Electronic Supplementary Material (ESI) for Materials Horizons.

Supporting information

Magnetic fields affect hot electrons in silicon-based photodetectors at telecommunication wavelengths

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Characterization of barrier height of the fabricated Ni/n-Si DTTM device

In order to estimate the barrier height of the Ni/n-Si DTTM device, herein, we used the modified Fowler theory [1] to estimate the barrier height of the device. According to modified Fowler's theory, the quantum efficiency (η_i) should comply with the following relationship:

$$\eta_i \approx C_F \frac{(h\nu - q\phi_b)^n}{h\nu} \tag{1}$$

where C_F is the Fowler emission coefficient, $h\nu$ is the photon energy, n=2 for most metals, and $q\phi_b$ is the Schottky barrier height. This equation describes the number of carriers with enough energy to overcome the barrier height and, thereby, contribute to the photocurrent. In general, the spectral response of an active antenna-based device can be considered as that of a Schottky diode in the absence of a specific plasmon resonance. The responsivity depends only on the quantum efficiency (η_i) of the photoemission process, which can be approximated by the modified Fowler equation. To investigate the quantum transmission probabilities, according to equation (1), we calculated the quantum transmission probability-wavelength diagrams for the Schottky devices having various barrier heights. Next, when the Schottky barrier is formed by an active antenna rather than a continuous film, the responsivity of active antenna-based devices will show a Fowler equation modified by the plasmon absorption spectrum [1],

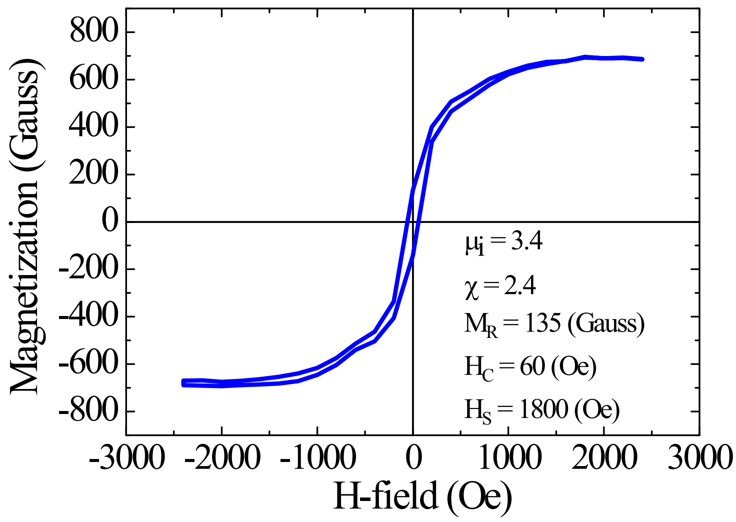
$$R(\lambda) = \eta_i S(\lambda) \tag{2}$$

where the R(λ) is the spectral responsivity, and the S(λ) is the absorption spectrum. With the extended Fowler relation, we can fit the experimental responsivities [Figure 2(b) in manuscript] with equation (2), using a known absorption spectrum for S(λ) [Figure 1(c) in manuscript]. Therefore, we can estimate that the barrier height of the Ni/nSi DTTM device (H065P13) is in the range of 0.40 and 0.45 eV.

Magnetic property of DTTM-based Ni–Si device

We used a superconducting quantum interference device (SQUID) to measure the magnetic hysteresis curve of the DTTM-based Ni–Si device featuring a 10-nm Ni film. **Supplementary Figure S1** reveals the behavior of the DTTM-based Ni–Si device graphically; the relationship between the magnetization and the external magnetic field was non-linear. The DTTM-based Ni–Si device exhibited magnetic hysteresis, with initial relative permeability and initial susceptibility of 3.4 and 2.4, respectively. The initial susceptibility is the susceptibility obtained from the tangent slope at or near zero Oersted. The initial relative permeability is the initial susceptibility plus one; its value for a non-magnetic (or paramagnetic) materials trends toward 1. In **Supplementary Figure S1**, all the domains in the Ni crystal were aligned (magnetic saturation) when the magnetic field strength reached 1800 Oe. When the magnetic field was not longer applied to the sample, the magnetization did not completely decrease to zero, due to the interactions between the dipole moments; the residual magnetism (M_R) was 135 Gauss. To remove the residual magnetism, we required a coercive force—the reversing magnetic field necessary for demagnetization—of 60 Oe. Therefore, we confirmed that the DTTM-based Ni–Si device featuring even a very thin (10 nm) Ni film retained a soft ferromagnetic property, as evidenced by the narrow magnetic hysteresis loop in

