

Manipulation and assembly of nanowires with holographic optical traps

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We demonstrate that semiconductor nanowires measuring just a few nanometers in diameter can be translated, rotated, cut, fused and organized into nontrivial structures using holographic optical traps. The holographic approach to nano-assembly allows for simultaneous independent manipulation of multiple nanowires, including relative translation and relative rotation.

Semiconductor nanowires [1, 2] are emerging as versatile building blocks for the assembly and fabrication of a wide range of nanoelectronic and nanophotonic devices [3–5]. To date, the properties of simple nanowire-based devices have been determined using nanowires deposited on the surface of a substrate either at random or else by directed assembly controlled by flowing fluids or electric fields [6–8]. These latter approaches represent a significant advance over random assembly, yet remain limited in that the end-to-end registry and three-dimensional (3D) orientation of nanowires are not controlled, thus precluding the rational assembly of more complex architectures with interesting and potentially useful functional properties. Here we describe the use of holographic optical traps (HOTs) [9] as a general approach for parallel manipulation and assembly of nanowires in 3D. The HOT technique can create hundreds of independently controlled optical traps that can manipulate mesoscopic objects in 3D [10, 11]. We demonstrate that cadmium sulfide (CdS) nanowires with cross-sections at least as small as 20 nm can be isolated, translated, rotated and deposited onto a substrate with HOT arrays. We also exploit spatially localized photothermal and photochemical processes induced by the well-focused traps to cut nanowires and to fuse junctions. These capabilities have been used to assemble nontrivial structures, thus demonstrating the substantial potential for assembling and subsequently investigating the functional properties of complex and previously inaccessible structures.

We synthesized CdS nanowires by the laser ablation technique via the gold catalyzed vapor-liquid-solid (VLS) growth mechanism [1]. The nanowires range from 50 to 150 nm in diameter with lengths ranging from 10 to 40 μm . The nanowires were then suspended in ethanol by mild sonication. Deionized water was added to the suspension (20% v/v) prior to the experiments to prevent the rapid evaporation of the solution, which can lead to the deposition of nanowires on the bottom glass surface. These samples then were charged into slit pores roughly 40 μm thick formed by bonding the edges of #1 glass coverslips to the surfaces of microscope slides.

Sealed nanowire samples were mounted for observation and manipulation on the stage of a Nikon TE-2000U microscope outfitted with a 100 \times NA 1.4 Plan Apo oil-

