Ultrafast Optics in High Magnetic Fields:
Cooperative recombination of a quantized electron-hole plasmas

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\textit{cond-matter/0607022};
2005 \textit{NHMFL Research Highlight}

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Motivation: Quantum Optics in Solids

Quantum Optics in High Magnetic Fields:

- Is light excitation in solids really quantum, as in quantum optics of atoms?
- Unique experimental frontiers for condensed matter physics

LL Level Carrier Density

\[ N = \frac{eB}{h} \sim 10^{12} \text{ cm}^{-2} \text{ at } 25 \text{ T} \]

Superintense fs pulse

Superstrong B-field

\[ T_1 \sim 1000 \times T_2 \]

0D in QWs

Vs

0D in Atoms

\[ T_1 = 2T_2 \]

DOS
**Introduction:** Spontaneous Emission in polar semiconductors

- **Very fast (< pulse width):** Laser Pulse Excitation
- **Fast:** Intraband relaxation & Decoherence ($T_2 \sim 1$ ps)
- **Slow:** Recombination/ Luminescence ($T_1 \sim 1$ ns)

- **Conduction Band**
- **Valence Band**
- **Phonon/e-e Scattering**
Magneto-PL in 2D high density plasmas

Undoped InGaAs/GaAs superlattice

Pump creates 2D electron hole pair gas

Rapid intraband thermalization → quasi Fermi levels $F_c$ and $F_v$

Electron hole pair recombination From LL’s
**Instrumentation:** Ultrafast Spectroscopy at the NHMFL

Cell 3, 2003

Cell 5 Magnet top

Fiber Inlet

Optical Window

cryostat
Ultrafast Spectroscopy at the NHMFL

Bruker 66v Spectrometer

17.5 T SCM

31 T Bitter magnet

Laser Routing Optics

Cell 3

Cell 5
Ultrafast Spectroscopy at the NHMFL

Cell 3, 2006

Cell 5 Front

Ti:S Oscillator

SCM
Ultrafast Spectroscopy at the NHMFL

Instrumentation at the NHMFL Optics cell:

- chirped pulse amplifier (CPA)
- optical parametric amplifier (OPA)
- Ti:sapphire laser \textit{(Coherent Mira)}
- resistive magnet (31 T)
- Superconducting magnet (17.5 T)
- Streak Camera

Current Capabilities

- 150 fs, 1kHz repetition rate,
  - time-resolved pump-probe over 0.06 – 4.0 eV energy scales
  - Time-resolved luminescence with >7 ps resolution
  - 4.2 – 300 K
- 150 fs, 76 MHz repetition rate
  - pump-probe time resolved spectroscopy up to 31 T
    (710nm to 900nm)
Superfluorescence

✓ Luminescence mechanisms
✓ Previous studies
✓ Characteristics
✓ Experiments
  • Method
  • Results
    - Field dependence
    - Power dependence
    - Spot size dependence
    - Directional control
    - Random directionality
Emission mechanisms in semiconductors

1) Spontaneous emission (PL) \[ \propto N \]

2) Amplified spontaneous emission (ASE) \[ \propto N \]

3) Cooperative spontaneous emission \[ \propto N^2 \text{ (Homogeneous)} \]
   \[ \propto N^{1.5} \text{ (Inhomogeneous)} \]

3a) Superradiance
   \[ \tau_N = T_2 \]
   No threshold
   Coherence (dipole moment) established externally

3b) Superfluorescence
   \[ \tau_N < T_2 \]
   Threshold behavior
   Coherence established spontaneously
Cooperative Emission

Ensemble of $N$ interacting dipoles

$$d \rightarrow p = Nd$$

Spontaneous decay time

$$\tau_N \propto \frac{T_1}{N}$$

Radiated energy

$$N\hbar\omega$$

Emission intensity

$$I_{SF} \propto \frac{N\hbar\omega}{\tau_N} \propto N^2$$

$T_1$: Spontaneous Decay time of independent 2-level atom
$\tau_c$: Spontaneous Decay time of $N$ 2-level atoms
$T_2$: Coherence time; the polarization of dipole moments are in phase
Previous studies on Cooperative Emission Processes

