Semiconductor nanowires (NWs) are promising candidates for a large variety of novel optoelectronic devices [1, 2]. These applications include efficient solar cells [3], nanoscale sensors [4], and nanoscale lasers [5–7]. The nanowire geometry offers several intrinsic advantages for realizing nanoscale lasers. It naturally provides a waveguide with effective reflectivity’s at the end facets forming a laser cavity. Strong confinement of the guided mode, in addition to material gain of the semiconductor, enables lasing in nanowires which are even thinner than the vacuum wavelength of the emitted light [5]. Stimulated emission from single semiconductor NWs on a low refractive index substrate has been demonstrated upon optical pumping in the single or multiphoton absorption regime at room temperature [8, 9]. Furthermore, arrays of NWs, in which individual wires are either aligned vertically [10–13] or randomly oriented [11, 14, 15] also showed lasing. It has been theoretically predicted that inter-wire coupling might play an important role for lasing in arrays [16–18]. However, experimental proof of the influence of coupling on the emission properties of NW arrays remain absent. Here, we present results of comparative experimental investigations on stimulated emission from vertically aligned NW arrays consisting of wires with different lengths, diameters, and spacing between the wires, shedding light onto the role of coupling effects.

Vertically aligned, single crystalline ZnO NW arrays were synthesized using a vapor phase transport (VPT) technique within a horizontal tube furnace [19, 20]. Briefly, the source material, a mixture of ZnO and carbon powder (molar ratio 1:1), was evaporated in the hot zone of a tube furnace at 1050 °C for the growth of the NW arrays. The vapor was transported by a carrier gas (7 sccm Ar + 10 sccm O2) downstream towards
the growth substrates following the temperature gradient. The NWs were grown self-catalytically using the vapor–liquid–solid (VLS) approach epitaxially on top of a 500 nm Al-doped ZnO seed layer covering a Si substrate [21]. The pressure within the tube was kept constant at 10 mbar for the synthesis of the NW arrays, and the growth substrates where placed at different positions in the tube furnace holding different growth temperatures and yielding into different NW geometries.

The properties of the arrays, such as length, diameter, and inter-wire spacing were evaluated using a scanning electron microscope (SEM), as shown in figures 1(a) and (b). The diameters and lengths of the wires used in the experiments varied in the range from 100 to 850 nm and from 1.4 to 14 µm, respectively. The growth method used for the array production features correlation between the wire diameter and the spacing between the wires. The measured dependence of the inter-wire spacing, and the wire diameter is shown in figure 1(c). It reveals that thinner wires are closer to each other, which is explained by the competitive growth mode for our synthesis parameters [22].

The NW arrays were optically excited by 805 nm central wavelength, 45 fs duration, 1 kHz repetition rate, up to 0.7 mJ energy pulses delivered by a Ti:Sa amplifier system. Considering a bandgap of 3.37 eV for ZnO, the excitation of electrons from the valence to the conduction band is achieved by three photon absorption [23, 24]. The beam was focused by a lens, down to a beam diameter of ~0.75 mm on the target, with a maximum intensity of 1.5 TW cm\(^{-2}\). The intensity on the target was controlled using a variable retardation plate and a thin film polarizer. The emission in the near UV (NUV) was collected with a 4 x microscope objective (NA 0.4) and imaged onto the entrance slit of a spectrometer (Ocean Optics USB 4000). All experiments were carried out at ambient conditions and room temperature.

The spectrally integrated near ultra-violet (NUV) signal and its spectral width (at full-width half-maximum (FWHM)) were estimated from the measured emission spectra in order to study the dependence of the emission intensity and the lasing onset in arrays as a function of average NW length, diameter and inter-wire distance. These values are summarized in figure 2 for different intensities of the pump pulse and two different NW arrays. Two characteristic emission line shapes are observed in the experiment. The broadband spontaneous emission has a spectral width almost independent of the pump intensity over the entire range of pumping intensities used in the experiments (figure 2(a)). The emission spectra reveal a sudden narrowing at a certain threshold pump intensity (figure 2(b)). This spectral narrowing manifests the transition from spontaneous emission to stimulated emission [25]. This transition is further confirmed by an increasing slope in the log–log scale dependence of the emission yield on the pumping intensity at the threshold value (figure 2(c)) and by time-resolved measurements, as found in [11].

