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Graphene-controlled Terahertz Plasmonic Laser

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Abstract: For the first time, we demonstrate that monolayer graphene can control surface-plasmon modes in terahertz lasers. Integrating a graphene-coated, multi-band filter, varied *in situ* by electrochemical gating, individual modes can be switched on and off.

OCIS codes: (250.0250) Optoelectronics; (140.5965) Semiconductor lasers, quantum cascade; (240.6680) Surface plasmons; (160.4670) Optical materials; (160.4760) Optical properties

1. Introduction

Graphene – a two-dimensional hexagonal lattice of carbon atoms – has a long list of extraordinary properties, such as incredible strength, flexibility and electrical conductivity [1]. It also has fascinating optical properties, including the ability to support surface plasmons (SP). Most interestingly for plasmonic research, the Fermi level in graphene (and therefore its conductivity and plasmon properties) may be adjusted by a variety of methods, for example electrical gating or surface doping. Our objective is to harness this quality to create plasmonic waveguides with dynamic and user-defined properties. More specifically, we demonstrate that graphene is capable of supporting terahertz (THz) SP modes and changing the properties of such waveguides. In this way, the emission characteristics of a THz quantum cascade laser (QCL) can be controlled. The THz QCLs we report have novel, integrated aperiodic distributed feedback (ADFB) gratings, providing a multi-band filtering 'signature' [2]. We aim to directly alter the filtering strength and therefore reveal the influence of graphene on the SP mode. Use of a multi-band signature makes changes in filtering strength readily discernable. This demonstration opens the door to the creation of re-writable plasmonic microstructures by utilising the tunable nature of the plasmonic modes in graphene.

2. Device fabrication, characterisation and results

Terahertz QCLs, based upon semi-insulating SP waveguides (180 µm wide, 6 mm long, Fig. 1a), were fabricated from a $GaAs/Al_{0.15}Ga_{0.85}As$ bound-to-continuum active region (AR), then characterised in pulsed operation (10 kHz pulse rate, 1 µs pulse width) at ≤ 10 K. The ADFB grating, with a multi-band filter response $\rho(f)$, was subsequently introduced into the upper layers of the QCL waveguides by focussed ion beam (FIB) milling. The grating elements are narrow and shallow (< 1µm) slits penetrating the metallic uppermost waveguide layers, interrupting the guided SP mode and creating a discontinuity in its complex modal refractive index. For a given ADFB filter design, individual $\rho(f)$ band strengths are proportional to the modal refractive index contrast $|\Delta n|$ between the milled and un-milled waveguide regions, and influence the response of the compound Fabry-Perot (FP) plus $\rho(f)$ system. With sufficient $|\Delta n|$ the laser emission exhibits a clear spectral signature of $\rho(f)$. Figure 1b shows typical emission spectra from a FP QCL (QCL *A*), prior to FIB milling. The intended $\rho(f)$ of the ADFB filter introduced to this laser is presented in Fig. 1c and was calculated using the approximate Fourier transform relationship with the real space relative permittivity of the grating, using $|\Delta n| = 0.1$ [2]. In reality, $|\Delta n|$ is unknown for QCL *A* but is expected to be lower than 0.1 due to the observed lack of switchable, single mode selectivity (Fig. 1d).

After initial device characterisation large area, high quality, monolayer (up to 99% by area) graphene was introduced as an overlayer to the ADFB grating (Fig. 1a). Use of chemical vapour deposition (CVD) allowed the creation of extensive graphene sheets, large enough to cover the entire millimetre-scale grating area. The intention behind the introduction of graphene was to impose greater control over THz QCL emission by dynamically adjusting $|\Delta n|$, thereby controlling the $\rho(f)$ signature. Since the graphene monolayer thickness is ~0.3 nm (over two orders of magnitude smaller than the slit lengths and depths), it only fills < 0.01% of the slit volume. Changes in the slit refractive indices (and hence $|\Delta n|$) are therefore attributable to the existence of intra-slit graphene-supported SPs. Figure 1e shows the graphene-modified emission spectra of QCL *A*. We see a reduced filtering effect, with FP mode suppression is less pronounced than Fig. 1d. Consequently, there is no longer a one-to-one relationship between the dominant lasing modes and the $\rho(f)$ bands at lower bias (V_{QCL}), whereas at high V_{QCL} the emission becomes almost indistinguishable from the original FP QCL.

