Highly efficient electrospray process for carbon nanotube-based laser pulse formers

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Abstract: We demonstrate laser pulsation of CNTs deposited by electrospray process that ensures target-localized and homogeneous deposition of CNTs with dramatically improved deposition efficiency (>95%). Resultant pulses have 3.95-MHz repetition rate and 190.9-fs pulse width.

OCIS codes: (160.4236) Nanomaterials, (230.4320) Nonlinear optical devices, (140.4050) Mode-locked laser

1. Introduction:

With notable advantages over conventional materials, recently, single-walled carbon nanotubes (CNTs) have emerged as one of the powerful candidates to realize next generation photonic devices and systems. [1-3] So far, laser pulsation with nonlinear elements have been actively researched with CNT-incorporated mode-lockers using diverse schemes. [4-5]

To date, since a straight forward CNT spray method guarantees intact optical nonlinearities as well as polarization independence with homogeneous and randomized deposition of nanostructures, respectively, the majority of pulsed lasers rely on a conventional CNT spray process. This has allowed for remarkable femto-second laser pulses passively generated by CNTs. However, the deposition efficiency of the conventional spray method is very low, to a degree that most CNTs are lost during the process. [6] Furthermore, large droplet size of the sprayed CNT suspension can induce agglomeration of the CNTs. This in turn can lead to deformation of the original morphologies of the individual nanostructures, which are directly correlated with the nonlinear properties of the CNTs.

In this work, we demonstrate a passive laser pulse formation with electrosprayed CNTs, providing a target-localized deposition and homogeneous dispersion controlled by the spray geometry as well as nanometer-scaled droplet size. The electrospray shows notable advantages including, (i) reduced droplet size with a very narrow size distribution, (ii) self-dispersion of the droplets, which prevents detrimental agglomeration of the nanostructures, (iii) controllable motion of the droplets with electric field tuning, and (iv) very high deposition efficiency. [7] The deposition efficiency is improved up to 95 %. Resultant pulses have 3.95-MHz repetition rate and 190.9-fs pulse width.

2. Fabrication of laser pulse former incorporating electrosprayed CNTs.

The passive pulse former is prepared with single-walled CNTs that are electrosprayed onto a side-polished optical fiber to have the interaction with the evanescent field of a broadened propagating mode. In contrast with the conventional interaction scheme in which light directly penetrates the CNT layer, the present scheme circumvents thermal damage of CNTs as well as alignment problems with a non-blocking operation. [4,8,9] With electrospraying fine sub-droplets of a CNT suspension can be produced in a nanometer scale. This is achieved by applying electrostatic charge on the surface of the original drops on the nozzle tip. When the charge reaches a critical point, where the Coulomb force is stronger than the surface tension of the drops, the CNT suspension breaks apart into a cloud of tiny, highly charged droplets. During their progress toward the collect electrode (substrate) with opposite charge, the droplets are separated by Coulomb repulsion into nanometer-scaled sub-droplets by repeated separation. (see Fig. 1(a)). [10,11] Fracture of the drops can be encouraged by the potential built between the nozzle and a grounded collecting plate accompanying directional acceleration with a spray angle of about 30°. Compared with conventional sprays, electrospray dramatically improves the deposition efficiency with a localized spray within a targeted area while maintaining homogeneity. As can be seen in Fig. 1(b), the present electrospray system consists of a CNT suspension feeder, a metallic nozzle, a grounded plate, and a high voltage supplier. The sprayed area is determined by the distance (D) between the nozzle tip and the substrate. This distance is minimized to 5 cm in order to maximize both deposition efficiency and homogeneity. In the electrospray procedure, the prepared CNT suspension is transferred into a feeder and continuously supplied by a syringe pump (KD Scientific, 781200) at a flow rate of 13 μm/min. A high voltage (8 kV) is applied between the nozzle tip and the substrate (distance: 5 cm), resulting in a highly homogeneous and target-localized CNT deposition onto the flat surface of the side-polished fiber. Figure 2(a) shows a prepared side-polished fiber with sprayed CNTs. The morphology of the deposited CNTs is analyzed with a scanning electron microscope (SEM, S-4100, Hitachi). The image in Fig. 2(b) reveals that there is no significant agglomeration. Also, CNTs are randomized, thus avoiding polarization dependence of the nonlinear absorption for the linearly polarized mode in the laser cavity.

