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Interfacial transport homogenization for nanowire ensemble photodiodes by using a tunneling insertion

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Interfacial transport inhomogeneity critically degrades the performance of nanowire ensemble photodiodes. In this work, an ultrathin Al2O3 insertion is introduced to improve the photoresponse including stable response, a high on/off ratio, and a quick response ascent/descent. Homogeneous tunneling across the insertion dominantly controls the transport fluctuation originated from the inconsistent interfacial states of individual nanowires. The present work demonstrates a progressive practical application of nanowire ensemble devices. © 2013 American Institute of Physics. [http://dx.doi.org/10.1063/1.4795317]

Optoelectronic functionalities of semiconductor nanowires have been intensively studied. A representative prototype is a Schottky photodiode (PD) based on a single nanowire,1–4 whose interfacial transport is crucial. In practical applications, a PD necessarily integrates with multiple nanowires,5,6 and the photoresponse performance relies on the interfacial transport of individual nanowires in the ensemble. Unfortunately, electrical inhomogeneity occurs, because individual transport conditions are inconsistent with multiform geometries,7 instantaneous interfacial states,8 etc. This situation negatively affects the stability and reproducibility of the photoresponse and other functionalities. Therefore, homogenizing the interfacial transport of nanowire ensemble devices is highly significant. Modification by introducing a dielectric insertion is a valuable approach for thin-film PDs, which can adjust barrier heights, passivate surface states, and prevent metallic diffusion.9–12 Similarly, a core-shell nanowire synthesis has been adopted in a nanowire PD.13 However, few works have focused on the unfavorable influence of interfacial transport inhomogeneity and relevant modifications. The homogenization function of insertions for performance stability in nanowire ensemble devices needs to be well understood.

In this work, an ultrathin Al2O3 layer is inserted between the ZnO nanowires and the Schottky contact of a nanowire ensemble PD. The performance is improved to exhibit a stable on/off ratio of 1600 and quick decay time of 0.1 s, without steep fluctuations under illumination. Theoretical models are applied to analyze interfacial transport, demonstrating that homogenized tunneling across the insertion benefits the stability.

The ZnO nanowires used in this work were grown by vapor transport method and dispersed into a nanowire ensemble for the fabrication process. The nanowire ensemble PDs were fabricated by applying lateral contacts as in a previous work,15 and the Al2O3 insertion was carried out by atomic layer deposition (ALD; BENEQ TFS200). In this process, an Al film was evaporated on the nanowire ensemble to form an electrical contact on one lateral side. A structurally matched alumina layer was formed through a transfer step and selective anodic oxidation. A 2 nm-thick Al2O3 was deposited, prior to Au Schottky contact formation on another lateral side of nanowires. The nanowire ensemble PD was thus fabricated by introducing an ultrathin Al2O3 insertion. As a counterpart, an unmodified Schottky PD without the insertion was also prepared. Corresponding morphology was observed by atomic force microscopy (AFM; Veeco IMS). Current–voltage (I–V) and photoresponse properties were measured by a semiconductor parameter analyzer (Agilent 4156C).

In the PD architecture, individual nanowires are electrically circuited in parallel [Fig. 1(a)], which is geometrically similar to practical devices integrated with multiple nanowires.5 The elementary cross-sectional structure of the modified PD is asymmetrical along the diametrical direction of the nanowires, as shown in the inset of Fig. 1(a). Al2O3/Au forms a modified contact on the top side, and Al forms ohmic contact on the bottom side. An alumina layer insulates the two contacts and separates each nanowire. Patterned SiO2 beneath Au is used to configure the window of one cell.

Photo-generated carriers in the nanowires are dominated to drift under the diametrical distribution of the electrical field.3,16 Considering that the nanowire geometries are not the same, the field distribution and induced carrier transport of individual nanowires are inhomogeneous.7 The active dangling bonds on the surface lead to transient changes in the interfacial states. These facts result in the interfacial transport inhomogeneity of the nanowire ensemble PD without a modification.

In the case of the three-dimensional morphology of nanowire ensembles, a high-quality modification through the Al2O3 insertion requires a rigorous regularity of structure at the nanoscale level. The fabrication15 affords a well planarization to fulfill the deposition requirement. AFM is used to evaluate the morphology before ALD. The topography [Fig. 1(b)] shows only a few nanometers in profile at the top
side of the nanowire. Therefore, a uniform Al$_2$O$_3$ insertion can be deposited to modify the interfacial transport and photoresponse performance of the nanowire ensemble PD.

The photoresponse, measured under switching illumination, is investigated. On/off ratios [Fig. 2(a)] are described as the ratio of photocurrent to dark current at reverse 2 V. An unstable performance is found in the unmodified PD, which commonly occurs in other reported Schottky PDs. In contrast, the modified PD exhibits stable response with an on/off ratio of 1600. The smoothing photocurrent is achieved with the Al$_2$O$_3$ insertion and contributes to the stability under illumination.

Both the ascent and descent of photoresponse in the modified PD are also improved. Decay time is summarized at varied reverse bias in Fig. 2(b). The decay time of the modified PD indicates a rapid recovery of approximately 100 ms. The distinct behavior excludes the slow equilibrium of interfacial states involved with ambient molecule absorption/desorption. Consequently, the stable and quick photoresponse, which agrees with the temporal trace of illumination, illustrates the modification of Al$_2$O$_3$ insertion for practical applications.

