Thresholdless Coherent Light Scattering from Subband-polaritons in a Strongly-Coupled Microcavity.

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We study a “strongly-coupled” (SC) polariton system formed between the atom-like intersubband transitions in a semiconductor nanostructure and the THz optical modes that are localised at the edges of a gold aperture. The polaritons can be excited optically, by incoherent excitation with bandgap radiation, and we find that they also coherently scatter the same input laser, to give strikingly sharp, multiple-order sidebands (SB’s) at the input beam, generating strikingly sharp, multiple-order sidebands (SB’s), at \( \omega_{SB} \sim \lambda \sim 100 \mu m \) free-space wavelength. Upper left inset: - Raw spectra of the light backscattered from the nanostructure, embedded in a THz microcavity which is symbolised here by external mirrors. THz polaritons are created (A) by photo carriers, excited by a near IR laser, \( \omega_{THz} \), cascading down through the subband system until they reach the polariton state. The same near IR laser (B), is also coherently scattered from these \( \omega_{THz} \) polaritons, and generates “sidebands” at \( \omega_{SB} = \omega_{THz} + n \omega_{THz} \). Upper left inset: Raw spectra of the light backscattered from the top of the device, as the \( \sim 50 \mu m \) diameter \( \lambda \sim 744nm \) laser spot is scanned along the centre of the ridge, across one of the slots, at distances from the centre of the slot of 0 \( \mu m \) (squares), 20\( \mu m \) (triangles), 40\( \mu m \) (circles) and 100\( \mu m \) (inverted triangles).

Fig 1. A single period of the nanostructure, embedded in a THz microcavity which is symbolised here by external mirrors. THz polaritons are created (A) by photo carriers, excited by a near IR laser, \( \omega_{THz} \), cascading down through the subband system until they reach the polariton state. The same near IR laser (B), is also coherently scattered from these \( \omega_{THz} \) polaritons, and generates “sidebands” at \( \omega_{SB} = \omega_{THz} + n \omega_{THz} \). Upper left inset: - Raw spectra of the light backscattered from the top of the device, as the \( \sim 50 \mu m \) diameter \( \lambda \sim 744nm \) laser spot is scanned along the centre of the ridge, across one of the slots, at distances from the centre of the slot of 0 \( \mu m \) (squares), 20\( \mu m \) (triangles), 40\( \mu m \) (circles) and 100\( \mu m \) (inverted triangles).

In a strongly-coupled (SC) system, the electron and photon sub-systems are so strongly coupled to each other that they form a new hybrid entity, a polariton, where the coupling is characterised by a vacuum-Rabi energy, \( \hbar \Omega_{VR} \), that exceeds the original natural linewidths of both the electron and photon modes. Viewed in the time domain, the excitation energy cycles coherently, at a rate \( \hbar \Omega_{VR}^{-1} \) between electronic and photonic forms, before being lost, either by optical emission or by other non-radiative loss channels [1].

Here we study a SC system whose polaritons are formed from the THz photon modes of a tightly-confined metal-semiconductor microcavity, that are hybridised with the electronic “Intersubband transitions” (ISBT’s) in a semiconductor nanostructure. These polaritons carry an optical dipole, so when we create a population of them incoherently, (with an interband near-infrared pump laser, \( \omega_{NIR} \) (fig.1)), we find that they coherently scatter that same input beam, generating strikingly sharp, multiple-order sidebands (SB’s), at \( \omega_{SB} = \omega_{NIR} + n \omega_{THz} \) (n=-2,-1,0,+1,+2) in the spectrum of backscattered light.

The devices studied were fabricated from epilayers comprising many (~175) repeats [2,3] of a GaAs/ [Al,GaAs] semiconductor nanostructure module [fig 1]. The epilayers were fabricated into ~10\( \mu m \) thick gold-epilayer-gold sandwich waveguides that used the near ideal gold conductivity [4,5] to confine the THz fields into layers ~10 times thinner than the \( \lambda \sim 100 \mu m \) free-space wavelength. Three device structures were studied, identified here as “\( \lambda \sim 80\mu m \)” [3], “\( \lambda \sim 100 \mu m \)” [2] and “\( \lambda \sim 120 \mu m \)” [3] corresponding to the approximate THz free-space wavelengths they emitted when they were electrically driven, in separate experiments [2,3], as standard quantum cascade lasers (QCL’s).
Fig 2(a) T~14K Background-subtracted spectra taken, from the “λ~120 μm” (solid line, right hand vertical axis), and “λ~80 μm” (dotted line, left hand vertical axis) devices. Both spectra show coherent sideband features at ω₀NIR = eℏωNIR/t_πω(ℏ/2,1,1/2,2/2), whose linewidths are set by the spectrometer resolution (Δω~ 1.4nm in this case). (b) Comparison between the T~ 14K Anti-Stokes GaAs Raman line, [enhanced by x100 on this plot] and a typical n=+1 sideband feature. (c) Temperature dependence of the SB intensity measured with a near IR wavelength of 810nm and ~1mW power entering the “λ~80 μm” device; squares, the n=-1 SB intensity (right hand axis), filled circles, the n=2 SB intensity (left hand axis); open circles, output of a QCL made from the same heterostructure [3] (arb units).