3. Experimental result

In the case of the conventional spray method, a 30 ml of CNT suspension is sprayed for 30 minutes to coat the substrate for the targeted nonlinear photonic devices. On the other hand, employing the electrospray scheme to realize the same devices, a CNT suspension of 260 μl is sprayed for 20 minutes, resulting in dramatic improvement of the deposition efficiency of up to 95 %. In order to demonstrate the ultrafast functionality of the electrosprayed CNTs, as can be seen in Fig. 3, we experimentally evaluated a pulsed fiber laser that includes a CNT-deposited device as a laser pulse former. Figure 4(a) depicts the obtained optical spectrum of the mode-locked laser with a spectral width of 13.87 nm at a center wavelength 1559.1 nm. Assuming transform-limited $sech^2$ pulses that have a time-frequency product of 0.315, the estimated pulse duration is 190.9 fs. Thus, the electrosprayed CNTs exhibit highly improved pulse quality. This is directly correlated with the degree of dispersion of the individual CNTs, and therefore the intensity of their high-speed nonlinearity. Figure 4(b) verifies the pulse train measured with an oscilloscope, showing that the repetition rate of output pulses is 3.95 MHz with an evenly distributed peak intensity. Figure 5(a) illustrates the changes of spectral width and spectral peak power with respect to the current level of the pump laser diode (LD). The full width at half maximum (FWHM) decreases at about 0.04 nm/mA, and the spectral peak power increases at about 0.032 dB/mA as the LD power increases. Figure 5(b) depicts the changes of the center wavelength of the output as well as the estimated pulse peak power according to the pump level adjustment. The figure shows that the peak position is very stable, and the peak power is inversely proportional to the pump level due to temporal pulse broadening with a rate of 0.2125 fs/mA. Note that negligible pulse wavelength shift is induced with errors from maximum-point-reading. The peak power of our pulsed laser at a pump current of 200 mA is 12.1 kW. However, as the pump power is increased, the peak power decreases due to nonlinear pulse broadening and/or breaking in the fiber, which has an intracavity energy limit to maintain the soliton-shaped “clean” pulses.

4. Conclutsion

We demonstrate CNT-based laser pulse former by employing an efficient electrospray method. The scheme achieves a 95 % improvement of CNT deposition efficiency compared with conventional CNT spray methods. The CNT deposition process is optimized by controlling the charging voltage, nozzle-substrate distance, flow rate, and CNT dispersion in the solvent. Ultrahigh-quality dispersion of the individual nanomaterials with the electrospray provides maximized ultrafast nonlinearity of the CNTs, resulting in an outstanding temporal pulse duration of 191 fs passively achieved by a CNT pulse former. We expect that this scheme will provide a practical way to realize diversified ultrahigh-speed nonlinear photonic devices by simply upgrading the nanomaterial-deposition process for future photonic and optoelectronic systems.

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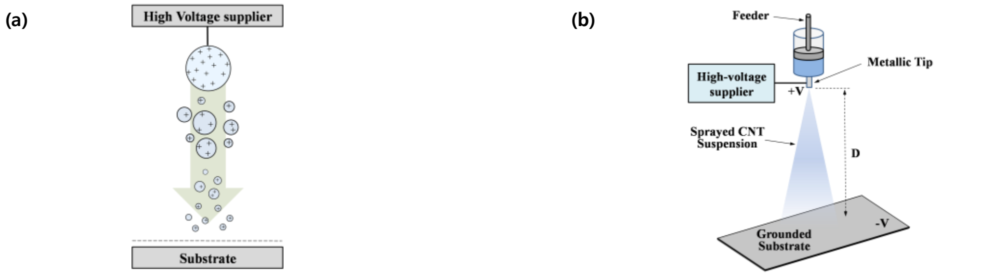

Fig. 1. Schematic explanation of (a) the operation principle, and (b) the experimental setup of the electrospray scheme. In (b), the spray angle is 30, and D is the nozzle-substrate distance.

