Superfluorescence from dense electron-hole plasmas in high magnetic fields

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Abstract. Cooperative spontaneous recombination (superfluorescence) of electron-hole plasmas in semiconductors has been a challenge to observe due to ultrafast decoherence. We argue that superfluorescence can be achieved in quantum-confined semiconductor systems and present experimental evidence for superfluorescence from high-density photoexcited electron-hole plasmas in magnetized quantum wells. At a critical magnetic field strength and excitation fluence, we observe a clear transition in the band-edge photoluminescence from omnidirectional output to a randomly directed but highly collimated beam. Changes in the linewidth, carrier density, and magnetic field scaling of the emission spectra correlate precisely with the onset of random directionality and are consistent with cooperative recombination.

1. Introduction

Superfluorescence (SF) [1] is one of the very few observable quantum phenomena in which a macroscopic ensemble of initially incoherent quantum oscillators demonstrates cooperative, coherent behavior due to an efficient mutual self-phasing. In this process, a system of N inverted atoms is initially incoherently prepared, but macroscopic coherence builds up self-consistently, starting from vacuum fluctuations, via exchange of photons. The resultant macroscopic dipole decays superradiantly [2] after a certain delay time t_d, producing a burst of highly-directional coherent radiation whose duration scales as $1/N$ in the simplest case, and peak intensity can be proportional to N^2. The duration of the SF pulse and the delay time are shorter than the times of incoherent spontaneous emission T_1 and phase relaxation T_2 in the medium. This is the distinctive feature of SF which makes this phenomenon so different from superluminescence or standard lasing. In a sense, SF places a fundamental upper limit on the rate of stimulated radiative transitions in matter. Note the principal difference between SF and superradiance [2,3]: the latter develops in a system in which the coherent macroscopic polarization has been initially excited by an external laser field.

SF has been observed in atomic systems and rarefied impurities in crystals but still unobserved in semiconductor systems where ultrafast decoherence tends to destroy optical coherence. Here, we present experimental evidence for SF from cooperative recombination of high-density electron-hole (e-h) plasmas in semiconductor quantum wells (QWs) placed in a strong perpendicular magnetic field. Strong-field quantization and Fermi degeneracy increase the density of states and suppress decoherence processes, making SF possible. We measure emission as a function of pump laser fluence

F and the magnetic field B. As the laser fluence increases, the band-edge photoluminescence peak evolves into strong multiple lines, whose energies correspond to excited Landau levels. At high enough magnetic fields, the widths of these peaks show a significant narrowing followed by a slight broadening with increasing fluence, signaling a crossover from amplified spontaneous emission (ASE) to SF. As we enter the SF regime, the emitted radiation shows the expected random directionality from shot to shot, reflecting the fluctuation-driven origin. This inherent randomness is a hallmark of SF: the fluctuations of the polarization grow from initially incoherent quantum noise and reach a macroscopic level. Both the emission direction and the delay time between the pump pulse and the SF pulse should vary from shot to shot. These observations thus constitute the first realization of SF from free carriers in a semiconductor.

2. Superfluorescence versus spontaneous emission

The key parameter governing the growth rate of cooperative emission is the coupling strength between the electromagnetic field and optical polarization, expressible as the cooperative frequency ω_c. To observe SF from a system of *e-h* pairs in a semiconductor QW, $\omega_c = \left(8\pi^2 d^2 N\Gamma c/(\hbar n^2 \lambda L_{QW}\right)^{1/2}$ must be greater than $2/T_2$ (or $2/(T_2T_2^*)^{1/2}$ for a system with inhomogeneous dephasing time $T_2^* < T_2$) [4,5]. Here d is the dipole moment of the transition, N is the 2D *e-h* density within the spectral bandwidth of SF emission, Γ is the overlap factor of the radiation with the active QW region, c is the speed of light, • is the Planck constant, n is the index of refraction, λ is the wavelength of the radiation in vacuum, and L_{QW} is the total width of the QW system.

SF develops with the growth rate $g \approx \omega_c^2 T_E/4 \propto N$ when the cooperative frequency is smaller than the field dissipation rate: $1/(T_2T_2^*)^{1/2} < \omega_c/2 < 1/T_E$. When the cooperative frequency becomes greater than the field dissipation rate, $\omega_c/2 > 1/T_E, 1/T_2$, the growth rate reaches its maximum value $g \approx \omega_c/2 \propto N^{1/2}$. Here T_E is the photon lifetime for a given field mode determined by the geometry of a sample. The SF pulse duration is $\tau_{SF} \approx 2/g \ll T_2$, while the delay time is longer than τ_{SF} by a logarithmic factor of order 10. An incoherent process of ASE exists in the regime of low inversion density and long photon lifetime, i.e., $1/T_2^*$, $1/T_2 >> \omega_c/2 > 1/T_E$. Its growth rate is slower than the dephasing rate: $g_{ASE} \approx (\omega_c^2/4)\min\left[T_2, T_2^*\right] - 1/T_E << 1/T_2$. Even in the high-gain regime $g_{ASE}L/c >> 1$, where L is the length of the inverted region (typically 0.5 mm), ASE leads to the formation of relatively long pulses of duration ~ Ln/c ~ 6 ps > T_2 emitted in all electromagnetic modes simultaneously, i.e. without shot-to-shot fluctuations of pulse direction. The high-gain regime of ASE is often called superluminescence.