Supplementary Figure S1; thus, the hot electrons in the DTTM-based Ni–Si device would experience a stronger Lorentz force in Si than those in the DTTM-based Au–Si device, resulting in a more substantial decrease in responsivity and a more obvious difference in IQE variation between the parallel and perpendicular cases for the DTTM-based Ni–Si device.

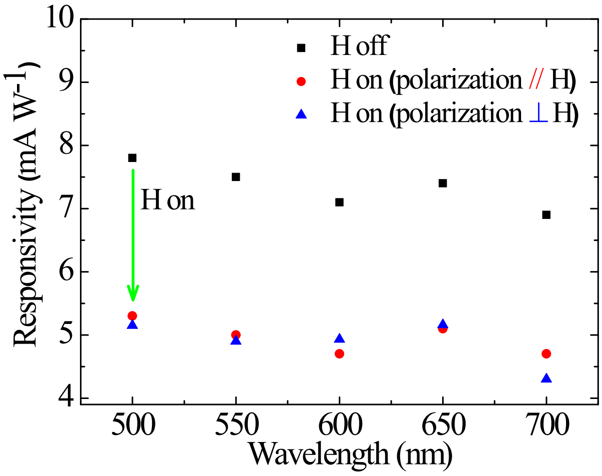


Supplementary Figure S1. Hysteresis curve of the Ni-H065P13 device.

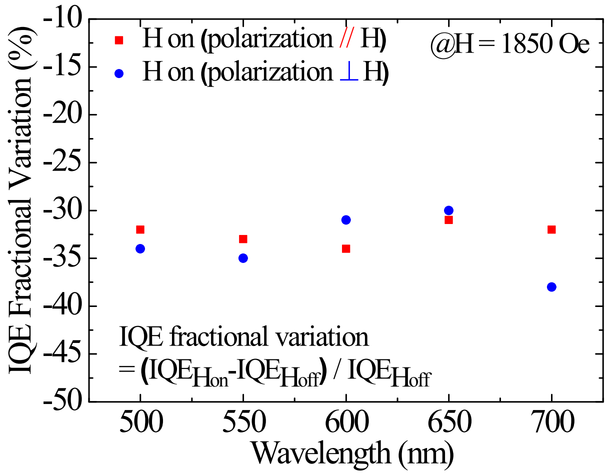
The behavior of the DTTM-based device in visible light regime

