THz Superradiant Emission from 1550-nm Pulse-Pumped Quantum-Dot Arrays

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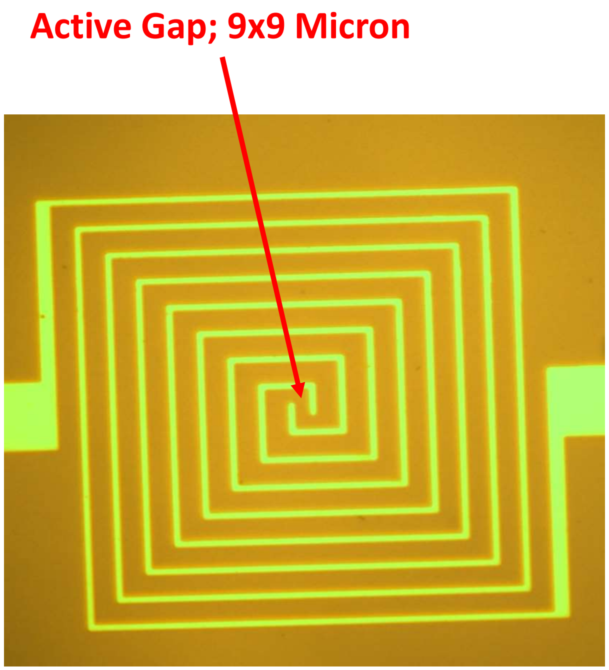
Outline

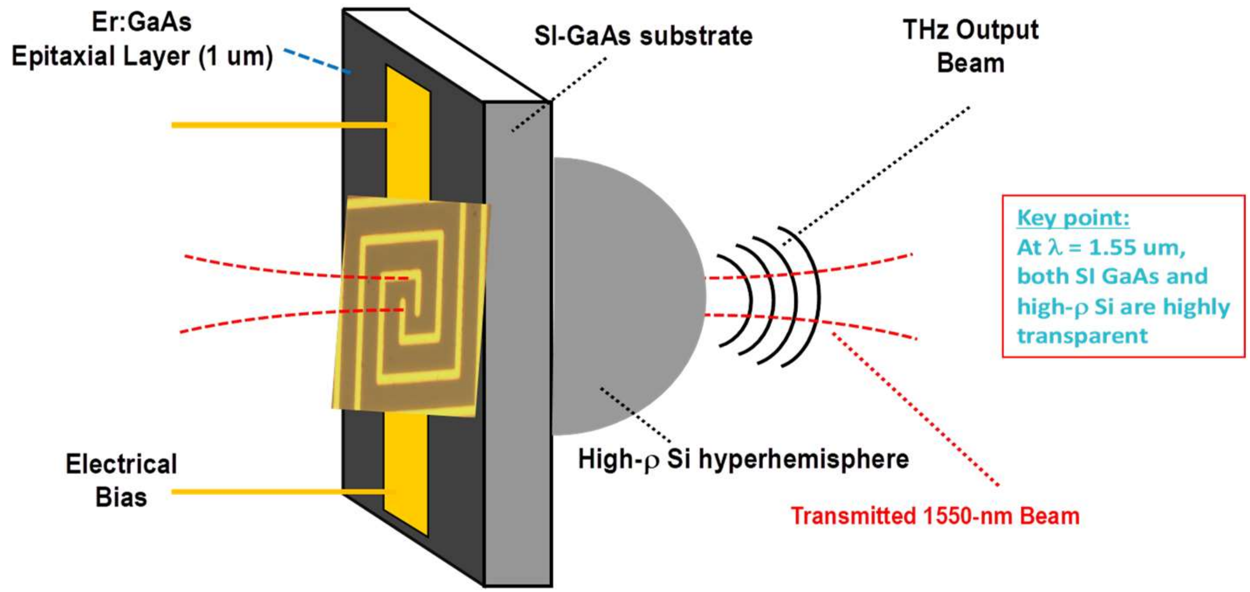
- Performance of 1550 nm-driven ultrafast photoconductive switch
- ErAs quantum dot array and 1550 nm resonant absorption
  - GaAs:Er epitaxial layer grown with MBE,
  - filled with ErAs quantum dots due to high Er concentration (>10^{20}/cm^3)
- 1550-nm fs-pulse response: evidence for superradiance
  - cooperation among quantum dots with two-energy-level systems
- Conclusion

Device Fabrication: Electrical and THz Performance

Device Structure for DC and THz Characterization
(Archimedean Square Spiral)



Experimental Set-Up

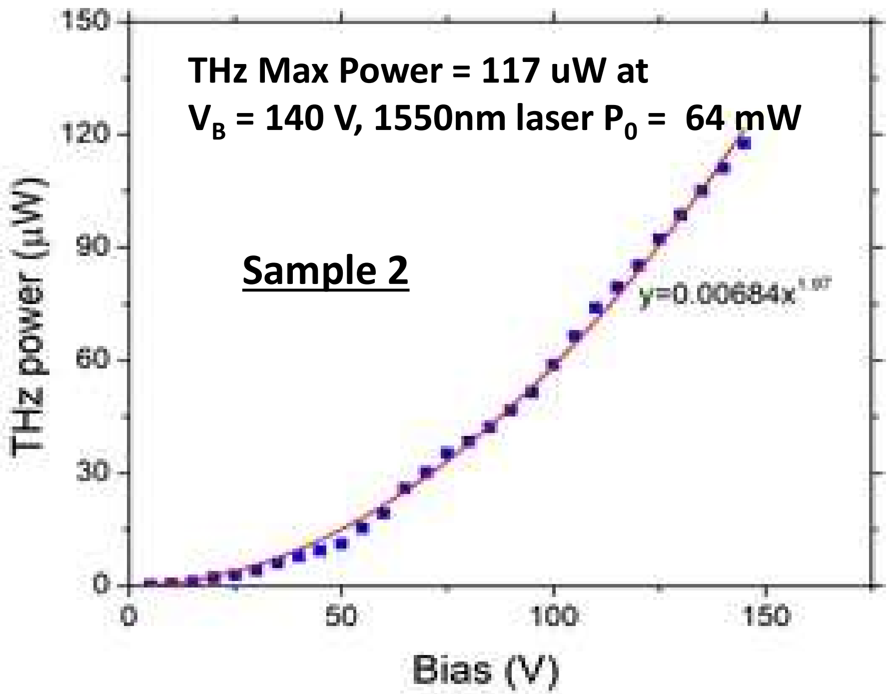


1550-nm fiber mode-locked laser:

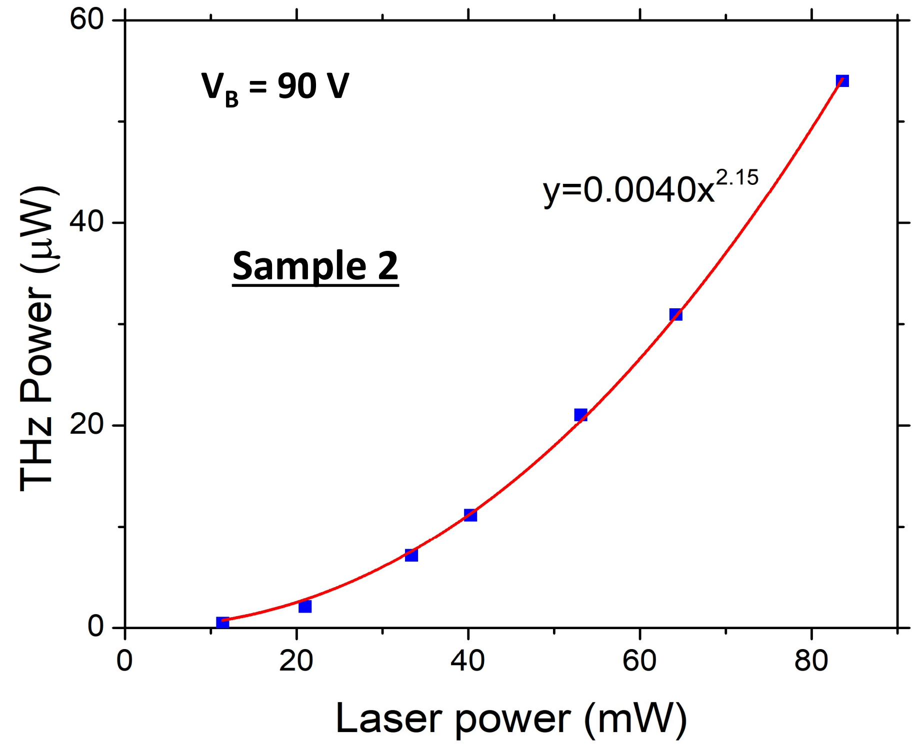
- **≈90 fs pulses width**
- **100 MHz repetition rate**

Quantum-Dot THz Pulsed Source (Sample#2)