Each period of the the “λ~100μm” nanostructure in fig. 1 [2] comprises 4.9/7.9/2.5/6.6/4.1/15.6/3.3/9.0 nm thick layers of Al induced GaAs/GaAs and supports ~5 confined electron states. The 15.6 nm well is doped at 1.9 x 10¹⁰ cm⁻², giving an areal electron density, n = 3 x 10¹⁰ cm⁻² and a Fermi energy of ~1meV, so only the lowest subband is occupied at equilibrium. The 1.52 eV photoluminescence peak implies an effective bandgap close to that of bulk GaAs and the |2> => |1> ISBT had a modelled energy of E₁₂ =15.8 meV and a transition dipole of |z₁₂| = 2.3 nm. The waveguide is ~10μm x 50 μm x 834 μm long. It has a periodic array (fig. 1 inset) of 6 30 μm x 8 μm slots, spaced by Λ = 31 μm, etched into it’s top surface. These were originally designed to outcouple the THz fields when the structure was used as a QCL [4].

The devices were illuminated normally, with a tuneable continuous-wave titanium:sapphire near-infrared laser, ω₀NIR, with a 50μm spot size and a linewidth ΔωNIR < 0.1 meV. The backscattered light was polarisation filtered before being analysed with a 0.25m grating monochromator and a standard, background-subtracting, photon-counting setup whose cooled photomultiplier had a dark count < 10 cps. The sharpness of the SB’s meant careful attention to the system’s mechanical and laser wavelength stability was needed to see them.

Backscattered spectra were typically dominated by an elastically scattered background peak and a λ =815 nm / 1.52 eV photoluminescence peak (not shown), but they also featured strikingly sharp “sideband” (SB) peaks. The data presented in figures 1 - 3 were taken with the spectrometer slits opened up to improve the signal-to-noise ratio, but separate trials (not shown) always found SB linewidths that were resolution-limited, down to the 0.3nm/0.5meV working limit of the spectrometer. For a given device, the SB features stayed at a fixed frequency interval from ω₀NIR, as the Ti:sapph laser was tuned over a wide wavelength range (fig.3).

All the SB’s disappeared if ω₀NIR was tuned below the ~1.52 eV/ 815 nm effective bandgap of the nanostructure. Both 1st and 2nd order peaks vanished [fig.2 (c)] by ~120K, similar to the maximum operation temperature of comparable QCL’s [3]. The optical polarisation of the SB’s matched the polarisation of the ω₀NIR input to better than 99%, whether it was linear or circular.

When a small bias was applied across the waveguide it acted as a photocative detector which could be calibrated to measure the fraction of the ω₀NIR beam that made it through the illuminated slot in the top gold layer. When the laser spot was scanned along the centre of the waveguide, across a given slot, (Fig 1 inset) the SB intensity scaled with this photocurrent.

The SB’s were reproducible over months of measurement and through numerous changes to the optical setup. They were never seen in control experiments, when the ω₀NIR spot was focussed onto (i) un-metallised epilayer samples, (ii) onto GaAs test wafers, (iii) onto highly scattering parts of the cryostat mount or (iv) onto the gold contacting layers adjacent to the slots in the structure of fig.1.

The way the SB’s tune with ω₀NIR [figs. 3(b) and 3(c)] immediately implies that they are generated by a form of coherent scattering. We rule out normal phonon Raman scattering because the SB’s are ~100 x more intense, at least 12 times sharper, and appear at the wrong energy offsets [fig. 2(b)] compared with typical LO phonon Raman lines.

Coherent scattering processes generate SB’s whose lineshape is a convolution of the linewidths of the input laser (<0.01meV) and that of the scattering excitation. Therefore, the sharpness of the SB lines [< 0.5meV] compared with an estimated bare ISBT linewidth of ~ meV [6] argues that they cannot be due to standard electronic Raman scattering from the ISBT.

