## Plasmonic lasers at deep sub-wavelength scales

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Laser science has been successful in producing high-power, fast and small coherent light sources [1-9]. Examples of recent advances are microscopic lasers that can reach the diffraction limit, based on photonic crystals [3], metal-clad cavities [4] and nanowires [5–7]. However, such lasers are restricted, both in optical mode size and physical device dimension, to being larger than half the wavelength of the optical field, and it remains a key fundamental challenge to realize ultracompact lasers that can directly generate coherent optical fields at the nanometre scale, far beyond the diffraction limit [10,11]. A way of addressing this issue is to make use of surface plasmons [12,13], which are capable of tightly localizing light, but so far ohmic losses at optical frequencies have inhibited the realization of truly nanometre-scale lasers based on such approaches [14,15]. A recent theoretical work predicted that such losses could be significantly reduced while maintaining ultrasmall modes in a hybrid plasmonic waveguide [16]. Here we report the experimental demonstration of nanometre-scale plasmonic lasers, generating optical modes a hundred times smaller than the diffraction limit. We realize such lasers using a hybrid plasmonic waveguide consisting of a high-gain cadmium sulphide semiconductor nanowire, separated from a silver surface by a 5-nm-thick insulating gap. Direct measurements of the emission lifetime reveal a broad-band enhancement of the nanowire's exciton spontaneous emission rate by up to six times owing to the strong mode confinement [17] and the signature of apparently threshold-less lasing. Because plasmonic modes have no cut-off, we are able to demonstrate downscaling

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of the lateral dimensions of both the device and the optical mode. Plasmonic lasers thus offer the possibility of exploring extreme interactions between light and matter, opening up new avenues in the fields of active photonic circuits [18], bio-sensing [19] and quantum information technology [20].

Surface plasmon polariton (SPP) are the key to breaking down the diffraction limit of conventional optics as they allow the compact storage of optical energy in electron oscillations at the interfaces of metals and dielectrics [11-13]. Accessing sub-wavelength optical length scales introduces the prospect of compact optical devices with new functionalities as it enhances inherently weak physical processes, such as fluorescence and Raman scattering of single molecules [19] and non-linear phenomena [21]. An optical source that couples electronic transitions directly to strongly localized optical modes is highly desirable as it would avoid the limitations of delivering light from a macroscopic external source to the nano-scale, such as low coupling efficiency and difficulties in accessing individual optical modes [22].

Achieving stimulated amplification of SPPs at visible frequencies remains a challenge due to the intrinsic ohmic losses of metals. This has driven recent research to examine stimulated SPP emission in systems that exhibit low loss, but only minimal confinement, which excludes such schemes from the rich new physics of nanometre scale optics [14, 15]. Recently, we have theoretically proposed a new approach hybridizing dielectric waveguiding with plasmonics, where a semiconductor nanowire sits atop a metallic surface, separated by a nano-scale insulating gap [16]. The coupling between the plasmonic and waveguide modes across the gap enables energy storage in non-metallic regions. This hybridization allows SPPs to travel over larger distances with strong mode confinement [23] and the integration of a high quality semiconductor gain material with the confinement mechanism itself.

In this Letter, we utilize the "hybrid plasmonics" approach to experimentally show the laser action of SPPs with mode areas as small as  $\lambda^2/400$ . The truly nano-scale plasmonic laser devices consist of Cadmium Sulphide (CdS) nanowires [24] on a silver film, where

the gap layer is Magnesium Fluoride (MgF<sub>2</sub>, Fig. 1a). The close proximity of the semiconductor and metal interfaces concentrates light into an extremely small area as much as 100 times less than a diffraction limited spot [16] (Fig. 1b). To show the unique properties of hybridized plasmon modes, we compare the plasmonic lasers directly with CdS nanowire lasers on a quartz substrate, similar to typical nanowire lasers reported before [5-7]. In what follows, we will refer to these two devices as *plasmonic* and *photonic* lasers respectively.

