

Polarization-dependent reflectivity from dielectric nanowires

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The presence of GaN nanowires grown primarily normal to the surface of a sapphire substrate has a dramatic influence on the polarization dependence of laser light reflectivity at $\lambda = 1550$ -nm wavelength. Even at 12% substrate surface coverage, there is a factor of 2 enhancement in polarization dependence of reflectivity relative to bulk sapphire at values of incident angle greater than $\phi = 72^\circ$. © 2003 American Institute of Physics. [DOI: 10.1063/1.1598283]

The recently reported ability to control the growth of GaN nanowires on conventional substrates¹ opens up the possibility of a new class of nanostructured material with new types of functionality not normally found in bulk material. In this letter, we demonstrate an enhanced polarization dependence of reflectivity due to the presence of GaN nanowires on a sapphire substrate.

Controlled growth of single-crystal GaN nanowires on a sapphire substrate is achieved using chemical vapor deposition. The catalyst used is monodispersed gold clusters with a range of diameters.

The preparation of the GaN nanowires proceeds by first depositing a 3-nm-thick layer of Au on a clean sapphire substrate via e-beam evaporation. Surface mobility of gold ensures that the deposited metal film is discontinuous. Following metal deposition, the substrate is placed in a quartz tube at one end of a furnace heated to 900 °C. The Au nanoparticles that form are used as a catalyst for subsequent GaN growth. Pure Ga (99.9999%, Alfa Aesar) is placed downstream in the quartz tube. A typical growth sequence involves the flow of anhydrous NH_3 gas through the quartz tube at a rate of 100 sccm for about ten min. The synthesis of GaN is based on the vapor–liquid–solid growth mechanism, where the Ga vapor first diffuses into the gold catalytic particles and then grows out and reacts with NH_3 to form GaN once the Ga/Au alloy reaches supersaturation. Continued addition of Ga into the Ga/Au nanoparticle feeds the GaN growth, and hence one anticipates the diameter of the GaN nanowire is directly linked to the Au catalyst particle size. Growth of GaN nanowires takes a few minutes, after which the system is allowed to cool down and the sample is removed from the quartz tube. Following this procedure, the sapphire substrate is coated with a layer of material that has a white appearance. This layer of material was confirmed to consist of single-crystal GaN nanowires by scanning electron microscopy (SEM), transmission electron microscopy (TEM), and selected area electron diffraction.

Figure 1 shows a typical SEM image of the high-yield GaN nanowires grown from the gold catalyst. These nanowires cover the substrate surface and appear to be homogeneous in diameter. There is some disorder both in the separation between nanowires and their orientation with respect

to the substrate normal. Detailed TEM and SEM examination show that these nanowires have average diameters near 70 nm and length near 10 μm , indicating an aspect ratio exceeding 100:1. The estimated fraction coverage of wires is $\alpha = 0.10$ (10%).

Because the band gap of both GaN and sapphire are much larger than the photon energy of $\lambda = 1550$ -nm wavelength light, one expects little, if any, absorption for light incident at this wavelength. However, the nanoscale structure of the GaN should influence the polarization dependence of reflectivity at large scattering angles. To explore this possibility, we developed an anisotropic dielectric layer model of reflectivity of linearly polarized light. The incident angle and reflected angle is ϕ with respect to the substrate normal (see inset in Fig. 2). The fractional coverage of dielectric nanowires on the sapphire substrate is α . The GaN nanowires form an effective thin film on the surface of the sapphire substrate. The dielectric constant of the effective thin film parallel to the interface is ϵ_{\parallel} , and the dielectric constant perpendicular to the interface is ϵ_{\perp} . The ratio of anisotropic dielectric constants used for calculating the theoretical curves is $\epsilon_{\parallel}/\epsilon_{\perp} = 1/8$.