THz Power vs Bias Voltage



THz Power vs 1550 laser Power

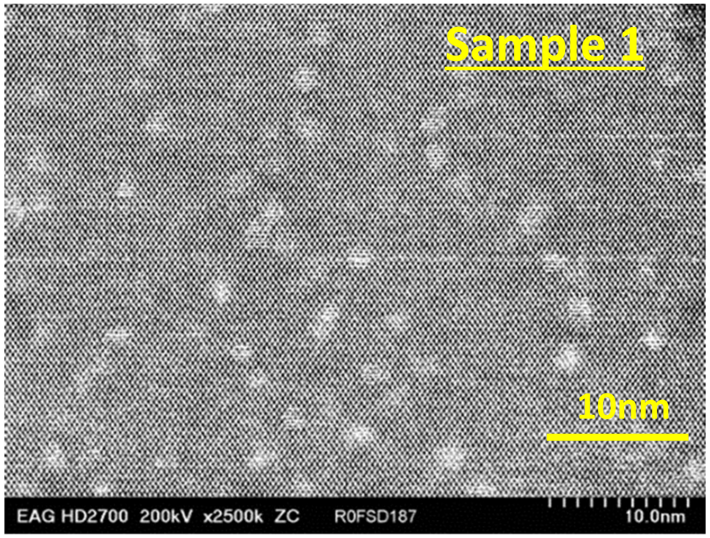


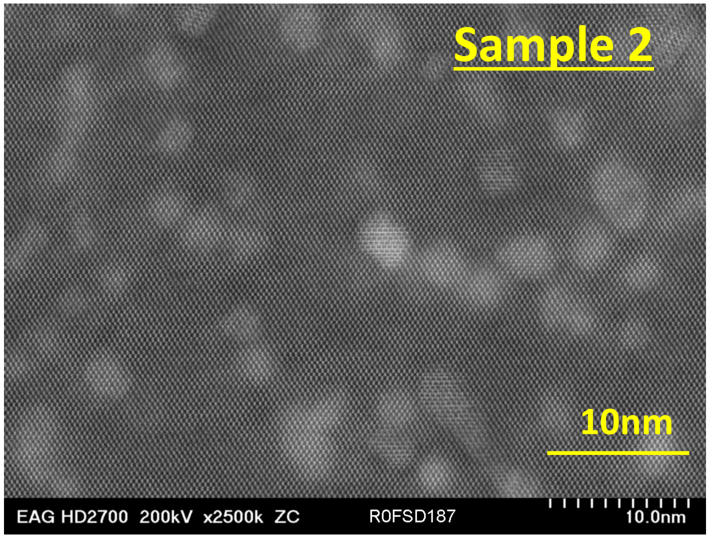
- **THz power was measured with a calibrated pyroelectric detector+IR filter**

First Growths of Heavily-Doped GaAs:Er

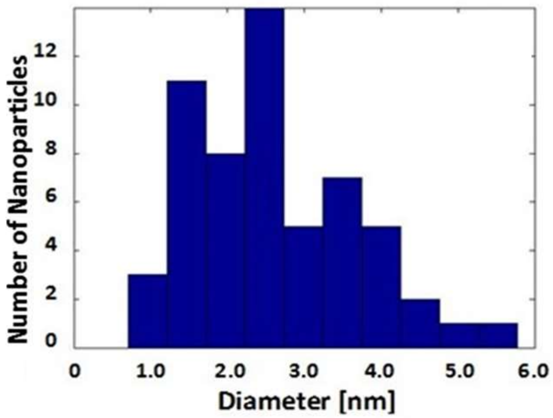
- Growth carried out by MBE at NIST (Dr. Rich Mirin) at three doping concentrations
- Sample#1: $N_{Er} \approx 4.4 \times 10^{20}$ cm^{-3}, Er/Ga = 2% by volume; Sample#2 : $N_{Er} \approx 8.0 \times 10^{20}$ cm^{-3}, Er/Ga = 4% by volume;
- Control Sample: $N_{Er} \approx 3.0 \times 10^{18}$ cm^{-3}.
- Growth temperature ≈600^{o}C; Growth rate 0.65 ML/sec

Cross-Sectional TEM Images





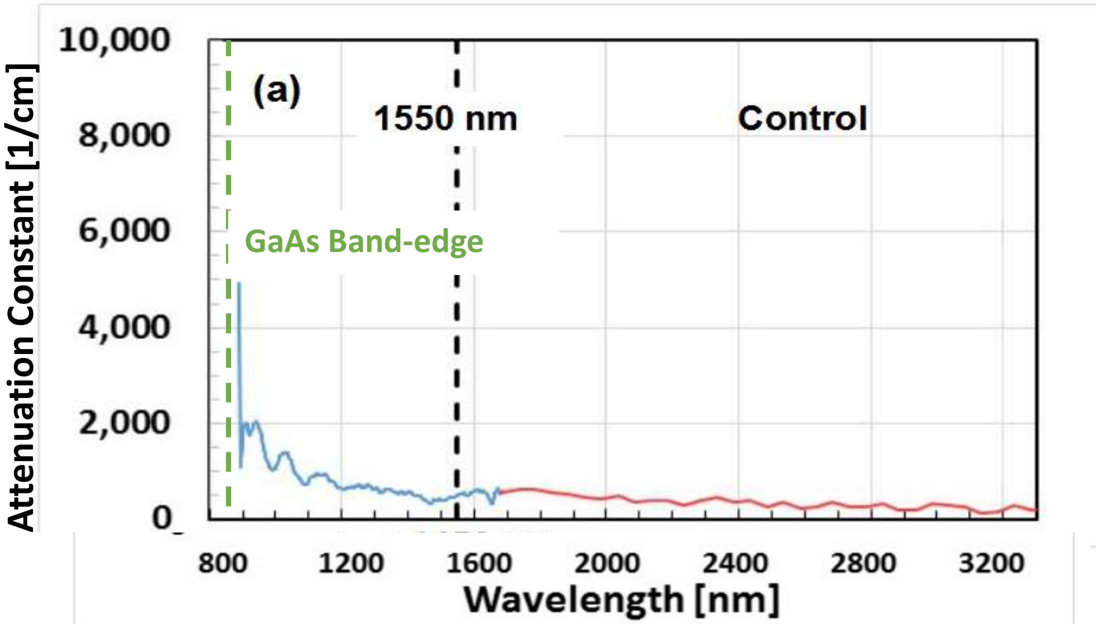
2.5 nm

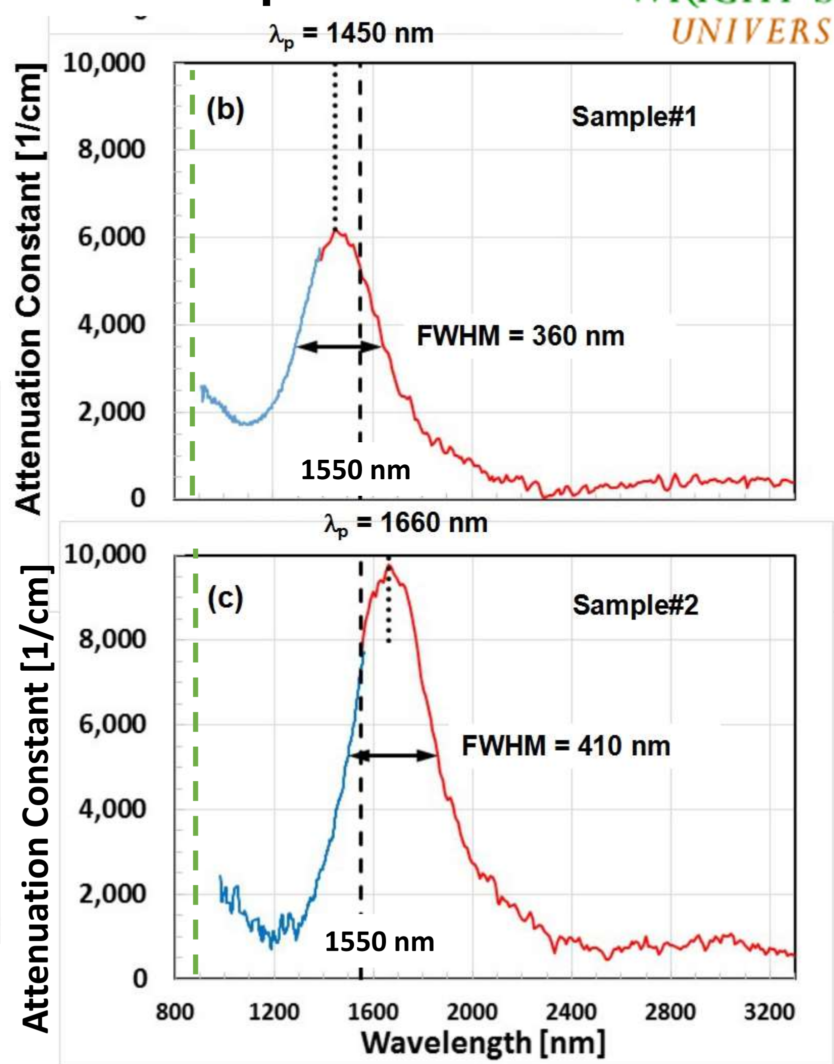


For Er Doping Density = 8.0×10^{20} cm^{-3}; Density of Quantum Dots ≈ 5×10^{18} cm^{-3}

