, photovoltaics, photocatalysis, and surface imaging. In terms of photodetection, the internal photoemission process based on hot carriers enables convenient infrared photodetection, room-temperature and zero-bias operation, and high tunabilities on the working wavelength, bandwidth, and polarization dependence.

Due to the advantages of hot-carrier devices, increasing interest is given to hot-carrier photodetectors, especially those based on hot electrons. For example, Knight et al. successfully observed the hot-electron photoelectric response in the near-infrared band using the surface plasmon effect of Au nanoantenna arrays deposited on an n-type silicon substrate. Sobhani et al. constructed a periodic Au nanograting on n-type silicon substrate for hot-electron photodetector, achieving a strong and narrowband absorption, where the photoresponsivity and internal quantum efficiency are 0.6 mA/W and 0.2% at zero bias, respectively. Li et al. combined a metamaterial perfect absorber with hot-electron photodetector in which the metal used to generate the hot electrons can be thinner than the diffusion length of the hot electrons, resulting in the enhanced photoresponsivity up to 3 mA/W at 1300 nm. Wen et al. proposed a disordered ultrathin Au-covered n-type silicon nanohole (Au/SiNH) structure with an unbiased photoresponsivity of 1.5−13 mA/W at the wavelength range of 1100−1500 nm. Apparently, the photoresponsivity and quantum efficiency are still low, restricted by the severe thermalization losses of hot electrons during the transport process, the relatively high Schottky barriers, and the nonideal experimental fabrications. Besides, the higher Schottky barrier in the hot-electron device also determines that the detection cutoff wavelength is limited and hard to be extended to longer wavelengths. Considering that the Schottky barriers (φSB) formed by noble metals (e.g., Au, Ag, Cu, and Al) and p-type silicon are normally lower than those with n-type silicon, e.g., Au/p-Si is ~0.32 eV and Au/n-Si is ~0.8 eV. Due to the low barrier height, the dark current is large unless under the low-temperature operation; thus, the reports concerning the hot-hole photodetections are very rare. However, hot-hole device could have advantages in the other key performance parameters including the responsivity (Res.) and the detection cutoff wavelength (λcut-off). For example, the significantly enhanced responsivity of 1 A/W can be obtained at the relatively low biases of 275 mV in the hot-hole system of Au grating on p-Si substrate. Therefore, it is worth to study the hot-hole devices, as well as make a thorough comparison of the hot-electron and hot-hole
electrons (right) emission e below and above the Fermi level lie in the energy band bending, density of occupied states junction. Concerning the intrinsic physics, distinct di of the realistic hot-carrier device based on the Au/Si Schottky transport toward the metal include the light-excitation of hot carriers in metal, the The working processes of the hot-carrier photodetectors PHYSICAL PROCESS OF HOT CARRIERS ■ PHYSICAL PROCESS OF HOT CARRIERS The working processes of the hot-carrier photodetectors include the light-excitation of hot carriers in metal, the transport toward the metal–semiconductor (M/S) interface, and the emission over the Schottky barrier into semiconductors. The internal photoemission process plays the key role of below-band-gap photodetection, since the Schottky barrier height is normally lower than the band gap of semiconductor ($E_g$)\(^{30,31}\) moreover, the hot electrons and hot holes are emitted into various bands of semiconductors driven by the bended energy band. Therefore, it is of significance to comparably address the physics accounting for the energy band, hot-carrier generation, transport, and emission processes in hot-electron and hot-hole devices.