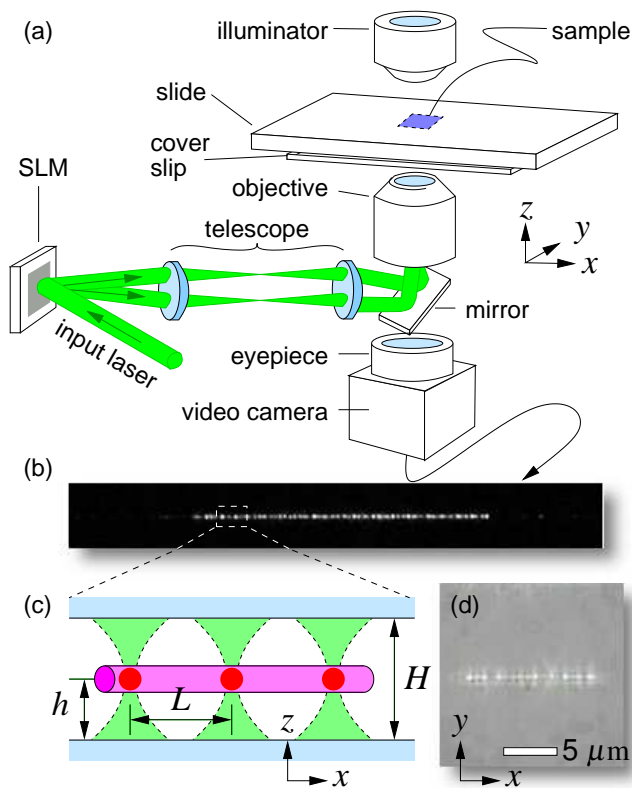


FIG. 1: Holographically trapping semiconductor nanowires. (a) The light from a frequency-doubled solid-state laser is imprinted with a computer-generated hologram by a phase-shifting spatial light modulator (SLM) before being relayed to the input pupil of a high-numerical-aperture objective lens, which focuses the light into an array of optical traps, shown in (b). (c) An individual semiconductor nanowire can be localized by multiple optical traps, whose intersection with the wire typically is visualized by intense laser-induced fluorescence, as in (d).

immersion objective. This lens is used both to collect bright-field images of the dispersed nanowires and also to focus light from a continuous wave (CW) frequency-doubled Nd:YVO₄ laser operating at 532 nm (Coherent Verdi) into optical traps (Fig. 1(a)). To create a large number of diffraction-limited optical traps, we utilized the holographic optical tweezer (HOT) technique, as described previously. Our implementation uses a liq-

uid crystal spatial light modulator (SLM) (Hamamatus X8260 PPM) to imprint a computer-designed phase-only hologram encoding the desired array of traps [11] onto the laser beam's wavefronts. Each trap in the array can be translated independently in three dimensions by projecting a sequence of holograms encoding the sequence of intermediate trapping patterns. Interactive assembly was performed with a BioRyx 200 holographic optical trapping system (Arryx, Inc.), also operating at 532 nm, with an integrated MicroPoint pulsed laser cutter (Phononic Instruments) operating at 440 nm.

In our approach, nanowires dispersed in a fluid medium on the stage of a light microscope are organized into structures by projecting computer-designed patterns of diffraction-limited optical traps using the dynamic HOT technique [9–11] (Fig. 1). HOT micromanipulation relies on a generalization of the single-beam optical gradient force traps known as optical tweezers that can capture mesoscopic objects in 3D [12]. An individual optical tweezer is not effective for trapping highly asymmetric structures, however, and appears to be incapable of moving our semiconductor nanowires at laser powers below 0.1 W. Increasing the power to increase the trapping force also induces rapid heating and the evolution of vapor bubbles whenever the focal point passes through a nanowire, and to visible changes in the nanowires themselves, including bending, formation of nodules, and even scission. This is consistent with heating due to absorption in the substantial photon flux passing through the micrometer-scale focal volume.

To exert more force on the nanowires while minimizing radiative damage, we project large numbers of holographic optical traps along the length of each nanowire. The image in Fig. 1(d) shows a freely floating semiconductor nanowire ca. 15 μm long captured by an array of holographic optical traps with an inter-trap separation of 0.4 μm . Once aligned and localized in the array of traps, the nanowire can be translated at speeds up to $u = 10 \mu\text{m}/\text{sec}$ by moving the array across the field of view (e.g., Fig. 2(a)) or by moving the sample stage relative to the array. This upper bound can be used to estimate the optical trapping force. The drag on a cylinder of length L and radius a traveling through an unbounded fluid of viscosity η at low Reynolds number is [13]

$$F_\infty = 4\pi\eta (\epsilon + 0.193\epsilon^2 + 0.215\epsilon^3) Lu, \quad (1)$$

where $\epsilon = [\ln(L/a)]^{-1}$. This sets a lower limit on the optically applied force of 0.2 fN/trap for the ca. 80 nm diameter CdS nanowire used in this measurement. The actual drag is substantially enhanced by the need to satisfy no-flow boundary conditions at the nearby coverslip, which is $h \approx 0.5 \mu\text{m}$ away from the nanowire's center. To lowest order in a/h , the corrected drag is [14],