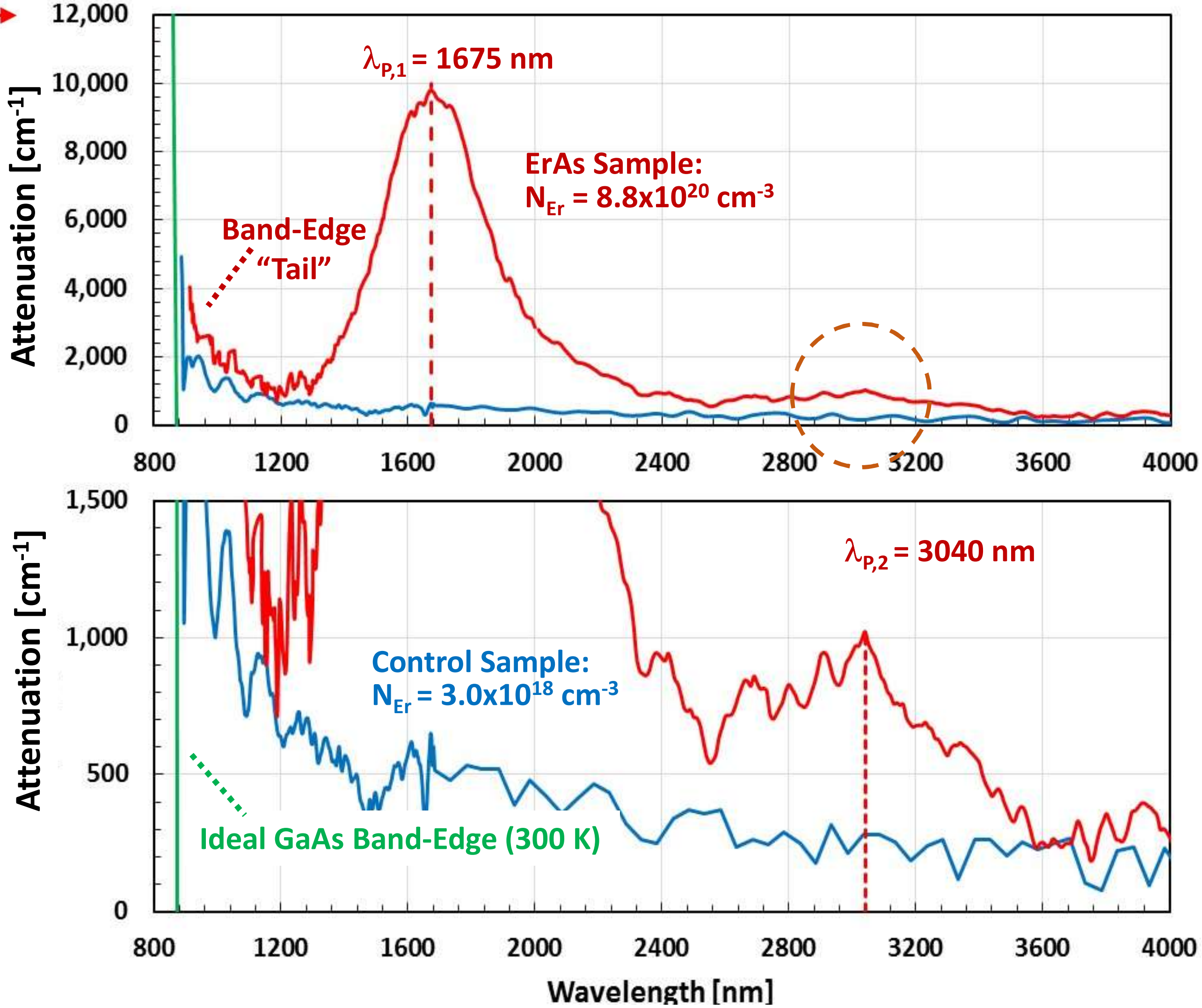
Optical Characterization of Samples

- Transmission measurements made with fiber (λ < 1650 nm) and free-space grating spectrometers (λ > 1650 nm)
- Normalized to a double-side polished SI-GaAs substrate





Further Analysis of IR Spectrum



- The first peak at ~1675 nm is almost symmetric and narrow
- There is a second peak at ~3040 nm
- Can't be explained with the surface plasmon resonance model*

*E. R. Brown, et. al. PRL. 9, 077403, (2003)

Quantum-Mechanical Modeling

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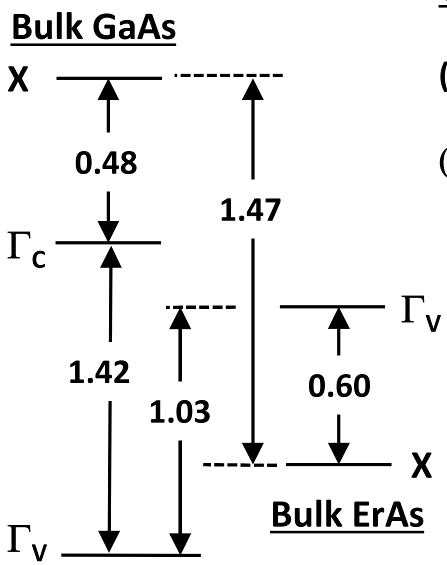
Solving Schrodinger Eqn for Bound States: Envelope Function Approximation (EFA)

- First Assumption: Quantum dots are perfect spheres of radius R, with "hard-walled" potential $V(r<R) = V_0$, where V_0 is the bandgap of GaAs (1.42 eV)
- Second Assumption: Envelope function "character" is maintained across the heterointerface



Two separate quantum-dot Schrodinger Equations to solve:

(1) X point (electron), potential depth = 1.47 eV, $m^*(ErAs) = 0.25m_e$; $m^*(GaAs) = 0.32m_e$

(2) Γ point (hole), potential depth = 1.03 eV, $m^*(ErAs) = 0.235$; $m^*(GaAs) = 0.50m_e$

Parameters taken from:

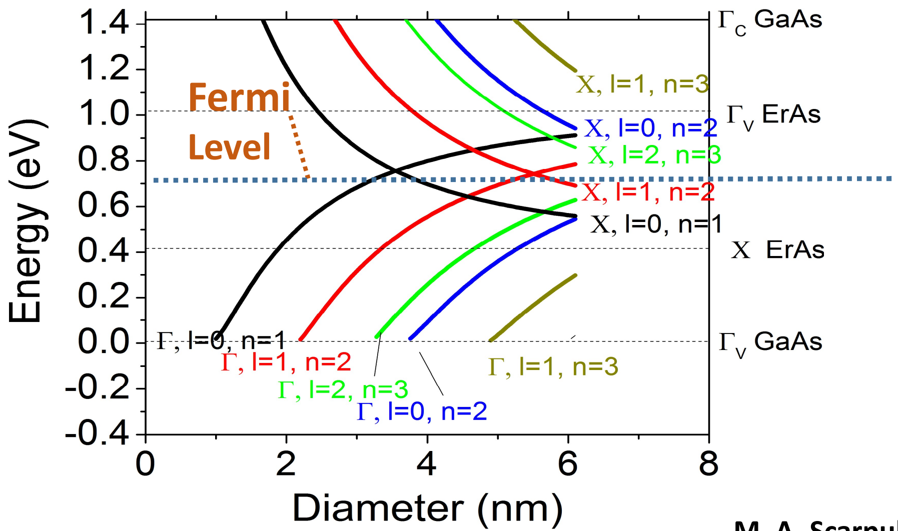
(1) http://www.ioffe.ru/SVA/NSM/Semicond/GaAs/bandstr.html#Basic
(2) M. A. Scarpulla, et. al. APL. 92, 173116 (2008).
(3) J. K. Kawasaki, R. Timm, K. T. Delaney, E. Lundgren, A. Mikkelsen, and C. J. Palmstrøm, Phys. Rev. Lett. 107, 036806 (2011)

EFA Schrodinger Eqn: $$-\frac{\hbar^2}{2m^*}\nabla^2\psi + V(r)\psi = En\psi$$

Boundary Conditions:

$$\psi_{l,n}(r<R) = \psi_{l,n}(r>R), \quad (1/m_{ErAs})d\,\psi_{l,n}(r<R)/dr|_{r=R} = (1/m_{GaAs})d\,\psi_{l,n}(r>R)/dr|_{r=R}$$

Energy-Level ("Fan") Diagram



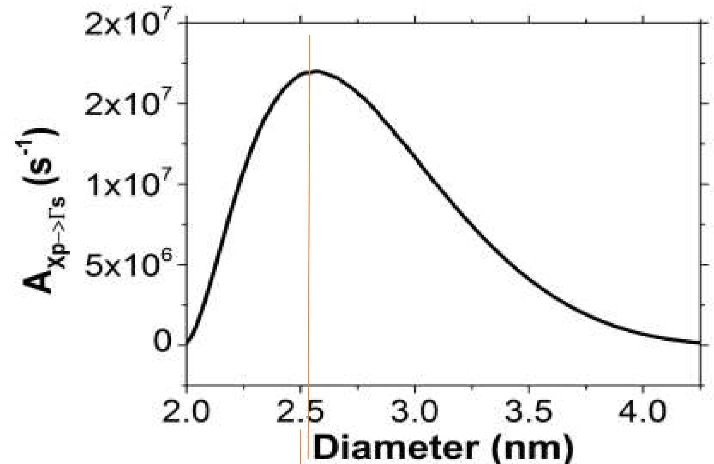
M. A. Scarpulla, et. al.
APL. 92, 173116 (2008).

