Research article

Mel F. Hainey, Jr.*, Takaaki Mano, Takeshi Kasaya, Tetsuyuki Ochiai, Hirotaka Osato, Kazuhiro Watanabe, Yoshimasa Sugimoto, Takuya Kawazu, Yukinaga Arai, Akitsu Shigetou and Hideki T. Miyazaki*

Near-field resonant photon sorting applied: dual-band metasurface quantum well infrared photodetectors for gas sensing

<https://doi.org/10.1515/nanoph-2020-0456>

Received August 6, 2020; accepted September 17, 2020;

published online October 8, 2020

Abstract: Two photodetectors for measuring transmission and two bulky, separated narrowband filters for picking a target gas absorption line and a non-absorbing reference from broadband emission are typically required for dual-band non-dispersive infrared (NDIR) gas sensing. Metal–dielectric–metal (MDM) metasurface plasmon cavities, precisely controllable narrowband absorbers, suggest a next-generation, nanophotonic approach. Here, we demonstrate a dual-band MDM cavity detector that consolidates the function of two detectors and two filters into a single device by employing resonant photon sorting—a function unique to metasurfaces. Two MDM cavities sandwiching a quantum well infrared photodetector (QWIP) with distinct resonance wavelengths are alternately arranged in a subwavelength period. The large absorption cross section of the cavities ensures ~95% efficient lateral sorting of photons by wavelength into the corresponding detector within a near-field region. The flow of incident photons is thus converted into two independent photocurrents for dual-band detection. Our dual-band photodetectors show competitive external quantum efficiencies up to 38% (responsivity 2.1 A/W, peak wavelength 6.9 μm) at 78 K. By tailoring one resonance to an absorption peak of NO_2 (6.25 μm) and the other to a non-absorbing reference wavelength (7.15 μm), NDIR NO_2 gas sensing with

10 ppm accuracy and 1 ms response times is demonstrated. Through experiment and numerical simulation, we confirm near-perfect absorption at the resonant cavity and suppressed absorption at its non-resonant counterpart, characteristic of resonant photon sorting. Dual-band sensing across the mid-infrared should be possible by tailoring the cavities and quantum well to desired wavelengths.

Keywords: infrared photodetector; metasurface; nanophotonics; NDIR sensing; photon sorting; quantum well.

1 Introduction

Non-dispersive infrared (NDIR) sensing, widely applied in industrial diagnostics and process control, measures gas concentrations based on mid-infrared absorption lines unique to molecules [1, 2]. In conventional NDIR, broadband emission traverses through the gas specimen and two wavelengths, a target gas absorption line and a non-absorbing reference, are picked out by bulky, spatially separated narrowband filters and passed to two wideband detectors for transmission measurement [1, 2]. The target gas concentration is determined from the detectors' signal ratio to cancel the drift in incoming intensity. In general, relatively slow thermal detectors are applied, measuring concentration from the change in temperature after converting absorbed light into heat. However, NDIR for transient phenomena demanding fast (~1 ms) and strong response, such as *in-situ* combustion engine diagnostics [3, 4], requires photon detectors based on electron transitions in semiconductors.

Metal–dielectric–metal (MDM) metasurface plasmon cavities, precisely controllable narrowband absorbers [5, 6] ($\Delta\lambda/\lambda_{\text{peak}} \leq 0.1$, where $\Delta\lambda$ is the full-width half maximum and λ_{peak} is the peak wavelength of the cavity), offer a simpler NDIR approach [7] without external filters; vertically stacked MDM cavities can exhibit near-perfect absorption of normally incident light via magnetic coupling [5]. Using an

*Corresponding authors: Mel F. Hainey, Jr. and Hideki T. Miyazaki, National Institute for Materials Science, Tsukuba, Ibaraki 305-0047, Japan, E-mail: HAINYJR.Mel@nims.go.jp, melhaineyjr@gmail.com (M.F. Hainey), MIYAZAKI.Hideki@nims.go.jp (H. T. Miyazaki). <https://orcid.org/0000-0003-3878-099X> (M. F. Hainey, Jr.)

Takaaki Mano, Takeshi Kasaya, Tetsuyuki Ochiai, Hirotaka Osato, Kazuhiro Watanabe, Yoshimasa Sugimoto, Takuya Kawazu, Yukinaga Arai and Akitsu Shigetou, National Institute for Materials Science, Tsukuba, Ibaraki 305-0047, Japan

MDM detector resonant at 4.28 μm , Lochbaum et al. [8] recently demonstrated filterless NDIR sensing of CO_2 gas. However their system was based on an incomplete NDIR configuration; only a single wavelength was used, thus, subject to intensity drift. Furthermore, the MDM cavities were used only as narrowband absorbers atop a wideband thermal detector, therefore the response time was limited.

Instead of a single type of MDM cavity, metasurfaces combining multiple types of cavities can realize more complex interactions with light. Resonant photon sorting in metasurfaces made of paired MDM cavities offers a route to recover dual-band functionality necessary for NDIR [9–12]. When two cavities with distinct fundamental resonances are alternately arranged in a subwavelength period, incident photons with different wavelengths are laterally sorted into corresponding cavities within a near field. Because each deep subwavelength cavity has an absorption cross section much larger than its geometrical area [13–17], each cavity realizes nearly perfect absorption of light at its resonance wavelength, without significant interference from its counterpart. Sorting efficiencies, the fraction of the absorption in the resonant cavity over the total absorption in the two cavities, as high as 95% have been observed [9, 12, 18].

