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Effective Surface Passivation of InP Nanowires by Atomic-Layer-Deposited Al₂O₃ with POₓ Interlayer

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Supporting Information

ABSTRACT: III/V semiconductor nanostructures have significant potential in device applications, but effective surface passivation is critical due to their large surface-to-volume ratio. For InP such passivation has proven particularly difficult, with substantial depassivation generally observed following dielectric deposition on InP surfaces. We present a novel approach based on passivation with a phosphorus-rich interfacial oxide deposited using a low-temperature process, which is critical to avoid P-desorption. For this purpose we have chosen a POₓ layer deposited in a plasma-assisted atomic layer deposition (ALD) system at room temperature. Since POₓ is known to be hygroscopic and therefore unstable in atmosphere, we encapsulate this layer with a thin ALD Al₂O₃ capping layer to form a POₓ/Al₂O₃ stack. This passivation scheme is capable of improving the photoluminescence (PL) efficiency of our state-of-the-art wurtzite (WZ) InP nanowires by a factor of ~20 at low excitation. If we apply the rate equation analysis advocated by some authors, we derive a PL internal quantum efficiency (IQE) of 75% for our passivated wires at high excitation. Our results indicate that it is more reliable to calculate the IQE as the ratio of the integrated PL intensity at room temperature to that at 10 K. By this means we derive an IQE of 27% for the passivated wires at high excitation (>10 kW cm⁻²), which constitutes an unprecedented level of performance for undoped InP nanowires. This conclusion is supported by time-resolved PL decay lifetimes, which are also shown to be significantly higher than previously reported for similar wires. The passivation scheme displays excellent long-term stability (>7 months) and is additionally shown to substantially improve the thermal stability of InP surfaces (>300 °C), significantly expanding the temperature window for device processing. Such effective surface passivation is a key enabling technology for InP nanowire devices such as nanolasers and solar cells.

KEYWORDS: Indium phosphide, semiconductor nanowires, surface passivation, atomic layer deposition, photoluminescence
stability. More critically, this native surface passivation is generally not maintained following the deposition of standard dielectric layers (e.g., SiO₂ or Al₂O₃) on the InP surface, as required for the fabrication of many types of devices. More recent attempts to passivate InP nanowire surfaces have encountered similar difficulties. Wet chemical treatments have been shown to provide some degree of surface passivation, but this is generally not stable. Münch et al. found that atomic layer deposition (ALD) of HfO₂ significantly degraded surface passivation regardless of surface pretreatment or deposition conditions. Dhaka et al. reported similar surface degradation following ALD of Al₂O₃, AlN, TiN, GaN, and TiO₂ on InP nanowires and pillars. Some improvement was observed for very thin Al₂O₃ films below a critical thickness of 2–3 nm, though carrier lifetimes remained well below 1 ns. Zhong et al. recently reported an increase in photoluminescence (PL) intensity and lifetime following plasma-enhanced chemical vapor deposition (PECVD) of SiNₓ on InP nanowires; however, they also observed a significant decrease in the open-circuit voltage of nanowire solar cells with the same films, making the significance of this result somewhat ambiguous. In sum, despite the acknowledged importance of surface recombination in InP nanowire devices, there remains no clearly established, stable, and effective surface passivation method for this material.

In attempting to devise an effective surface passivation scheme for InP nanowires, it is instructive to look back at historical work on the development of InP metal–insulator–semiconductor (MIS) devices. As noted above, early work showed that the deposition of standard dielectrics on InP tended to result in interfaces with poor electrical properties. It was conjectured that this might be linked to thermally induced degradation of the InP surface observed by various authors at temperatures ranging from 150 to 225 °C and thought to be linked to the desorption of surface P known to occur at least above 200 °C. Following this line of thought, or inspired by alternative structural considerations, a number of groups investigated the use of various P-rich layers, including deposition of Pₓ, POₓ, AlPₓOᵧ, InPₓOᵧ, POₙₓNᵧ films (usually by PECVD), growth of P-rich anodic oxides, or deposition of other dielectrics in P over-pressure to improve the interface characteristics of InP MIS structures. Such approaches appear to have been relatively successful, in most cases leading to significant improvements in MIS electrical characteristics. Since few of these groups employed PL characterization, it is not clear whether any of these approaches resulted in passivation improvements, or at least avoided degradation, relative to the native InP surface (those few who did perform such a comparison observed relative degradation), but such approaches nevertheless show significant promise as a path toward effective surface passivation of InP.