From the SEM image shown in Fig. 1, we can see that the separation between the dielectric nanowires is quite random and the effective dipole moments of the nanowires are not equal. It is, therefore, a valid assumption that the elec-

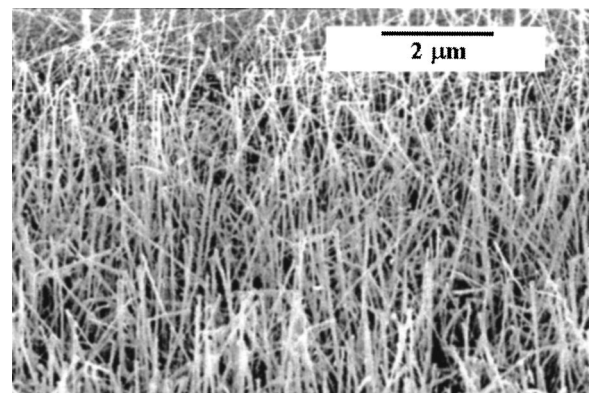


FIG. 1. A typical SEM image of the high yield GaN nanowires grown from the gold catalyst. These nanowires cover the substrate surface and appear to be homogeneous in diameter. Detailed TEM and SEM examination show that these nanowires have diameters near 70 nm and an aspect ratio exceeding 100:1.

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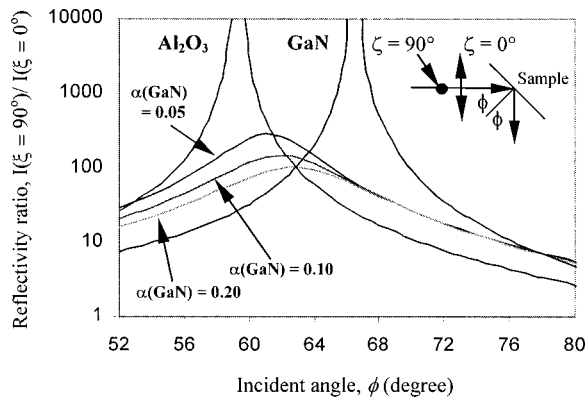


FIG. 2. The ratio of reflection intensity $I(\zeta=90^\circ)/I(\zeta=0^\circ)$ as a function of incident angle ϕ for a sapphire substrate with the indicated fractional coverage of GaN nanowires. Also shown is the result for a pure sapphire substrate and a pure GaN substrate. The inset shows the reflection geometry.

tromagnetic field reradiated from the nanowire dielectric layer loses phase information from the incident electromagnetic wave. Hence, we model the reflected electromagnetic wave from the sapphire interface as incoherent with respect to the electromagnetic wave from the nanowire layer. For polarization in the plane of the substrate normal and incident electromagnetic wave propagation direction ($\zeta=0^\circ$), the collected light intensity from the sample plus the substrate $I(\zeta=0^\circ)$ is obtained from the incoherent scalar addition of the electromagnetic field intensity from the sample and sap-