We optically pump these laser devices at a wavelength of 405 nm and measure emission from the dominant I<sub>2</sub> CdS exciton line at 489 nm [25]. At moderate pump intensities (10-60 MWcm<sup>-2</sup>), we observe the onset of amplified spontaneous emission peaks. These correspond to the longitudinal cavity modes that form when propagation losses are compensated by gain allowing plasmonic modes to resonate between the reflective nanowire end-facets (see Supplementary Information, Fig. 2a). The large amount of gain needed to compensate both cavity and propagation losses produces a strong *frequency pulling* effect [26] causing the Fabry-Perot modes to be much more closely spaced than expected for a passive cavity (Fig. 2c). A detailed discussion of this effect can be found in the Supplementary Information. The clear signature of multiple cavity mode resonances at higher pump powers demonstrates sufficient material gain to achieve full laser oscillation, shown by the non-linear response of the integrated output power with increasing input intensity (Fig. 2b). The occurrence of these cavity resonances indicates that the laser mode's *plasmonic coherence* is determined by cavity feedback and not by its propagation distance.

Surmounting the limitations of conventional optics, plasmonic lasers not only support nano-scale optical modes, their physical size can also be much smaller than conventional lasers, i.e., plasmonic lasers operate under conditions where photonic modes cannot even exist [7]. Plasmonic lasers maintain strong confinement and optical mode-gain overlap over a broad range of nanowire diametres (Fig. 2e - f) with only a weak dependence on the nanowire diametre (see Supplementary Information). While hybrid modes do not experience mode cut-off, the threshold intensity increases for smaller nanowires due to

the reduction in the total gain volume. Conversely, photonic lasers exhibit a strong dependence of the mode confinement on the nanowire diametre (Fig. 2g - h), resulting in a sharp increase in the threshold intensity at diametres near 150 nm due to a poor overlap between the photonic mode and the gain material. Moreover, actual photonic lasers suffer mode cut-off as the leakage into the quartz substrate prevents lasing for nanowire diametres less than 140 nm [7]. This is to say, the observation of plasmonic lasing for nanowire diametres of just 52 nm confirms the role of the hybrid plasmonic mode where a purely dielectric nanowire mode cannot exist.

Plasmonic modes often exhibit highly polarised behaviour as the electric field normal to the metal surface binds most strongly to electronic surface charge. We have detected the signature of lasing plasmons from the polarisation of scattered light from the nanowire end-facets (see Supplementary Information), which is in the same direction as the nanowire. Conversely, the polarisation of scattered light from photonic lasers is perpendicular to the nanowire. This distinction provides a direct confirmation of the plasmonic mode.

We find a strong increase of the spontaneous emission rate when the gap size between the nanowire and metal surface is decreased. Lifetime measurements reveal a Purcell factor of more than 6 for a gap width of 5 nm and nanowire diametres near 120 nm (Fig. 3), where hybrid plasmonic modes are most strongly localized [16]. This enhancement factor corresponds to a mode that is 100 times smaller than the diffraction limit, which agrees well with our mode size calculations (see Supplementary Information). While the enhanced emission rate, or Purcell effect [17], is usually associated with high quality micro-cavities [3, 4], we observe a broad-band Purcell effect arising from mode confinement alone without a cavity [20].

We next examine the physics underlying the gain mechanism in the plasmonic lasers, which combines exciton dynamics, the modification of spontaneous emission [17] and mode competition (see Supplementary Information). While photo-generated excitons have intrinsic lifetimes of up to 400 ps [25], they recombine faster at the edge of the

nanowire near the gap region due to strong optical confinement mediated by the hybrid plasmon mode (Fig. 3). The exciton diffusion length in bulk CdS is about a micometre [27], which is much larger than the nanowire diametre. Therefore, the distribution of exciton recombination quickly adjusts itself to match the hybrid mode's profile (see Fig. 1b). The fast diffusion and the enhanced emission rate into the hybrid plasmonic mode lead to preferential plasmon generation. In this way, the proportion of light that couples into the laser mode, known as the *spontaneous emission factor*,  $\beta$  can be high (Fig. 3b) [28]. The measured emission rates and a simple emission model show that the  $\beta$ -factor of the plasmonic mode is as high as 80% for a gap width of 5 nm (see Supplementary Information). For gap widths below 5 nm, the exciton recombination is too close to the metal surface, causing rapid non-radiative quenching [29]. While nanowires placed in direct contact with the metal surface show the highest spontaneous emission rates, these devices exhibit weak luminescence and do not lase. Our calculations support these observations indicating a sharp reduction in the  $\beta$ -factor below gap widths of 5 nm.