In this work, we adopt a novel approach inspired by this early work on InP MIS technology to develop an effective dielectric passivation scheme for InP nanowires. Specifically, we investigate the use of a deposited phosphorus oxide (POₓ) film to passivate the InP surface. Since phosphorus oxides are known to be hygroscopic and consequently unstable under ambient conditions (indeed uncapped POₓ films were observed to visibly degrade upon exposure to atmosphere), we encapsulate this layer with a thin ALD Al₂O₃ capping layer to form a POₓ/Al₂O₃ stack. We deposit both layers in an ALD reactor at room temperature (25 °C) using a plasma-assisted process in order to avoid thermal degradation of the InP surface. Using this novel surface passivation scheme, we are able to increase the PL internal quantum efficiency and PL lifetime to levels representing unprecedented performance for undoped InP nanowires. The investigated passivation scheme displays excellent long-term stability and is additionally shown to improve the thermal stability of InP surfaces, significantly expanding the temperature window for InP device processing.

The InP nanowires studied in this work were grown by catalyst-free selective-area vapor-phase epitaxy on Zn-doped InP (111)A substrates using a SiNₓ mask patterned by nanoimprint lithography. Growth was performed at 750 °C from trimethyl indium (TMI) and phosphine (PH₃) (PH₃/TMI molar ratio = 82) for 20 min. The wires were not intentionally doped. High-resolution transmission electron microscopy (TEM) showed that the wires were pure wurtzite phase, consistent with photoluminescence spectroscopy. The wires had a length of ~2.7 μm and a hexagonal cross-section with an average diameter of ~180 nm and were arranged in a square array with a 500 nm pitch. Figure 1 shows top-down and tilted SEM images of the nanowire array.

Deposition of the POₓ/Al₂O₃ thin film passivation stacks was performed in an Oxford Instruments FlexAL ALD reactor.

Figure 1. (a) Top-view and (b) 30° tilt SEM images of as-grown WZ InP nanowire array. (c) Bright-field TEM image of single nanowire with POₓ/Al₂O₃ passivation stack. (d) Bright-field and (e) HAADF scanning TEM images taken around the center of the same nanowire, which show the bilayer structure of the deposited amorphous POₓ/Al₂O₃ stack. (f) Expanded partial view of panel e.
Immediately prior to loading samples for deposition, they were immersed for 1 min in a 1% aqueous HF solution to remove the native oxide, followed by rinsing in deionized water. Phosphorus oxide (PO	extsubscript{x}) deposition was performed at 25 °C by exposing the samples alternately to trimethyl phosphate (TMP), (CH	extsubscript{3})	extsubscript{3}PO	extsubscript{4}, and an O	extsubscript{2} plasma, with separating N	extsubscript{2} purges, in an ALD-like manner. The deposition of ALD Al	extsubscript{2}O	extsubscript{3} capping layers was performed in situ immediately following PO	extsubscript{x} deposition from trimethyl aluminum [TMA, Al(CH	extsubscript{3})	extsubscript{3}] and O	extsubscript{2} plasma at the same temperature. More information on the deposition processes is given in the Supporting Information (SI). The use of a room-temperature deposition process was found to be critical to achieving the best passivation quality. Deposition of PO	extsubscript{x}/Al	extsubscript{2}O	extsubscript{3} stacks at higher temperatures (50 or 100 °C) resulted in consistently lower PL intensity and shorter PL lifetimes for both planar and nanowire InP samples compared to that obtained at room temperature (see SI). The thickness of the PO	extsubscript{x} and Al	extsubscript{2}O	extsubscript{3} layers was ∼5 nm and ∼16 nm, respectively (each process was run for 100 cycles), as determined by in situ spectroscopic ellipsometry on planar InP surfaces. The PO	extsubscript{x} layer appeared to be transparent within the measured range (<5 eV) with a refractive index of 1.67 at 632 nm.