By incorporating an infrared semiconductor detector as the dielectric layer of these MDM cavities [9, 10], two fast response detectors for different wavelengths, integrated in the footprint of one, can be fabricated. Because enhanced intensity in the cavities enables us to minimize the semiconductor thickness, high responsivities (signal) with reduced dark currents (noise) can be realized [19, 20], leading to enhanced signal-to-noise ratio, detectivity. Thus by employing resonant photon sorting, NDIR detection with no filters, with complete dual-band functionality, can be realized in a single device. Although a preliminary photon sorting detector based on paired trench cavities has been reported [18], neither clear dual-band operation nor sufficient responsivities for real-world application could be shown.

Here, we demonstrate a dual-band photodetector for NDIR gas sensing based on resonant photon sorting consisting of two electrically isolated, interdigitated stripe MDM cavities sandwiching quantum well infrared photodetectors (QWIPs), metasurface QWIPs [21–25], with different resonance wavelengths. Metasurface QWIPs are based on intersubband transitions (ISBT) in quantum wells. An intense vertical electric field, essential for ISBT absorption [26, 27], is provided by the MDM cavity through rotation of the incident horizontal electric field to vertical, and intensity enhancement [14, 28, 29]. High responsivities [23], fast response times [24, 30], and detectivities

approaching theoretical limits [21, 24, 25, 31], sufficient to replace toxic HgCdTe presently dominant at wavelengths beyond 5 μm , have been demonstrated.

Our dual-band metasurface QWIPs show a maximum unpolarized responsivity of 2.1 A/W at a peak wavelength of 6.9 μm (external quantum efficiency: 38%), comparable to previously demonstrated single-band stripe metasurface QWIPs [21]. Tailoring one resonance to an absorption peak of NO_2 (6.25 μm) and the other to a non-absorbing reference wavelength (7.15 μm), we determine NO_2 gas concentration with 10 ppm accuracy and 1 ms response times, suitable for transient gas sensing applications demanding fast and accurate detection such as combustion engine diagnostics. Through systematic experiments and calculations, resonant photon sorting behavior, nearly perfect absorption at the resonant cavity, and suppressed absorption in the non-resonant cavity are confirmed and graphically visualized. Our dual-band metasurface QWIPs represent a high-performance, flexible approach to photodetection, consolidating the functions of two separate detectors and filters into a single device. Both the cavity and QWIP resonance can be fully controlled, enabling fully engineered detection across the mid-infrared spectrum.

2 Results

The QWIP layer used in this study consists of a single 4 nm GaAs quantum well layer sandwiched by two 50 nm $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barrier layers and two 48 nm GaAs contact layers [21, 25, 32], a total thickness $T = 200$ nm, which exhibits a photoconductive responsivity peak at 6.7 μm [21, 25]. Stripe cavities consist of a $t_{m1} = 100$ nm thick Au stripe and a thick Au substrate (650 nm, but set at $t_{m2} = 200$ nm for calculations) sandwiching the QWIP layer. Each stripe cavity is called a QWIP stripe from here on. Stripes with widths of L_1 and L_2 ($L_1 < L_2$) are alternated with a period $P/2$ (Figure 1A). The QWIP layer between the QWIP stripes is removed [21, 25] for electrical isolation between neighboring stripes. The resonance wavelengths λ_1 and λ_2 , determined by L_1 and L_2 , respectively, should fall within the absorption range of the quantum well (6.0–7.4 μm). The resonance occurs only for x-polarized incidence.

For demonstration of NO_2 sensing afterward, resonance wavelengths of $\lambda_1 = 6.25$ μm and $\lambda_2 = 7.15$ μm are chosen to match the NO_2 absorption peak and provide a non-absorbing reference wavelength, respectively. Near-perfect absorption ($\sim 99\%$) at these wavelengths is predicted in cavities with $L_1 = 0.87$ μm and $L_2 = 1.02$ μm for a period $P = 3.1$ μm (Figure 1B), based on calculations assuming the QWIP layer to be a homogeneous dielectric

Figure 1: Design and fabrication of dual-band metasurface quantum well infrared photodetectors (QWIPs). A) Cross section showing critical dimensions of individual QWIP stripes. E_x and k indicate the electric field and wave vector of incident light. Quantum well (QW) position is indicated by lines in the semiconductor (blue) layer. B) Simulated spectra showing absorption peaks of QWIP stripes with L_1 and L_2 and target wavelengths $6.25 \mu\text{m}$ (blue) and $7.15 \mu\text{m}$ (orange). The red curve is NO_2 absorbance at 100 ppm-m in 1 atm N_2 at 25°C (right axis). C) Colorized SEM image of QWIP stripes in a dual-band device. Gold indicates the Au substrate and top stripe. Blue indicates the QWIP layer.

with a refractive index $n = 3.05 + 0.10i$ taken from preliminary experiments [21, 25]. All calculations in this work are based on rigorous coupled wave analysis (RCWA). Several dual-band devices with QWIP stripes around these L_1 and L_2 values were fabricated following previously reported methods [21, 25] (Figure 1C).

Two interdigitated single-band detectors, called channel 1 (Ch. 1) and channel 2 (Ch. 2), make up a dual-band metasurface QWIP, sometimes simply called a device. In each device, QWIP stripes with the same L value are connected to an electrode pad at one end for independent biasing and current collection. When a bias voltage is applied across the QWIP layer and light within absorption range impinges on each detector, photocurrent is generated (Figure 2A). Based on the principles of resonant photon sorting [9–12], light with wavelengths of λ_1 (blue) and λ_2 (red) should be selectively absorbed in the corresponding resonant cavity (L_1 or L_2).