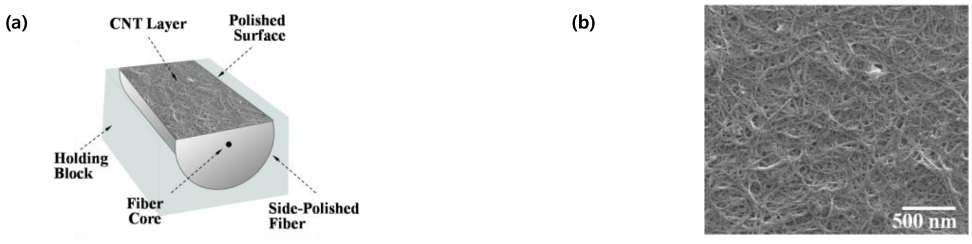

Fig. 2. (a) Prepared side-polished fiber deposited with CNTs onto a flat surface resulting in evanescent field interaction of the traveling mode with CNTs. (b) SEM image to evaluate the nano-morphology of the electrosprayed CNT layer.

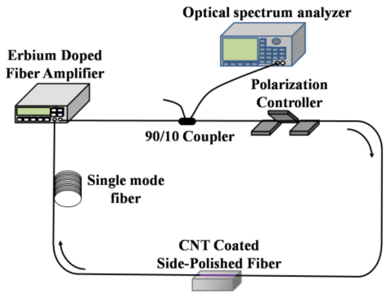

Fig. 3. Structure of a passively pulsed fiber ring laser mode-locked by a nonlinear photonic device incorporating electrosprayed CNTs. Ultrafast recovery time of CNTs enhances the short pulsation of the laser.

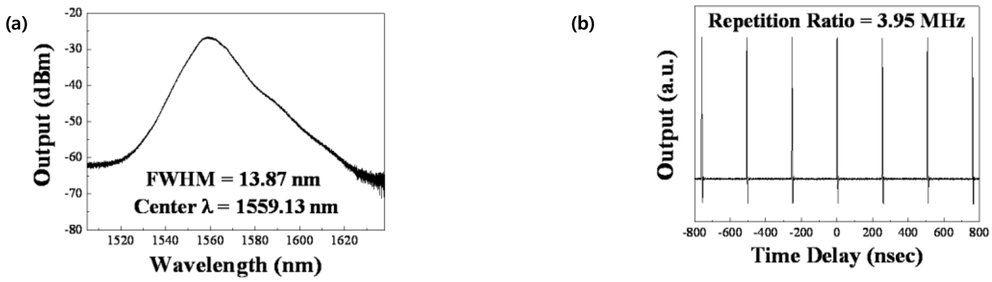

Fig. 4. (a) Optical spectrum of the pulsed laser output, and (b) its pulse train. The spectral width and the repetition rate are 13.87 nm and 3.95 MHz, respectively. The estimated temporal pulse duration is 190.9 fs.

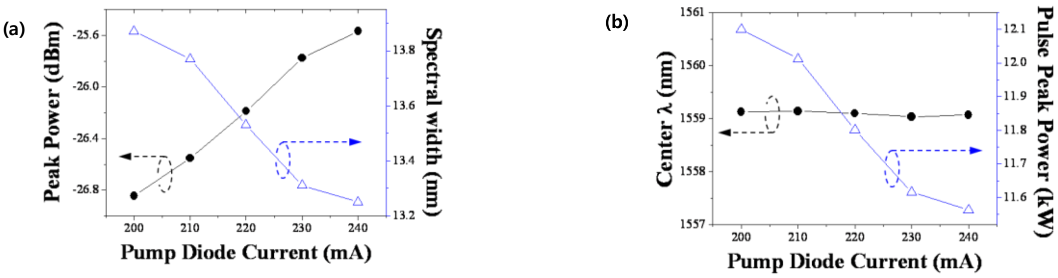

Fig. 5. (a) The correlation of spectral characteristics with pump current. The spectrum becomes sharper as the pump power is increased. (b) Estimated pulse peak powers in the time domain that are inversely proportional to the pump power. (b) also shows the stability of the center wavelength with respect to the pump power tuning.