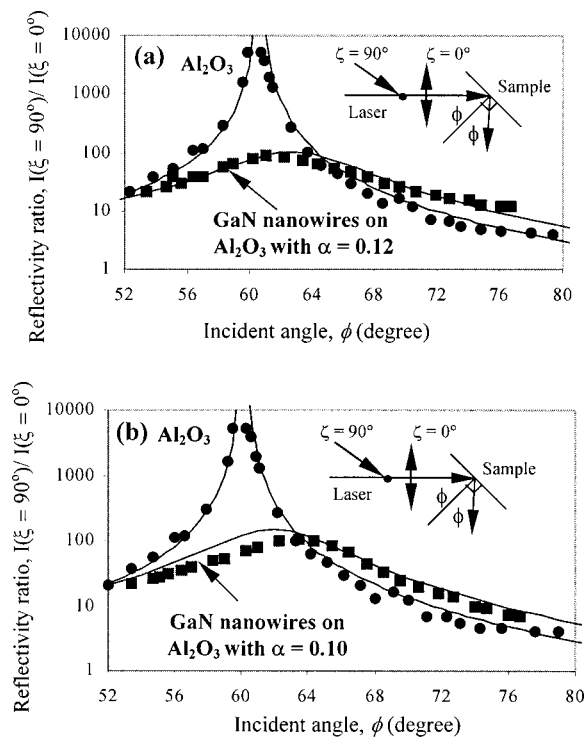


FIG. 3. (a) Squares are a result of measuring the ratio of reflection intensity $I(\zeta=90^\circ)/I(\zeta=0^\circ)$ as a function of the incident angle ϕ for a sapphire substrate with $\alpha=0.12$ fractional coverage of GaN nanowires (sample A). Dots are the result of measuring of the same ratio but for a pure sapphire substrate. Solid lines are the predictions of the model calculation. (b) Squares are the result of measuring the ratio of reflection intensity $I(\zeta=90^\circ)/I(\zeta=0^\circ)$ as a function of the incident angle ϕ for a sapphire substrate with $\alpha=0.10$ fractional coverage of GaN nanowires (sample B). Dots are the results of measuring the same ratio, but for a pure sapphire substrate. Solid lines are the predictions of the model calculation.

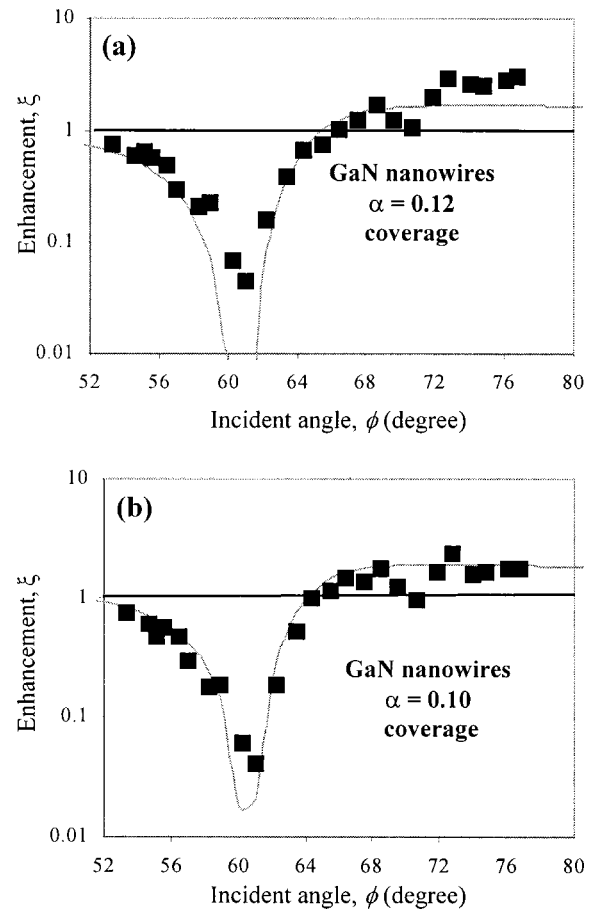


FIG. 4. (a) Measured enhancement ratio $\xi(\phi)$ is shown as squares, and the predictions of the model calculation are shown as a solid line when GaN nanowire coverage is $\alpha=0.12$ (sample A). For values of incident angle $\phi > 72^\circ$ the enhancement ratio $\xi(\phi) > 2$. (b) Measured enhancement ratio is shown as squares and the predictions of the model calculation as a solid line when GaN nanowire coverage is $\alpha=0.10$ (sample B).

phire substrate weighted for fractional coverage. For polarization perpendicular to the plane of the substrate normal and incident electromagnetic wave propagation direction ($\zeta=0^\circ$), the dielectric nanowires are not polarized and consequently the reflected electromagnetic wave intensity $I(\zeta=90^\circ)$ is the same as reflection from a pure sapphire substrate.