To characterize the interfacial transport, I–V properties are measured. Rectified dark currents are observed in the unmodified and the modified PDs, as shown in Fig. 3(a). A thermionic emission (TE) model is applied to examine the forward currents and is expressed by

![FIG. 1. (a) Top photomicroscopic image of one cell of the modified PD. The inset is the schematic of the cross-sectional structure. (b) Topography of the surface before depositing Al$_2$O$_3$.](image1)

![FIG. 2. (a) On/off ratio profiles of the unmodified PD and the modified PD at reverse 2 V bias with switching illumination. The dotted line is the temporal trace of illumination. (b) Decay time, plotted at varied bias voltages, is defined as the time required for the current to decrease from 90% to 10% of the maximum.](image2)

![FIG. 3. (a) I-V curves of the unmodified and the modified PDs in the dark and under illumination. (b) The photocurrent of the modified PD re-plotted on ln(I/V) versus 1/V, and on ln(I/V$^2$) versus ln(1/V) is shown in the inset. The grey areas approximately describe the two transport regimes.](image3)
where \( n \) is the ideality factor, \( I_s \) is the reserve saturation current, and \( R \) is the series resistance considering the insertion. The ideality factors calculated from the slope of \( \ln(I) \) versus \( V \) are 1.73 and 13.8, respectively. The bit deviation from unity in the unmodified PD implies partial thermionic transport due to the interfacial states at the Schottky contact and dangling bonds on the ZnO nanowires. A high value of this factor in the modified PD corresponds to the present insertion. The large band offset between Al\(_2\)O\(_3\) and ZnO makes carriers tunnel through the ultrathin insertion rather than transport over it.

The electrical parameters of both PDs are listed in Table I. The reverse current is further inhibited in the modified PD, which is attributed to interfacial passivation and an additional barrier of the insertion. The reduced rectification and the increased turn on voltage are caused by the introduced resistivity. These results indicate that the Al\(_2\)O\(_3\) insertion modifies the intrinsic interfacial transport of the nanowire ensemble PD.

Significant photoresponse is observed under illumination in both PDs. The current substantially increases at reverse bias. In contrast to the TE-related current of the unmodified Schottky PD (not shown here), the current under illumination is involved with tunneling in the modified PD. In this study, two tunneling models are applied for analysis, including direct tunneling and Fowler-Nordheim (F-N) tunneling, respectively, expressed as

\[
I = I_s \exp \left( \frac{q(V - IR)}{n k T} - 1 \right),
\]

(1)

where \( d \) is the barrier width, \( m_e \) is the effective carrier mass, and \( \Phi \) is the barrier height. Two distinct regimes are distinguished, and the reverse current under illumination is replotted in Fig. 3(b). At low bias (0–0.2 V), the linear slope agrees with Eq. (2) in the inset of Fig. 3(b). Meanwhile, a relationship of Eq. (3) at relatively high bias (0.8–2 V) indicates F-N tunneling transport. This behavior suggests that the photoresponse of the modified PD originates from the tunneling transport.

To elucidate the interfacial transport homogenization for the nanowire ensemble PDs, the photoresponse mechanisms are discussed. In the unmodified PD, photo-generated carriers are separated by the built-in field of the Schottky diode and the external bias, as illustrated in Fig. 4(a). Electrons drift to Al contact and become the minor photocurrent, whereas holes migrate to the interface of ZnO nanowires. Several holes discharge the negative charges of absorbed oxygen. The discharged oxygen changes the interfacial states and decreases the barrier height. Thus, induced interfacial transport over the lowered barrier contributes to the response under illumination. The potential barrier is transiently affected by the unbalanced ionized charges near the interface. Inconsistent interfacial conditions, under which individual nanowires settle, result in transport inhomogeneity. The steep inhomogeneity critically distorts the photocurrent, thereby degrading the photoresponse performance.

In the modified PD, the Al\(_2\)O\(_3\) insertion attenuates the electron wave function in Al\(_2\)O\(_3\) before ZnO nanowires and forms a valence band barrier rather than a Schottky barrier, as illustrated in Fig. 4(b). Photo-generated carriers separate along a potential gradient. The holes accumulate at the Al\(_2\)O\(_3\)/ZnO interface because of the valence band offset. These holes can directly transmit through the ultrathin barrier at a specific bias, which follows direct tunneling. At high bias, the shape of the barrier transits to a triangle. This condition prompts the holes to tunnel through the thinner section of the barrier, which can be interpreted by F-N tunneling. The tunneling transport at the interface contributes to the response, with only dependence on the external bias and the thickness of the insertion. The transport of individual nanowires is thus homogeneous at a fixed bias attributed to the uniformity and conformity of the Al\(_2\)O\(_3\) insertion. This transport homogeneity results in stable response. The Al\(_2\)O\(_3\) insertion also isolates the ZnO nanowire ensemble from ambient atmosphere, resulting in a quick response. Consequently, the Al\(_2\)O\(_3\) insertion modifies the photoresponse mechanism based on the homogeneous tunneling and improves the performance with stable and quick photoresponse of nanowire ensemble PDs.

In conclusion, interfacial transport homogenization is performed by an ultrathin Al\(_2\)O\(_3\) insertion within a ZnO nanowire ensemble PD. The homogeneous tunneling across the uniform insertion layer, following direct and F-N models, effectively controls the photoresponse fluctuations under inconsistent interfacial conditions of individual nanowires. Photoresponse stability is thus achieved to meet practical photodetection as well as a high on/off ratio beyond three orders of magnitude and a rapid recovery of 100 ms. The introduced insertion eliminates the negative influence of electrical inhomogeneity on
devices and suggests developing potentials for diverse applications based on nanowire ensembles.

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