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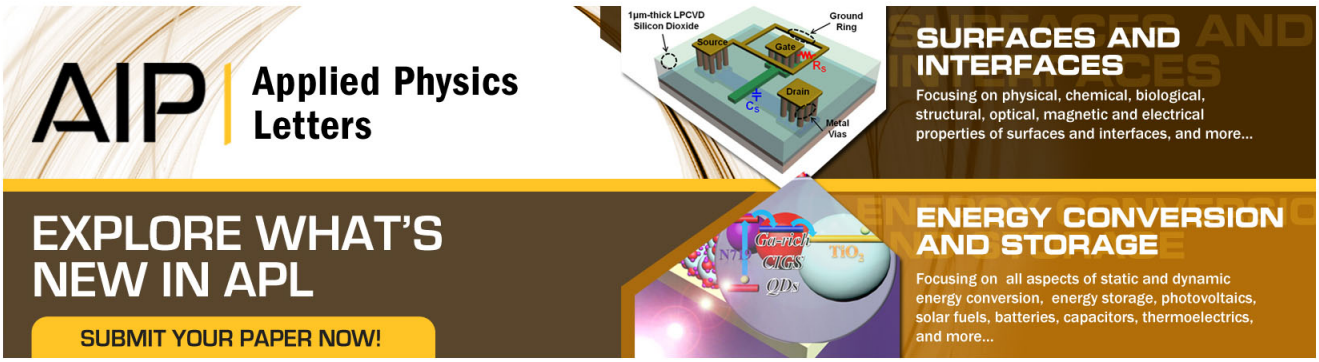

Ultrafast all-optical gated amplifier based on ZnO nanowire lasing

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We present an ultrafast all-optical gated amplifier, or transistor, consisting of a forest of ZnO nanowire lasers. A gate light pulse creates a dense electron-hole plasma and excites laser action inside the nanowires. Source light traversing the nanolaser forest is amplified, partly as it is guided through the nanowires, and partly as it propagates diffusively through the forest. We have measured transmission increases at the drain up to a factor 34 for 385-nm light. Time-resolved amplification measurements show that the lasing is rapidly self-quenching, yielding pulse responses as short as 1.2 ps. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4733972]

All-optical computing is potentially much faster than conventional electronic computing. For the development of ultrafast all-optical computing, ultrafast all-optical transistors and logic gates are required. However, nonlinear optical effects on which ultrafast all-optical components must be based are almost invariably very weak in conventional materials, thereby limiting possible applications. Solutions are therefore pursued in specially designed materials, such as plasmonic nanorod metamaterials[1] and periodically poled lithium niobate crystals,[2] where these nonlinear effects are greatly enhanced.

Lasing in ZnO nanowires forms an interesting opportunity in this context. ZnO nanowires have been shown to exhibit strong laser action between about 385 nm and 390 nm under optical excitation.[3–6] This laser action requires gain lengths of a few micrometers only. Indeed, modal gain lengths in this range have recently been measured inside single ZnO nanowires.[7] Time-resolved measurements on lasing ZnO nanowires have been performed using several techniques: optical injection probing,[8,9] Kerr gating,[10,11] sum-frequency gating,[12] and by using a streak camera.[13–15] All reports show that under strong excitation, the laser action lasts very short: its duration can be shorter than 2 ps. This short lasing time contrasts with the luminescence lifetimes of tens or hundreds of picoseconds that were measured below the laser threshold.

Here, we present an ultrafast all-optical gated amplifier consisting of a forest of ZnO nanowire lasers. Its operation at room temperature is analyzed by time-resolved amplification measurements. A source light pulse is strongly amplified by a forest of ZnO nanowire lasers if it arrives shortly after an excitation gating pulse. We have measured an on-off ratio of 34 at the drain and a pulse response as short as 1.2 ps.

Coupling a light pulse into a single nanowire is cumbersome and suffers from severe losses. In order to obtain robust amplification, we have chosen to use a dense forest of ZnO nanowires as gated amplifier. Nanowire forests exhibit exceptionally small reflectivity losses, so virtually all input light propagates into the forest.[16–19] Using a nanowire forest as gated amplifier also has the virtue that the source light strongly scatters at the nanowires and the seed film. The resulting diffusive motion of the source photons through the forest increases the average time photons spend inside the sample from values around 100 fs to values around 1 ps.[20] The increased length over which gain takes place critically enhances the total amplification of the transistor.

A scanning-electron-microscope (SEM) image of our transistor is shown in Fig. 1. This is the same nanowire forest as used for earlier light diffusion measurements.[20] The nanowires were epitaxially grown on a sapphire substrate, using the carbothermal method.[21] First, a porous ZnO seed film was created. On top of that, the nanowires were grown, with their crystal *c*-axes parallel to the wires. The wires of our transistor are all about 20 μm long. Their diameters vary between 100 and 500 nm with an average of 250 nm. We measured the nanowire density to be 0.85 μm^{-2} and the ZnO filling fraction 0.08.