$$F(h) = \frac{F_\infty}{\ln\left(\frac{2h}{a}\right)}, \quad (2)$$

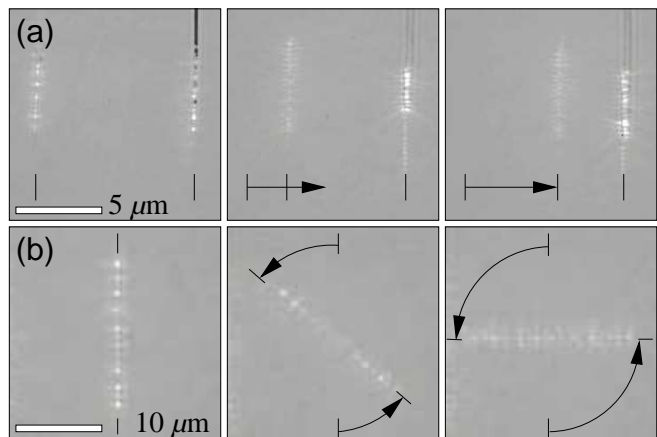


FIG. 2: Translation and rotation of semiconductor nanowires by holographic trap arrays. (a) Two free-floating semiconductor nanowires translated toward each other with parallel arrays of holographic optical traps. One wire is held stationary in one line of traps while the other is translated by moving a second line of traps in discrete steps of 700 nm. The traps in each line are separated by 0.4 μm and each trap is powered by 3 mW. (b) Rotating a semiconductor nanowire by rotating an array of traps in discrete steps of 5°. The optically trapped CdS nanowires in these sequences appear bright because of photoluminescence excited by the strongly focused optical traps. Because these images are created with a filter that blocks the bandgap emission of CdS [16], the luminescence can be attributed to emission from defect sites in the CdS material [17].

which would increase the estimate for the trapping force by at least a factor of two.

Although these estimates suggest that a single optical tweezer should be able to manipulate a nanowire, a point-like trap's symmetry allows a nanowire to rotate into an orientation that minimizes drag, and thus to escape from the trap. The spatially extended trapping potential provided by the holographic optical tweezer array maintains the nanowire's orientation and thus makes controlled translation possible. As few as two traps can capture and translate a nanowire, although more stable trapping is observed for multiple traps arranged in a line. Comparable trapping and orientation control has been demonstrated for single CuO nanorods [15] in a linear optical tweezer created with a cylindrical lens. Our HOT approach offers the additional benefit of manipulating multiple nanowires simultaneously and independently in complex ways, as described below.

Figure 2(a) and the associated video show two CdS nanowires being manipulated by two arrays of traps projected simultaneously with a single computer-generated hologram. One nanowire is held stationary while the second is advanced in steps of 0.7 μm by projecting an appropriately designed sequence of holograms at 1 s intervals. Similar sequences also can be used to rotate a nanowire precisely, as shown in Fig. 2(b). The video of

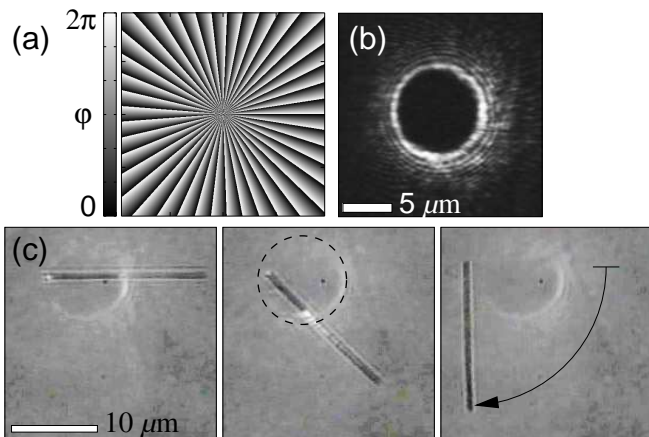


FIG. 3: Rotating a semiconductor nanowire with the orbital angular momentum flux of a helical mode of light. (a) When transmitted to the SLM, the helical phase mask $\varphi(r, \theta) = \ell\theta$ transforms the wavefronts of a TEM_{00} laser mode into an ℓ -fold helix. This helical beam focuses into the ring-like optical trap, shown in (b). The orbital angular momentum density in this trap can be used to rotate a semiconductor nanowire, as shown in the sequence of photographs in (c), which are separated by 1 sec intervals. The dashed circle in shows the position of an $\ell = 30$ optical vortex at 1 W.