Energy Band Bending. Different semiconductors in contact with metal cause distinguished energy band bendings and junction barriers. For junction composed by metal and n-type semiconductor (hot-electron case), $\Phi_{SB} = W - \chi$ (Au/n-Si: 0.8 eV), whereas for that by metal and p-type semiconductor (hot-hole case), $\Phi_{SB} = E_g - (W - \chi)$ (Au/p-Si: 0.32 eV)\(^{32}\) where $W$ is the work function of metal and $\chi$ is the electron affinity of the crystal semiconductor. Due to the structural defects and the higher Schottky barrier [Au/n-type (p-type) amorphous silicon: 1.2 (0.52) eV]\(^{32}\) the amorphous silicon is less used in hot carrier photodetection. Figure 1a shows the energy band diagrams of typical hot-hole (left) and hot-electron (right) devices, which show that the hot holes and hot electrons are emitted into the valence band and conduction band of semiconductors, respectively. Such a specific energy band plays a primary role in forming the initial energy distributions of the respective hot carriers as well as the forthcoming carrier transport behaviors.

Hot-Carrier Generation. Electrons in the low-energy level of metal are raised to the higher by absorbing photon energy, leaving behind holes, i.e., the hot electrons and hot holes, respectively. However, the generated hot electrons and holes are not distributed uniformly in the energy spectrum and has to be carefully addressed. For the low-energy photons in the near-infrared band, the hot-carrier energy distribution can be calculated by the simplified electron distribution joint density of states: the product of the electron density of states (EDOS)
Figure 2. (a) Schematic diagram of the hot-carrier device with the normalized electric field at working wavelength. (b) Profile of the hot-carrier generation rate along the z-axis with the optical response inserted. Contour map of the hot-hole (left) and hot-electron (right) transport probability (c) and the flux (d) of hot carriers with energy E reaching the Schottky interface at position z.

at the initial and the final electron energies. For the generation rate in the excitation process, the assumption of an absorbed photon generates only one hot electron–hole pair is reasonable for the low-energy photons and the resistive loss arising from the finite carrier life time is also considered. With the EDOS, incident photon energy (incident light of λ = 1200 nm), Fermi distribution function, Figure 1b shows the asymmetric initial energy distributions relative to $E_f$ of hot holes (left) and hot electrons (right). The detailed calculation can be found in Section 2 of the Supporting Information. The shaded areas indicate the proportion of the hot carriers with energy higher than $\varphi_{SB}$ (for Au/p-Si: $\varphi_{SB} = 0.32$ eV; for Au/n-Si: $\varphi_{SB} = 0.8$ eV). Although the number of the generated hot holes and hot electrons are the same, the energy of hot holes with peaked $D$ and the proportion of hot holes with energy over $\varphi_{SB}$ are larger than those of hot electrons. This is because most electrons occupy the states with energies below $E_F$ whereas holes left after photoexcitation occupy preferentially high energy levels relative to $E_F$ as shown in Figure 1b. The above analysis implies that there are more hot holes that can overcome the barrier for photocurrent detection.

Hot-Carrier Transport. In the hot-carrier transport process, only if hot-carrier lifetime is long enough or the MFP is greater than the distance from the position of hot-carrier generation to the M/S interface, hot carriers can arrive at the interface for collection. Employing the calculation method of MFP reported in ref 37, energy-dependent MFP is shown in Figure 1c, which indicates that the MFPs of hot holes are larger (less) than that of hot electrons when hot-carrier excess energy relative to the Fermi level ($E_e = E - E_F$) $< (>) \sim 1.2$ eV due to the differences in the energy losses of electron–electron and electron–phonon scatterings for electrons and holes in Au. Typically, the MFP of hot holes (electrons) with excess energy of $\varphi_{SB}$ is $\sim 47$ (22) nm, i.e., the hot holes have a higher probability to reach Schottky interface than that of hot electrons before been dissipated via thermalization. In the case of metal nanoparticles (NPs), the effect of the MFP is ignorable for the hot carriers generated at metal NPs/semiconductor interface. However, for the hot carriers generated away from the nanoparticle surface, they could undergo electron–electron and electron–phonon scatterings and be thermalized before injecting into semiconductor.