Taking advantage of the fact that the photocurrent in the optical telecommunication regime (photon energies below the band gap of Si) could be generated only by hot electrons in the metal, we could further modulate the performance of the Si-based photodetector by applying an external magnetic field. In contrast, in visible light regime (photon energies above the band gap of Si), the photocurrent was dominated by the electron/hole pairs generated in Si, with the hot electrons in the metal contributing in only a very minor manner to the photocurrent. The photogenerated excitons in Si were separated by the built-in electric field created at the metal–Si interface, with the electrons and holes transported through the Si and metal, respectively, to the electrodes to further generate photocurrent. In contrast to the hot electrons, excitons would not be driven by the electric field of the incident light; in other words, there would be no difference in directionality between the cases of parallel and perpendicular polarization. Therefore, we would not be able to modulate the behavior of excitons by applying an external magnetic field, unlike like the situation for the hot electrons. **Supplementary Figure S2** displays the responsivity of the DTTM-based Ni–Si device operated in the visible regime before and after applying an external magnetic field of 1850 Oe. For both the parallel and perpendicular cases, the decreases in responsivity were caused by the charged particles (electrons and holes) experiencing the Lorentz force in Si after being separated by the built-in electric field. In contrast to the situation in Figure 4a, we observed no significant difference in the decrease in responsivity between the parallel and perpendicular cases when applying a magnetic field to the device. For example, the responsivity of the DTTM-based Ni–Si (H065P13) device was 7.8 mA W^{-1} at a wavelength of 500 nm in the absence of a magnetic field; after applying a magnetic field, the responsivity decreased to 5.3 and 5.15 mA W^{-1} for the parallel and perpendicular cases, respectively. **Supplementary Figure S2b** reveals that the corresponding IQE fractional variations were –32 and –34% for the parallel and perpendicular cases, respectively. Furthermore, Figures **S2c** and **S2d** present the responsivities and corresponding IQE fractional variations, respectively, of the Au–Si based DTTM device in the visible regime before and after applying an external magnetic field of 1850 Oe. Similar to the behavior of the Ni–Si based DTTM device, there were no significant differences in the decrease in responsivity in the visible regime between the parallel and perpendicular cases. From these results, we confirm that the difference in responsivity in the NIR regime when applying an external magnetic field between the parallel and perpendicular cases arose from the effect of hot electrons. In other words, we have obtained evidence that the phenomena described in Figures 4, 5, and 6 originated from the behavior of hot electrons; thus, we have verified our proposal regarding the initial directionality of hot electrons being dominated by the polarization of the incident light.

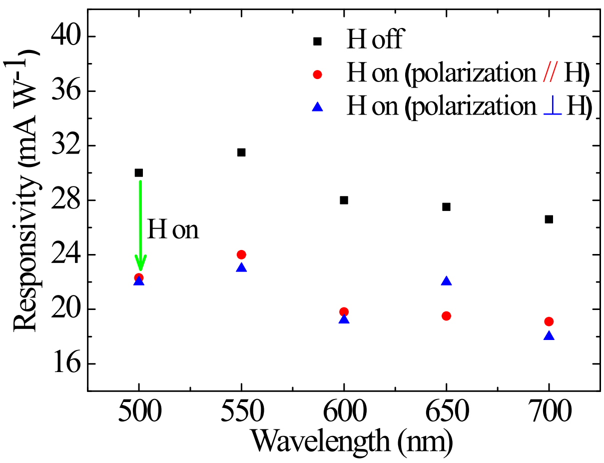
(a)



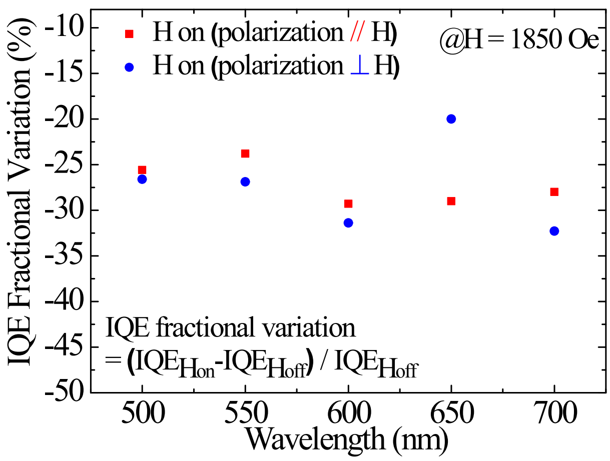
(b)



(c)



