Nanowire network–based multifunctional all-optical logic gates

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All-optical nanoscale logic components are highly desired for various applications because light may enable logic functions to be performed extremely quickly without the generation of heat and cross-talk. All-optical computing at nanoscale is therefore a promising alternative but requires the development of a complete toolbox capable of various functional properties. We demonstrate nanoscale all-optical switches by exploiting the polarization-dependent light emission property of crossbar InP and AlGaAs nanowire networks. These networks can perform various logic operations, such as AND, OR, NAND, and NOR binary logic functions. Furthermore, on the basis of these logic operations, our networks successfully enable all-optical arithmetic binary calculations, such as $n$-bit addition, to be conducted. Our results underscore the promise of assembled semiconductor nanowire networks as a building block of on-chip all-optical logic components for future nanophotonics.

INTRODUCTION

Current limitations of modern integrated circuits, resulting from the prolonged scaling of the transistors, are propelling intense activities in the field of nanophotonics (1, 2). In this respect, integration of all-optical nanoproces-sors into future integrated circuits may drive the computers to perform their functions ever more effectively compared to their modern electronic counterparts (3). For this purpose, it is important to realize a complete toolbox filled with chip-integrable all-optical nanoscale components (for example, light sources, switches, waveguides, modulators, and logic gates). Over the last few decades, the main research trends in this area have focused toward the development of all-optical logic devices based on nonlinear optical effects in silicon waveguide circuits (4–6) and semiconductor optical amplifiers (7, 8). More recently, nanowires (NWs) have emerged as promising alternative candidates to fulfill the toolbox. From the basic science perspective, these NW structures are fascinating since they provide new means to construct all-optical logic components owing to their extraordinary properties including strong waveguiding effects (9–11), optical nonlinearity (12, 13), and plasmonic effects (14, 15) combined with their subwavelength diameter. Because of these properties in particular, various NW-based components have been demonstrated in the field of nanophotonics, such as electrically driven lasers (16), electro-optical modulators (17), and also, as noted, all-optical logic gates (11, 14, 15). Among those components, logic gates form the core of the data processing and are therefore of particular importance in the construction of all-optical nanoprocessor circuits.

The NW-based all-optical logic gates have thus far exploited the stimulated scattering of exciton–polaritons in CdS NWs (11) and plasmonic interference effects in Ag NW networks (14, 15). As such, the realization of the conditions suitable for all-optical switching requires complex fabrication steps and delicate measurement schemes. For instance, since the switching in the CdS NW-based device is dependent on the pump-excited polaritons inside the NWs, the operation occurs only at low temperature (inside a liquid nitrogen–cooled cryostat) and requires a gallium ion milling fabrication step to obtain efficient light coupling within the NW waveguides (11). On the other hand, Ag NW-based logic gates demand interference conditions obtained by accurately positioning each individual NW with a micromanipulator setup. Hence, the practicality of these approaches is largely limited by the fabrication process, and therefore, it is of great importance to demonstrate new approaches better suited for the realization of future all-optical nanoproces-sor circuits capable of complex logic operations. In this study, to achieve this target, we use a scalable fabrication process without any lithography steps to prepare crossbar networks of InP and AlGaAs NWs. The fabricated semiconductor NW network operates as a polarization-controlled all-optical wavelength switch. The polarization-controlled operation mode of the switch is based on the anisotropic structure of the NWs, which makes their emission extremely sensitive to the excitation light polarization direction (18). Thus, the linear optics explained the operation principle of our switch well, in contrast to all previously reported NW-based all-optical logic gates (11, 14, 15). Furthermore, InP and AlGaAs NWs are selected because their bandgap energy differs considerably, making the all-optical wavelength switching very easily resolvable (wavelength shift as large as more than ~200 nm, that is, from ~665 to ~890 nm). Compared to GaAs NWs, our InP and AlGaAs NWs are also found to show photoluminescence (PL) emission at room temperature due to the reduced nonradiative surface recombination channels in InP (19) and in situ doping of AlGaAs. Our crossbar NW structure allows multiple parallel all-optical logic functions (for example, AND, OR, NAND, and NOR) to be realized. In particular, we perform various binary arithmetic calculations all-optically (such as $n$-bit addi- tion) with these logic gates.