Hot-Carrier Emission. Only if the kinetic energy associated with momentum normal component of hot carriers reaching the M/S interface are greater than $\varphi_{SB}$ that hot carriers can emit into semiconductor and be collected as photocurrent for detection. Figure 1d compares the emission efficiency of hot holes (left) and hot electrons (right) reaching the M/S interface as a function of hot-carrier energy without considering the interface carrier reflection ($\eta_{w/o_ref}$). It is found that the emission efficiency of hot holes is much larger than that of hot electrons due to the lower barrier height of hot holes than that of hot electrons. With considering the hot-carrier reflection, the efficiencies ($\eta_{w_ref}$) of hot holes and hot electrons will both be reduced, since some hot carriers in the allowed momentum space will be reflected back into the metal. Moreover, the emission efficiency of hot holes is decreased in a more significant way (compared to that of hot electrons) due to the special momentum distribution and reflection determined by the different effective masses of holes and electrons in Si. Despite the emission efficiency of hot holes is still higher than that of hot electrons, e.g., for hot holes and hot electrons with $E_e = 1.5$ eV, $\eta_{w_ref} = 0.16$ and 0.11, respectively, which implies that more hot holes can be successfully injected into semiconductor to cause the higher photocurrent and photoresponsivity.

The above analysis indicates that the hot-electron and hot-hole photoconversion devices indeed show fundamentally distinguished physics, which regulate the operations of the
devices. In the next section, we further propose a realistic device to explore the differences in the optoelectronic performances of hot-hole and hot-electron photodetectors.

![Figure 3. Contour maps of the hot-hole (left) and hot-electron (right) flux accumulated at the Schottky interface (a) and the emission efficiency (b) as a function of energy $E$ and diffusion angle $\theta$. (c) The hot-hole (left) and hot-electron (right) emission efficiency with the consideration of anisotropic momentum distribution as a function of energy $E$. (d) Energy collection and loss distribution in the hot-hole (top) and hot-electron (bottom) devices.](image)

**REALISTIC HOT-ELECTRON AND HOT-HOLE DEVICES**

For a more comprehensive study on comparing the hot-electron and hot-hole devices, we now investigate a realistic planar Tamm system, which can be modulated to collect the energy by hot electrons or hot holes.\(^{17-19}\) Recently, Tamm plasmon has been studied in the hot-carrier-based photodetection\(^ {20}\) and organic narrowband near-infrared photodetectors.\(^ {20}\) The system is under planar Tamm configuration composed of three pairs of alternating SiO$_2$/Si under distributed Bragger reflector (DBR) design on an Au layer ($d_{Au} = 200$ nm), as shown in Figure 2a. The refractive index of Si (SiO$_2$) is 3.52 (1.62) for the design of DBR with the central wavelength of $\lambda_{DBR} = 1057$ nm. At the Tamm plasmon resonance of $\lambda_{tamm} = 1200$ nm, the electric field is highly confined near the Schottky interface and decays exponentially toward the Au layer with a narrow reflection dip and perfect absorption (inset of Figure 2b). The spatial distribution of the hot-carrier generation rate ($G$) is shown in Figure 2b, which shows that most of the hot carriers are generated in the region close to the Schottky interface with $\delta_0 = 12$ nm (i.e., the distance at which the generation rate drops to $1/e$ of the maximum). The calculation of absorption efficiency ($A$) and detailed derivation of the hot-carrier generation rate $G(z, \alpha)$ can be found in Section 1 of the Supporting Information.\(^ {37}\) According to the physical process of hot-carrier generation, a pair of hot electron and hole is generated upon absorbing a photon, so the generation rate of hot electrons and hot holes are identical in the device.