Responsivities R for unpolarized incidence for three devices—Device 1, 2, and 3 with $(L_1, L_2) = (0.85, 1.04)$, $(0.89, 1.07)$, and $(0.92, 1.11)$ in μm , respectively—evaluated at 78 K

are compared with simulated results in Figure 2B. Simulated R was determined as

$$R(\lambda) = \frac{A_{\text{ISBT}} \times g}{2} \times \frac{\lambda e}{hc}, \quad (1)$$

where A_{ISBT} is the ISBT absorption of the quantum well calculated for x -polarization [21, 25], g ($=2.6$) the photoconductive gain previously determined for unpaired etched QWIP stripes [21], λ the wavelength, e electron charge, h Planck's constant, and c the speed of light. The factor of two indicates that the stripe cavity has responsivity only for x -polarized incidence. The term $A_{\text{ISBT}} \times g/2$ shows the external quantum efficiency for unpolarized incidence [26, 27]. Simulated R in this work was based on previously determined experimental R of the QWIP layer itself evaluated by Brewster-angle incidence [21, 25].

For all three devices, the solid lines in Figure 2A and values in Table 1 indicate that the QWIP stripes with L_2 have maximum experimental peak responsivities (R_{peak}) as high as 2.1 A/W ($L_2 = 1.04 \mu\text{m}$), comparable to the maximum reported value for single-band stripe metasurface

Figure 2: A) Schematic of device operating principle. One device consists of two detectors (Ch. 1, Ch. 2) generating independent photocurrents (I_1 , I_2), composed of electrically isolated interdigitated quantum well infrared photodetector (QWIP) stripes with different dimensions (L_1 , L_2) selectively absorbing at their corresponding resonance wavelengths (λ_1 , λ_2). B) Experimental and simulated unpolarized responsivity of QWIP stripes from three dual-band devices. Brackets indicate which detectors correspond to which output signal in a device.

QWIPs ($R = 2.3$ A/W, background-limited detectivity $D_{BG}^* = 3.3 \times 10^{10}$ cm $\sqrt{\text{Hz}}/\text{W}$) [21], and R_{peak} gradually decreases with increasing L . The QWIP stripes with L_1 have much lower R_{peak} than those with L_2 , and exhibit increasing R_{peak} with increasing L . Fairly good agreement between experiment and simulation (dashed lines, Figure 2A) near 7 μm , and overestimated R in the simulation compared to experiment near 6 μm is consistent with what Ref. 21 reported on etched stripe metasurface QWIPs. This characteristic relationship between experiment and simulation was attributed to changes in the R profile of the QWIP layer due to the metasurface fabrication and insensitive regions at the etched surfaces of the stripes [21].

In each device, when one QWIP stripe exhibits a peak responsivity R_{peak} at its resonance wavelength λ_1 or λ_2 (bold R_{λ_1} , R_{λ_2} , Table 1), its counterpart shows greatly reduced R at the same wavelength (non-bold R_{λ_1} , R_{λ_2} , Table 1), leading to a high contrast in R at these wavelengths (Figure 2A, Table 1). Simulated spectra show similar trends. Resonant

absorption in each QWIP stripe proceeds with minimal interference from its counterpart.

Numerical simulation clarifies how photons are sorted into resonant QWIP stripes. $|E_z|^2$ in Device 2 for x -polarized incidence at simulated A_{ISBT} peak wavelengths $\lambda_1 = 6.30$ μm and $\lambda_2 = 7.15$ μm (Table 1) is shown in Figure 3A, B. Strong $|E_z|^2$ is observed in only the resonant QWIP stripes, manifesting the selective coupling of incident light into the corresponding cavities (Figure 3A, B).

Power flow [13, 14, 16, 25] clearly visualizes how photons with different wavelengths are routed into the corresponding cavities (Figure 3C, D). Stream lines of the time-averaged Poynting vector for x -polarized incidence show that the vertically incident light starts to deflect around $z = 800$ nm and is finally collected into the resonant cavities almost perfectly (Figure 3C, D). The adjacent QWIP stripe exhibits negligible absorption. The solid black curves and the black arrows at the top of Figure 3C, D illustrate each cavity's sorting efficiency [9, 12, 18] over one

Table 1: Peak wavelengths and corresponding resonant (bolded) and non-resonant responsivity values of quantum well infrared photodetector (QWIP) stripes in different dual-band devices.

Device	Channel	L_1, L_2 (μm)	Peak wavelength λ_1, λ_2 (μm)	R_{λ_1} (A/W)	R_{λ_2} (A/W)
Device 1	Ch. 1	0.85	6.03 (6.10)	0.38 (1.08)	0.046 (0.060)
	Ch. 2	1.04	6.90 (7.00)	0.068 (0.037)	2.13 (2.30)
Device 2	Ch. 1	0.89	6.23 (6.30)	0.72 (1.50)	0.056 (0.067)
	Ch. 2	1.07	7.06 (7.15)	0.11 (0.050)	2.08 (1.99)
Device 3	Ch. 1	0.92	6.40 (6.50)	1.19 (1.90)	0.081 (0.050)
	Ch. 2	1.11	7.22 (7.35)	0.15 (0.098)	1.78 (1.54)

Values from simulations are indicated in parenthesis.