Superfluorescence: Cooperative emission of a burst of intense radiation upon the formation of spontaneous coherence

- **Superradiance (Dicke, 1954)**
  - Coherent preparation

- **Incoherently prepared Dicke SR**
  - Coherence establishes spontaneously
  - Coherence volume: \( V \leq \lambda^3 \)

- **Superfluorescence (Bonifacio and Lugiato, 1975)**
  - Incoherent preparation, \( V > \lambda^3 \), Fresnel \# \sim 1
  - Observed in HF by Skribanowitz, et al., *PRL* 30 (1973)
  - Observed in Cesium by Gibbs et al., *PRL* 39 (1979)
Problem: $T_2, T_2^*$ are too short in semiconductors for $\tau_N (\sim T_i/N) < T_2$

- $T_2 \sim 1$ ps; $T_2^* < 1$ ps, $T_i \sim 1$ ns

Idea: use fs laser excitation and strong magnetic fields to increase DOS to accommodate a high density e-h plasma

Relevant parameters
- Cooperative frequency:

$$\omega_c = \left[ \frac{8\pi^2 d^2 N_{e-h} \Gamma c}{\hbar n^2 \lambda L_{QW}} \right]^{1/2}$$

- Regimes for SF (SR) vs. ASE:

  - ASE: $1/(T_2 T_2^*)^{1/2} \gg \omega_c/2$
  - SF: $1/(T_2 T_2^*)^{1/2} < \omega_c/2$

- Gain (growth rate) in the SF regime:

$$g \approx \omega_c/2 \propto N_{e-h}^{0.5}$$

- Emitted Intensity:

$$I_{SF} \sim \hbar \omega N_{e-h} \frac{\omega_c}{2} \propto N_{e-h}^{1.5}$$
SF in QWs: Some Computed Parameters

- **System:** InGaAs QWs

- **Requisite carrier density:** \( N_{e-h} > 5 \times 10^{11} \text{ cm}^{-2} \) within SF FWHM

- **Cooperative frequency:** \( \omega_c \propto \sqrt{N_{e-h}} \quad :3 \times 10^{13} \text{ s}^{-1} \sim 10^{13} \text{ s}^{-1} \)

- **Coherence length:** \( L_c \approx c \cdot \tau_{SF} \cdot \ln\left[\frac{I_{SF}}{I_{SE}}\right] \sim 500 \text{ mm} \)

- **Radiation time:** \( \tau_{SF} \sim 0.5-1 \text{ ps} \)

- **Delay time:** \( \tau_d \sim \tau_{SF} \ln\left[\frac{I_{SF}}{I_{SE}}\right] \sim 5-10 \text{ ps} \)
Superfluorescence characteristics

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<tr>
<th>SF Characteristic</th>
<th>Underlying physics</th>
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<td>Shot-to-Shot Random directionality</td>
<td>Coherence established stochastically</td>
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<td>Pulsed, delayed</td>
<td>$\tau_{SF} &lt; T_2$, coherence buildup</td>
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<td>Linewidth increase with $N$</td>
<td>$\Delta\omega \sim 1/\tau_{SF}$, $\tau_{SF} \sim 1/N$</td>
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<td>Super-linear increase ($\sim N^{1.5}$)</td>
<td>Collective effect &amp; Propagation</td>
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<td>Threshold behavior with $N$</td>
<td>Density; $T_1/N &lt; T_2$</td>
</tr>
<tr>
<td>Exponential growth with area</td>
<td>$e^{gl}$</td>
</tr>
</tbody>
</table>

Diagram:
- $T_1$: Spontaneous Decay time of independent 2-level atom
- $\tau_c$: Spontaneous Decay time of $N$ 2-level atoms
- $T$: Coherence time: the polarization of dipole moments are in phase

Collective regime
Previous Studies of Magneto-Plasmas


Many-body physics:
- Band gap renormalization
- Effective mass renormalization
- Line broadening

spontaneous emission regime:
10 ns, 11.5 T

What happens at:
- Higher densities?
- Higher magnetic field?