The nanowire forest transistor is gated by a 125-fs, 800-nm pulse from an amplified Ti:sapphire "Hurricane" laser. Since 800-nm photons have an energy of 1.55 eV and the band gap of ZnO is 3.37 eV at room-temperature, 800-nm absorption in ZnO is a three-photon process.[22–25] At high intensities, this three-photon excitation leads to the formation of a dense electron-hole plasma, whereby optical gain and possibly laser action take place.[25–28] The advantage of using three-photon excitation is that the nanowire forest can be excited over its entire thickness in an approximately homogeneous way. Alternatively, gating by ultraviolet above-band-gap pulses is possible, but the small penetration depth of ultraviolet light (50 nm) in ZnO limits the fraction of the system that is excited and thereby the amplification that is obtained.

In our experiment, a source pulse traverses the nanowire forest with a tunable delay with respect to the 800-nm gating pulse. As source pulses we used 125-fs pulses of four wavelengths: 385 nm, 395 nm, 405 nm, and 415 nm, each with 2-nm bandwidth. These pulses were created via white-light generation in a sapphire crystal and subsequent sum-frequency generation by mixing this white light with 800-nm light in a beta barium borate crystal, allowing frequency selection by angle tuning.[29] If the source pulse arrives at the nanowire forest transistor after it has been gated by the

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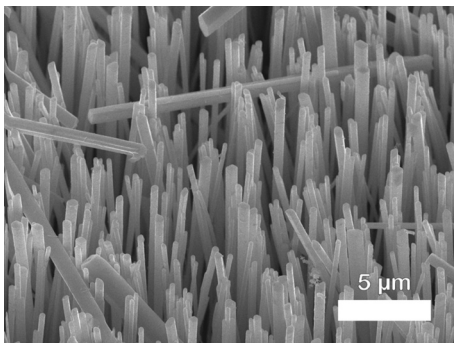

FIG. 1. SEM image of our ultrafast all-optical gated amplifier: a forest of 20 μm long ZnO nanowire lasers. Reprinted with permission from M. A. M. Versteegh, R. E. C. van der Wel, and J. I. Dijkhuis, Appl. Phys. Lett. **100**, 101108 (2012). Copyright © 2012 American Institute of Physics.

excitation pulse, the stored electronic energy is drained and the source light is amplified. This amplification occurs partly as source light is guided through the excited wires, and partly as scattered source light diffuses through the excited forest. Transmitted source light is collected by a lens and measured by a photodiode and a lock-in amplifier. To calibrate our results, we measured the fraction of transmitted light that is collected by the lens. We divided our measurement results by that fraction to get the total transmission.

Results of the ultrafast signal response of our optical transistor for all four source wavelengths are presented in Fig. 2. For gate fluences above 200 J/m^2, the transistor opens up. We observe increased signal transmission when the source pulse arrives at the nanowire forest after the gate pulse. The amplification of the signal rapidly increases with gate fluence and we observe a strong wavelength dependence. At 629 J/m^2, we observe for 395-nm source light a factor 7 increase in transmission, from 0.25 to 1.7. For 385 nm, an on-off ratio as high as 5.3/0.15 = 34 is found.

Fast decay of the amplification becomes visible for fluences above 300 J/m^2. The response becomes faster with increasing gate fluence. At 629 J/m^2, the duration of the 385-nm amplification is only 1.2 ps (full width at half maximum). The remarkable strength of the observed amplification, in combination with its very short duration, makes this ZnO nanowire forest especially suitable as an ultrafast optical UV amplifier.

For zero delay, the transmission of the source pulse is reduced (Fig. 2). This is caused by two-photon absorption of a source photon and a gate photon, simultaneously present inside the sample. This effect forms the mechanism of our ultrafast bulk ZnO all-optical shutter.[29] Inside a ZnO nanowire forest, the same phenomenon can be used to measure the photon diffusion.[20]

The fast decay of amplification we observe for high gate fluences signifies rapid decay of charge carriers. This fast carrier decay can be ascribed to laser action. The amplification in our experiment (Fig. 2) is so strong that lasing must occur in the nanowire forest, mediated by the electron-hole plasma. This interpretation is confirmed by the emission spectra shown in Fig. 3, which were measured on a similar ZnO nanowire forest. The sharpening of the emission peak for increasing excitation intensity agrees with a gradual transition from spontaneous emission to lasing.