After photoexcitation, to better understand the difference in hot-hole and hot-electron transport processes, Figure 2c compares the spatial distributions of the hot-hole (left) and hot-electron (right) transport probabilities, $P_{trans}(z, E)$, defined by eq S5.2 in the Supporting Information. Here, the hot-carrier thermalization loss is described by the exponential attenuation model, i.e., $\exp(-d/l_{MFP})$, which indicates that the thermalization loss is determined by the transport distance ($d$) and MFP. The exponential attenuation model can be employed in the system of metal NPs or other complex metal nanostructures. Since the MFP of hot holes is larger than that of hot electrons for $E_c < \sim 1.2$ eV, hot holes have a higher probability to reach the Schottky interface than hot electrons at the same position of the device. Accordingly, Figure 2d shows that the flux $N_{int}(z, E)$ [defined by eq S6.2 in the Supporting Information] of hot holes with high energy greater than $\phi_{SB}$ is much larger than that of hot electrons and the energy distribution of hot holes accumulated at the Schottky interface is more uniform than that of hot electrons, regulated by the specific initial energy distribution and transport probability. Besides, from the hot-carrier overall transport efficiency $\eta_{trans}$ defined by eq S7 in the Supporting Information, the efficiencies of hot holes and hot electrons are 30.59 and 28.73%, respectively, showing the advantage of hot holes in the hot-carrier transport process. For metals with a short mean free path (i.e., Ni with $l_{MFP} \sim 6$ nm\(^{-1}\)), it is not appropriate for the hot-carrier photocconversion.

Since the diffusion distance and the momentum distribution of carriers along various diffusion angles are different, the influences of the diffusion angle $\theta$ on the transport and emission processes should be considered with particularly addressing the differences between hot-electron and hot-hole devices. Figure 3a compares the angular flux distributions of hot holes (left) and hot electrons (right) reaching the interface. It is found that most hot carriers are concentrated...
in a range with small diffusion angles, since the propagation path is larger under a large diffusion angle, leading to more hot carriers from thermalization. Moreover, the angular distribution of hot holes is relatively uniform compared to that of hot electrons, with a broader-band distribution of the hot holes, due to differences in the initial energy distribution and transport probability. The calculation process can be found in Section 2 of the Supporting Information.

Figure 3b plots the emission efficiencies ($\eta_{emi}$) of hot holes (left) and hot electrons (right) versus $\theta$ and $E - E_c$. Due to the lower barrier height of the hot-hole system, the emission efficiency of the hot-hole device is much larger than that of the hot-electron counterpart. In addition, considering the requirement of the kinetic energy and momentum for hot-carrier interfacial transfer, there exists a critical angle. Over the critical angle, the hot carriers cannot transfer across the barrier into the semiconductor, i.e., the emission efficiency is 0. From the greatly differed effective masses of electrons and holes in Si and the barrier heights of hot-electron and hot-hole junctions, it is reasonable that the critical angles of hot holes are larger than that of hot electrons, so more hot holes can cross over the barrier compared to that of hot electrons. Taking $E_c = 1.2$ eV as an example, the critical angle of hot holes is $\sim 48^\circ$, which is larger than that ($\sim 35^\circ$) of hot electrons.

Figure 3c shows the emission efficiencies of hot holes (left) and hot electrons (right) with the anisotropic momentum distribution at the interface. It is clear that $\eta_{emi}$ of the hot hole is further greater than that of hot electrons. The detailed calculation can be found in Section 3 of the Supporting Information.

| Table 1. Performance Comparison of the Proposed Hot-Hole, Hot-Electron, and Other Reported Devices Based on the Same Working Principle$^a$ |
|---|
| device structure | responsivity | dark current/dark current density | detectivity | refs |
| planar Au/n-Si | 1.72 mA/W@1200 nm | $3.56 \times 10^{-7}$ A/cm$^2$ | $5.095 \times 10^7$ Jones | this work |
| planar Au/p-Si | 27.49 mA/W@1200 nm | $11.32$ A/cm$^2$ | $1.444 \times 10^7$ Jones | this work |
| Au nanoislands/n-Si | 30 mA/W@1064 nm | $10^{-7}$ A | N/A | 20 |
| Au grating/p-Si | 3.5 mA/W@1200 nm | $5 \times 10^{-3}$ A/cm$^2$ | $8.745 \times 10^7$ Jones (cal.) | 21 |
| Au stripe/p-Si | 13 mA/W@1550 nm | $11.32$ A/cm$^2$ (cal.) | $6.826 \times 10^7$ Jones (cal.) | 22 |
| GaAs nanocone/MLG | 12 mA/W@1550 nm | $30$ A/cm$^2$ (1.5 $\times 10^{-7}$ A) | $3.871 \times 10^7$ Jones (cal.) | 24 |
| graphene/Si | 1.73 mA/W@850 nm | $\sim 10^{-6}$ A | $1.83 \times 10^{11}$ Jones | 44 |
| graphene/Si | 29 mA/W@850 nm | $\sim 10^{-8}$ A | $3.9 \times 10^{10}$ Jones | 45 |