For low pump intensities, the broadband spontaneous emission centered at ~385 nm originates from combined action of exciton and free electron recombination [26]. Previous investigations of spectral dynamics in NW array pumped via three-photon absorption suggest that, for pump intensities around the lasing threshold, the density of the excited carriers well exceeds the Mott density at room temperature, such that the emission then originates from electron–hole plasma (EHP) recombination [11, 27]. In short wires the gain is not enough to compensate the cavity losses. Thus, no lasing is observed even at the highest pump intensities used in the experiment, which are close to the threshold for permanent degradation of the arrays. Usually, lasing in individual nanowires, with a diameter less than 1 µm, shows Fabry–Perot mode structure (FPM) [7], whereas the mode structure in microwires includes also whispering gallery modes [28]. In the case of NW arrays, FPM are not visible because the mode structure in the spectrum is washed out due to statistical variations of wire length in the arrays [29]. Increasing the pump intensity above the threshold results in a broadening (figure 2(d)) and red shift of the central emission wavelength (figure 2(c)) in the lasing spectrum [29], and a saturation of the emission yield (figure 2(c)). The broadening can be explained by a widening of the spectral gain profile [23]. The observed redshift for higher pump intensities is attributed to the band gap renormalization in the EHP [23]. The saturation of the three-photon-absorption process explains the saturation in the NUV emission yield [24]. Note that all these spectral changes of NW arrays with increasing pump intensity have been observed for individual wires as well. Therefore, we suppose that the observed lasing thresholds in arrays can be explained and qualitatively described by non-interacting individual NWs with the average length and diameter of the array.

Stimulated emission in a single ZnO NW becomes dominant, if the roundtrip loss in the wire is compensated by the optical gain. This threshold gain \(g_\text{th}\) is given by:

\[
\Gamma_{g_\text{th}} = \alpha_\text{w} + \frac{1}{2L} \ln(R_1R_2),
\]

with the confinement factor \(\Gamma\) describing the overlap of the guided mode inside the wire with the gain material [7]. The loss on the right-hand side is the sum of the waveguide loss \(\alpha_\text{w}\), and the mirror losses, which are determined by the reflectivity on the wire-air (R1) and wire-substrate end facet (R2), and the length of the wire L [30]. Since the diameter of the NW is in the order of the wavelength, the mode confinement, the end facet reflectivity, and the transverse mode structure depend strongly on the wire diameter [7, 9]. We assume that the transversal mode structure is dominated by the HE\(_{11}\) and/or TE\(_{01}\) mode for wire diameters below and above 200 nm, respectively [31]. Figure 3(a) provides an estimation of the reflectivity and confinement factor for different ZnO wire diameters. The values for wire diameters up to 400 nm have been taken from [7] and extrapolated for larger diameters assuming the plain wave field structure instead of a mode. For the reflectivity on the ZnO-Si-substrate interface a constant value of 0.19 was assumed.

We calculated the threshold gain for NWs with different lengths and diameters using equation (1), the quantities for reflectivity and confinement factor given in figure 3(a) and neglecting the waveguide losses \(\alpha_\text{w}\). The estimated threshold gain, when varying the wire length from 1 to 14


µm and the diameter from 100 to 900 nm, is depicted in the contour plot.

Figure 3(b). Furthermore, the experimentally investigated arrays showing stable laser oscillations are indicated by blue dots, while arrays with absent lasing are depicted in red. Thus, only arrays consisting of wires with average diameters >200 nm and/or lengths >4 µm showed stimulated emission in the experiments. The corresponding gain thresholds for arrays showing lasing are in the range of 0.1–0.5 µm⁻¹, which is in a good agreement with previously reported numbers for single ZnO NW [7] with geometrical parameters, similar to the average geometrical parameters of wires in our array samples. Figure 3(b) provides, in fact, a rough estimate for the gain achievable in ZnO NW arrays under conditions of three-photon absorption. For example, the absence of lasing in experiments with NW arrays having the average diameter ≈300 nm and the length ≈2 µm implies that the gain value remains below the calculated threshold value ≈1 µm⁻¹. Therefore, our results might benchmark the onset of population inversion by three-photon absorption in intense off-resonant ultrashort laser pulses like the ones developed in [23, 24]. The threshold gain calculated using equation (1), for

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**Figure 1.** (a) and (b) SEM images of two different arrays of vertically aligned thick and thin ZnO NWs with 430 and 150 nm mean diameters, respectively. (c) Measured inter-wire distance as a function of the NW diameter for samples grown under different conditions. The red line is a guide to the eye and reveals that diameter and inter-wire distance are coupled. The error bars show the standard derivative of measured nanowire parameters.