The laser threshold is commonly manifested as a kink between two linear regimes of the output power versus pump intensity response (log-log scale). However, it is known that lasers with strong mode confinement do not necessarily exhibit such behaviour so that the laser threshold may be obscured [28, 30]. Since the plasmonic lasers exhibit strong mode confinement, we indeed observe this *smearing* of the threshold pump intensity (Fig. 4). The photonic lasers that we have measured, on the other hand, show the distinctive kink in the output power, which is in agreement with recently reported Zinc Oxide nanowire lasers [7]. Since both plasmonic and photonic lasers in this work utilize the same gain material, we conclude that the smeared response in output power arises from local electromagnetic field confinement. We therefore attribute this distinct behaviour to the increased spontaneous emission factor of the hybrid plasmon mode. A high spontaneous emission factor is often associated with low threshold laser operation where undesired emission modes are suppressed. While the linear response, shown in Fig. 4 for a nanowire diametre of 66 nm, may therefore give the impression of a lower threshold [28], in reality, the laser threshold only occurs once cavity losses are compensated (see Supplementary Information).

The demonstration of deep sub-wavelength plasmonic laser action at visible frequencies paves the way to new sources that produce coherent light directly far below the diffraction limit. Extremely strong mode confinement, which is evident from the increase of the spontaneous emission rate by up to 6 times, and the resulting high spontaneous emission factor are key aspects for the operation of deep sub-wavelength lasers. We have also shown that the advantage of plasmonic lasers is the ability to down-scale the physical size of devices, as well as the optical modes they contain, unlike diffraction limited lasers. Furthermore, the use of metals in plasmonics could provide a natural route toward electrical injection schemes that do not interfere with mode confinement [6]. The impact of plasmonic lasers on optoelectronics integration is potentially significant as the optical fields of these devices rival the smallest commercial transistor gate sizes and thereby reconcile the length scales of electronics and optics.

## **Methods**

The technical challenge of constructing the plasmonic lasers is ensuring good contact between the nanowire and planar optical film. Low film roughness ( $\sim$ 1 nm rms) and accurate deposition ( $\sim$ 10%) allow <2 nm gap width. Crystalline CdS nanowires exhibit extremely low surface roughness (see Supplementary Information), hence the contact is limited by film roughness. Nanowires were grown using chemical vapour deposition of CdS powders on Si substrates by self assembly from a 10 nm Au film seeding layer leading to random nanowire diametres, d = 50 - 500 nm. CdS nanowires were deposited from solution by spin coating onto pre-prepared films with varying MgF<sub>2</sub> thicknesses of 0, 5, 10 and 20 nm, along with control devices of nanowires on a quartz substrate.

A frequency doubled, mode-locked Ti-Sapphire laser (Spectra Physics) was used to pump the plasmonic and photonic lasers ( $\lambda_{pump} = 405$  nm, repetition rate = 80 MHz, pulse length = 100 fs). An objective lens (20x, NA = 0.4) was used to focus the pump beam to a 38  $\mu$ m diametre spot on the sample. All experiments were carried out at low temperature, T < 10 K, utilizing a liquid-He cooled cryostat (Janis Research). Individual spectra were

recorded using a spectrometer with a resolution of 0.25 nm and a liquid- $N_2$  cooled CCD (Princeton Instruments). The lifetime measurements were conducted under very low pump conditions to avoid heating and exciton-exciton scattering effects using time-correlated single photon counting (PicoQuant: PicoHarp 300, Micro Photon Devices APD 40 ps timing resolution). A 490  $\pm$  10 nm band pass filter was used to filter out ambient light and pass light from the  $I_2$  CdS exciton line near 489 nm. An emission model was used to describe the enhanced lifetime data (Purcell effect), which relates to deep sub-wavelength mode confinement (see Supplementary Information).