RESULTS AND DISCUSSION

InP and AlGaAs NWs are grown on silicon substrates by using a Au nanoparticle–assisted vapor-liquid-solid (VLS) growth method inside a horizontal-flow metalorganic vapor phase epitaxy (MOVPE) reactor (see the Supplementary Materials for detailed information). The fabrication of (opto)electronic devices from VLS-grown NWs...
typically involves complex processing steps [such as focused ion beam milling (11), micromanipulation (14), electron beam lithography (20), etc.] due to unorganized location and tilting of the NWs. To address this constraint, we use a scalable nanocombing method to align the randomly pointing NWs in the horizontal plane, which does not need the abovementioned complex fabrication steps (21, 22).

In this way, the as-grown NWs are transferred onto a glass substrate using the nanocombing technique (see fig. S1). It should be noted that this nanocombing step is performed in this work without any micromanipulator setup, illustrating the simplicity of our approach. We estimate that ~95% of NWs are aligned along the same direction within an alignment error of ±10° after the single combing step (see fig. S2). Furthermore, with this method, it is easy to realize crossbar NW junctions by simply combing InP NWs along one direction and the other (that is, AlGaAs) NWs along the perpendicular direction for the fabrication of multifunctional logic gates. Note that, after combing the InP NWs, we deposit a 100-nm-thick SiO₂ layer to isolate the InP NWs from the AlGaAs NWs. Because of the random nature of the process, it is difficult to make every InP NW perpendicular to AlGaAs NW and simultaneously be overlapping; however, it is easy to find many crossbar NW junctions on the target substrate, as shown in Fig. 1A. In such a ~6 μm × 6 μm area, we find six pairs of well-aligned InP and AlGaAs crossbar NW junctions. It verifies that this easy nanocombing method is effective and helpful for the realization of large-area crossbar NW networks.

Polarization-dependent PL measurements are performed on crossbar NW junctions. Figure 1B shows the room temperature PL spectra of an exemplary NW junction measured from a single measurement spot by using the excitation wavelength of 532 or 730 nm with varying excitation laser polarization directions (see details in the Supplementary Materials). Under the 532-nm laser excitation, two PL peaks located at 665 and 890 nm are observed, which we attribute to AlGaAs and InP NWs, respectively. Under the 730-nm laser excitation, only a single PL peak is observed at the wavelength of 890 nm. Note that, in this case, no PL is created at the wavelength of 665 nm because the band-to-band transition energy of AlGaAs NWs is larger than the photon energy of the excitation laser. PL quantum efficiency (PLQE) of the NWs is an important measure to
consider for the assessment of optical properties of our crossbar NW structures. So far, the external QEs of NWs have typically been reported with a range of $10^{-5}$ to $10^{-2}$ (23). In this work, by comparing the PL intensity of our NWs to that of Rhodamine 6G, we estimate the PLQE of our NWs to be $\sim 10^{-4}$, still sufficient for the results presented later in this work.

To verify the observed PL transitions, a structural analysis is performed by high-resolution scanning transmission electron microscopy (HRSTEM) and energy-dispersive x-ray (EDX) measurements (see the Supplementary Materials for details). As shown in Fig. 1C, the structure of InP NWs is predominantly zinc-blende (ZB) with frequent twin planes along the whole NW length (rotationally twinned ZB InP NWs). Compared to the room temperature PL peak of bulk ZB InP at the wavelength of $\sim 920$ nm, the $\sim 30$-nm blue shift in the PL peak position of the InP NWs can be attributed to these frequent twin faults (24–27). On the other hand, PL properties of AlGaAs NWs depend strongly on the elemental compositions and the crystal quality. As shown in fig. S3, the crystal structure of AlGaAs NWs is predominantly ZB with a small concentration of crystal defects. Figure 1D shows a typical EDX line scan of AlGaAs NWs along the [111] crystal direction, revealing an initial Al composition of 80% within a very short region of the NW tip. The high Al composition in the vicinity of Au catalyst is attributed to the incorporation of extra Al left in the Au droplet to the NW body during the cooling period. Thereafter, an Al composition of $\sim 30\%$ (±5%) is measured from various NWs along the whole NW body. The calculated bandgap energy for AlGaAs with the Al composition within the error margins corresponds to the emission wavelengths ranging from 665 to 714 nm and is overall in the same range as the observed PL peaks. Hence, the above structural characterization agrees with the PL measurement results.

