Silicon-nitride photonic circuits interfaced with monolayer MoS\textsubscript{2}

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(Received 6 June 2015; accepted 18 August 2015; published online 2 September 2015)

We report on the integration of monolayer molybdenum disulphide with silicon nitride microresonators assembled by visco-elastic layer transfer techniques. Evanescent coupling from the resonator mode to the monolayer is confirmed through measurements of cavity transmission. The absorption of the monolayer semiconductor flakes in this geometry is determined to be 850 dB/cm, which is larger than that of graphene and black phosphorus with the same thickness. This technique can be applied to diverse monolayer semiconductors for assembling hybrid optoelectronic devices such as photodetectors and modulators operating over a wide spectral range.

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Integrated electro-optical circuits are a promising alternative to traditional silicon electronics for low-power and high-speed information processing.\textsuperscript{1} Hybrid electro-optical devices optimized for light modulation and detection can be assembled at fundamental atomic-scale thickness limits by integrating two-dimensional (2D) atomic-scale materials such as graphene with on-chip photonic circuits.\textsuperscript{2–4} Although the electro-optical properties of 2D materials can be further exploited for high-performance optoelectronics by assembling layered heterostructures\textsuperscript{9,10} or for polarization-sensitive valley-dependent electronic devices.\textsuperscript{11} Interfacing TMDCs with optical waveguides and resonators is required to take advantage of these layer-sensitive structures in photonic circuits.

Emission of 2D TMDCs can be controlled by interfacing layers with optical cavities.\textsuperscript{12,13} Lasing from TMDCs coupled with photonic cavities has been recently demonstrated,\textsuperscript{14,15} but the coupling of TMDCs to planar photonic circuits has not been explored. Absorptive or refractive integrated TMDC-photonics devices such as modulators or photodetectors require evanescent coupling of optical modes with monolayer materials. Strong absorption can reduce the device footprint and increase modulation depth or sensitivity. Here, we show that visco-elastic transfer techniques\textsuperscript{16} can be used to assemble an on-chip silicon nitride integrated photonic circuit architecture evanescently coupled to monolayer molybdenum disulphide (MoS\textsubscript{2}). We measure strong absorption of the monolayer (850 dB/cm) in the evanescent field larger than that for graphene (670 dB/cm)\textsuperscript{17} or the equivalent thickness of black phosphorus (2000 dB/cm for 11 nm thick).\textsuperscript{18} The integration of 2D semiconductors with silicon-based photonic structures demonstrates a path for assembling on-chip functional hybrid electro-optical devices based on monolayer materials and their heterostructures.

Evasive coupling of nanomaterials in hybrid photonic devices can be confirmed by measuring absorption. Direct measurement of absorption loss from monolayer TMDCs is challenging because of the small overlap of atomic-scale layers with evanescent fields of guided optical modes. As demonstrated with graphene, integrated photonic structures such as Mach-Zehnder interferometers and microresonators can be exploited for measuring coupling since they are sensitive to single-layer absorption.\textsuperscript{2,3,17} Monolayer MoS\textsubscript{2} is even more attractive for this mode of measurement because of its enhanced band edge absorption of 5%–10% for normal incident light\textsuperscript{19,20} which is several times larger than the broadband absorption of graphene.\textsuperscript{17} Since the $Q$-factor of a photonic resonator is closely related to the absorption losses in its mode, microring resonators with high $Q$-factors are a sensitive system for measuring the absorption of monolayer materials to optical modes (Fig. 1(a)). For demonstration of photonic coupling, we use the canonical monolayer MoS\textsubscript{2}, although the technical approach directly translates to other TMDCs without modification.

Since the direct bandgaps of various 2D TMDCs are distributed from 1.1 eV (MoTe\textsubscript{2}) to larger than 2.0 eV (WS\textsubscript{2}), a material platform for TMDC hybrid photonics needs low visible and infrared light absorption. Silicon nitride (SiN) is a wide bandgap semiconductor material transparent to both visible and infrared wavelengths. Because of its large refractive index ($n \approx 2.0$) contrast to SiO\textsubscript{2} ($n \approx 1.45$) and its compatibility with CMOS processing, SiN is very attractive for linear and non-linear photonics in wavelength regimes where traditional silicon photonics architecture would absorb.\textsuperscript{21} Here, we use stoichiometric Si\textsubscript{3}N\textsubscript{4} deposited by low pressure chemical vapor deposition on silicon substrates. Because MoS\textsubscript{2} is observed to have poor adhesion to SiN, a layer of...
15 nm PECVD SiO$_2$ is deposited over the SiN to improve adhesion of the MoS$_2$ after device patterning.