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## **Figure Captions**

**Figure 1 The deep sub-wavelength plasmonic laser. a** The plasmonic laser consists of a Cadmium Sulphide semiconductor nanowire atop a Silver substrate, separated by a nanometre scale MgF<sub>2</sub> layer. This structure supports a new type of plasmonic mode [16] whose mode size can be 100 times smaller than a diffraction limited spot. The inset shows a scanning electron microscope image of a typical plasmonic laser, which has been sliced perpendicular to the nanowire's axis to show the underlying layers. Panel **b** shows the electric field distribution and direction |E(x,y)| of a hybrid plasmonic mode at a wavelength  $\lambda = 489$  nm corresponding to the CdS I<sub>2</sub> exciton line [25]. The cross-sectional field plots (along broken lines in field map) illustrate the strong overall confinement in the gap region between the nanowire and metal surface with sufficient modal overlap in the semiconductor to facilitate gain.

Figure 2 Laser oscillation and threshold characteristics of plasmonic and photonic lasers, a Laser oscillation of a plasmonic laser, d = 129 nm, h = 5 nm (longitudinal modes). The four spectra for different peak pump intensities exemplify the transition from spontaneous emission (21.25 MWcm<sup>-2</sup>) via amplified spontaneous emission (32.50 MWcm<sup>-2</sup>) to full laser oscillation (76.25 MWcm<sup>-2</sup> & 131.25 MWcm<sup>-2</sup>). **b** Shows the nonlinear response of the output power to the peak pump intensity. The relationship between mode spacing,  $\Delta\lambda$ , and nanowire length, in c indicates a high group index of 11 due to the high material gain. The pictures on the left correspond to microscope images of a plasmon laser with d = 66 nm exhibiting spontaneous emission, amplified spontaneous emission and laser oscillation, where the scattered light output is from the end-facets. d Threshold intensity of plasmonic and photonic lasers versus nanowire diametre. The experimental data points correspond to the onset of amplified spontaneous emission, which occurs at slightly lower peak pump intensities compared to the threshold of gain saturation (see Supplementary Information). Amplified spontaneous emission in hybrid plasmonic modes occurs at moderate pump intensities of 10-60 MWcm<sup>-2</sup> across a broad range of diametres. Plasmonic lasers maintain good mode confinement over large range of diametres as shown in e (d = 200 nm) and remain confined even for a 50 nm diametre wire as shown in **f**. While the photonic lasers have similar threshold intensities around 10 MWcm<sup>-2</sup> for all nanowires larger than 200 nm shown in g, a sharp increase in the threshold occurs for diametres near 150 nm, due to the loss of confinement within the nanowire and subsequent lack of overlap with the gain region [7] shown in h. The solid lines show a numerical fit to a simple rate equation model (see Supplementary Information). Below 140 nm, the photonic mode is *cut-off* and lasing could not be observed at all. In contrast, plasmonic lasers maintain strong confinement and optical mode-gain overlap for diametres as small as 52 nm, a diametre for which a photonic mode does not even exist. The relatively small difference in the thresholds of plasmonic and photonic lasers can be attributed to high cavity mirror losses, which are on the same order of magnitude as plasmonic losses.

**Figure 3 The Purcell effect in plasmonic and photonic lasers. a** The emission rates of photonic and plasmonic nanowire lasers with different MgF<sub>2</sub> gap widths as a function of

the nanowire diametre following the calculated trend. The optimal confinement condition of hybrid plasmonic modes is found near d=120 nm, where the hybridization of nanowire and SPP modes is strongest giving the highest emission rate [16]. The emission rates of nanowires placed in direct contact with the metal surface are given in the Supplementary Information. Panel **b** shows the Purcell factors determined from a numerical fit of the emission rate measurements to a simple emission model (see Supplementary Information). Near optimal confinement (d=120 nm  $\pm 20$  nm), the average Purcell factor for devices with 5 nm gaps is more than 6, which is considered high for a broad band effect. We have also calculated the  $\beta$ -factor from the emission model fit by accounting for the possible emission pathways. The  $\beta$ -factor reaches a maximum of 80 % for a gap width near 5 nm. At gap widths smaller than 5 nm, non-radiative quenching to the metal surface causes a sharp drop in the hybrid plasmon  $\beta$ -factor, which subsequently eliminates the possibility for lasing. The error bars show standard deviation of emission rates from data collected within 25 nm nanowire diametre ranges.