![Fig. 2. NW-based all-optical n-bit full adder.](image)

(A) The optical image showing that, after a single combing, 10 AlGaAs and InP crossbar NW junctions exist in 30 µm × 30 µm area. The dashed white boxes highlight the locations where 10 parallel NAND and NOR logic gates can be constructed. (B) The truth tables of NOR and NAND logic gates using the polarization-dependent PL property of NWs. (C) The rules for obtaining the addition operation result from NAND and NOR logic gate outputs. (D) An example showing PL mapping of a single NW junction when the addition operation is performed [that is, 24 (110102) is added to 16 (011102)]. The addition operation result is obtained by following the rules shown in (C) (28).
As shown in Fig. 1B, the relative magnitude of the PL peaks depends strongly on the polarization direction of the excitation laser due to the one-dimensional structural confinement of NWs (18). When the polarization direction of the 532-nm excitation laser is parallel to the AlGaAs alignment axis (that is, horizontal; Fig. 1A), the PL signal emerges at the wavelength of 665 nm. In contrast, when the laser polarization direction is aligned along the InP alignment axis (that is, vertical; Fig. 1A), the PL signal is observed at the wavelength of 890 nm (28). On the other hand, when the laser at the 730-nm wavelength is used, the PL peak appears strongly with the laser polarization direction parallel to the InP alignment axis, while the PL peak nearly disappears at the corresponding vertical direction. The degrees of polarization (DOP) of 94 and 65% are obtained for InP and AlGaAs PL peak intensities, respectively, as shown in fig. S4. The slightly smaller DOP in the case of AlGaAs is attributed to the larger AlGaAs NW diameter than that of InP NWs (as described in the Supplementary Materials), therefore making these AlGaAs NWs less sensitive to the excitation laser polarization direction. Nevertheless, the crossbar NW junction allows the polarization direction of the laser to select whether InP or AlGaAs NW emits PL. Since their PL wavelengths (that is, 665 nm for AlGaAs NWs and 890 nm for InP NWs) are different, our crossbar NW junction is capable of operating as polarization-controlled all-optical wavelength switch (under 532-nm laser excitation) and polarization-controlled all-optical intensity switch (under 730-nm laser excitation). The dimensions of our structure are in subwavelength scale (NW diameter smaller than 120 nm), thereby suggesting that our crossbar NW junctions could be used in future on-chip nanophotonic networks. Further, we note that the observed switching effect originates from the linear optics, in contrast to previously reported all-optical NW-based switches (11, 14).

Using the polarization-dependent PL wavelength switch, we construct all-optical nanoscale NAND and NOR logic gates (depicted in Fig. 2A). Figure 2B shows the truth tables of our proposed all-optical NAND and NOR logic gates. For the NAND function, the excitation laser wavelength and the polarization direction mark the logic gate input state \((A, B)\), respectively, while the PL signal from the NW junctions returns the logic gate output (that is, \(C\), with \(j = 1\) implying the NAND function in the following discussion). The NAND function can be realized by defining the PL signal at the wavelength range covering only the bandgaps of AlGaAs and InP NWs as the NAND logic gate output [that is, \(\lambda_{PL} \in (620 \text{ nm}, 690 \text{ nm}) \wedge (860 \text{ nm}, 940 \text{ nm})\)]. As a result, if the PL signal appears in this wavelength range, the NAND gate returns an output of 1. In addition, an output of 0 (that is, a very weak PL signal) is only obtained if the NAND gate is excited by the 730-nm laser (corresponding to the input state of \(A_1 = 1\) with the polarization direction along the AlGaAs alignment axis (corresponding to the input state of \(B_1 = 1\)). For the NOR function, on the other hand, the gate input is coded similarly in the excitation laser wavelength \((A_j)\) and the corresponding laser’s polarization direction \((B_i)\), with the difference that the polarization direction in the gate input \(B_i\) is opposite to that of the NAND function. As illustrated in Fig. 2B, the PL signal at wavelength range, covering only the AlGaAs NWs, defines the NOR gate output (that is, \(C_{ji}\), with \(j = 2\) for the NOR function). As a result, the gate output of 1 is obtained if the PL signal emerges in this wavelength range [that is, \(\lambda_{PL} \in (620 \text{ nm}, 690 \text{ nm})\)]. Otherwise, the NOR gate returns an output of 0. Based on this definition, only the 532-nm laser excitation wavelength and the polarization direction along the alignment axis of AlGaAs NWs (corresponding to the input state of \(A_1 = 0\) and \(B_1 = 0\)) allows the NOR gate to return an output of 1 (28). Moreover, it is noteworthy to mention that, by defining our NW-based logic gate output similarly but the input differently, the NW junction can also be used for the construction of OR and AND logic functions (see fig. S5). Hence, this further highlights the multifunctional performance of our NW-based all-optical logic gates.