Figure 3: Photon sorting over two periods P for Device 2 ($L_1 = 0.89 \mu\text{m}$, $L_2 = 1.07 \mu\text{m}$, and $P = 3.1 \mu\text{m}$) at $\lambda = 6.30 \mu\text{m}$ and $\lambda = 7.15 \mu\text{m}$, respectively. A, B) Calculated $|E_z|^2$ in Device 2. E_x and k indicate the electric field and wave vector of incident light. C, D) Power flow in Device 2. Stream lines are spaced every 100 nm at $z = 1100 \text{ nm}$. Solid black lines and black arrows describe the sorting efficiency of each cavity over one period P (white arrows). Dashed lines and red arrows define the absorption by the quantum well.

period P . These graphical sorting efficiencies of 95% at both $\lambda_1 = 6.30 \mu\text{m}$ and $\lambda_2 = 7.15 \mu\text{m}$ are consistent with the numerically obtained absorption in individual cavities.

However, this does not simply show the light absorbed in the quantum well. Most stream lines terminate in the Au stripe and Au substrate without any contribution to the photocurrent. The dashed black curves and red arrows at the top of Figure 3C, D define the fraction of total absorbed incidence collected in the quantum well over one period P . These fractions agree with the calculated A_{ISBT} corresponding to the simulated spectra of Figure 2B (22 and 26% for L_1 and L_2 , respectively, in Device 2), but are lower than the total sorting efficiency of the cavity, similar to previous reports for other metasurface QWIPs [21, 25]. Nevertheless, the single quantum well design of our QWIP layer [25] allows us to realize high g and quantum efficiencies, leading to the high R_{peak} values predicted from Equation (1).

The observed contrast in R between QWIP stripes and photon sorting behavior from power flow suggest our dual-band device can operate effectively as an NDIR sensor. As mentioned previously, we chose NO_2 as our target molecule for demonstration. Emission control of NO_2 , the most toxic component of NO_x , remains a critical topic for automotive engineering research [33–35]. A typical combustion engine emits NO_2 on the order of 10^1 – 10^3 ppm within a cycle of 50–80 ms, demanding ppm accuracy and ms response time to investigate the generation process of the NO_2 gas [34, 35].

Presently, state-of-the-art NO_x analysis with 5 ppm accuracy [36] and 2 ms response times [34] for engine development is conducted by chemiluminescence [34], using a chemical reaction of NO and O_3 . However, the present technique requires extraction of gas from the exhaust port to a separate reaction chamber, and also a NO_2 -to- NO converter before the chamber.

Our aim here is to demonstrate similar performance using NDIR NO_2 sensing with our dual-band detector, which integrates the functions of two bandpass filters and two infrared photodetectors into a single, much smaller, monolithic device. Compared to chemiluminescence systems, NDIR can use the gas channel from the exhaust port as the optical path for directly measuring NO_2 without any need for a special reaction chamber and a gas converter.

In Device 2, Ch. 1 corresponds to the NO_2 absorption wavelength at $6.25 \mu\text{m}$, while Ch. 2 is used as the reference at $7.15 \mu\text{m}$ (Figure 4A). Based on calibration experiments for known NO_2 concentrations, the NO_2 concentration can be determined from the ratio of photocurrent I_p in Ch. 1/Ch. 2 (Supplementary information S1, Figure S1A) with a standard deviation of 18 ppm (Figure S1B). To clearly demonstrate ms response in our detectors, we introduce a pulse of NO_2 (Figure 4B, Methods) and simultaneously observe changes in Ch. 1 and Ch. 2 response to track NO_2 concentration. Figure 4C shows a sharp decrease in I_p in Ch. 1 from the introduced NO_2 followed by a more gradual

Figure 4: Dual-channel sensing using a dual-band metasurface quantum well infrared photodetector (QWIP) (Device 2). A) Experimentally determined resonance of Ch. 1 ($L_1 = 0.89 \mu\text{m}$) is matched to NO_2 absorption peak at $\lambda = 6.25 \mu\text{m}$. Ch. 2 ($L_2 = 1.07 \mu\text{m}$) is the non-absorbing reference. NO_2 absorbance at 100 ppm-m in 1 atm N_2 at 25°C (right axis). B) Experimental setup for fast response NO_2 concentration measurement. C) Response to rapid concentration changes in each channel of Device 2. Photocurrent I_p in Ch. 2 (top) and Ch. 1 (middle) measured over multiple NO_2 pulses. The signal ratio (bottom) gives NO_2 concentration. D) Fitting the rise associated with a single pulse to determine the time constant.

return to the original signal, while I_p in Ch. 2 remains nearly constant. Fitting to the rise of this curve gives a time constant of 3.2 ms (Figure 4D).

This speed is only limited by the gas supply system, not the detector. Using another experimental setup with a much faster gas supply, we confirmed a 0.83 ms response time with the same detectors (Supplementary information S2, Figure S2), clearly indicating that our device performance already approaches state-of-the-art benchmarks.

For further optimized NO_2 sensing, we can increase the relatively low R of Ch. 1 (Figure 4A) by redesigning the quantum well for maximum sensitivity at $6.25 \mu\text{m}$. In addition, since the peak wavelength in both channels can be precisely controlled by the design of the MDM cavity and quantum well, our dual-band can be applied for other gases.

3 Discussion

Our dual-band device consolidates the function of two single-band detectors in the footprint of one; each QWIP stripe in a dual-band device operates as if their counterpart

is not present. Here, we examine the influence of the counterpart stripe more straightforwardly and quantitatively by comparing paired QWIP stripes from Devices 1 to 3 with unpaired QWIP stripes having the same (or very close) L values but no counterpart from our previous study [21] on single-band devices. Unpaired single-band devices were designed ($P = 2.4 \mu\text{m}$) for optimized R [21].