Figure 1 shows TEM images of an InP nanowire after deposition of such a PO	extsubscript{x}/Al	extsubscript{2}O	extsubscript{3} thin film stack. From the bright-field images the films appeared to be amorphous, as also indicated by X-ray diffraction measurements of similar film stacks on planar Si surfaces. The distinct bilayer structure of the passivation stack is clearly apparent in the contrast of the high-resolution TEM images. High-resolution TEM imaging of the PO	extsubscript{x}/Al	extsubscript{2}O	extsubscript{3} stack was unfortunately found to be impossible due to a pronounced beam sensitivity of the PO	extsubscript{x} layer, which was observed to undergo rapid partial crystallization and delamination from the nanowire surface when exposed to higher fluxes of the electron beam (see TEM images in SI). This also precluded accurate analysis of the composition profile by energy-dispersive X-ray spectroscopy (EDX).

To establish the composition of the deposited layers, depth-resolved (sputtered) X-ray photoelectron spectroscopy (XPS) measurements (Thermo Scientific K-Alpha system) were performed on PO	extsubscript{x}/Al	extsubscript{2}O	extsubscript{3} stacks deposited in parallel on HF-etched, polished (100) InP wafer substrates. Figure 2a shows the compositional depth profile determined from these measurements, while Figure 2b shows the main photoelectron peaks at several representative depths. Consistent with the TEM measurements, the profile exhibits a distinct bilayer structure. Beneath the capping Al	extsubscript{2}O	extsubscript{3} there is a P-rich oxide layer, as expected. Surprisingly, however, this layer also appears to contain significant Al (Al–P–O ratio ≈ 0.5:1:4). This is despite the low deposition temperature and absence of postdeposition annealing which could promote Al diffusion. Near the substrate interface, indium oxides are also present in low concentrations, which can be attributed to the oxidizing effect of the O	extsubscript{2} plasma during the initial deposition cycles when the PO	extsubscript{x} film is still nucleating. The carbon concentration in the phosphate layer was ∼0.7%, close to the detection limit. Note that the relative concentration of both In and Al in the PO	extsubscript{x} layer is likely to be overestimated due to known selective sputtering of P with respect to both elements (this also accounts for the apparent nonstoichiometry of the InP substrate bulk: measurements of unsputtered InP wafers both as-received and after HF etching showed stoichiometric composition using the same sensitivity factors—see SI).

Figure 2. (a) Relative atomic composition determined by XPS as a function of sputtering time for the optimized PO	extsubscript{x}/Al	extsubscript{2}O	extsubscript{3} passivation stack on a polished InP (100) substrate. (b) P 2p, In 3d	extsubscript{5/2}, Al 2p, and O 1s photoelectron spectra for sputter times of 20, 240, and 330 s, respectively, corresponding to the Al	extsubscript{2}O	extsubscript{3} bulk, PO	extsubscript{x} bulk, and near-interface regions, respectively.

The apparent composition of the phosphate layer, as well as the binding energy of the bulk P 2p oxide peak (135.2 eV), are most consistent with a phosphorus-rich aluminum polyphosphate structure Al(PO	extsubscript{3})	extsubscript{x}, analogous with In(PO	extsubscript{3})	extsubscript{x}, most likely incorporating excess oxygen in the form of OH groups (see SI for additional discussion of the XPS data).