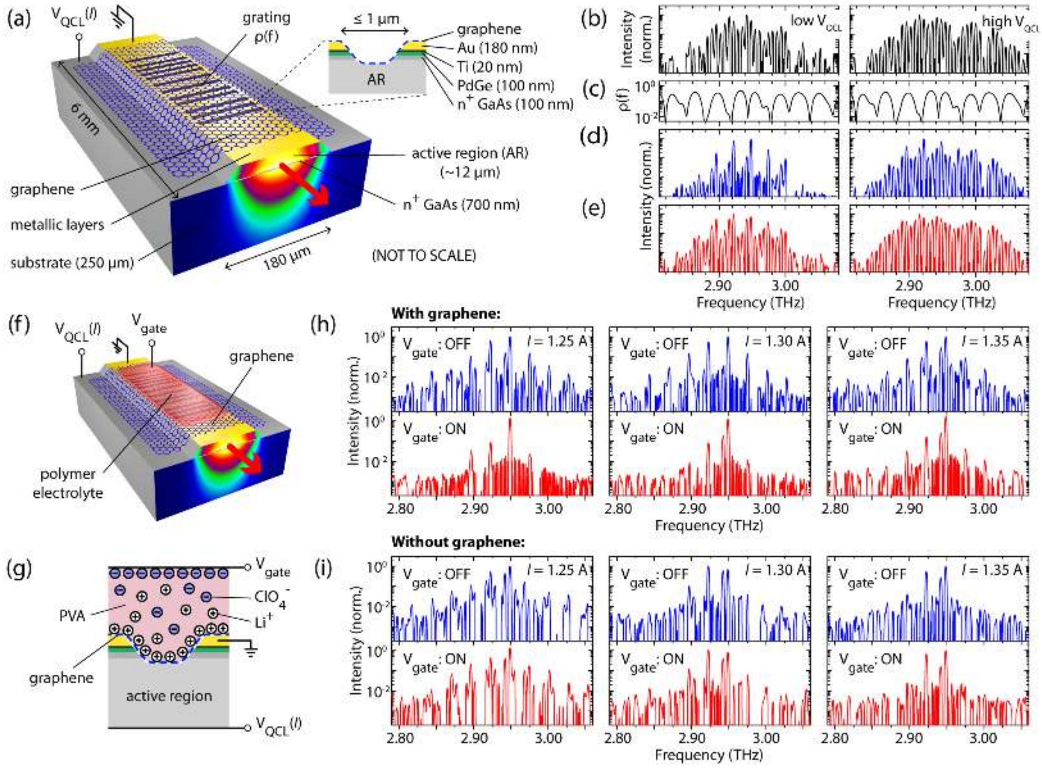

Fig. 1. **(a)** Schematic of a THz QCL with an ADFB grating and graphene overlayer. **(b)** Emission spectra from QCL *A* prior to FIB milling. Dips around 2.97 and 3.02 THz are due to strong atmospheric water absorption lines. **(c)** Spectral reflectivity response ρ(f) of the ADFB grating, assuming $|\Delta n| = 0.1$. **(d)** Emission spectra from QCL *A* after FIB milling. **(e)** Modified emission after graphene deposition, displaying an increased favorability for the original FP cavity modes in (b). **(f)** and **(g)** The grating microstructure was coated in a thin layer of polymer electrolyte (PVA + $LiClO_4$), which could be electrically biased (V_{gate}) to form Debye layers at each electrode. **(h)** and **(i)** Measured emission spectra of QCL *A* at three driving currents (*I*) just above lasing threshold, at $V_{gate} = 0$ V (blue) and 1 V (red), **(h)** with and **(i)** without an underlying graphene monolayer. Application of $V_{gate} = 1$ V improves the filter strength in the presence of graphene (fewer lasing modes observed), but has no effect in the absence of graphene.

After establishing the crucial first step that monolayer graphene can successfully interact with the THz optical modes in a QCL, the optical influence of the graphene was then varied by electrochemical gating. An electrolytic film, consisting of a synthetic polymer (PVA) and a lithium salt ($LiClO_4$), was deposited over the graphene-covered microstructure (Fig. 1f). The graphene surface doping (and hence conductivity) is changed by applying a bias (V_{gate}) across the electrolyte (Fig. 1g), influencing the supported plasmon modes (and consequently $|\Delta n|$) and altering the ρ(*f*) band strengths. The resulting change in emission is most clearly discernable around the onset of lasing in QCL *A*. Figure 1h shows such emission spectra, with V_{gate} off (= 0 V, blue) and on (= 1 V, red). Under electrolyte biasing we see a stronger filtering effect, the multi-moded (FP) nature of the spectra are drastically reduced and only the most highly favoured ADFB-dictated modes reach lasing threshold. Similar measurements were performed on QCL *A* with electrolyte but no graphene (Fig. 1i). Unsurprisingly the resultant spectra differ from Fig. 1h, (graphene removal changes $|\Delta n|$), but more importantly the weak spectral filtering remains unchanged by V_{gate}.

3. Conclusions

This first demonstration that a graphene monolayer, incorporated into an active plasmonic waveguide, can interact with a guided optical mode is a crucial foundation for future graphene-based THz QCLs. Furthermore, *in situ* adjustment of the graphene conductivity and therefore plasmonic properties opens up a wealth of possible device architectures. By substituting of electronic gating for electrochemical doping, one can envisage independently controlled graphene regions and hence plasmonic microstructures capable of being re-written for any desired spectral response. This work was primarily supported by the EPSRC, UK and acknowledges support from Ministry of Education, Science and Technology, Korea.

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