Figure 5 shows experimental total absorption (A_{TOT}) for paired QWIP stripes from Devices 1 to 3 (black) and corresponding unpaired QWIP stripes (colored). Maximum values of A_{TOT} , corresponding to resonant absorption in one of the paired QWIP stripes show near-perfect correspondence in position, magnitude and overall profile with unpaired QWIP stripes with the same L . At the resonance wavelengths of paired QWIP stripes, each resonant stripe acts like an unpaired QWIP stripe, absorbing almost all light, as if their counterpart was not present.

Highly selective resonant absorption in paired QWIP stripes is further confirmed from measured R . In Figure 6A, experimental R spectra of paired (solid) and unpaired (dotted) QWIP stripes again show nearly identical peak position, magnitude, and overall profile. In dual-band devices,

Figure 5: Comparison of paired quantum well infrared photodetector (QWIP) stripes from dual-band devices (black) and unpaired QWIP stripes with identical L (colored). A) One period of each structure shown schematically for comparison. B–D) Experimental total absorption (A_{TOT}) taken from s -polarized reflection (Incidence angle $\theta = 26^\circ$ in the y - z plane) of Devices 1, 2 and 3 along with corresponding unpaired QWIP stripes. L of the unpaired QWIP stripes corresponds with either L_1 or L_2 in the paired QWIP stripe. An unpaired QWIP stripe with $L = 0.93 \mu\text{m}$ is shown to be compared with the paired QWIP stripe with $L_1 = 0.92 \mu\text{m}$ due to lack of an unpaired QWIP stripe at the exact dimensions. All A_{TOT} is calibrated, thus independent of the device area.

each QWIP stripe maintains its original performance as a single-band device without essential degradation.

Away from resonance, a critical difference between the paired and unpaired QWIP stripes, also characteristic of photon sorting, can be observed (Figure 6B). In all three dual-band devices, around its counterpart's resonance wavelength (black vertical lines), R in a non-resonant QWIP stripe is significantly suppressed (filled circles) compared to an unpaired QWIP stripe with the same L (unfilled circles)—behavior also observed in previous reports on photon sorting [9, 12, 18]. Instead, these photons

are absorbed in the resonant QWIP stripe. This can be interpreted that the persistent absorption of the resonant QWIP stripe and the high contrast in R between the paired QWIP stripes are supported by suppressing the absorption of the non-resonant counterpart. Combined, the enhanced resonant absorption and suppressed non-resonant absorption lead paired QWIP stripes to behave as if their counterpart was not present, allowing us to consolidate two single-band detectors in the footprint of one.

Throughout this work, low experimental R in the QWIP stripes with L_1 resonant around $6 \mu\text{m}$ has been noticeable. To

Figure 6: A) Experimental unpolarized R of paired quantum well infrared photodetector (QWIP) stripes from dual-band devices (solid) and unpaired QWIP stripes with identical L (dotted). One period of each structure is shown schematically for comparison. An unpaired QWIP stripe with $L = 0.93 \mu\text{m}$ is shown to be compared with the paired QWIP stripe with $L_1 = 0.92 \mu\text{m}$ due to lack of an unpaired QWIP stripe at the exact dimensions. B) Close-up of A) for respective devices. Non-resonant R in paired QWIPs around their counterpart's resonance wavelength (black vertical lines). Black filled circles indicate values of experimental non-resonant R in paired QWIP stripes. Black unfilled circles indicate values of non-resonant R corresponding unpaired QWIP stripes (dotted lines). All experimental R is calibrated, thus independent of the device area.

increase R at this wavelength range, we considered increasing the number of stripes resonant at shorter wavelengths in one detector. We fabricated dual-band devices with two L_1 QWIP stripes and one L_2 QWIP stripe, with a larger $P = 3.75\ \mu\text{m}$ to accommodate the third stripe (Supplementary information S3, Figure S3). However, we could not realize significantly increased R in the QWIP stripes with L_1 , but instead observed a red-shift of λ_1 from 6.05 to $6.40\ \mu\text{m}$. These results are presumed to indicate the near-field coupling between two L_1 QWIP stripes (Figures S4, S5). Because adding an additional stripe did not lead to enhanced R , we should instead redesign our quantum well to improve R around $6\ \mu\text{m}$.

Finally, we would like to remark on the optical cross section of each QWIP stripe. In the power flow in Figure 3 and Figure S5, the numerically visualized absorption cross section of each stripe is much larger than its geometrical area. This is also evidenced in the experimental responsivity spectra. In Figure 6A, unpaired QWIP stripes exhibited a very similar R spectrum to that of paired QWIP stripe, irrespective of the difference in P (2.4 and $3.1\ \mu\text{m}$ for the unpaired and paired QWIP stripes, respectively). Also in Figure S4B, the QWIP stripe with L_2 in a three-stripe device showed a very similar R with that of a QWIP stripe from a one-stripe device despite an even greater difference in P (2.4 and $3.75\ \mu\text{m}$ for one- and three-stripe devices, respectively).

Responsivities are independent of device areas. Therefore, R for one- (unpaired), two- (paired), and three-stripe devices show the photocurrents for the same area. As P increases from 2.4 to $3.1\ \mu\text{m}$ and $3.75\ \mu\text{m}$, the number of resonant QWIP stripes per area correspondingly decreases. Nonetheless, devices with fewer resonant stripes maintain a nearly constant R . This means that resonant QWIP stripes in two- and three-stripe detectors with a larger P are generating greater photocurrent per stripe than one-stripe detectors; i.e., they are collecting incident light from wider areas. Experimentally observed consistent R for one-, two-, and three-stripe devices is direct evidence of the numerically visualized optical cross sections.