Steady-state and time-resolved photoluminescence (PL) measurements were performed to characterize the electrical and optical quality of the nanowires and the effectiveness of the surface passivation. In all cases the nanowires were measured as upright arrays on the growth substrate with excitation and detection parallel to the nanowire growth axis. A “positive aging” effect was noted for the passivated nanowire samples, whereby PL intensity and lifetime were observed to increase substantially over a period of weeks and months during sample storage under dark, ambient conditions. Such an effect has previously been noted for other semiconductor/dielectric interfaces, for example, Al	extsubscript{2}O	extsubscript{3}-passivated Si. It was found that this effect could be accelerated by postdeposition annealing at moderate temperatures. For instance, a 1 min anneal at 250 °C in N	extsubscript{2} shortly after PO	extsubscript{x}/Al	extsubscript{2}O	extsubscript{3} deposition was sufficient to increase the PL lifetime to a level similar to that obtained after months of aging at room temperature (see SI). Unless otherwise mentioned, the PL measurements of the PO	extsubscript{x}/Al	extsubscript{2}O	extsubscript{3}-passivated nanowires reported below represent stabilized values measured more than 7 months after PO	extsubscript{x}/Al	extsubscript{2}O	extsubscript{3} deposition, without annealing.

The stabilized passivation layers were found to significantly enhance the PL efficiency of the nanowires. The integrated steady-state PL intensity at room temperature (295 K) was...
observed to increase by a factor of ∼10−20 for the passivated wires compared to the as-grown wires depending on excitation intensity (Figure 3b), with the greatest increase observed at low excitation. The PL spectrum, shown in Figure 3a, is typical of pure WZ InP, with an emission peak close to 1.42 eV at room temperature. Interestingly, the dependence of PL intensity $I_{PL}$ on excitation intensity $I_{exc}$ for both as-grown and passivated wires closely follows a power law ($I_{PL} \propto I_{exc}^n$) over at least 7 orders of magnitude in PL intensity, with average exponents $n$ of 1.52 and 1.41, respectively [the local value of $n$ may be evaluated from the slope of log($I_{PL}$) vs log($I_{exc}$)]. These values can be considered as ideality factors in the one-diode equivalent circuit of the wires. While the physical interpretation of such apparent ideality factors should be treated with caution, increasing values in the range of 1 < $n$ < 2 are consistent with increasing nonradiative recombination associated with bulk or surface states.

In addition to the relative increase in PL efficiency apparent from Figure 3b, it is desirable to quantify the absolute PL internal quantum efficiency (IQE). This quantity relates directly to the efficiency potential of light-emitting devices and to the open-circuit voltage potential of photovoltaic devices. Yoo et al. have previously proposed that the PL IQE may be determined by fitting the room-temperature excitation-dependent PL intensity at high excitation levels with an idealized "rate equation" describing the recombination dynamics. Using this approach, Gao et al. have reported an IQE of ∼50% for as-grown, undoped WZ InP nanowires of a similar diameter (200 nm) to our own, which constitutes the highest IQE reported for such structures. If we apply the same analysis to our own data at similar high excitation levels, we derive an IQE of 61% for the as-grown wires and 75% for the passivated wires (see SI for details). Gratifying as such values would be, they are clearly not reliable given the much larger (~10 times) relative difference in PL intensity between the same samples, and thus serve rather to highlight the general unreliability of this method for determining the PL IQE.

