Nanoimprinted Perovskite Metasurface for Enhanced Photoluminescence

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Recently, hybrid halide perovskite has emerged as promising materials for advanced optoelectronic devices such as photovoltaics, photodetectors and light emitting diodes (LEDs). All-dielectric metasurface and semiconductor nanostructures with high-refractive-index has attracted attention due to low-loss and highly-efficient resonance. Here we report novel organolead halide perovskite metasurfaces, created by cost-effective nanoimprint technology, which demonstrated enhanced emission properties. Such metasurfaces could be used to create distributed feedback lasers and integrate with LEDs for higher efficiency.

The $MAPbI_3$ perovskite thin film was formed on thermally grown Si dioxide with Si substrate by spin-coating. The sample was then nanoimprinted with a Si nanograting mold under a temperature of 100 ^{o}C and a pressure of 7 MPa for 30 mins (Fig. 1a). The mold was fabricated using electron beam lithography with hydrogen silsesquioxane and then followed by plasma etching to form nanograting patterns (Fig. 1b). Fig. 1c shows the perovskite metasurface with negative replicated nanogratings. The results have demonstrated that although perovskite is an ionic solid and does not have a glass transition behavior like polymer, nanoimprint technology could successfully pattern perovskite which provides a convenient and cost-effective method for fabricating perovskite metasurface. Fig 2 shows measured transmission (Fig. 2a) and reflectance (Fig. 2b) of perovskite reference thin-film and nanograting metasurface with a structure width, period and depth of 270 nm, 600 nm and 300 nm respectively. Nanograting metasurface demonstrated much reduced transmission and reflection for $\lambda < 800$ nm suggesting strong light absorption, while the reflection peak around 1000-1100 nm corresponds to the excitation of optical resonance. The enhancement of photoluminescence (PL) with nanograting structure was revealed in Fig. 3a. The unpatterned perovskite thin-film shows a PL peak at 763 nm with a FWHM of 44 nm while the nanograting metasurface shows much improved PL properties. The metasurface with a structure width of 225 nm and a period of 350 nm demonstrated an 8-fold improvement in PL intensity and a 2-fold reduction in FWHM. Our previous results have demonstrated up to 70-fold PL enhancement under non-linear multi-photon excitation. The enhanced photoluminence was due to improved light absorption, excitation of optical resonance, and the photonic crystal cavity formation as suggested by the emergence of narrow cavity resonance in PL spectra. Fig. 3b and Fig. 3c show the temperature dependent PL of perovskite metasurface and reference thin-film respectively. With a decrease of temperature from 300 K to 100 K, increased intensity and narrowed linewidth were observed with a red shift of PL peak, which requires further study. By improving nanostructure design and fabrication process, lasing via distributed feedback is expected with direct nanoimprint of perovskite. Highly efficient perovskite LEDs is also expected by incorporating the metasurface for enhanced emission.