In addition, these results reveal another essential conclusion—the optical cross section of a cavity is not necessarily unique to the cavity itself, but depends on the environment of the cavity. When another cavity is placed between equivalent cavities, the original cavities can have much larger cross sections.

4 Conclusion

In summary, we have demonstrated dual-band, dual-channel photodetection based on resonant photon sorting

in metasurface QWIPs made of paired stripe cavities. Individual QWIP stripes with responsivities up to $2.1\ \text{A/W}$ at $6.9\ \mu\text{m}$ at $78\ \text{K}$, comparable with previous high-detectivity, single-band devices have been demonstrated. Our devices operate as dual-band integrated NDIR sensors combining the function of two filters and two detectors in the footprint of a single device. Tailoring one detector to the specific absorption wavelengths of NO_2 , concentration measurement with $10\ \text{ppm}$ accuracy and $1\ \text{ms}$ response time, comparable with state-of-the-art chemiluminescence systems, has been demonstrated. Through electric field distribution, power flow, and comparison with unpaired stripes, we observed how photon sorting behaviors lead to efficient resonant absorption from a wider area and suppressed non-resonant absorption in the paired stripes. These behaviors ensure high R is maintained even for devices with larger numbers of different cavities.

Conventionally, dual-band infrared photodetectors have been achieved by sequential epitaxial growth of semiconductor crystals such as toxic HgCdTe and QWIPs [26, 27, 37] with different compositions. The responsivity spectrum of each band has been determined by the band structure of the individual crystal. In contrast, resonant photon sorting realizes dual-band detection with a high flexibility with a single semiconductor crystal; the responsivity spectra are defined by the metasurface design. By employing photon sorting, the flow of the incident photons are laterally manipulated in the near field region, and finally converted to flow of electrons in a desired channel.

Engineering of both electrons and photons, as in metasurface QWIPs, would suggest a promising frontier in optoelectronic devices. Although intensive engineering of photonic structures was demonstrated in this work, tailoring electronic structures would be another important topic. For instance, by replacing the photoconductive QWIP with a photovoltaic QWIP [31, 38], room temperature operation with sufficient detectivity could be achieved, greatly increasing the functionality of our devices.

5 Methods

5.1 Design, fabrication, and characterization of metasurface QWIPs

The QWIP layer grown by molecular beam epitaxy on a GaAs substrate was transferred to an Au substrate by wafer bonding, and processed to QWIP stripes by electron beam drawing and inductively coupled plasma etching. Three metasurface QWIPs containing 32 paired stripes in a $100 \times 100\ \mu\text{m}$ area were assembled on an 8-pin ceramic package.

The devices were installed in a cryostat with ZnSe windows, and their responsivity spectra were measured with an FTIR by feeding the amplified current signal to the external port. The spectral responsivity was quantified based on a calibrated HgCdTe detector. No corrections other than the detector areas were applied to the R values. RCWA simulation was performed using home-built software. L_1 and L_2 values used in the calculations were determined from SEM measurements. The quantum well layer was treated as an anisotropic material having the ISBT absorption in the vertical direction and Drude free-carrier absorption in the lateral directions.

The reflection spectra ($Refl$) used for determining A_{TOT} were measured with an infrared microscope equipped with a Cassegrain objective lens connected to the FTIR. A relatively small incidence angle distribution centered at 26° was realized with an aperture. Given the thickness of our Au substrate, we consider transmission to be negligible, so $A_{TOT} = 1 - Refl$.

One-stripe and three-stripe detectors were designed based on the same model as the two-stripe detectors [21, 25]. More details on the simulation, material parameters, fabrication, and characterization can be found elsewhere [21, 25].

5.2 Power flow visualization

The power flow was visualized by tracing the time-averaged Poynting vector [13, 14] from $z = 1100$ nm, where incident light still forms a plane wave, until the magnitude decreased to 10^{-2} . Note that the poynting vector expresses the absorbed power flow. Graphical absorption efficiencies shown in Figure 3 and Figure S5 show the ratio of the absorption in each cavity to the total absorption, rather than to the total incidence. Based on this ratio, we can graphically determine the sorting efficiency of each cavity.

5.3 Gas concentration measurement

Absorbance data for gases were taken from a commercial database (HANST, Infrared Analysis, Inc.). Detailed information about NO_2 calibration can be found in Supplementary information S1. A pulse of NO_2 was realized by injecting pressurized NO_2 gas (0.952% in N_2 , 0.5 MPa) with a high-speed solenoid valve (CKD, AB31-02-3) into N_2 carrier gas flowing at a rate of 160 L/min. Signals from the two detectors were current amplified and recorded with a resolution of 40 μs .

For the sub-1-ms demonstration in the Supplementary information S2, HFC-152a (1,1-difluoroethane) from a spray can was orthogonally ejected into the light path without using the gas cell.

The time constant τ was determined by fitting the waveform to an exponentially converging function: $y(t) = y_0 + y_1[1 - \exp\{-(t - t_0)/\tau\}]$, where t is time, t_0 starting time, y_0 baseline, and y_1 step height. For the NO_2 in Figure 4D, $y_0 = 0$ ppm, $y_1 = 2480$ ppm, $t_0 = 0.0346$ s, and $\tau = 0.00324$ s. For HFC-152a in Figure S2C, $y_0 = 27.98$ nA, $y_1 = -4.34$ nA, $t_0 = 0.3564$ s, and $\tau = 0.000829$ s.