Our aligned NW structure allows light to perform arithmetic functions, such as addition operation. The result of an addition operation of any two bits (that is, \(A = A_r \ldots A_2 A_1\) and \(B = B_r \ldots B_2 B_1\)) can be obtained from the outputs of \(n\) pairs of parallel NAND and NOR logic gates. To simplify the situation, we first describe how our all-optical NAND and NOR logic gates enable the result of 1-bit addition operation to be unambiguously achieved. In case \(0_2\) is added to \(0_1\) (with subscript 2 defining the number as a binary number), the inputs of NAND and NOR gates (that is, \(A_1 = 0\) and \(B_1 = 0\)) will result in gate outputs of \(C_{11} = 1\) and \(C_{12} = 1\), respectively. On the other hand, if \(0_2\) is added to \(1_2\) (or \(1_2\) to \(0_2\)), the gate inputs of \(A_1 = 0\) (or 1) and \(B_1 = 1\) (or 0) will produce gate outputs of \(C_{11} = 1\) and \(C_{12} = 0\). Furthermore, the NAND and NOR logic gates will only return outputs of \(C_{11} = 0\) and \(C_{12} = 0\) when both gate inputs are 1 (that is, \(A_1 = 1\) and \(B_1 = 1\)). Therefore, the result (that is, \(D = D_2 D_1\) of the
two 1-bit addition operations can be unambiguously obtained from
the gate outputs since three different gate output configurations are
possible. By following the same order as above, the gate output con-
figuurations are defined to return a result of $D = 00_2$, $D = 01_2$, and
$D = 10_2$, as shown in Fig. 2C, which is consistent with the binary addi-
tion operation result (28). Note that a more complex situation is an
addition operation of two $n$-bit numbers ($A$ and $B$ as above), since
the possibility of carry information needs to be considered. Neverthe-
less, the carry information can be accounted for, and the result of the
addition operation can still be obtained from the outputs of $n$ pairs of
NAND and NOR logic gates by following the rules shown in Fig. 2C.

Figure 2D shows an example of the logic gate outputs, when a single
crossbar NW junction performs an addition operation of two 5-bit
numbers ($11010_2 + 01110_2$). In this calculation, the operation of a single
NAND and NOR gate is also presented under all four different
input conditions ($A_i, B_i$). As shown in PL maps of the NAND gate,
the PL signal (threshold value for PL $\sim 0.3$) emerges with the (0,0),
(1,0), and (0,1) gate inputs, while no PL signal arises under (1,1) input
state, in agreement with our proposed all-optical NAND function de-
scribed above. The operation of the NOR gate is proven similarly. As
shown in Fig. 2D, the PL signal (threshold value for PL $\sim 0.6$) is
generated only when the NOR gate input state is (0,0). Since all the
other gate input states return an output of 0, the realization of the
NOR function is experimentally verified. The result of the exemplary
addition operation can also be resolved by following the rules shown
in Fig. 2C, with the final carry information placed as the largest bit
(28). Applying the rules of Fig. 2C to PL maps, the correct number of
$101000_2$ is obtained for the $11010_2 + 01110_2$ binary number addi-
tion operation.

In addition to a high-sensitivity commercial micro-PL ($\mu$-PL)
system used in the previous demonstration, we build an optical system
that reads the gate output with a silicon photodiode to prove that our
all-optical NW-based logic gates can be realized at a system level.
Figure S6 shows the schematics of the proof-of-principle system-
level demonstration. The logic gate input is coded, as above, into the
excitation wavelength and the polarization direction, and a silicon
photodiode measures the gate output. Figure 3 shows the results. As
shown in Fig. 3, both the NOR and NAND gates return an output of
1 when the photodiode gives a signal larger than 10 $\mu$A (threshold
limit), in agreement with the truth tables shown in Fig. 2B.

It should be noted that $n$-bit subtraction and 2-bit multiplication
operations can also be performed with our multifunctional all-optical
NW logic gates. Subtraction operation, for instance, requires only the
largest bit of the $n$-bit addition operation to define the sign of the
number (that is, $A = -2^{-m}A_n + 2^{-m-1}A_{n-1} + \ldots + 2A_2 + A_1$), while
the 2-bit multiplication operations can be performed with the NW-based
all-optical logic circuit as shown in Fig. S7. As the multifunctionality of
these NW structures combined to scalable fabrication process suggests,
these nanostructures are interesting alternative candidates to be used
in future all-optical nanoprocessors.