Energy (eV)

Emission (a.u.)
Experimental Details

**Optics Set-up**

- **Excitation:** Free Space
- **Collection:** Multimode fiber
  - Transmission
  - Edge collection
    - Collection angle: 40°
- **Sample:** 15 period In$_{0.2}$Ga$_{0.8}$As/GaAs QWs

**Diagram**:
- CPA: 775 nm, 1 kHz, 150 fs
- $\Delta E \sim 150$ meV
- $B < 31$ T
- 4.2 K
- Fiber
- Barrier
- Well
- Spectrometer
- CCD
Quantum Well sample used in this work

- 15 Layers of QWs [8 nm Well/15 nm Barrier]: No inter-well coupling
- **Exciton Bohr radius** $a_B \sim 10$ nm
- Pseudomorphic strain: Large HH-LH splitting (~80 meV)
- Nominally undoped
Quantum Well Sample used in this work

B=0: 2D

\[ l_B = \frac{\hbar}{\sqrt{eB}} = \frac{25.66}{\sqrt{B[T]}} \text{ nm} \]

at 25 T

\[ l_B \sim 5 \text{ nm} < a_B \sim 10 \text{ nm} \]

B=25 T: 0D
From exciton to plasma

Y.D. Jho et al., PRB 72, 45340 (2005).

Low power CW absorption: Excitonic

High power PL: Plasmonic

• Anti-crossing at Dark-Bright Exciton Mixing

• No Dark-Bright Exciton Mixing
Field Dependence

$0.62 \text{ mJ/cm}^2, 10 \text{ K}, 0.5 \text{ mm diameter}$

- Threshold behavior
- Signal Strength: $I_{SF} \sim B^{3/2} \sim N_{e-h}^{3/2}$
- Line-width increase: shortening of SF
- S enhanced as H2 and LH LLs cross
Making Superradiant Laser by Magnetic Field

- Intense
- Threshold
- Spectrally narrow
- Directional

Conditions:
- $\lambda = 775$ nm = 1.6 eV
- 150 fs
- 10 K
- $\phi = 0.5$ mm
Power dependence

20 T, 10 K, 0.5 mm diameter

- Signal Strength: $I_{SF} \sim I^{3/2} \sim N_{e-h}^{3/2}$
- Saturation: $I_{SF} \sim N_{e-h}$
- Suggests multiple SF bursts per shot as LLs ‘reload’
Field Scaling at different pump fluence

- complex relaxation dynamics: depletion from higher to lower LL
- suggests multiple SF bursts per shot as LLs ‘reload’
Power Scaling Vs Spot Size

-Observe 3/2 scaling & line broadening only when spot-size = \( L_c \).

Coherence length \( L_c \approx c \cdot \tau_{SF} \cdot \text{Ln}[I_{SF}/I_{SE}] \sim 0.5 \text{ mm} \)
Coherent direction control

25 T
> 3x0.5 mm$^2$ spot
0.05 mJ/cm$^2$

Growth rate with area:
expansional both for ASE & SF

$L=3$ mm & $l=0.5$ mm:
$e^L / e^l \sim 20$
Single pulse-two fiber scanning: saturated SF regime

Random directionality

25 T
0.5 mm diameter
9.7 mJ/cm²
Single pulse-two fiber scanning: ASE regime

Equal directionality

25 T
> 3x3 mm² spot
0.02 mJ/cm²
**Conclusion:** Evidence for Superfluorescence

- Random directionality
- Linewidth increase with $N$
- Threshold behavior
- Exponential growth with area
- Super-linear increase ($\sim I^{1.5}$)
- Very short pulse ($\tau_{SR} \sim 0.5$ ps), delayed ($t_d \sim 5-10$ ps)

**Future Plan:**
Streak camera commissioning underway

*This work suggests that the coherence of high density electron-hole plasmas can be controlled using a combination of strong magnetic fields and ultrafast excitation*
Future Plan: Superfluorescence

Streak-Camera—resolved luminescence

Conduction band

Valence band

SF, SE, ASE
Future Plan: Superfluorescence
Future Plan: Coherent control of Superradiance