Acknowledgments: The authors acknowledge helpful discussion with an anonymous company, K. Miyano, H. Miyazaki, Y. Jimba, Y. Sakuma, T. Noda, A. Ohtake, D. Tsuya, N. Ikeda, E. Watanabe, M. Iwanaga, and K. Sakoda, and the technical assistance of SIJTechnology, Inc. This work was supported by JSPS KAKENHI Grant Numbers JP15H02011,

JP17H01275, and JP19H00875, Iketani Science and Technology Foundation, the Center for Functional Sensor and Actuator, the National Institute for Materials Science (NIMS), and the NIMS Nanofabrication Platform in Nanotechnology Platform Project sponsored by the Ministry of Education, Culture, Sports, Science and Technology, Japan.

Author contribution: All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.

Research funding: This work was supported by JSPS KAKENHI Grant Numbers JP15H02011, JP17H01275, and JP19H00875, Iketani Science and Technology Foundation, the Center for Functional Sensor and Actuator, the National Institute for Materials Science (NIMS), and the NIMS Nanofabrication Platform in Nanotechnology Platform Project sponsored by the Ministry of Education, Culture, Sports, Science and Technology, Japan.

Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

References

- [1] J. Hodgkinson and R. P. Tatam, "Optical gas sensing: a review," *Meas. Sci. Technol.*, vol. 24, no. 1, p. 012004, Jan. 2013.
- [2] D. Popa and F. Udrea, "Towards integrated mid-infrared gas sensors," *Sensors*, vol. 19, no. 9, p. 2076, 2019.
- [3] M. S. Peckham, A. Finch, and B. Campbell, "Analysis of transient HC, CO, NO_x and CO_2 emissions from a GDI engine using fast response gas analyzers," *SAE Int. J. Engines*, vol. 4, no. 1, pp. 1513–1522, Apr. 2011.
- [4] M. Duckhouse, M. Peckham, B. Mason, E. Winward, and M. Hammond, "In-cylinder CO_2 sampling using skip-firing method," in *Volume 2: Emissions Control Systems; Instrumentation, Controls, and Hybrids; Numerical Simulation; Engine Design and Mechanical Development*, 2, November, 2018, <https://doi.org/10.1115/ICEF2018-9570>.
- [5] G. Lvque and O. J. F. Martin, "Tunable composite nanoparticle for plasmonics," *Opt. Lett.*, vol. 31, no. 18, pp. 2750–2752, 2006.
- [6] X. Liu, T. Starr, A. F. Starr, and W. J. Padilla, "Infrared spatial and frequency selective metamaterial with near-unity absorbance," *Phys. Rev. Lett.*, vol. 104, no. 20, p. 207403, 2010.
- [7] S. Ogawa and M. Kimata, "Wavelength- or polarization-selective thermal infrared detectors for multi-color or polarimetric imaging using plasmonics and metamaterials," *Materials*, vol. 10, no. 5, p. 493, May 2017.
- [8] A. Lochbaum, A. Dorodnyy, U. Koch, and S. M. Koepfli, "Compact mid-infrared gas sensing enabled by an all-metamaterial design," *Nano Lett.*, vol. 20, no. 6, pp. 4169–4176, Jun. 2020.
- [9] C. Koechlin, P. Bouchon, F. Pardo, et al., "Total routing and absorption of photons in dual color plasmonic antennas," *Appl. Phys. Lett.*, vol. 99, no. 24, p. 241104, 2011.
- [10] J. Le Perchec, Y. Desieres, N. Rochat, and R. Espiau De Lamaestre, "Subwavelength optical absorber with an integrated photon sorter," *Appl. Phys. Lett.*, vol. 100, no. 11, p. 113305, 2012.