Figure 4 The signature of threshold-less lasing due to high spontaneous emission factor. The dependence of measured output power over pump intensity highlights clear differences in the physics underlying the plasmonic (h = 5 nm) and photonic lasers using a multi-mode lasing model [30]. In particular, our fitting parameter,  $x_0$ , is related to the gain saturation of individual longitudinal laser modes and their lateral mode area. A higher value of  $x_0$  corresponds to a smaller mode area and a higher  $\beta$ -factor. Photonic lasers exhibit a clear transition between spontaneous emission and laser operation characterized by a change in the gradient of input peak pump intensity versus output power, corresponding to the laser threshold. For a 150 nm diametre nanowire, the parameter  $x_0 = 0.026$  is in agreement with a recent nanowire laser study [7]. Plasmonic lasers, however, show a strong dependence of  $x_0$  on the nanowire diametre; a large multimode plasmonic laser (d = 232 nm) shows a somewhat smeared transition region ( $x_0 = 0.074$ ), while smaller single mode plasmonic lasers (d = 129 nm and d = 66 nm) have much less visible changes in gradient ( $x_0 = 0.150$  and  $x_0 = 0.380$  respectively). The large value of  $x_0$  observed in the plasmonic lasers is associated with so-called threshold-less

operation and attributed to the strong mode confinement giving rise to a high spontaneous emission factor. Nevertheless, the onset of amplified spontaneous emission peaks in all devices occurred at non-zero threshold intensities as shown in Fig. 2d.

# **Manuscript Figures**

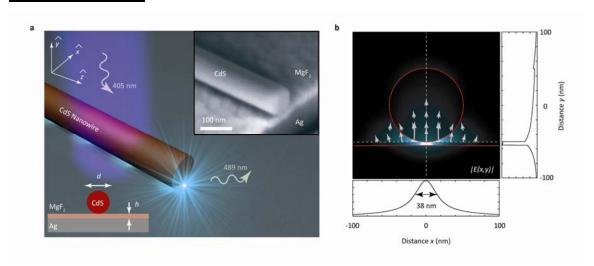


Figure 1

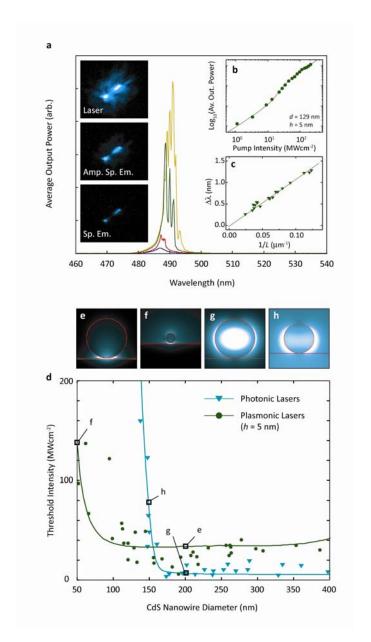


Figure 2

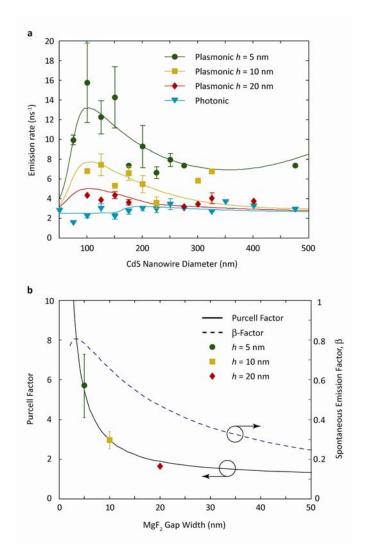


Figure 3

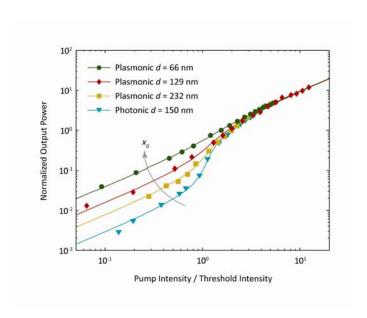


Figure 4