In our experiments, *e-h* pairs are created high in the band having very short T_2 (~ a few fs), then thermalize on Landau levels (LLs) during the first ~100 fs and become degenerate with a long T_2. Obviously, the ensemble of degenerate *e-h* pairs created in this way is initially completely incoherent. In a low density limit they emit a spontaneous emission peak with an inhomogeneous Gaussian full width of ~9 meV. Spontaneous emission is emitted isotropically in all directions. When inversion and net gain are reached, the ASE process develops. It leads to the emission of ASE pulses of duration Ln/c ~ 6 ps. Since the pumped area is a circle, ASE proceeds in all directions along the plane simultaneously. In the high gain regime, the spectrum narrows down to 2 meV FWHM, which is still larger than $2/T_2$. Note that ASE radiates away only a half of the inversion, bringing it to zero. Remaining *e-h* pairs recombine with the time T_1 ~ 1 ns. When the energy density of states is high enough (so they can be condensed tightly in the energy spectrum) and their volume density is high enough, the cooperative frequency ω_c becomes larger than $2/(T_2T_2^*)^{1/2}$. Then, the *e-h* pairs become cooperated after a short delay time and emit a SF pulse or a sequence of pulses, depending on the pump fluence and the size of the pumped area. At very high pump powers the line becomes broadened due to the shrinking pulse duration, until eventually saturation (due to the filling of all available states) halts further decrease in pulse duration.

3. Experimental results and discussion

The samples were grown by molecular-beam epitaxy on GaAs substrates, and consisted of a GaAs buffer layer followed by 15 layers of 8-nm $In_{0.2}Ga_{0.8}As$ QW separated by 15-nm GaAs barriers and a 10-nm GaAs capping layer. We used a 150 fs, 775 nm Ti:Sapphire regenerative amplifier system to measure the photoluminescence as a function of laser fluence and magnetic field up to 9.7 mJ/cm^2 and 25 T, respectively. A laser fluence of 0.01 mJ/cm^2 generated a carrier density of ~ 10^{12} *e-h* pairs/cm^2 in our structure. The pulse was focused into a spot of variable size with a circular lens. Emission was collected by using optical fibers from the opposite face and cleaved edges of the sample at 4.2 K. Details of the experiment can be found in [6].

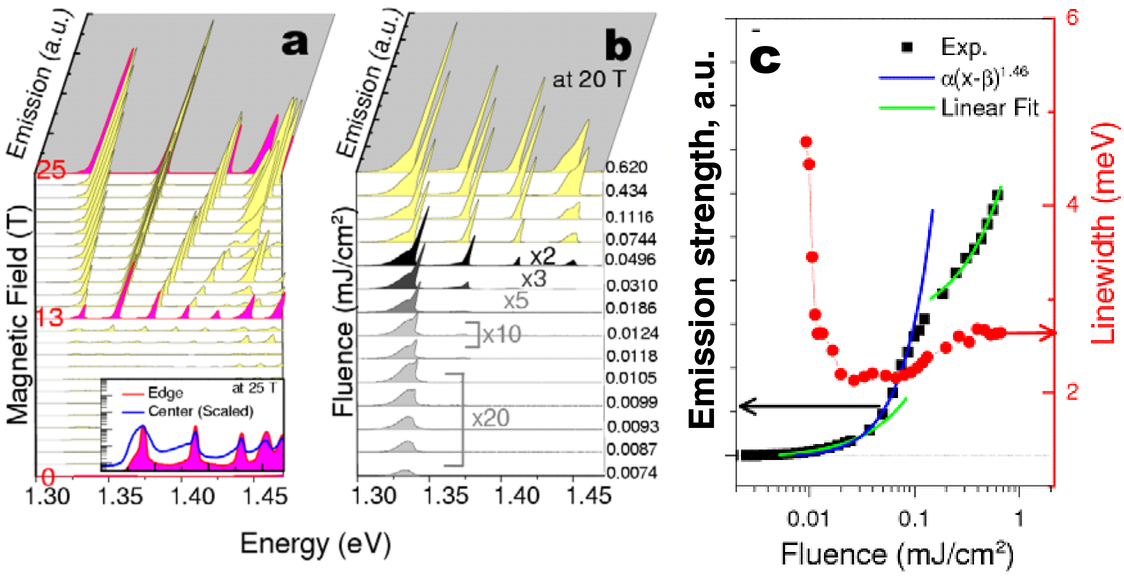

Figure 1. Emission spectra as a function of (a) magnetic field for a fixed pump fluence of 0.62 mJ/cm^2, and (b) pump fluence for a fixed 20 T field. (c) Emission strength and linewidth of the sharp Lorentzian peak from the 0th LL as a function of laser fluence. The linewidth decreases initially and then increases both versus magnetic field and versus pump fluence.