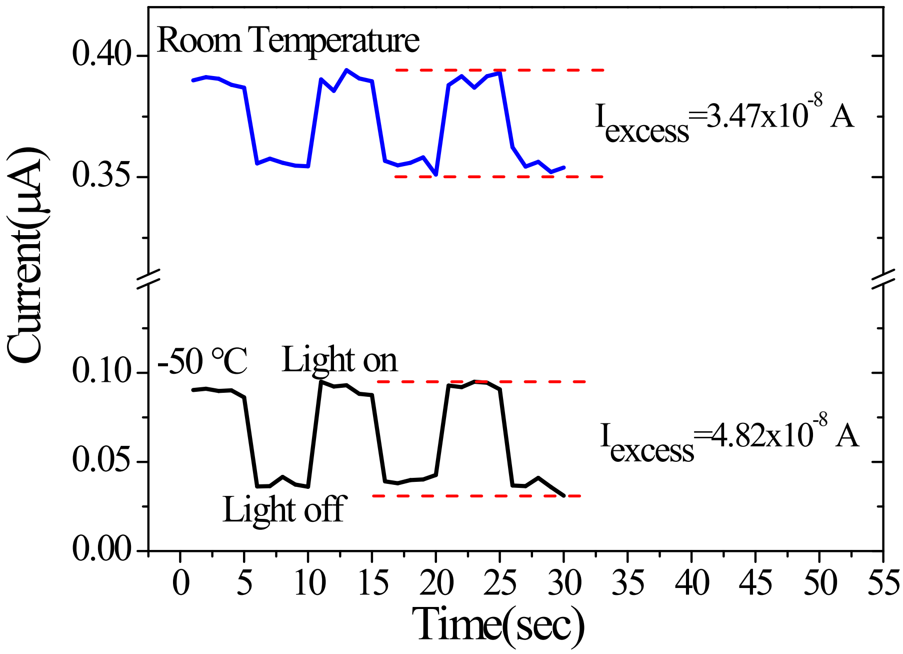
(d)



Supplementary Figure S2. (a, c) Responsivity spectra (visible regime) of (a) Ni-H065P13 and (c) Au-H065P13 devices; black squares represent the responsivities in the absence of an applied external magnetic field; red circles and blue triangles represent responsivities when an external magnetic field of 1850 Oe was applied to the devices in the parallel and perpendicular cases, respectively. (b, d) Correlation between IQE fractional variation and wavelength for the (b) Ni-H065P13 and (d) Au-H065P13 devices.

Elimination of thermal effects on DTTM Ni–Si device

In general, the increase of current in a Schottky-based device at high temperature is related to the thermal generated carriers (thermal electrons), which are generally increased with temperature. Herein, we measured the current of the DTTM device at low temperature (e.g. -50°C) to reduce thermal impact on the device. As shown in the **Supplementary Figure S3**, the measured short circuit current (I_{SC}) of the Ni/n-Si DTTM device in the dark and under the illumination of near IR light having a wavelength of 1300 nm with power density of 1.74 μW/mm^2 at room temperature (25°C) and -50°C, respectively. We found that the DTTM device exhibits significant photoresponse at -50°C; furthermore, the measured excess current (I_{excess}) of the device at -50 °C is even larger than that at room temperature. We attribute the results to the increasing of mean free path for hot electrons in Ni film at lower temperature [2]. Therefore, we suggest the photoresponse of the DTTM device is the generation of hot electrons but not thermal electrons.