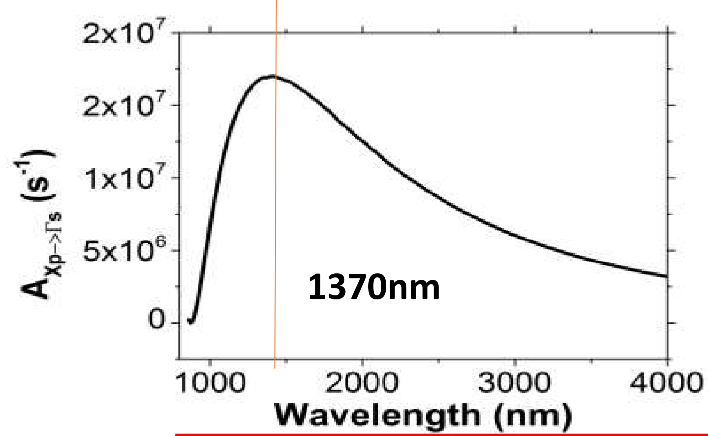
Optical Selection Rules:

(1) "Optically active" transitions are those for which $\Delta l = 1$ (conserve total momentum (electron + photon)

(2) But the ground state *must be occupied* (i.e., lie below the Fermi level)

Γs , Xp transition



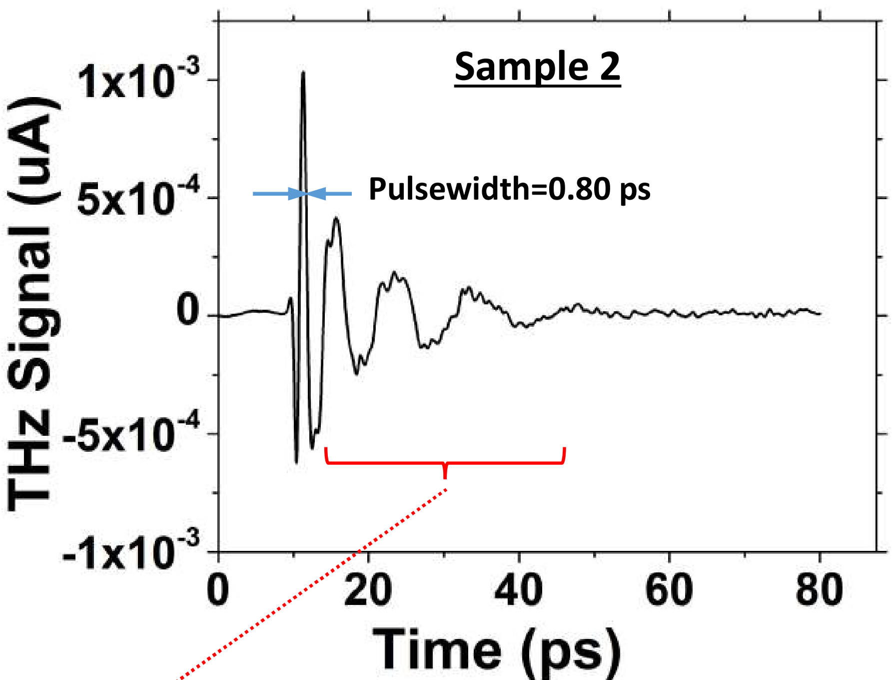


Spontaneous lifetime ~60 ns

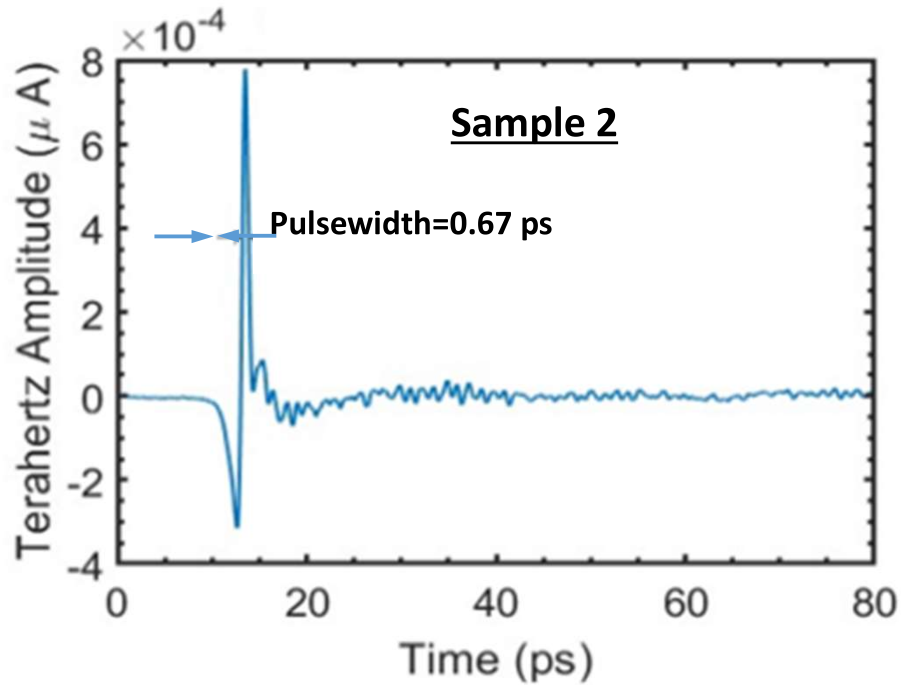
1550 vs 780 nm Pump Performance (Sample#2)

With TeTechS kit

1550-nm TDS Waveform



780-nm TDS Waveform



Peak-to-peak separation increases from about 5 to 10 ps

Traditional ErAs:GaAs Cross-Gap Ultrafast Photoconductor

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Physical Picture: superradiance of quantum dots

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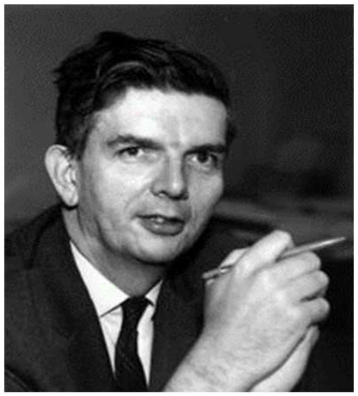