SB features which are superficially similar to the ones we see here have previously been generated by exploiting optical frequency mixing effects which arise due to non-linear components of the optical polarisability in the material of a QCL [7,8]. However, this mechanism requires the presence of a spectrally sharp THz optical field inside the structure, and this could only be happening here if we had somehow managed to produce an optically-pumped...
between the detected sideband intensity and the comparable QCL threshold. 

observed in Raman and non-linear frequency mixing processes are seen [fig.2(a)] contrasts sharply with what is excited [2], its threshold current density, \( J_{\text{th}} \approx 435 \text{ A cm}^{-2} \), device of fig. 1 was configured as a QCL and electrically power is increased. When a comparable structure to the entering the slot [Fig.3(a)], single, dispersionless, atom-like Lorentzian line, with a strong electron correlation modes in these devices. The ISBT energy is independent of the detector system’s photon noise floor as the power entering QCL (mW) is scanned . (c) tuning behavior (right hand axis) of the \( n=+2 \) (squares), \( n=+1 \) (circles), \( n=-1 \) (diamonds) and \( n=-2 \) (triangles) SB peaks from the \( \sim 100 \mu \text{m} \) structure. Spectra (left hand axis), of the \( n=+2 \) SB’s peaks from the same sample at the input wavelength denoted by the corresponding triangle data point on the tuning line. All data taken at \( T\sim 14 \text{K} \).

To understand the origin of the sidebands we first need to define the SB generation efficiency, \( \eta_{\text{SB}} \), as the ratio between the detected sideband intensity and the \( \delta \Omega_{\text{NR}} \) power entering the slot [Fig.3(a)], \( \eta_{\text{SB}} \) climbs linearly from the detector system’s photon noise floor as the \( \delta \Omega_{\text{NR}} \) pump power is increased. When a comparable structure to the device of fig. 1 was configured as a QCL and electrically excited [2], its threshold current density, \( J_{\text{th}} \approx 435 \text{ A cm}^{-2} \), corresponded to \(-4.8 \times 10^{10} \text{ sec}^{-1} \text{ m}^{-2} \) ISBT transitions throughout the epilayer’s 175 periods. In our optical experiments (fig.3) the SB first appears with \(-0.1 \text{mW} \) entering the \( 2.4 \times 10^{-5} \text{ m}^{2} \) slot, an areal excitation rate \(~1.7 \times 10^{25} \text{ sec}^{-1} \text{ m}^{-2} \) , i.e. \(~2500 \) times smaller than the comparable QCL threshold.

Finally, the ease with which the \( n\pm2 \) higher order processes are seen [fig.2(a)] contrasts sharply with what is observed in Raman and non-linear frequency mixing experiments.

The model correctly reproduces the “radiating” modes (fig. 4(a) squares) that were originally intended to outcouple [4] the THz when biased as a QCL. However, at large transition dipole, \( z_1 \), that has already been shown to generate SC with giant \( \hbar \Omega_{VR} \) energies in planar structures [10,11], especially with the wider wells used in THz devices [12]. In fact, \( \hbar \Omega_{VR} \) values have been achieved that not only exceed the linewidths, but are also are comparable with the transition energy itself [13,14], the so-called ultra-SC (USC) condition[15].

In our non-planar devices, the photon modes are confined in all 3 dimensions, so to estimate the degree to which they couple with the ISBT’s we must first compute their mode shapes and volumes. This is done with a standard finite-difference-time-domain (FDTD) calculation, on a 125 nm mesh, which treats the gold as a perfect conductor and the semiconductor as an insulator with a dielectric constant of 13.3 [16]. It models the ridge structure of fig 1 as an infinite array of slots so that periodic boundary conditions allow the modes to be plotted in terms of the superlattice wavevectors, \( 2\pi/\Lambda \), where \( \Lambda = 31 \text{ nm} \) is the slot repeat distance (fig.4)

The “radiating” mode designed to out-couple radiation when electrically driven as a QCL Red triangles:- the “localised” family of modes. Dotted (solid) lines are light lines in the vacuum (semiconductor). (b) energy distribution, across a single slot in the periodic structure, of the “localised” modes. (c) Schematic 3D distribution of the fields in the “localised” mode concentrated at the edges of the slots in the structure.

almost the same energy (12.5 meV/\(-3 \text{ THz} \)) there is another family of “localised” THz modes [fig. 4(a) triangles] whose field distributions are tightly localised at the slot edges, similar to the ultra-confined modes recently reported [17,18] in other sub-wavelength structures. These originate from a vertical \( 1/4 \) wave “organ–pipe” resonance, with a node...
at the lower gold layer and an antinode at the slot opening, giving a resonance roughly corresponding to a free-space wavelength $\lambda \approx 4n\lambda$, where $n$ is the semiconductor refractive index and $h \approx 10 \mu m$ the slab thickness. The field localisation means that photon modes on adjacent slots oscillate almost independently, so they are practically mono-energetic and dispersionless in the photonic superlattice plot (fig. 4(a)), and all the THz modes in the family can strongly couple to the electronic ISBT's at the same time. This contrasts with the anti-crossing behaviour previously seen in dispersive 2D systems [11-15]. The field localisation around the slot edge also gives very weak outcoupling to the free-space THz modes, raising the Q factor to $\approx 1100$, compared with $\approx 57$ for the "radiating" modes.