Resonators suitable for measuring material absorption must simultaneously exhibit both well-resolved changes in $Q$ from the layer transfer process and ideally tuned external coupling for a spectral measurement. These conditions require tailoring the microresonator for the range of expected material absorption. We estimate the expected resonator loss caused by MoS$_2$ with finite element modeling using COMSOL Multiphysics. The monolayer MoS$_2$ is approximated as a rectangular cross-section of thickness 0.65 nm above the SiO$_2$-capped SiN device (Fig. 1(a), inset). The resonator is simulated using an axisymmetric model with a SiN cross-section and channel are consistent with previous observations of exciton optical properties will not be strongly modified by the resonator and optical loss in the integrated hybrid system will be determined by the classical dispersive absorption of the material in the evanescent field.

Ring resonators of the desired geometry are patterned with high-resolution electron beam lithography using a JEOL 9300 operating at 100 kV with a shot pitch of 3 nm (Fig. 1(c)). Positive resist (GL2000) was used for pattern definition and as the etching mask following development. This writing procedure creates channels on both sides of the waveguides that the transferred monolayer bridges (Fig. 1). The ring resonators are coupled to planar bus waveguides with the same cross-sectional dimensions as the ring (375 nm × 250 nm). A pair of gratings couple the two ends of the bus waveguide for input and output coupling. The device is carefully designed so both can be in the field of view of a 100× objective.

Visco-elastic transfer is employed for deterministic assembly of monolayer integrated photonic devices. Monolayers of MoS$_2$ are mechanically exfoliated from a bulk crystal with a gel film and positioned to the desired location using a 3-axis translation stage. The gel is lowered into contact with the substrate and slowly peeled away, leaving the monolayer flake on top of the ring resonator as confirmed by optical microscopy. Monolayers can be placed with micrometer accuracy on top of photonic structures. Fig. 1(d) shows a representative device covered by a monolayer of MoS$_2$. The MoS$_2$ layer is suspended over the trenches surrounding the ring. The nearby SiN on either side of the trench helps support the intact monolayer so that it remains wrinkle-free with minimal strain near the coupling region.

Photoluminescence (PL) characterization is used to confirm successful monolayer transfer. Fig. 2(a) shows spatially resolved PL from the device shown in Fig. 1(d), and a representative spectrum from a single ~1 μm spot on (blue) and off (red) the ring is shown in Fig. 2(b). Each spectrum is taken with a scanning microscope with 100× objective and 40 μW excitation with a 532 nm laser. The PL clearly shows the expected spectrum of monolayer MoS$_2$ with the peak energy at about 1.88 eV, while the PL map confirms the layer coverage. The PL collected from on the ring and channel has a higher intensity, but it does not exhibit changes in linewidth caused by the cavity or any enhanced emission peaks corresponding to the cavity modes, both consistent with the low Purcell factor for this geometry. The slightly blue shifted peak and higher intensity of the spectra on the ring and channel are consistent with previous observations of

![FIG. 1. (a) A schematic of the experimental set-up. Inset shows the device profile. (b) Mode profile of the fundamental quasi-TE mode in the ring with MoS$_2$ monolayer on top. (c) SEM image of a representative device. (d) Optical image of a device with the ring covered by a monolayer MoS$_2$ over a circumference corresponding to $\phi = \pi/4$. The ring radius is 15 μm.](image)

![FIG. 2. (a) Photoluminescence intensity map using a 40 μW pump laser and analyzed with a spectrometer and CCD. The spectra are integrated between 600 nm and 720 nm. (b) Representative PL spectrum of MoS$_2$ on (blue, $x = 14 \mu m$, $y = 19 \mu m$) and off (red, $x = 20 \mu m$, $y = 15 \mu m$) the ring. The off-ring spectrum is shifted in y axis for clarity.](image)
suspended monolayers.\textsuperscript{23,24} Spatially resolved PL measurements indicate that the enhanced intensity near the ring and channels also has a contribution from additional absorption of the focused pump laser scattered by the etched structure, evidenced by the small unexpected emission collected when the pump is over the bare resonator.\textsuperscript{22}

We fabricated several devices with distributed angle-of-coverage ($\phi$) of monolayer MoS$_2$. Cavity transmission spectra are measured before and after monolayer transfer using a tunable diode laser with linewidth smaller than 1 GHz (which is much narrower than our >300 GHz resonance linewidths). The output from the grating coupler is spatially filtered for collection. The intrinsic quality factor due only to intracavity absorption and scattering loss is estimated from the transmission spectra assuming no backscattering. Fig. 3 shows a representative transmission spectrum of one device before and after monolayer transfer. The resonance center wavelengths shift after monolayer transfer because the mode effective refractive index changes. We analyze up to two adjacent quasi-transverse electric (TE) resonant modes for each device, limited by the tuning range of the laser, the free spectral range of the resonators, and the ability to isolate the spectral resonance from the interference background from the grating coupler scattering. The slight differences in $Q$-factor between adjacent resonator modes can be the result of different local mode profiles or the wavelength dependence of absorption. Since the adjacent resonances are close compared to the linewidth of the MoS$_2$ PL, we treat the adjacent resonances as two equivalent data points. The quantitative differences between them are expected to be less than the uncertainty of our following calculations, confirmed by the data in Fig. 4.