Robert Dicke: Princeton University, 1956

R. H. Dicke, “Coherence in spontaneous radiation process,” Phys. Rev. 93, pp. 99-110 (1954).

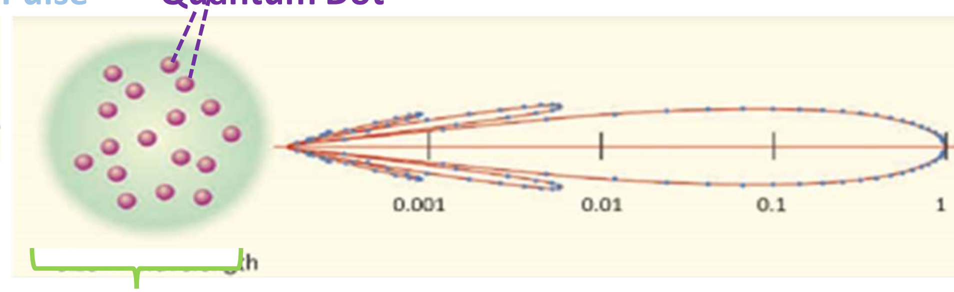
Observed in many atomic or spin systems

< 100 Femtosecond Pulse

Mode-Locked Laser

Ultrafast laser pulses Invert two energy level systems

Quantum Dot

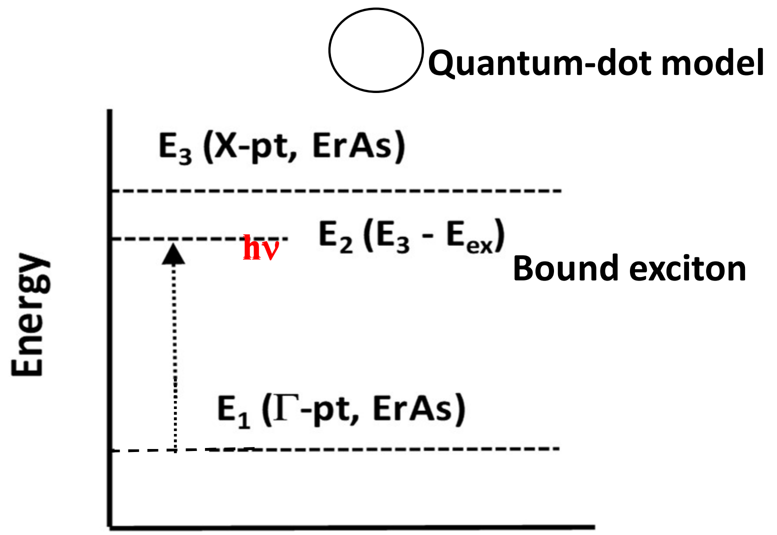


Sample Size >> Wavelength

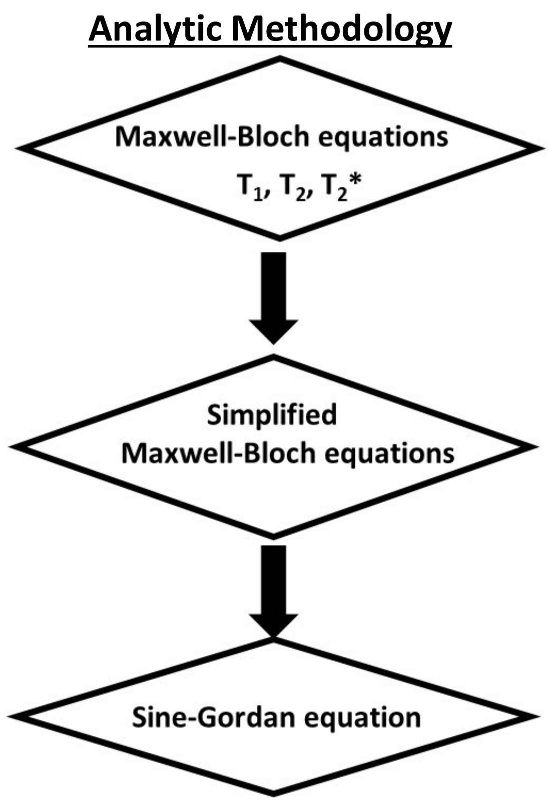
(1)“The Super of Superradiance,” M.O. Scully and A.A. Svidzinsky, Science 325 (5947), Sept 15, 2009.
(2)”Superradiance of quantum dots,” M. Scheibner, et. al. Nat. Physics 3, 106-110 (2007).

Likely Explanation: Superradiance

- A collection of atomic-like dipoles can behave *cooperatively in spontaneous emission if:*
  - *they are all excited by the same coherent excitation pulse, and*
  - *the inter-dipole separation is << the wavelength in the medium*



- Quantum dot size: a few nm <<wavelength: 1550 nm
- Multiple two energy level systems of quantum dots
- Atomic-like dipole ensemble cooperation makes the emission become superradiant

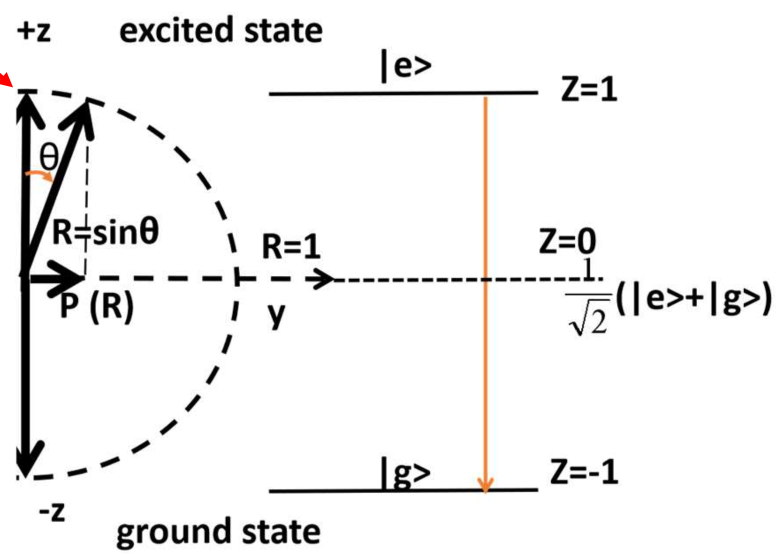


The nonlinear differential equation that also describes the motion of a pendulum w/ arbitrary initial conditions.

Coupling of Superradiant Gap to Planar THz Antenna

Pendulum model for inverted two-energy-level-Systems

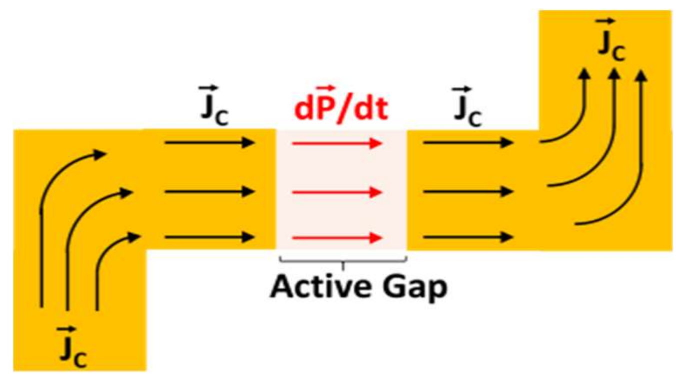
Initial inversion by ultrashort 1550 nm laser pulses



Z: population ; P: polarization; ε: optical field

P increases and decreases, even changes directions

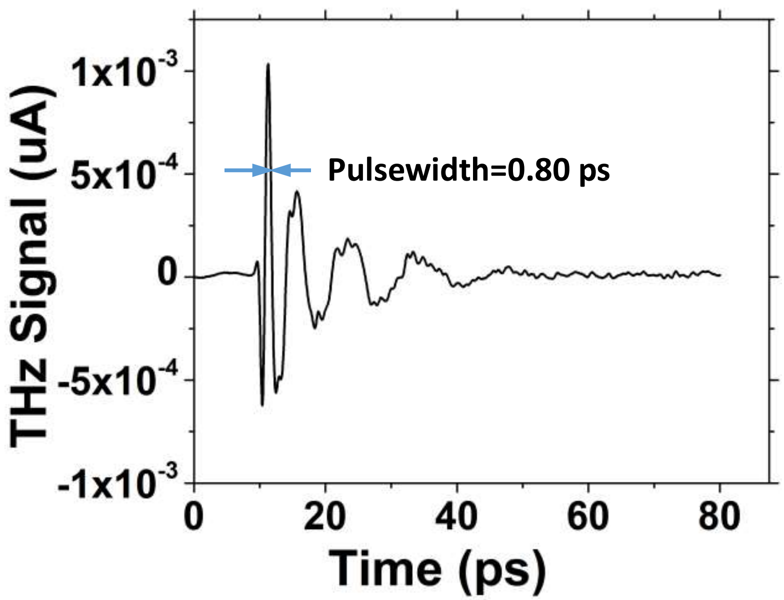
Polarization current has DC+AC components



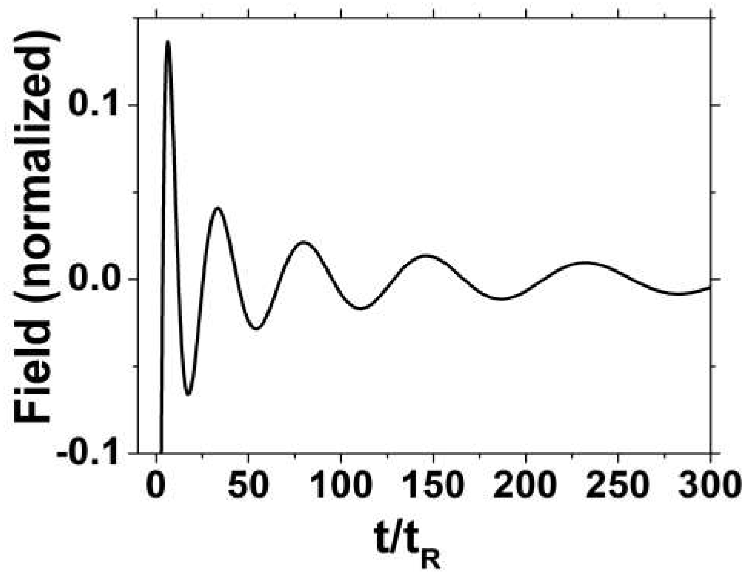
AC polarization current couples with THz antenna

Modeling Results

<u>Experimental Result (Sample #2 Device)</u>



<u>Solution for nonlinear Sine-Gordon Equation*</u>



Two important quantities required for the solution: (1) superradiance time constant T_R of ≈ 201 fs, and (2) the quantum-dot spontaneous lifetime T_{sp} ≈ 60 ns. $T_R << T_{sp}$ is an essential aspect of superradiant systems

*J. C. MacGillivray, and M. S. Feld., "Theory of superradiance in an extended, optically thick medium," Physics Review A. 14, 1169-1189 (1976).