An alternative method of determining the PL IQE which is widely used in the literature is to take the ratio of integrated PL intensity at room temperature (295 K) to that at low temperature (10 K in our case). In this approach it is assumed that the efficiency of radiative recombination is 100% at low temperature, where nonradiative processes are strongly suppressed. This assumption, though not always justified, is supported in our case by the $n = 1$ power dependence of the integrated PL intensity on excitation at 10 K for both the as-grown and passivated nanowires (Figure 3b). This provides strong evidence that the PL IQE determined in this way (Figure 3c) is reliable in the present case. Resulting PL IQE values of 3.5% for the as-grown and 27% for the passivated...
nanowires are obtained at the highest excitation levels. The latter represents an unprecedented level of performance for undoped WZ InP nanowires, taking into account differences in nanowire dimensions, as illustrated in Figure 3d. Time-resolved PL (TRPL) measurements provide additional support for the effectiveness of the investigated surface passivation scheme. As shown in Figure 4a, POx/Al2O3 passivation resulted in a significantly longer PL decay, evidencing an enhanced carrier lifetime due to reduced surface recombination. As observed previously for InP nanowires, this decay is nonexponential. We therefore choose to take the time constant characteristic of the initial, most rapid portion of the TRPL decay, which we designate as the excess carrier lifetime at the peak injection level of the measurement. This yields PL decay lifetimes of 1.8 and 5.4 ns, respectively, for the as-grown and passivated nanowires, indicating a 3 times increase in carrier lifetime due to passivation. Conversely, nanowires with only the Al2O3 layer (i.e., without the POx interlayer) displayed a significantly reduced PL lifetime of 0.3 ns, showing the importance of the POx layer for passivation. The PL decay characteristics were observed to be dependent on excitation intensity (Figure 4b). While such a dependence is expected, its exact form is rarely measured or reported. In Figure 4b we also show carrier lifetimes reported in the literature for undoped WZ InP nanowires of similar diameter to our own (160, 200, 220, 240, and 335 nm). None of these wires were deliberately passivated. It is interesting to
observe that the highest values fall just below, but very close to, the data for our own as-grown wires. Since the dimensions of our wires are similar, this suggests that the bulk quality of these wires is comparable to the state-of-the-art. The large increase in lifetime observed for the passivated wires, however, shows that the lifetime of the as-grown wires is still significantly surface-limited. This is contrary to the conclusions of some authors that surface recombination is practically negligible for such wires.\textsuperscript{49,58} The lifetimes observed for the passivated wires appear to be the longest so far reported for InP nanowires by a significant margin. Li et al.\textsuperscript{45} have reported lifetimes of up to 7.4 ns for WZ InP micropillars; however, these had a diameter greater than 1 μm, which significantly reduces the influence of surface recombination. Given the significantly larger (microscale) dimensions of these structures we do not include this result in the comparison of Figure 4b.

It is desirable to quantify the contribution of surface recombination in the passivated and unpassivated wires. This is not straightforward, because the contribution from bulk recombination processes for such WZ InP nanowires is not known. For a cylindrical geometry (and when diffusion is not limiting), the effective surface recombination velocity \( S_{\text{eff}} \) is given by\textsuperscript{60} \( S_{\text{eff}} = (D/4) (r_{\text{eff}}^{-1} - r_{\text{s}}^{-1}) \) where \( D \) is the nanowire diameter, \( r_{\text{eff}} \) is the effective excess carrier lifetime, and \( r_{\text{s}} \) is the bulk lifetime due to radiative and nonradiative processes. An upper limit on \( S_{\text{eff}} \) may be derived by assuming that all recombination occurs at the nanowire surface (\( r_{\text{s}} = \infty \)). If we take \( r_{\text{eff}} \) as equal to the highest measured \( r_{\text{min}} \) for the passivated wires (6.5 ns, see Figure 5), we derive an upper limit to \( S_{\text{eff}} \) of 690 cm/s. For the as-grown wires (\( r_{\text{min}} = 1.9 \) ns), we similarly derive an upper limit to \( S_{\text{eff}} \) of \( 2.4 \times 10^3 \) cm/s. A lower limit to \( S_{\text{eff}} \) for the as-grown wires may be derived by assuming that \( r_{\text{s}} \) is equal to the maximum measured lifetime for the passivated samples. This yields a lower limit of \( S_{\text{eff}} \) of \( 1.7 \times 10^3 \) cm/s for the as-grown wires. This value is an order of magnitude higher than the value of 170 cm/s reported by Joyce et al.,\textsuperscript{58} which they derived from transient terahertz photoc conductivity measurements of as-grown WZ/ZB InP nanowires with a range of diameters, but it is comparable to the value of \( 2.7 \times 10^3 \) cm/s which may be derived in the same way from the diameter-dependent TRPL data of Tedeschi et al.\textsuperscript{59} for pure WZ wires (see SI). Note that in general \( S_{\text{eff}} \) is expected to be dependent on dopant concentration.\textsuperscript{61}