Figure 1(a,b) shows spectra collected at the sample edge perpendicular to the excitation direction (a) versus *B* at a fixed *F* and (b) versus *F* at a fixed *B* (20 T) for a temperature of 10 K for an excitation spot size of 0.5 mm. A threshold is observed in both cases; inhomogeneously broadened photoluminescence peaks (~ 9 meV) are seen at each interband LL transition until *F* and *B* exceed a threshold value, whereupon a narrow peak (~ 2 meV) emerges from the high-energy side of the broad feature and dominates at high *F*. Identical spectra are seen when collecting light from the opposite side of the sample above the pump spot (left inset), although at a much lower efficiency. Increasing or decreasing the pump spot size resulted in qualitatively similar spectra for a given fluence; broad photoluminescence was observed at low fluence or field while a narrow blue-shifted peak observed at higher fluence with much higher signal strength. Thus, the observed behavior is not due to a spatially or spectrally inhomogeneous distribution of carriers and indicates the onset of stimulated emission.

The integrated strength of the 0th LL emission versus pump fluence (Figure 1c) was obtained from a Lorentzian lineshape analysis of the narrow blue-shifted feature. Below 0.01 mJ/cm^2, no narrow emission is observed. In the range 0.01-0.03 mJ/cm^2, the signal grows linearly (green lines) with both *B* and *F*, as expected for saturated ASE regime. Above 0.03 mJ/cm^2, the emission strength becomes *superlinear* (blue lines) with the integrated signal $S \propto F^{3/2}$, suggestive of a transition from ASE into multiple-pulse SF (see [6] for details). The linewidths (red circles), also plotted in Figure 1c, reveal a remarkable correlation with the emission strength. In the linear regime, the linewidth decreases monotonically with fluence until the emission becomes superlinear, where the linewidth begins to increase. A diminishing linewidth is expected for ASE as the gain increases. On the other hand, in the SF regime, *line broadening* is expected as the pulse duration becomes smaller than the inverse bandwidth of the stimulated emission peak.

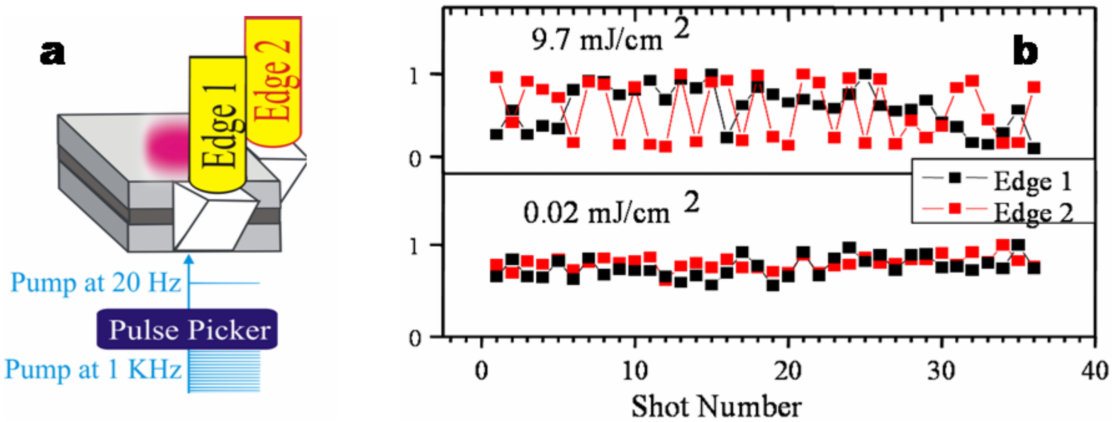

Figure 2. Single shot directionality of the emission. (a) Experimental schematic showing single-shot excitation and collection. (b) Normalized emission strength from the 0^{th} LL from 36 different pumping pulses at high (upper panel) and low (lower panel) pump fluences.

Compelling evidence for SF is found by investigating the stochastic nature of the emission direction. We collected SF simultaneously from two perpendicular edges (Figure 2a) using single pump pulse excitation of 0.5 mm in diameter. Figure 3b displays the maximum peak height from each edge (normalized to 1.0) versus shot number for two laser fluences. At a high fluence (upper panel), the maximum observed emission strength fluctuates as much as eight times the minimum value, far greater than the pump pulse fluctuation (~ 2%) implying that emission direction is highly collimated but randomly changing from pulse to pulse. At a lower pump fluence of 0.02 mJ/cm^2 (obtained with a 3 mm spot), qualitatively different behavior is seen: lower panel shows omnidirectional emission on every shot, as expected for ASE. Note that it is the disk-like geometry of the pumped active region that allowed us to observe the key evidence for SF: the growth of spatial coherence to a macroscopic level from initial quantum fluctuations and the resulting strong shot-to-shot fluctuations in the emission direction. Previous experiments almost exclusively employed a rod-like geometry, in which the only direct signature of SF is the macroscopic fluctuations of the delay time of the SF pulse. In a semiconductor system they would be manifested on the sub-ps scale and very hard to observe.

References

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