Conclusion

- Have shown that the typical ErAs nanoparticle (diam ~2.5 nm) is much smaller than excitonic Bohr radius (~13 nm), and is likely semiconducting thanks to quantum-confinement effect
- Have carried out characterization of ErAs-quantum-dot arrays in GaAs coupled to spiral antenna
  - 1550 nm driven ultrafast photoconductive switch
  - excellent THz power generation (at least 117 uW)
- When used as transmitter in TDS system, time-domain waveform displays “ringing” when driven with 1550 nm (≈100 fs pulses) , but no such ringing when driven at ≈780 nm.
- Superradiance of quantum dots with two energy levels
- Atomic-like dipole ensemble cooperation makes the emission become superradiant

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Acknowledgement

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Backup charts

Crystal and Band Structure of GaAs and ErAs

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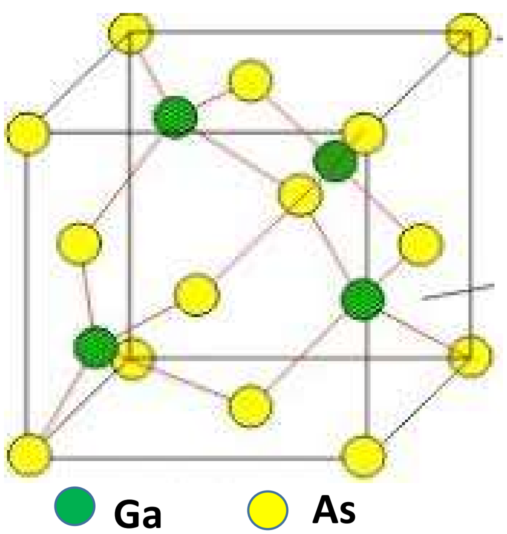
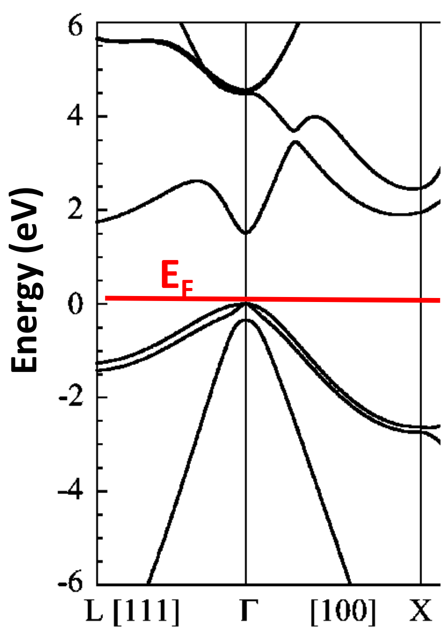
GaAs Properties

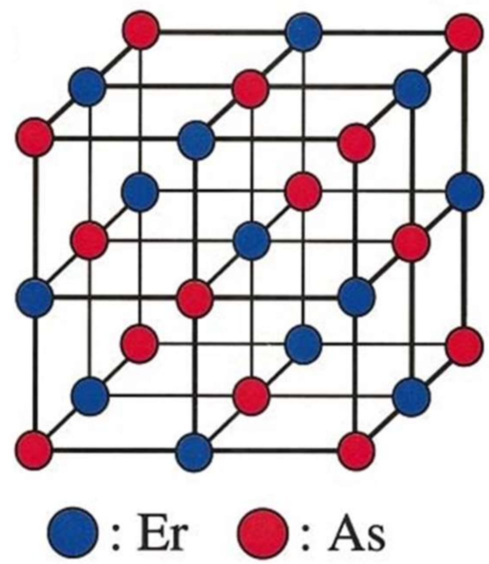
- Cubic, Zincblende Structure, a = 5.65 Ang
- Direct band gap at Γ point
- Band gap energy = 1.42 eV (semiconductor)
- Conduction band m* = 0.042 m_e

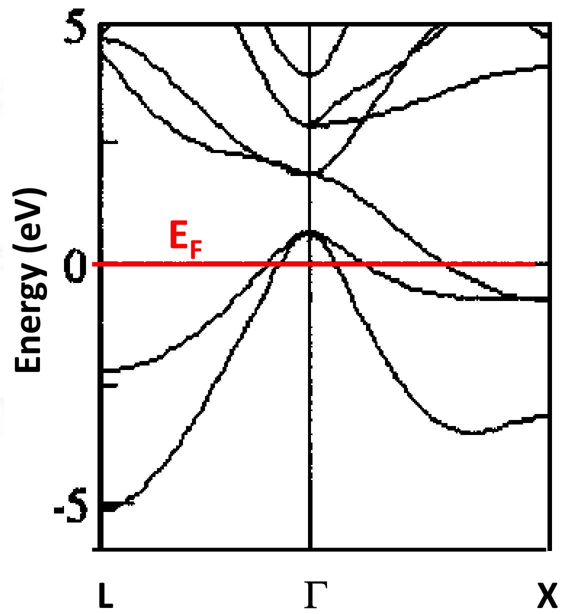




ErAs Properties

- Cubic, NaCl (rocksalt) Structure, a = 5.74 Ang
- No band gap; "conduction band" at X point falls below "valence band" at Γ by ≈0.60 eV (semimetallic)
- Valence band m* = 0.235 m_e





(the ErAs interface to GaAs maintains continuity of As Sub-Lattice, at least on the {100} facets)

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Photoconductivity spectrum

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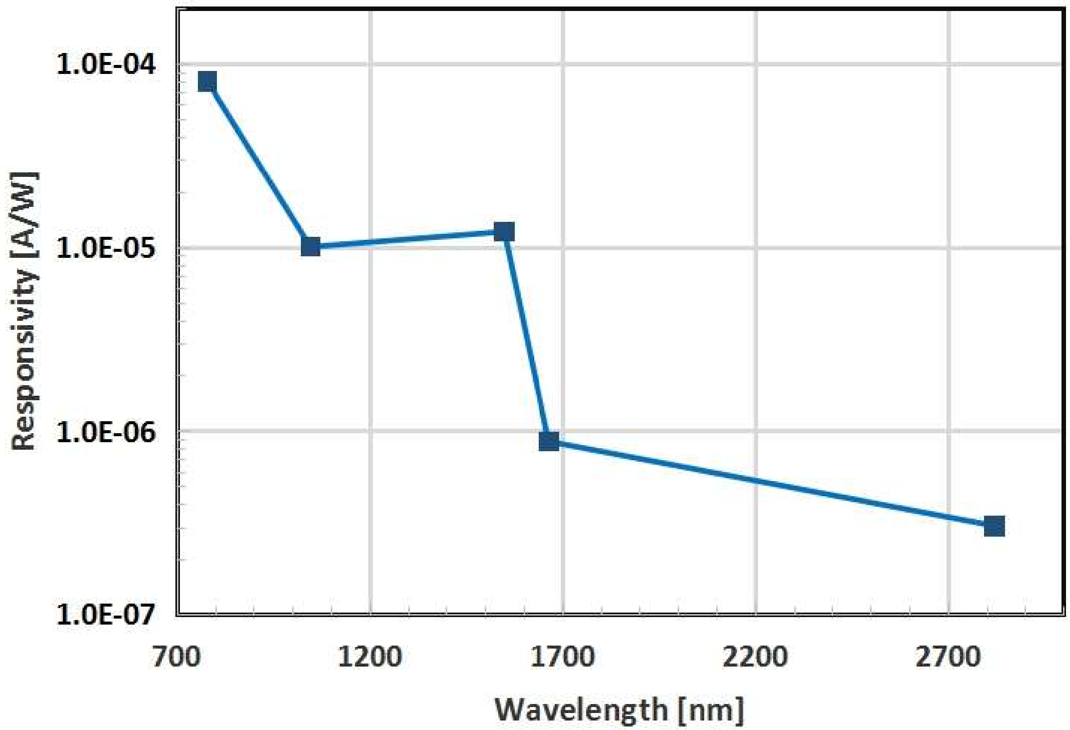
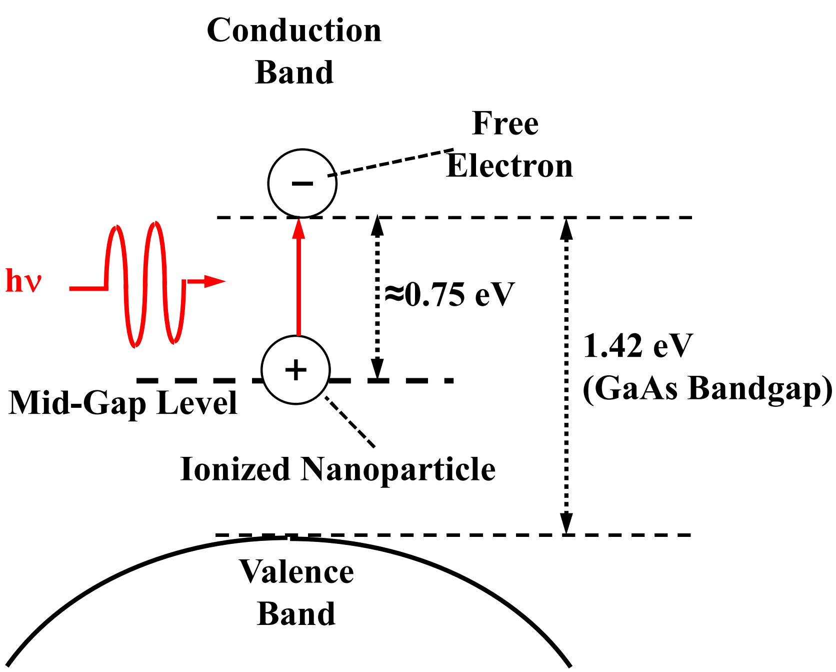
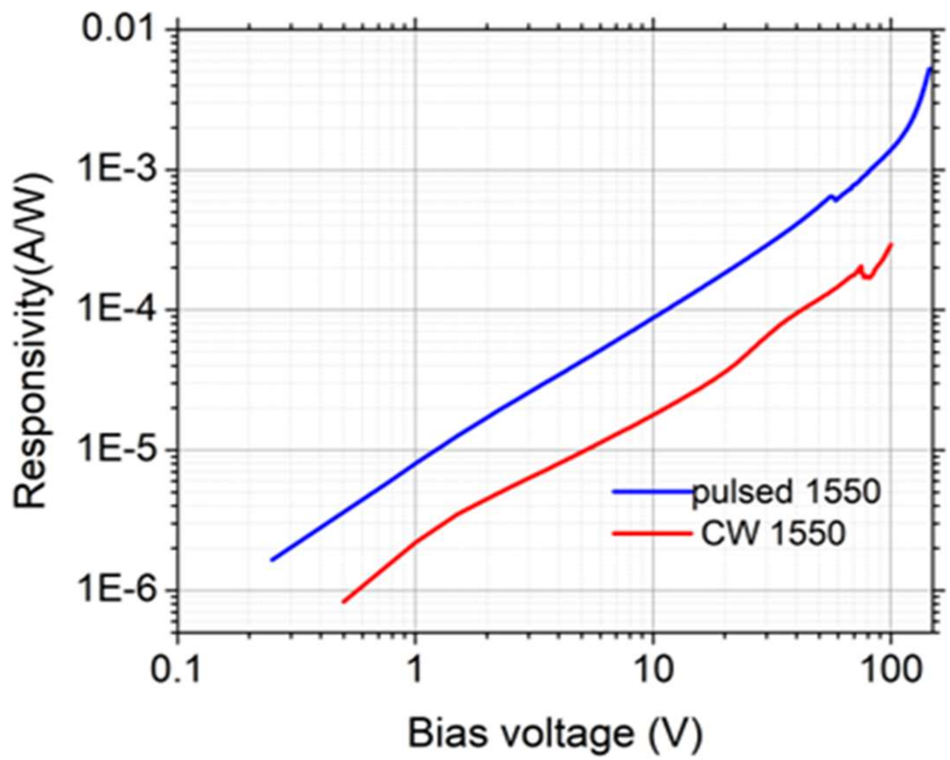