$^a$cal. indicates the result of substituting the relevant data in the reference into the formula used in this paper.
Table 2. Summary of Main Physical Differences and Consequent Results between Hot-Hole and Hot-Electron Devices

| Energy band bending | Carrier generation | Carrier transport | Carrier emission | EQE/Res. | Results | Results
|---------------------|--------------------|-------------------|------------------|----------|---------|---------|
|                     | More high-energy hot holes | More hot holes can cross over $\varphi_{SB}$ | Longer MFP → Larger transport probability | Lower $\varphi_{SB}$ → Larger injection efficiency | Larger, 2.84% / 27.49 mA/W | $\lambda_{cut-off}$ Longer, 3.875 μm |
| Hot-hole devices    | Less high-energy hot electrons | Less hot electrons can cross over $\varphi_{SB}$ | Shorter MFP → Smaller transport probability | Higher $\varphi_{SB}$ → Smaller injection efficiency | Smaller, 0.18% / 1.72 mA/W | $J_D$ Higher, 11.32 A/cm² |
|                     |                    |                   |                   | $D^*$ Lower, 1.444 $\times$ 10$^7$ Jones | $D^*$ Higher, 5.095 $\times$ 10$^9$ Jones |
| Hot-electron devices|                    |                   |                   |          |         |         |

momentum loss in hot-hole device is greater than that in hot-electron device due to different momentum distributions. As a result, the collection efficiency of hot holes is larger than that of hot electrons, leading to a higher responsivity and EQE.

According to the above analysis, the hot-hole device has advantages in the responsivity and EQE due to the lower barrier height and larger MFP. However, besides the responsivity and EQE, the dark current density and detectivity are also the key performance parameters of photodetectors. The dark current density under a weak bias can be obtained from the thermionic emission theory:

$$J_D = A^{**}T^2 \exp\left(-\frac{q\varphi_{SB}}{k_B T}\right)$$

where $A^{**}$ is the effective Richardson constant ($\sim 110$ A/(cm$^2$ K$^2$) for electrons in n-Si and $\sim 30$ A/(cm$^2$ K$^2$) for holes in p-Si), $T$ is the room temperature (300 K), $q$ is the elemental charge, and $k_B$ is the Boltzmann constant. Detectivity describes the ability of distinguishing weak light signals from the noise and can be written as $D^* = \text{Res.} / \sqrt{2 \alpha J_D}$ if the shot noise from the dark current dominates all noises. It is found that $J_D$ and $D^*$ of the realistic device with Au/n-Si [p-Si] are $3.56 \times 10^{-7}$ A/cm² and 5.095 $\times$ 10$^9$ cm Hz$^{1/2}$/W [Jones] [11.32 A/cm² and 1.444 $\times$ 10$^7$ cm Hz$^{1/2}$/W [Jones]], respectively. Due to negative exponential correlation between $J_D$ and $\varphi_{SB}$, although the hot-hole device has a $\varphi_{SB} = 0.32$ eV, which is only 2/5 of that of the hot-electron device (0.8 eV), $J_D$ is 8 orders of magnitude larger. Thus, although the Res. of hot-hole device (27.49 mA/W) is 16 times greater than that of hot-electron device (1.72 mA/W), $D^*$ by hot-hole strategies is 2 orders of magnitude smaller.