- [11] P. Bouchon, C. Koechlin, F. Pardo, R. Haïdar, and J.-L. Pelouard, "Wideband omnidirectional infrared absorber with a patchwork of plasmonic nanoantennas," *Opt. Lett.*, vol. 37, no. 6, pp. 1038–1040, 2012.
- [12] E. Lansey, I. R. Hooper, J. N. Gollub, A. P. Hibbins, and D. T. Crouse, "Light localization, photon sorting, and enhanced absorption in subwavelength cavity arrays," *Opt. Express*, vol. 20, no. 22, pp. 24226–24236, 2012.
- [13] C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles*, New York, Wiley, 1983.
- [14] H. T. Miyazaki and Y. Kurokawa, "How can a resonant nanogap enhance optical fields by many orders of magnitude?," *IEEE J. Sel. Top. Quantum Electron.*, vol. 14, no. 6, pp. 1565–1576, 2008.
- [15] P. Bouchon, F. Pardo, B. Portier, et al., "Total funneling of light in high aspect ratio plasmonic nanoresonators," *Appl. Phys. Lett.*, vol. 98, no. 19, p. 191109, 2011.
- [16] F. Pardo, P. Bouchon, R. Haïdar, and J.-L. Pelouard, "Light funneling mechanism explained by magnetoelectric interference," *Phys. Rev. Lett.*, vol. 107, no. 9, p. 093902, Aug. 2011.
- [17] D. Palaferri, Y. Todorov, A. Mottaghizadeh, G. Frucci, G. Biasiol, and C. Sirtori, "Ultra-subwavelength resonators for high temperature high performance quantum detectors," *New J. Phys.*, vol. 18, no. 11, p. 113016, 2016.
- [18] S. J. Kim, J.-H. Kang, M. Mutlu, et al., "Anti-Hermitian photodetector facilitating efficient subwavelength photon sorting," *Nat. Commun.*, vol. 9, p. 316, Dec. 2018.
- [19] J. Le Perche, Y. Desieres, and R. Espiau De Lamaestre, "Plasmon-based photosensors comprising a very thin semiconducting region," *Appl. Phys. Lett.*, vol. 94, no. 18, p. 181104, 2009.
- [20] A. Rogalski, "Infrared detectors: status and trends," *Prog. Quantum Electron.*, vol. 27, nos 2–3, pp. 59–210, 2003.
- [21] M. F. Hainey, T. Mano, T. Kasaya, et al., "Systematic studies for improving device performance of quantum well infrared stripe photodetectors," *Nanophotonics*, vol. 9, no. 10, pp. 3373–3384, Jul. 2020.
- [22] Y. Nga Chen, Y. Todorov, B. Askenazi, et al., "Antenna-coupled microcavities for enhanced infrared photo-detection," *Appl. Phys. Lett.*, vol. 104, no. 3, p. 031113, 2014.
- [23] Q. Li, Z. F. Li, N. Li, et al., "High-polarization-discriminating infrared detection using a single quantum well sandwiched in plasmonic micro-cavity," *Sci. Rep.*, vol. 4, p. 6332, May 2014.
- [24] D. Palaferri, Y. Todorov, A. Bigioli, et al., "Room-temperature nine- μm -wavelength photodetectors and GHz-frequency heterodyne receivers," *Nature*, vol. 556, no. 7699, pp. 85–88, 2018.
- [25] H. T. Miyazaki, T. Mano, T. Kasaya, et al., "Synchronously wired infrared antennas for resonant single-quantum-well photodetection up to room temperature," *Nat. Commun.*, vol. 11, p. 565, Dec. 2020.
- [26] B. F. Levine, "Quantum-well infrared photodetectors," *J. Appl. Phys.*, vol. 74, no. 8, pp. R1–R81, Oct. 1993.
- [27] H. Schneider and H. C. Liu, *Quantum Well Infrared Photodetectors*, vol. 2, Berlin Heidelberg, Springer, 2007.
- [28] G. Dolling, C. Enkrich, M. Wegener, J. F. Zhou, C. M. Soukoulis, and S. Linden, "Cut-wire pairs and plate pairs as magnetic atoms for optical metamaterials," *Opt. Lett.*, vol. 30, no. 23, pp. 3198–3200, Dec. 2005.
- [29] H. T. Miyazaki and Y. Kurokawa, "Squeezing visible light waves into a 3-nm-thick and 55-nm-long plasmon cavity," *Phys. Rev. Lett.*, vol. 96, no. 9, p. 097401, 2006.
- [30] A. Bigioli, D. Gacemi, D. Palaferri, et al., "Mixing properties of room temperature patch-antenna receivers in a mid-infrared ($\lambda \approx 9 \mu\text{m}$) heterodyne system," *Laser Photon. Rev.*, vol. 14, no. 2, p. 1900207, Feb. 2020.
- [31] A. Bigioli, G. Armaroli, A. Vasanelli, et al., "Long-wavelength infrared photovoltaic heterodyne receivers using patch-antenna quantum cascade detectors," *Appl. Phys. Lett.*, vol. 116, no. 16, p. 161101, 2020.
- [32] T. Mano, H. T. Miyazaki, T. Kasaya, T. Noda, and Y. Sakuma, "Double-sided nonalloyed ohmic contacts to Si-doped GaAs for plasmoelectronic devices," *ACS Omega*, vol. 4, no. 4, pp. 7300–7307, 2019.
- [33] N. Kato, Y. Hamada, and H. Kurachi, "Performance of thick film NO_x sensor on diesel and gasoline engines," *SAE Tech. Pap.*, no. 970858, pp. 199–206, 1997.
- [34] F. Leach, M. Davy, and M. Peckham, "Cycle-to-cycle NO and NO_x emissions from a HSDI diesel engine," *J. Eng. Gas Turbines Power*, vol. 141, no. 8, p. 081007, 2019.
- [35] K. S. J. Reavell, N. Collings, M. Peckham, and T. Hands, "Simultaneous fast response NO and HC measurements from a spark ignition engine," *SAE Tech. Pap.*, no. 971610, pp. 121–128, 1997.
- [36] C. D. Rakopoulos, A. M. Dimaratos, E. G. Giakoumis, and D. C. Rakopoulos, "Investigating the emissions during acceleration of a turbocharged diesel engine operating with bio-diesel or n-butanol diesel fuel blends," *Energy*, vol. 35, no. 12, pp. 5173–5184, 2010.
- [37] A. Rogalski, J. Antoszewski, and L. Faraone, "Third-generation infrared photodetector arrays," *J. Appl. Phys.*, vol. 105, no. 9, p. 091101, 2009.
- [38] L. Wang, S.-Q. Zhai, F. Wang, et al., "A polarization-dependent normal incident quantum cascade detector enhanced via metamaterial resonators," *Nanoscale Res. Lett.*, vol. 11, p. 536, Dec. 2016.

Supplementary Material: Details of dual-band detector calibration for NO_2 sensing. Details of fast response measurement realizing 1 ms response times. Fabrication and performance of the three-stripe detector, including numerical simulation.

The online version of this article offers supplementary material (<https://doi.org/10.1515/nanoph-2020-0456>).