The photoconductivity follows roughly the same spectral dependence as the IR spectrum

Pulsed and CW Photoresponsivity



Extrinsic Photoconductivity model



Pulsed responsivity is 4 times higher than CW

Extrinsic photoconductivity can't explain this!

Rule-of-Thumb for Quantum Dots

- **In non-metallic quantum dots, the diameter must be less than twice the *exciton* Bohr radius in medium**

$$r^* = (m_e/\mu^*) \cdot \varepsilon_r \cdot r_0$$

In ErAs: $m_e^* = 0.25 \cdot m_e$ (X pt) $m_h^* = 0.23\ m_e$ (Γ point), so that $\mu^* = 0.12 \cdot me$,

ErAs optical dielectric constant $\varepsilon_r = 15$; $r_0 = 0.053$ nm

So that $r^* = 124 \cdot r_0 = 6.5$ nm, or an exciton diameter of 13.0 nm

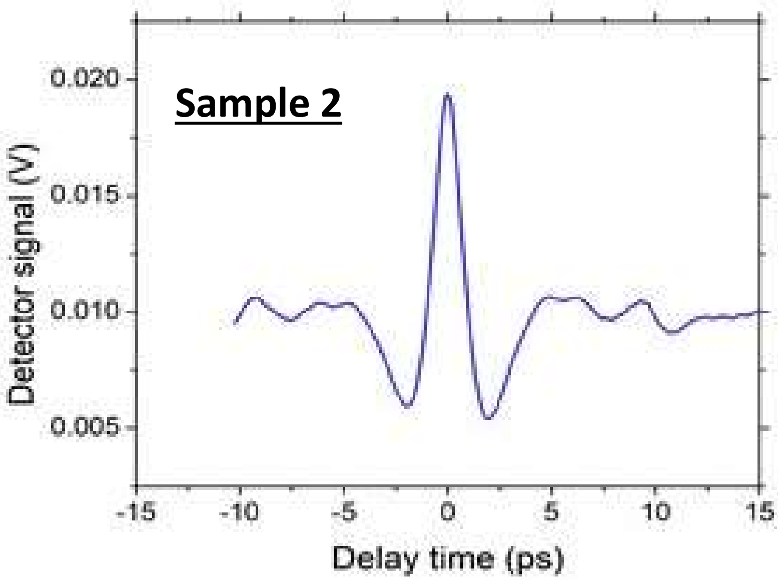
So the 2.5 (or so) nm ErAs nanoparticles are well into quantum-dot regime

- **But are the ErAs nanoparticles *semi-metallic* (as in bulk) or *semiconducting* ?**

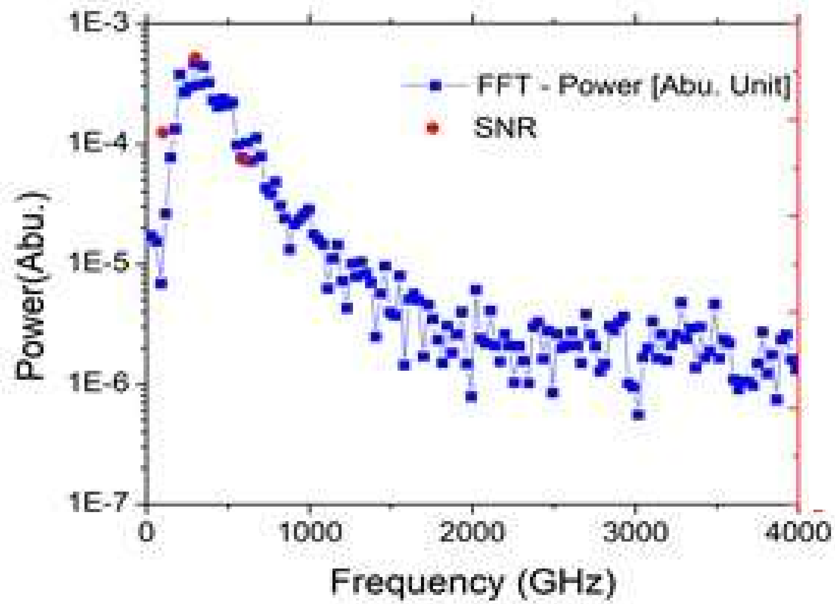
- **Interestingly, the exciton Bohr radius is comparable to the nanoparticle center-to-center separation ! (which could represent an inter-quantum-dot correlation mechanism)**

Quantum-Dot THz Pulsed Source (Sample#2)