this process demonstrates that both the separation and relative orientation of two nanowires can be controlled in this way, thereby providing the two basic capabilities required for building complex architectures.

The phase holograms used to create holographic optical traps also can modify the individual beams' wavefronts to create optical micromanipulators that do not require active updating to process nanowires. Specifically, a single static optical tweezer can be transformed into an optical vortex [18–21] by imposing a helical phase profile $\varphi(\mathbf{r}) = \ell\theta$ onto the trapping laser's wavefront. Here, $\mathbf{r} = (r, \theta)$ is a polar coordinate transverse to the beam's axis and ℓ is an integer winding number defining the wavefronts' helicity. The effect of this modulation is to transform a point-like optical tweezer into a ring-like trap whose radius scales linearly with winding number [21, 22], and whose photons each carry an orbital angular momentum, $\ell\hbar$, in addition to their intrinsic spin angular momentum [23], that can be transferred to objects illuminated by the ring of light [21, 24–26]. The resulting torque causes the nanowire in Fig. 3 to rotate, even though the trap itself is static. Arrays of optical vortices can be used to rotate large numbers of nanowires rapidly in parallel, although with less precise angular control than dynamic arrays of conventional optical tweezers.

We also have used HOT arrays to investigate other modes of manipulation that could be important for assembling complex structures. First, a trapped nanowire can be translated along the optical axis to the surface of a substrate. If the nanowire has not been stabilized, for

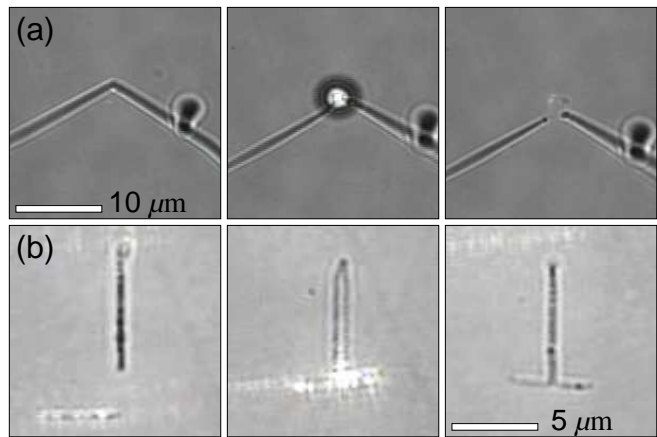


FIG. 4: Transforming nanowires with intense focused beams of light. (a) Cutting a semiconductor nanowire with an optical scalpel. A bent nanowire is brought to the focus of an optical trap powered by 0.5 W. An exposure time of 100 ms results in a clean cut at the bend. (b) Fusing two semiconductor nanowires into a free-floating assembly. The two nanowires are first trapped and then manipulated to form a T-junction. An optical trap powered by 100 mW is then focused on the junction for 1 s to non-destructively fuse the wires. The T-junction then floats freely once the traps are extinguished.

example with surfactant, this causes the nanowire to be deposited irreversibly through its van der Waals interaction with the substrate. In cases where the nanowires are stabilized, increasing the laser power in the trap array still can yield irreversible and site-specific deposition of nanowires with controlled orientation. Second, tightly focused optical traps at higher powers can be used to cut nanowires, as shown in Fig. 4(a). Here, a 0.5 W CW optical tweezer focused on a nanowire for ca. 100 msec acts as an optical scalpel. Finer cuts requiring substantially less power can be achieved with short laser pulses at shorter wavelengths [27].