**Figure 2.** (a) Emission spectra from a representative NW array do not showing the transition from spontaneous to stimulated emission. (b) Evolution of the emission spectra characteristic for NW arrays showing stimulated emission. (c) Pump intensity dependent integrated emission intensity and (d) FWHM of the NUV emission from a lasing ZnO NW array. The red lines are a guide to the eye illustrating the increase of the slope indicating the transition to stimulated emission.
the average wire parameters in the arrays, used in the experiments and showing lasing (blue dots in figure 3(b)), is plotted versus the measured pump threshold intensity in figure 4. The horizontal error bars are determined by uncertainties in estimating the pump threshold intensity from the experimental data and the vertical error bars account for variation of the gain threshold due to statistical variations of the wire parameters within different arrays. The data coincides with a linear regression. Therefore, the lower threshold gain values for an average single wire in the array corresponds to the lower pump intensity needed to achieve lasing. This indicates that single NW properties indeed determine the lasing capabilities of NW arrays in our experiments. Hence, the role of inter-wire coupling on lasing properties of an array remains an open question. Coupling effects are expected when wires are in close vicinity, enabling efficient interaction via the evanescent fields. The spatial scale of evanescent fields as a function of the NW diameter is estimated using the solution for eigenmodes in a sub-wavelength step index cylindrical waveguide [32]. It is expected that coupling effects are the strongest in arrays with thin wires, due to the lower field localization and the shorter inter-wire distance for thin grown wires (figure 1(c)). Moreover, it can be expected that coupling between the wires might enable lasing for wires with diameters less than 200 nm, where single wire lasing is not possible. For a single nanowire with a diameter of 100 nm (radius 50 nm), the magnitude of the evanescent field halves after 30 nm measured from the surface. This corresponds to less than half the mean free space of 100 nm between the nanowires. Furthermore, the efficiency of the inter-wire light coupling depends on the mutual phase shift of the guided modes and thus on the propagation constant $\beta$ [33]. The propagation constant $\beta$ for cylindrical nano-waveguides depends on the NW diameter and the refractive index [17], which, for excited ZnO NWs, is a function of the carrier density [23, 34]. For the investigated samples, diameters of individual wires and thus the propagation constant of neighboring wires are not identical, resulting in a significant phase mismatch which finally leads to a low coupling efficiency. Note, the coupling of the guided modes is considered in perfectly periodic NW arrays, where the formation of photonic crystal Bloch modes result in a photonic band structure [12, 35, 36].

Thus, we conclude that the onset of stimulated emission in ZnO NW arrays is mainly dictated by the geometrical parameters of the single wires in the array and influence of interwire coupling is negligible for disordered samples.

In conclusion, we have investigated the emission properties of VPT-grown vertically aligned ZnO nanowire arrays pumped via three-photon absorption, for a wide range of average wire diameters and lengths. Arrays with an average NW diameter of more than 200 nm and a length exceeding 4 $\mu$m showed stimulated emission exceeding a well-defined pump intensity threshold. All these array observations are in excellent agreement with estimations made for a single NW with average geometrical parameters of the array and with
published experimental results for single NWs with similar parameters [7]. Our experimental results suggest that coupling effects play a negligible role for lasing in the VPT-grown arrays, especially for thin wires with diameters less than 200 nm where the mode localization is relatively weak. This is explained by the phase mismatch between the modes in individual wires due to the statistical variations in the geometrical parameters and related to their dynamic variations of the refractive index during the pumping/emission process. Since lasing in NWs sets in when the density of the excited carriers exceeds a threshold value, determined by the material and geometry and independent from the excitation mechanism, it is possible to quantitatively compare the efficiency of different multiphoton absorption mechanisms using the evaluated lasing threshold as a diagnostic. Thus, we believe that these results are important for understanding and measuring the nonlinear absorption properties of semiconductor NWs.

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