Studies on the quantum efficiency of ZnO nanowires[30,31] show that below the laser threshold the external luminescence quantum efficiency is at most 10%–20%, which means that most of the charge carriers decay nonradiatively. Above the laser threshold, however, the external quantum efficiency rises to 60% and the internal quantum efficiency even to 85%.[30] Apparently, the laser action is so efficient that carrier decay by lasing dominates nonradiative losses, including nonlinear effects as Auger recombination. This increasing quantum efficiency is also observed in our

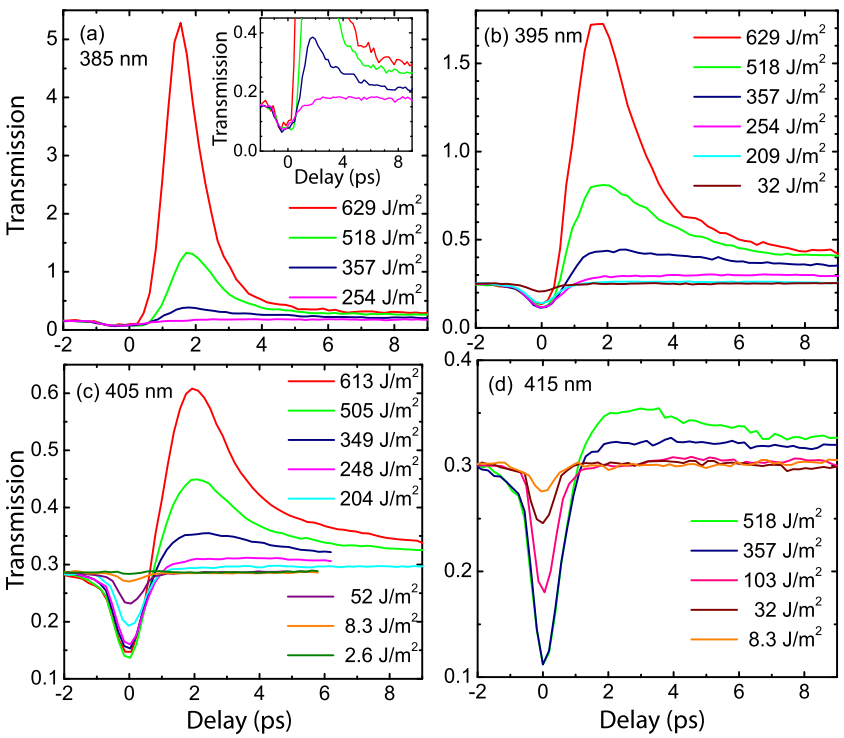

FIG. 2. Time-resolved amplification experiment on the ZnO nanowire forest shown in Fig. 1: Measured transmission of a source pulse vs delay with respect to the 800-nm excitation gate pulse for (a) 385 nm, (b) 395 nm, (c) 405 nm, and (d) 415 nm source wavelength and specified gate fluences.

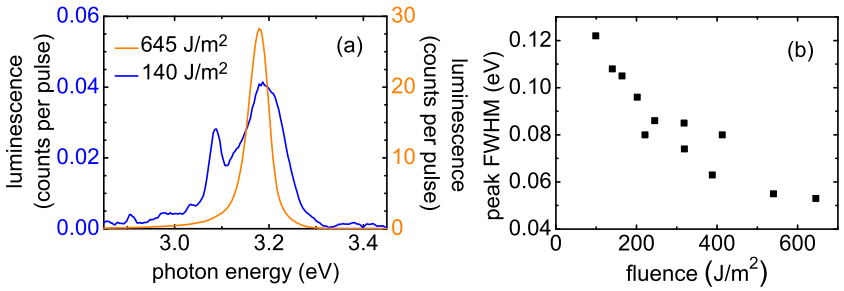

FIG. 3. Time-integrated photoluminescence experiment on a similar ZnO forest under 800-nm excitation. (a) Measured emission spectra, showing an electron-hole recombination peak centered at 3.2 eV and a second harmonic peak centered at 3.1 eV. The scale on the left corresponds to the data at 145 J/m^2. The scale on the right corresponds to the data at 645 J/m^2. (b) Width (FWHM) of the electron-hole recombination peak vs excitation fluence.

experiments, where we found that the time-integrated luminescence intensity due to electron-hole recombination (so excluding the second harmonic peak at 3.1 eV) increases more strongly than proportional to F^3, where F is the excitation fluence. We conclude that at high excitations, there is a self-quenching gain: strong stimulated emission causes the majority of the carriers to rapidly recombine radiatively, leading to an ultrafast reduction of the gain. The gain quenching is assisted by the fact that the reflectivities at the wire ends are small.[32,33] Self-quenching gain also explains the short lasing times in ZnO nanostructures reported in Refs. 8–15.

In conclusion, we have demonstrated that a forest of ZnO nanowire lasers acts as an ultrafast all-optical gated amplifier. Three-photon absorption of an 800-nm gating pulse leads to a net amplification of an incident 385-nm source pulse up to a factor 5 and an on-off ratio of 34 at the drain. The strong optical gain and lasing in the nanowire forest are rapidly self-quenching. The amplification time can be as short as 1.2 ps. The gating fluences needed are 300-600 J/m^2, low enough to allow gating of the fast transistor by high-repetition-rate mode-locked lasers. This nanowire all-optical UV transistor may have applications in optical computing and can be used in ultrafast pump-probe experiments. Nanowire forests of other direct semiconductors should permit nanowire optical transistors at other wavelengths.

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