Once nanowires have been cut to length and organized into specific configurations, forming junctions between them is critical for transforming these structures into electronic and photonic devices of the types that have been recently investigated and proposed [3, 7]. The HOT approach opens up new opportunities for creating such junctions. For example, the translation and rotation operations can be used to assemble two freely diffusing nanowires into a T-junction. Applying a high-power pulse (100 mW, 1 s) irreversibly fuses the nanowires to form a rigid T-junction that freely diffuses in solution when the HOTs are removed (Fig. 4(b)). These results highlight further the power of our approach; that is, it can be used to translate and rotate nanowires in a reversible manner, and also to irreversibly modify them through site-specific fusion, deposition and cutting.

Lastly, we have combined all of the manipulation steps described above to assemble a substantially more com-

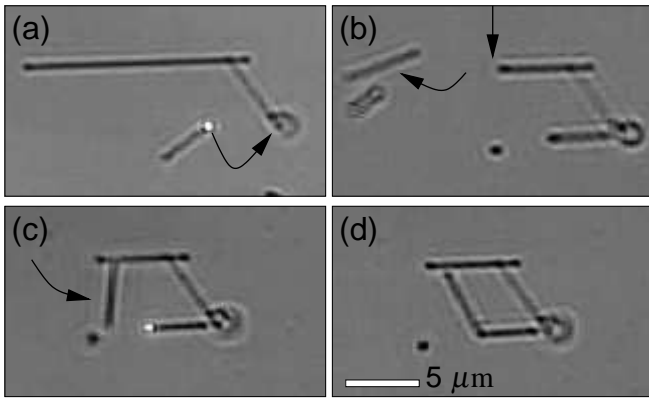


FIG. 5: Assembly of rhombus constructed from semiconductor nanowires using holographic optical traps. (a) A nanowire is translated towards an existing structure created earlier by trapping and fusing two nanowires. (b) The long nanowire is then cut with a pulsed optical scalpel. (c) The resulting free-floating nanowire piece then is brought back to the partially completed structure. (d) The structure is completed by fusing both ends of the fourth nanowire.

plex structure, as shown in Fig. 5. This interactive assembly was performed with a BioRyx 200 holographic optical trapping system. Figure 5(a) shows a nanowire segment being translated toward a pair of fused nanowires held in an optical tweezer array. After being translated and rotated into position, the additional segment is fused to the larger structure with a 0.5 W pulse of light distributed over 10 traps lasting 2 s. Next, the longer nanowire in the partially completed structure is cleanly cut (Fig. 5(b)) with a short-wavelength laser pulse (100 μJ , $\lambda = 440$ nm, 5 ns). The resulting free-floating nanowire segment is captured with multiple traps, and brought back to the optically trapped structure (Fig. 5(c)) to form a rhombus. Finally, additional laser pulses fuse the nanowires into a stable closed structure (Fig. 5(d)).

In summary, the results presented here demonstrate that holographically projected arrays of optical traps can be used to manipulate and assemble semiconductor nanowires into precisely organized two-dimensional and three-dimensional structures. In the future, it should be possible to optimize this process by tuning the laser wavelength to enhance the optical trapping force. The approach also will become substantially faster and more highly parallel with advances in holographic trapping technology. Optical assembly of functional subunits will facilitate hierarchical fabrication of larger systems, through processes that might exploit complementary techniques such as chemically-directed self-organization. The HOT technique also can be extended to bring together diverse nanoscale building blocks such as nanotubes [28] or nanoparticles [29], to utilize their unique properties in conjunction with those of nanowires. In addition, dynamic systems can be created by exploiting

the dynamically configurable nature of optical traps. We believe that the exciting opportunities provided by the HOT technique for nanofabrication with unprecedented and exquisite spatial control will be crucial for creating integrated and functional nanosystems in the future.

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