CONCLUSIONS
In summary, we demonstrate a method to construct a nanoscale all-
optical wavelength switch based on the polarization-dependent PL
properties of the crossbar InP and AlGaAs NW networks. The struc-
ture is formed without any lithography process by combing InP and
AlGaAs NWs along the two perpendicular directions, allowing the
emission wavelength of the crossbar structure to be tuned over 200
nm by altering the excitation laser polarization direction. This po-
larization-controlled behavior allows the fabrication of multiple
parallel all-optical logic gates with subwavelength dimension. By using
the fabricated multifunctional all-optical logic gates with AND, OR,
NAND, and NOR functions, our structure successfully enables the
light to perform the binary arithmetic operations such as $n$-bit addi-
tion. Overall, our results underscore that such a NW network is an
interesting candidate for future all-optical nanophotonic devices. In
particular, the reported NW building block provides a scalable alter-
native to be used in future all-optical nanoprocessor circuits. These
conclusions were also drafted in the doctoral dissertation of the first
author (28).

MATERIALS AND METHODS
NW growth
InP and AlGaAs NWs were fabricated on Si (111) substrates inside
a horizontal-flow atmospheric pressure MOVPE system. Trimethyl-
yaluminim (TMAI), trimethyl gallium (TMGa), tertiarybutylarsine
(TBA), trimethylindium (TMIn), and tertiarybutylphosphine (TBP)
were used as precursors for AlGaAs and InP NWs. Diethylzinc (DEZn)
was used for the doping of AlGaAs NWs to enhance their PL proper-
ties. Before growth, 40- and 60-nm-diameter gold (Au) nanoparticles
were from a colloidal solution (BBI International) were dropcasted
as catalysts for the VLS growth of InP and AlGaAs NWs, respectively.

NW combing
After synthesis, the as-grown NWs were transferred onto a glass
substrate by using a nano-combing method (that is, mechanically
sliding the NW substrate over the target substrate along one direc-
tion). This technique allows the randomly directed NWs on silicon
substrate to self-align in the horizontal plane and point toward one
direction. Thus, the crossbar NW structures of InP and AlGaAs NWs
examined in this work were fabricated by combing InP NWs along
direction and AlGaAs NWs along the perpendicular direction. Note that,
after combing the NWs of the first material, a 100-nm-thick plasma-enhanced chemical vapor deposition (PECVD) SiO$_2$
layer was deposited (PECVD 80+, Oxford Instruments) to isolate InP
NWs from AlGaAs NWs.

TEM characterization
HRSTEM measurements were carried out with a JEOL 2200FS dou-
bble aberration–corrected field emission gun microscope operated at
200 kV. Elemental composition in the AlGaAs NW body was deter-
mined using a TEM-integrated EDX spectroscopy tool. To prepare
a sample for TEM study, a 400-mesh copper grid (3 mm in diameter)
covered with a holey supporting film was used to slightly scratch the
surface of the Si substrate with as-grown NWs, allowing individual
NWs to be found for structural analysis.

PL measurement
All $\mu$-PL measurements performed in this work were carried out at
room temperature with WITec alpha300S scanning near-field optical
microscopy under confocal operation mode. In these measurements,
linearly polarized 532- and 730-nm laser sources were used for ex-
citation. Polarization-dependent PL measurements were performed
by rotating the incident light polarization direction with a half waveplate.
The polarization angle was defined as the angle between the horizontal
direction (in all the shown images) and the direction toward which the electric field of the linearly polarized excitation laser was pointing. The PLQE numbers reported in this work were evaluated by comparing the PL intensity of NWs to that measured from the known reference sample (that is, Rhodamine 6G).

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/7/eaar7954/DC1

Section S1. NW growth
Section S2. NW combing
Section S3. TEM characterization
Section S4. PL measurements
Section S5. NW-based all-optical AND and OR gates
Section S6. System-level demonstration of logic gate operations
Section S7. NW-based multiplication operation
Fig. S1. SEM characterization of NWs.
Fig. S2. NW combing statistics.
Fig. S3. TEM characterization of AlGaAs NWs.
Fig. S4. Polarization dependent PL properties of NWs.
Fig. S5. Truth tables of AND and OR logic gates using the polarization-dependent PL properties of crossbar NW networks.
Fig. S6. Schematics of our system-level demonstration of the logic gates.
Fig. S7. NW-based all-optical 2-bit multiplier.

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