THz Autocorrelation Function



THz Power Spectrum

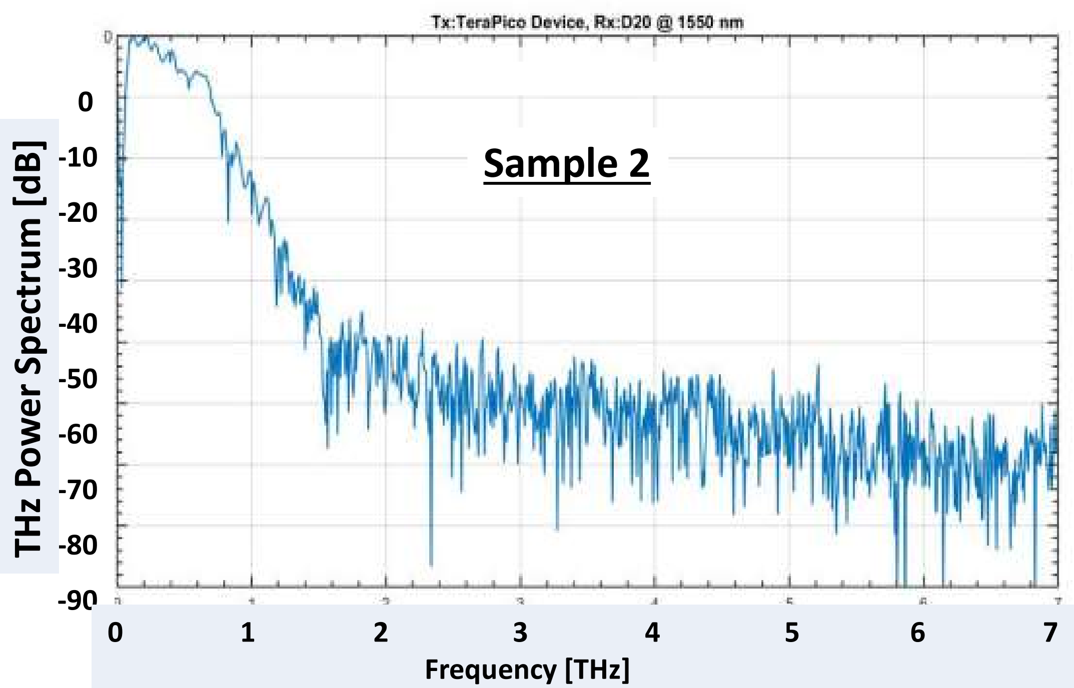


Measured with THz Michelson interferometer

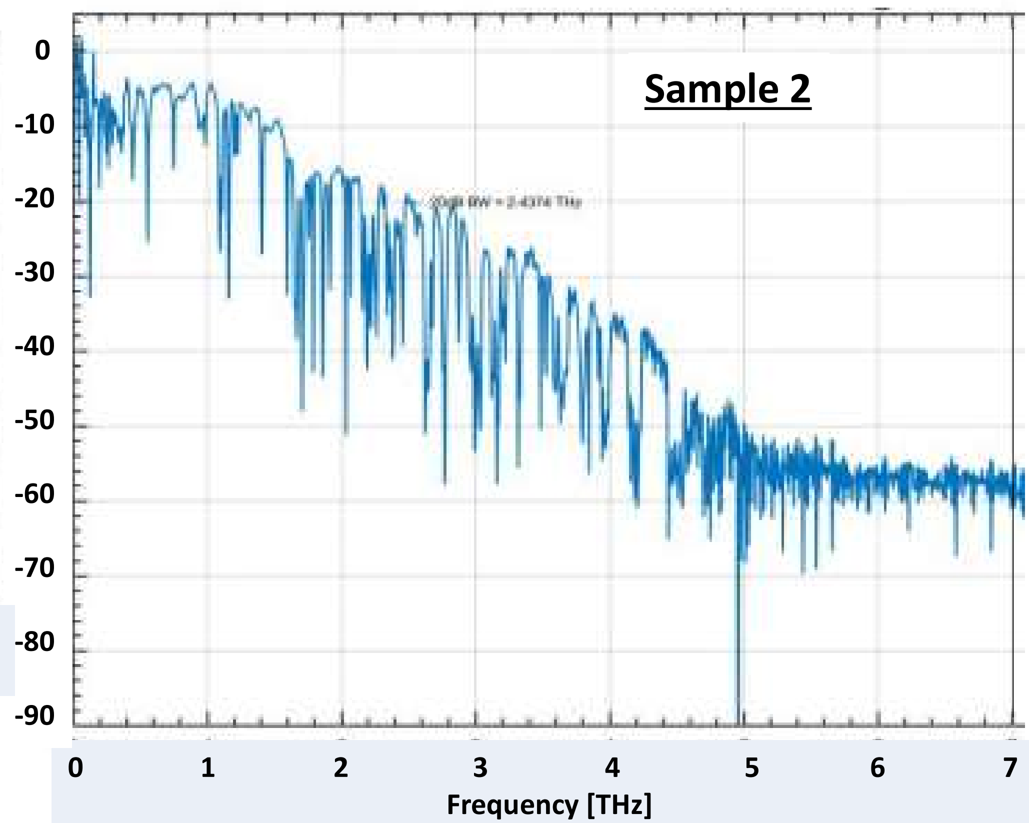
Vast majority of THz power between ~200 and 500 GHz

1550 vs 780 nm Pump Performance (cont) (Sample#2)

1550-nm TDS Power Spectrum

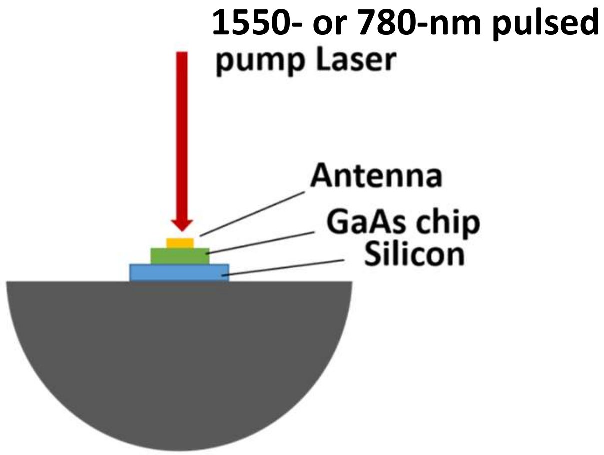


780-nm TDS Power Spectrum

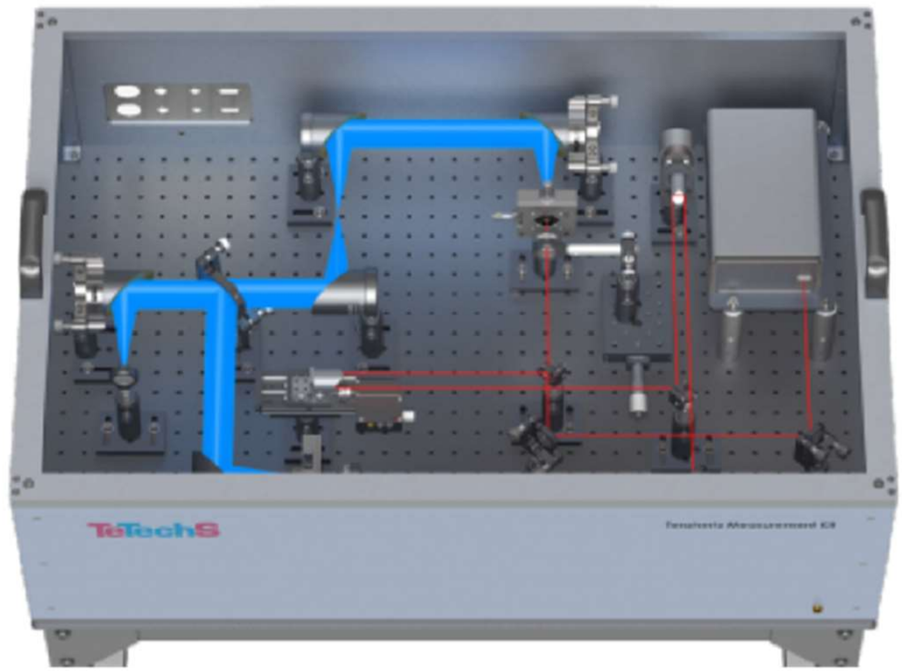


1550 vs 780 nm Pump in TDS System (Sample#2)

Device Coupled as Transmitter



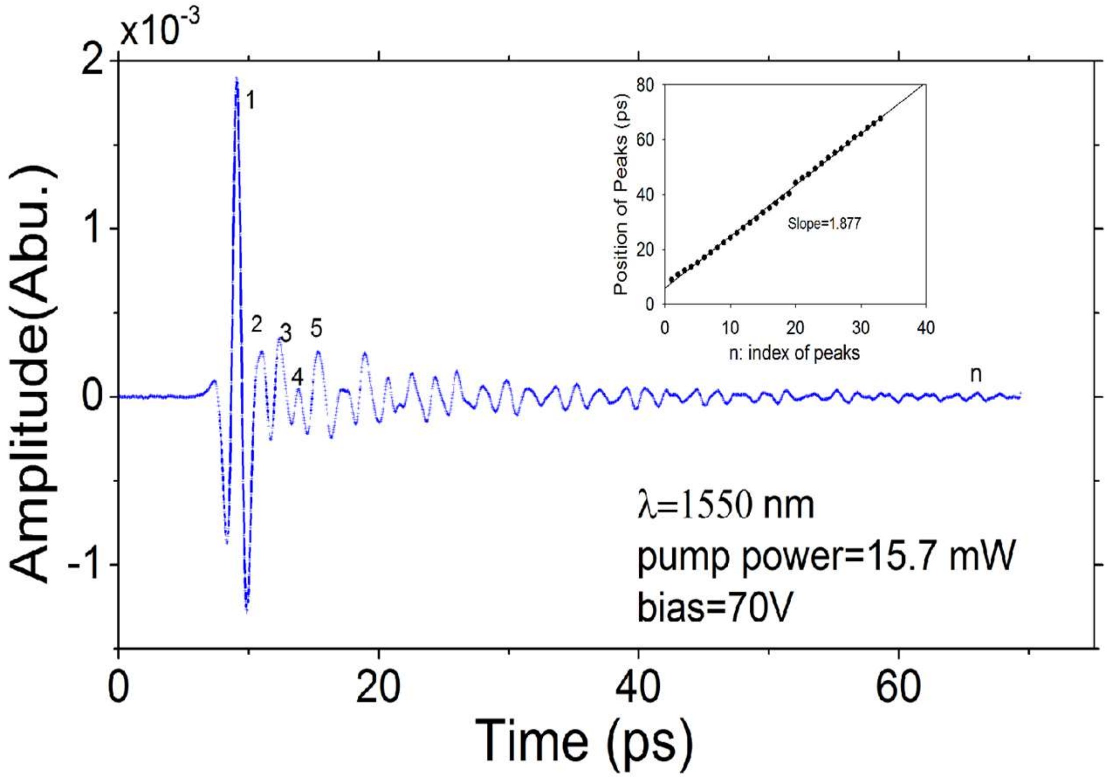
Instrument: TeTechS TDS "Kit"



Free-Space-Coupled Beam from either EDFA MLL at ≈1550 nm, or ≈780 nm (frequency doubled); Pulsewidth ~ 100 fs

1550 vs 780 nm Pump Performance (2nd Device)

Time-Domain Waveform 1550 nm Pump Pulses



- **There are ~30 peaks beyond primary one**
- **Average time separation between peaks ≈1.88 ps**

1550 vs 780 nm Pump Performance, Sample 1 (cont)

Time-Domain Waveform 780 nm Pump Pulses