Figure 2 shows the results of calculating the ratio of reflection intensity $I(\zeta=90^\circ)/I(\zeta=0^\circ)$ as a function of incident angle ϕ for a sapphire substrate with different fractional coverage of GaN nanowires. Also shown is the result for a pure GaN substrate. The inset in Fig. 2 shows the reflection geometry. The sapphire (refractive index $n_{Al_2O_3}=1.75$) and GaN (refractive index $n_{GaN}=2.3$) substrate have a peak in $I(\zeta=90^\circ)/I(\zeta=0^\circ)$ at the Brewster angle $\phi_B(Al_2O_3)=\tan^{-1}(n_{Al_2O_3})=60.3^\circ$ and $\phi_B(GaN)=\tan^{-1}(n_{GaN})=66.5^\circ$, respectively. As the fractional coverage α of GaN nanowires on sapphire increases, the peak in the ratio shifts to larger angles. In addition, the ratio increases with increasing α for large values of ϕ . This clearly shows that polarization is enhanced at large angles due to the presence of the dielectric nanowire structure.

We have explored the predictions of our model by performing experiments that measure the ratio $I(\zeta=90^\circ)/I(\zeta=0^\circ)$. In these experiments, the wavelength of linearly po-

larized laser light is $\lambda = 1550$ nm, and the spot size diameter of the laser beam on the sample is 1.2 mm. The GaN nanowire sample is grown within a circle of 3-mm diameter. The incident laser power is set to 1 mW. The system is calibrated by measuring the angular reflection from an Al mirror.

In Fig. 3(a), the squares show results of measuring the ratio of reflection intensity $I(\zeta = 90^\circ)/I(\zeta = 0^\circ)$ as a function of the incident angle ϕ for a sapphire substrate with an estimated $\alpha = 0.12$ fractional coverage of GaN nanowires (sample A). Dots are the results of measurement of the same ratio, but for a pure sapphire substrate. Solid lines are the predictions of the model calculation. In Fig. 3(b), the squares are the results measuring the ratio of reflection intensity $I(\zeta = 90^\circ)/I(\zeta = 0^\circ)$ as a function of the incident angle ϕ for a sapphire substrate with an estimated $\alpha = 0.10$ fractional coverage of GaN nanowires (sample B). Dots are results of measurement of the same ratio, but for a pure sapphire substrate. As in Fig. 3(a), solid lines are the predictions of the model calculation.

Figure 4(a) shows the enhancement ratio for experimental data as squares and the predictions of the model calculation as a solid line when coverage $\alpha = 0.12$ (sample A). We define an enhancement ratio $\xi(\phi) = [I_\phi(\zeta = 90^\circ)/I_\phi(\zeta = 0^\circ)]_{\text{GaN}}/[I_\phi(\zeta = 90^\circ)/I_\phi(\zeta = 0^\circ)]_{\text{Al}_2\text{O}_3}$. As anticipated by the model, the value of $\xi(\phi)$ near the Brewster angle for sapphire $\phi_B(\text{Al}_2\text{O}_3) = 60.3^\circ$ is suppressed. However, for

large incident angles, there is an enhancement in $\xi(\phi)$. For $\phi = 72^\circ$, the enhancement ratio is $\xi = 2.0$. Figure 4(b) shows the enhancement ratio for experimental data as squares and the predictions of the model calculation as a solid line when coverage $\alpha = 0.10$ (sample B). In this case, the enhancement ratio at $\phi = 72^\circ$ is $\xi = 1.68$.

In conclusion, we have demonstrated that, even at low surface coverage, the presence of GaN nanowires grown primarily normal to the surface of a sapphire substrate significantly enhances the polarization dependence of laser light reflectivity at $\lambda = 1550$ -nm wavelength. This demonstrates a functionality of nanostructured material different from previous experimental and theoretical work that has focused on enhanced resonant Raman scattering in single wall carbon nanotubes,² the theory of field enhancement in metal-filled carbon nanotubes,³ and optical reflection spectroscopy in ZnCdSe/ZnSe nanowire heterostructures.⁴

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