The reduction of the intrinsic $Q$-factor is used to extract the change in absorption loss per length $\Delta\alpha$ of the ring caused by the monolayer in the evanescent region. The intrinsic $Q$ of an axisymmetric resonator can be expressed in terms of the average loss per unit length $\alpha$

$$Q = \frac{10}{\ln(10)} \frac{2\pi n_g}{\lambda_0} \frac{1}{Q_i} - \frac{1}{Q_f},$$

where $\lambda_0$ is the resonance wavelength and $n_g$ is the effective refractive index of the guided mode. This internal loss comes from multiple channels such as scattering, bending loss (which is negligible for the size of resonators used), and material absorption loss. Assuming scattering, bending, and SiN loss are essentially unchanged by the layer transfer process, the difference in the effective absorption loss rate before ($\alpha_i$) and after ($\alpha_f$) monolayer transfer can be written

$$\Delta\alpha = \alpha_f - \alpha_i = \frac{2\pi n_g}{\lambda_0} \frac{10}{\ln(10)} \left( \frac{1}{Q_f} - \frac{1}{Q_i} \right),$$

where $Q_i$ and $Q_f$ are the $Q$-factors before and after placing monolayer MoS$_2$ on the ring. Subtracting the control measurement before monolayer transfer isolates the monolayer interaction from imperfections due to fabrication, surface roughness, and SiN quality of the bare resonators while allowing the separately processed devices to be compared with each other. As the ring is only partially covered by MoS$_2$, the total effective loss consists of the original total loss around the full ring plus additional absorption loss in the covered region caused by interaction with MoS$_2$ ($\alpha_{int}$)

$$2\pi R\alpha_f = 2\pi R\alpha_i + \phi R\alpha_{int},$$

where $\phi$ is the angle by which the monolayer covers the ring resonator and $R$ is the ring radius. We neglect the change in effective mode index and finite mode radial size since the corrections are smaller than the precision of the experiment. The relation between the measured $\Delta\alpha$ and material absorption is

$$\Delta\alpha_i = \alpha_f - \alpha_i = \alpha_{int} \frac{\phi}{2\pi}.$$

The change in effective absorption loss rate $\Delta\alpha$ is proportional to the coverage fraction $\phi/2\pi$ with a coefficient equal to the absorption per unit length of the monolayer in the evanescent field of this particular geometry.

The effective material absorption loss $\Delta\alpha$ is extracted from the $Q$-factors according to Eq. (2) (Fig. 4). A weighted linear fit yields $\alpha_{int} = 850 \pm 83$ dB/cm with an intercept of $23 \pm 8$ dB/cm for resonators with $R = 15$ $\mu$m. The above-zero
intercept is likely caused by scattering due to the discontinuous effective mode index at the boundary between covered and uncovered areas, which is not included in our axisymmetric simulation. The fitted $\chi_{int}$ is close to but unambiguously smaller than the $\chi_{int} = 1390 \text{dB/cm}$ expected from numerical simulations (Fig. 1(b)). Absorption and PL of monolayer MoS$_2$ are sensitive to strain and the substrate in ways that a numerical simulation cannot easily account for.\textsuperscript{23,26} The PL center wavelength on top of our SiN devices (658 nm) has slightly higher energy compared to typical results from the literature and from our monolayers on SiO$_2$ substrates, indicating sensitivity of the MoS$_2$ exciton to the substrate.\textsuperscript{23,25–27} The absorbance of monolayer MoS$_2$ is sensitive to substrate quality; substrate surface roughness can suppress the absorption or emission of the monolayer.\textsuperscript{25} Our PECVD-deposited capping layer is expected to have high surface roughness,\textsuperscript{26} which can lead to differences in absorption or emission of the monolayer.\textsuperscript{25} Our approach can be applied to combine atomic-scale semiconductors with pellucidal materials for hybrid opto-electronics. Our approach is anticipated because of the increased absorbance of monolayer MoS$_2$ (5%–10%) compared to graphene (2%–3%).\textsuperscript{19,20} The MoS$_2$ absorption is also larger than that measured for waveguide-integrated black phosphorus with a metric simulation. The fitted $\chi_{int}$ is close to but unambiguously larger than that for monolayer graphite (2%–3%).\textsuperscript{19,20} The MoS$_2$ absorption is also larger than that measured for waveguide-integrated black phosphorus with an effective mode index at the boundary between covered and uncovered areas, which is not included in our axisymmetric simulation. The fitted $\chi_{int}$ is close to but unambiguously larger than that for monolayer graphite (2%–3%).\textsuperscript{19,20} The MoS$_2$ absorption is also larger than that measured for waveguide-integrated black phosphorus with an effective mode index at the boundary between covered and uncovered areas, which is not included in our axisymmetric simulation. 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