Conduction band

Valence band

OPA

Superradiance: In-phase

Superradiance: Out-of-phase
**Future Plan**: Optically Detected Resonance

**Diagram**:
- **Conduction band** and **Valence band**
- OPA (Optical Parametric Amplifier)
- CPA (Chirped Pulse Amplifier)
- CPA-pump—OPA-probe
- PL (Photoluminescence)
- Delay Line
- Sample under B
- Slow Detector
- Spectrometer Monochromator
Future Plan: Autler-Townes Effect

Conduction band

Valence band

OPA1+OPA2

White-light

OPA-pump—White-light-probe

Sample under B

Spectrometer Monochromator

Slow Detector
Future Plan: Spin & anisotropy

Jho et al., in preparation

c-axis grown Wurtzite GaN

\[ \Delta R/R \]

\[ \tau (\text{ps}) \]

No flipping: 0.45 ps

I: (σσ) 1.43 ± 0.07 ps

II: (πσ) 4.22 ± 0.08 ps

Energy (eV)

Reflectance (a.u.)

Time Delay (ps)

\[ B \text{ per. to } Z-axis \]
**Future Plan** : MIT & FES in 2DEG

-PP for absorption shift
-Streak Camera for PL quenching

InGaAs 2DEG, \( n = 1.4 \times 10^{12} \text{cm}^{-2} \)
**Future Plan**: THz & Nano

*Jho et al., in preparation*

### Diagram Description

- **Depletion region**: 60 nm
- **Metal**
- **p-GaN**: 0.2 mm
- **p-AlGaN**: 30 nm
- **InGaN**
- **GaN**
- **Director lines**
  - **-20 V**
- **No strain**
- **Tensile**
- **Compressive**

### Graphical Data

**Graph (a)**
- **THz Signal**: $\Delta I / I \times 10^{-6}$
- **Time Delay (ps)**
- **Frequency (THz)**

**Graph (b)**
- **F.T. Amplitude (a.u.)**
- **Frequency (THz)**

### Nanophononics

**Standing waves**: $\tau = 2L/v$

$L = 30\text{nm} (\text{AlGaN layer}) \Rightarrow \tau \sim 8\text{ ps}$
A rich arena of fast optics lies ahead!

Cell 3, 2003

Cell 3, 2006

SCM

Optical Parametric Amplifier

Chirped Pulse Amplifier

Streak Camera
1. Why emission from high energy side?
   - $n$ modified by presence of dense electron-hole plasma
   - Guiding in the pumped region due to polariton modes

2. Facet feedback is *automatically* suppressed
   - No guiding outside pump – rapid divergence of the guided mode
   - In addition, ground state two level system outside of pumped region
     $\Rightarrow \alpha \sim 500 \text{ -} 1000 \text{ cm}^{-1}$
Further math


• Cooperative frequency:

\[ \omega_c = \left( \frac{8 \pi^2 d^2 N_{e-h} \Gamma c}{\hbar n^2 \lambda L_{QW}} \right)^{1/2} \]

- \( d \) Dipole moment
- \( N \) 2D e-h density within SF width
- \( \Gamma \) Overlap factor of the radiation with the active QW region
- \( c \) Speed of light
- \( \hbar \) Planck constant
- \( n \) Index of refraction
- \( \lambda \) Wavelength
- \( L_{QW} \) Total width of QWs
1. Dynamic spectral shift?
   - Nonlinear & Many body effects?

2. Field dependent oscillatory behavior
   - More evidence for underlying relaxation dynamics?
Temperature ($T$) Dependence

$$B_{TH} \propto B_0 + (\Gamma_0 + \gamma T + \frac{\Gamma_{LO}}{e^{E_{LO}/k_B T} - 1})^2$$

- $\gamma$ & $\Gamma_{LO}$: both similar to InAs/GaAs QD
  , Favero et al., PRB 68, 233301 (2003)
Preliminary Time-resolved Differrential Magneto-PL

$\text{CPA @ 775 nm } \sigma^- \text{, OPA @ 1.1 } \mu \text{m } \sigma^-$

Energy (meV)

Tail-states exist only for $< 150 \text{ps}$

Temp.$= 4.2 \text{ K}$
Field$= 25 \text{ T}$