Here, we make a comprehensive comparison of the responsivity, dark current density, and detectivity between the studied hot-electron, hot-hole, and other reported devices based on the same working principle in Table 1.20–22,24,44,45 It is found that the Res., $J_D$ and $D^*$ of the proposed hot-electron and hot-hole device are comparable to the reported values based on the similar M/S junction. Indeed, the detectivity is lower compared to that of the photodetector based on two-dimensional material.

The above study compares the differences of the photodetector performance at the fixed barrier height and the working wavelength. Since the Schottky barrier can be modified by engineering the interface and the resonant wavelength can be regulated by changing the structural parameters, we further compare the performances under various barrier heights and resonant wavelengths in the hot-hole and hot-electron devices as shown in Figure 4a–e. It is well known that the sum of the barrier height of the hot-electron and hot-hole devices is the band gap of the semiconductor; therefore, with the $\varphi_{SB}$ of Au/n-Si device being modified from 0.8 to 0.5 eV, $\varphi_{SB}$ of Au/p-Si device will be changed accordingly from 0.32 to 0.62 eV.43 Due to the negative exponential correlation between $J_D$ and $\varphi_{SB}$ and the decreased proportion of hot carriers with energy over $\varphi_{SB}$, $J_D$ and Res. will be decreased with the increase of $\varphi_{SB}$ (fixed $\lambda$), as shown in Figure 4a–c. Hence, in Figure 4d,e, $D^*$ exhibits a much more complicated dependence on $\varphi_{SB}$; therefore, we have to carefully design the hot-hole and hot-electron devices for a larger $D^*$. Similarly, since the absorption efficiency, hot-carrier energy distribution, transport efficiency, and injection efficiency change with incident photon energy, with the increase of the resonant wavelength (fixed $\varphi_{SB}$), the generated hot carriers with energy over $\varphi_{SB}$ and the emission efficiency in the interfacial transfer process are decreased, leading to the decreased Res. and $D^*$. Especially, when the resonant wavelength is greater than the cutoff wavelength, Res. and $D^*$ are both reduced to 0, i.e., the device fails to work. Particularly, the cutoff wavelength $\lambda_{cut-off}$ (3.875 μm) of the hot-hole device with $\varphi_{SB}$ of 0.32 eV is much longer than that (1.55 μm) of the hot-electron device with $\varphi_{SB}$ of 0.8 eV due to the lower barrier height, showing the advantage/ability of the
hot-hole device in long-wavelength photodetection. Considering the same Schottky barrier height in the range of 0.5–0.62 eV, due to different the momentum loss in the interfacial carrier transport process and the effective Richardson constant, the Res. and \( I_h \) of the hot-electron device are larger than those of the hot-hole device. As a result, \( D^* \) of the hot-electron device is less than that of the hot-hole device, showing the benefit of the collection of hot holes.

## CONCLUSIONS

In summary, based on the modified classical phenomenon model, we made a detailed comparison of the energy band bending, initial energy distribution, transport probability, and interfacial emission efficiency in the hot-hole and hot-electron generation, transport, and emission processes. The foremost physical differences and consequent effects are summarized in Table 2. In total, the hot-hole devices with lower barrier heights have advantages in responsivity and long-wavelength detection, whereas the hot-electron systems with higher barrier heights have advantages in dark current density and detection. The comparable study of the hot-hole and hot-electron systems presents the detailed devices physics as well as elucidates the performance differences for an optimal design of hot-carrier photodetectors.

## ASSOCIATED CONTENT

2 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.9b00267.

Calculation of hot-carrier generation, energy distribution, flux of hot carriers reaching the M/S interface and injection efficiency into semiconductor (PDF)

## AUTHOR INFORMATION

Corresponding Author

*E-mail: xflh@suda.edu.cn.

ORCID

Xiaofeng Li: 0000-0002-4115-3287

Author Contributions

\(^3\)Q.S. and C.Z. contributed equally to this work.

Author Contributions

Q.S., C.Z., and W.S. carried out the design and numerical simulation. X.L. conceived the design and supervised the research. All authors analyzed the data and wrote the manuscript. All authors have read and approved the final manuscript.

Notes

The authors declare no competing financial interest.

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