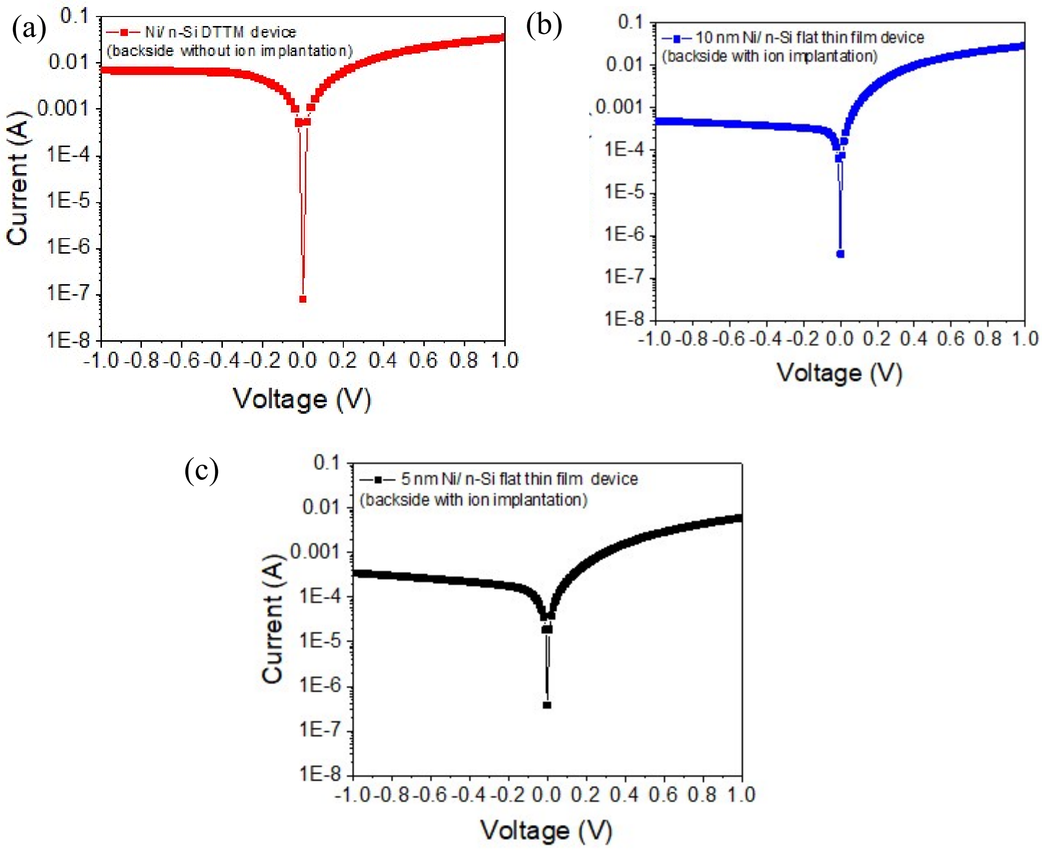


Supplementary Figure S3. Measured short circuit current of the Ni/n-Si DTTM device in the dark and under the illumination of near IR light having a wavelength of 1300 nm with power density of 1.74 $\mu W/mm^2$ at room temperature (25°C) and -50°C, respectively.

Effect of backside contact of the Ni/n-Si Schottky devices

The rectification properties of a Schottky-based device are affected by many factors, such as the quality of metal-semiconductor interfaces and ohmic-contact electrode on the backside of semiconductor. Herein, we found that the main reason behind the low rectification ratio of the Ni/n-Si DTTM device may be the fact that the backside electrical contact between the Ti thin film and the rough backside of n-Si substrate is not an ideal ohmic contact. The current of Ni/n-Si DTTM device in the reverse bias region is quite high, which implying the existence of Schottky contact between Ti film and the rough backside of n-Si substrate. We suppose that the Ni/n-Si DTTM device might be a kind of back-to-back Schottky device [3-4], which is composed of two Schottky contacts in series.

To further investigate this issue, we prepared the n-type Si substrates, whose doping level was approximately 10^{15} cm^{-3}, with heavily doped (phosphorus with dose of 10^{15}/ cm) on the backside of Si substrate, then we deposited flat thin Ni and Ti films as the front side and backside electrodes, respectively. As displayed in the **Supplementary Figure S4**, we measured the *I-V* characteristics of the Ni/ n-Si DTTM device without ion implantation and the flat Ni film/n-Si devices having different thicknesses of Ni films (5 and 10 nm) with ion implantation on the backside. We found both the flat Ni/n-Si devices perform much better rectification behaviors than the Ni/n-Si DTTM device. Furthermore, the flat Ni/n-Si device with 10-nm Ni exhibits the best rectification properties among all of the devices. Therefore, the results imply that the low rectification ratio of the Ni/n-Si DTTM device could be mainly attributed to the non-ideal backside electrode contact (Ti/n-Si) rather than the thickness of Ni film or the low Schottky barrier height between the Ni film and n-type Si substrate.



Supplementary Figure S4. *I-V* characteristics of (a) Ni/n-Si DTTM device without ion implantation on the backside and (b, c) flat Ni/n-Si devices having thicknesses of Ni film of (b) 10 and (c) 5 nm and with ion implantation process on the backside. All devices were measured at room temperature and in the dark.

Reference

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