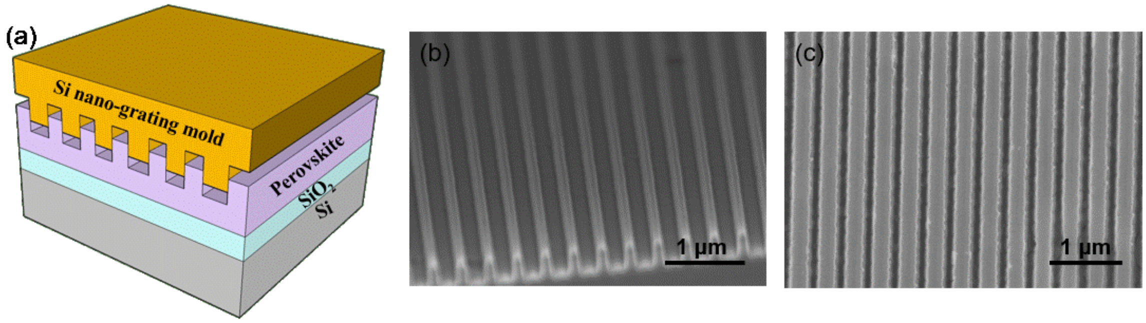
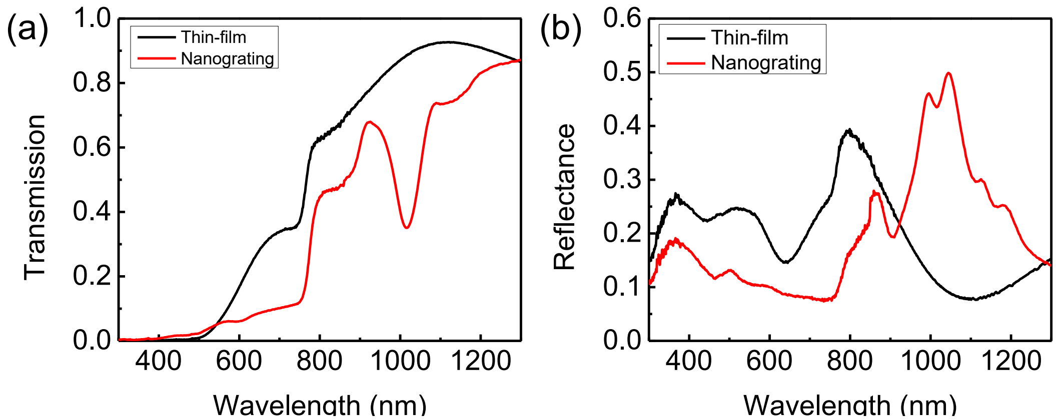


Figure 1: (a) Schematic of perovskite nanoimprint with a Si nanograting mold, (b) SEM graph of a Si nanograting mold, with structure width, period and depth of 140 nm, 350 nm and 240 nm, respectively, (c) SEM graph of nanoimprinted perovskite showing negative replication of the Si nanograting mold.



F*igure 2: (a) Transmission and (b) Reflectance of perovskite nanograting metasurface (structure width of 270 nm, period of 600 nm, and depth of 300 nm with 130 nm residue thickness) and reference thin-film without imprint (thickness 265 nm). The nanograting metasurface shows resonance in rear infrared region.*

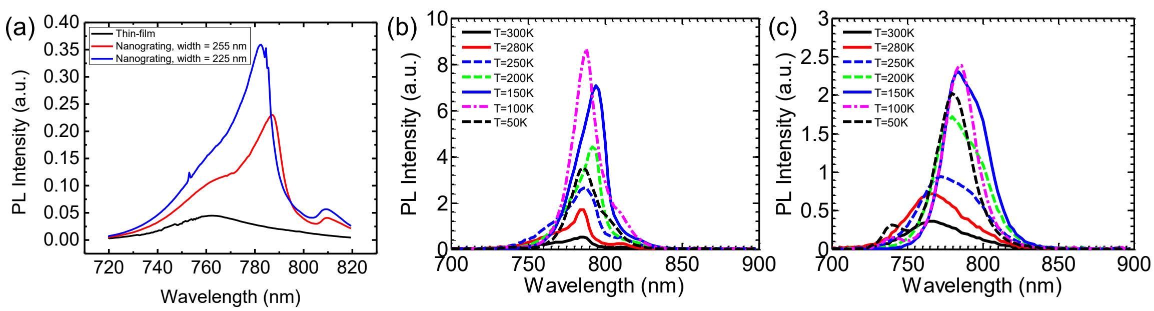


Figure 3: (a) Photoluminescence of perovskite nanograting metasurface (structure width of 255 nm and 225 nm, period of 350 nm and depth around 100 nm with residue thickness of 200 nm) and referenced thin-film (thickness 265 nm), with a pump power of 3 mW at room temperature. Corresponding temperature dependent photoluminescence of (b) nanograting metasurface with structure width of 225 nm and (c) referenced thin-film.