The computed volume of the "localised" mode, $V \approx 496 (\mu m)^3$, is only $\approx \lambda^3/2000$ of the $\lambda \approx 100 \mu m$ free space wavelength and $\approx 1/50$ of $\lambda^3$ in the semiconductor material. It’s frequency resonates closely with the modelled $E_{12} \approx 15 meV$ (fig. 1) ISBT energy and its electric field is mainly vertically polarised, so it couples strongly to the vertically polarised $z_{12}$ of the ISBT. Also, it’s half-height energy density (fig. 4b) is only $\approx 0.96 \mu m$ below the semiconductor-air interface, so overlaps well with the $\approx 1 \mu m$ penetration depth of the $\omega_{NR}$ incoherent pump light from the Ti:sapphire laser[16].

We compute $\hbar\Omega_{VR}$ for the "localized" photon mode by equating the classical stored electromagnetic energy, $V_{\epsilon_{0}}\epsilon_{0}E_{\text{vac}}^2$, with the quantum photon ground state energy, $\hbar\omega_{VR}/2$, to give a mean zero-point vacuum field of $E \approx 142 V \text{ m}^{-1}$. The interaction of a single electron ISBT oscillator with this field will give $\hbar\Omega_{VR} = 2E\epsilon_{0}z_{12}$, which, with $\epsilon_{0} = 13.5 \ [16]$, $z_{12} \approx 2.3 nm$, and $\hbar\omega_{THz} = 15 \text{ meV}$ gives $6.3 \times 10^{-7} \text{ eV}$. A factor $f=0.92$ of the mode energy lies inside the semiconductor, and $N \approx 10^6$ ISBT electrons lies within this volume, giving a total coupling energy of $\hbar\Omega_{VR} = 2E\epsilon_{0}z_{12} N \approx 1.0 \text{ meV}$. Even with no photoexcitation this is some $\approx 7\%$ of the ISBT energy, and will increase further under the experimental conditions. Assuming an interband carrier recombination time of $\approx 1 \text{ nsec}$, $\approx 1 \text{ mW}$ of absorbed laser power would triple the local electron concentration and increase $\hbar\Omega_{VR}$ by $\sqrt{3}$.

This $\hbar\Omega_{VR}$ value exceeds both the $\approx 0.5 \text{ meV}$ upper bound to the linewidth of the excitation responsible for the SB generation and the $\approx 15 \text{ meV}$ modelled linewidth of the localised photon mode. This confirms the SC nature of the electron-photon system, i.e. the SB’s arise from coherent scattering mechanism from polaritons whose linewidths lie between [6] the $\approx 1 \text{ meV}$ ISBT linewidth and the $\approx 15 \text{ meV}$ localised photon mode linewidth.

At higher optical excitation levels (not shown), the $\lambda \approx 100\mu m$ device $n=1$ SB conversion efficiency peaks at $\approx 5 \times 10^{-3} \text{ [at } 1 \text{ mW input power]}$, and then drops, because of sample heating (fig. 2(c)).

There are strong parallels between this SC system, and previous atom-cavity studies [19], where coherent output radiation was seen without the system needing to be driven into population inversion. Unfortunately, our attempts to detect the emitted optical component of the THz polaritons produced in these experiments (i.e. to demonstrate a THz analogue of a so called “inversionless laser”) were frustrated by the poor sensitivity of current THz detectors, and the very weak coupling of the “localised” modes to the outside world.

That said, we believe that this effect may prove more useful as a simple, passive coherent optical mixing devices than as a source of THz radiation. Although the conversion efficiencies and operating temperatures are low at the moment, this effect still has potential for frequency-shifting e.g. an optical bit stream by a fixed frequency interval that can be tightly specified at the design stage and would be data-transparent and operate across the full optical telecommunications bandwidth. Moving to the [In,Al,Ga],As materials system and optimising $\hbar\Omega_{VR}$ by judicious choice of doping levels, THz mode shapes and ISBT $z_{12}$ values may move the operating wavelengths, temperatures and efficiencies towards technologically useful values.

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