The passivated nanowires were found to be significantly more thermally stable than the as-grown wires. As mentioned earlier, the InP surface is known to be unstable at elevated temperatures, undergoing P-desorption and associated electrical degradation at temperatures above at least 200 °C.\textsuperscript{18−20,62} In fact, time-resolved PL measurements, which are extremely sensitive to surface quality, show that the onset of electrical degradation occurs already between 100 and 150 °C for “bare” (native oxide only) nanowire and planar InP surfaces, as shown in Figure 5 for such samples annealed at various temperatures for 1 min in N₂. In contrast, the PL lifetime of POₓ/Al₂O₃-passivated samples was initially lower but was observed to increase with annealing temperature between 100 and 300 °C, reaching values significantly higher (up to 6.5 ns) than those of the unannealed, unpassivated samples. Although the lifetimes of the passivated samples also declined at temperatures above 300 °C, they remained higher than those of the unpassivated samples annealed at the same temperatures, indicating an improvement in the thermal stability of the surface. Figure 5 also shows that the lifetime of samples coated with the same Al₂O₃ film without a POₓ interlayer remained well below those of the unpassivated surface regardless of annealing temperature up to 400 °C, emphasizing the critical role of the POₓ interlayer to passivation. The fact that similar trends are observed both for the undoped WZ nanowires and for heavily doped n-type (100) ZB InP is a nontrivial result and suggests the effectiveness of the investigated passivation scheme for InP surfaces in general. The improved thermal stability of POₓ/Al₂O₃-passivated surfaces should allow the use of higher thermal budgets for InP device fabrication without compromising surface electrical properties, thereby expanding the temperature window for InP device processing.

Finally, we note that the fact that we obtain good passivation of InP with a P-rich interfacial oxide runs counter to some recent thinking on InP surface passivation. It is not uncommon in the more recent InP passivation literature to find studies in which the goal is the elimination of P oxides, ostensibly for the purposes of surface passivation.\textsuperscript{54,63} For example, on the basis of a comparison of capacitance−voltage and XPS measurements of Al₂O₃ and H₂O interfaces with InP, Galatage et al. concluded that the formation of P- rich oxides was probably harmful for InP passivation.\textsuperscript{66} Density functional theory studies have been interpreted in a similar light.\textsuperscript{57} Our results suggest that, on the contrary, P-rich oxides may be beneficial for InP surface passivation.

In conclusion, we have reported evidence of the effective surface passivation of InP nanowire and planar surfaces by POₓ/Al₂O₃ thin-film stacks deposited at room temperature. The application of this passivation scheme allows us to achieve PL internal quantum efficiencies and lifetimes significantly higher than previously reported for wurtzite InP nanowires of similar diameter. The passivation displays excellent long-term stability and is additionally shown to significantly improve the thermal stability of InP surfaces. Further improvements in passivation may be possible by optimizing the surface pretreatment and POₓ/Al₂O₃ film thicknesses in combination with postdeposition annealing conditions. Such passivation layers are a key enabling technology for InP nanowire device applications such as nanolasers and solar cells.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.7b02972.

Details of POₓ/Al₂O₃ deposition process and POₓ optical properties, instability of POₓ layers during TEM, XPS spectra of planar InP surfaces at various stages of the passivation process, further discussion of XPS profile results, experimental details of PL measurements, evolution of POₓ/Al₂O₃ passivation with storage time, influence of deposition temperature on passivation quality, details of PL IQE extraction according to the method of Yoo et al., calculation of expected diameter dependence of PL IQE, detailed excitation dependence of time-resolved PL decay at low excitation levels, extraction of \( S_{\text{eff}} \) from the data of Tedeschi et al. (PDF)

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ABBRREVATIONS

ALD: atomic layer deposition; PL: photoluminescence; ZB: zinc blende; WZ: wurtzite; MIS: metal-insulator-semiconductor; PECVD: plasma enhanced chemical vapor deposition; TMII: trimethyl indium; TEM: transmission electron microscopy; SEM: scanning electron microscopy; HF: hydrofluoric acid; TMP: trimethyl phosphate; SE: spectroscopic ellipsometry; HAADF: high angle annular dark field; EDX: energy dispersive X-ray spectroscopy; XPS: X-ray photoelectron spectroscopy; TRPL: time resolved photoluminescence; IQE: internal quantum efficiency.

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