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13. ABSTRACT *(Maximum 200 words)*

We have used the UCSB Free-Electron Lasers to study nonlinear and nonequilibrium intersubband dynamics in GaAs/AlGaAs semiconductor heterostructures at Terahertz frequencies. We have observed extremely large, voltage-tunable, resonant optical nonlinearities in quantum wells, which can be successfully modeled by self-consistent theory which includes the electron-electron interactions. We have performed definitive measurements of the intersubband relaxation time for quantum wells with energy level spacings below the LO phonon energy. Finally, we have observed the collective intersubband absorption in a quantum well to shift from its "dressed" energy towards its undressed energy on excitation with intense Terahertz radiation.

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1. *"Far-Infrared Second-Harmonic Generation in GaAs/AlGaAs Heterostructures: Perturbative and Nonperturbative Response,"* by W. W. Bewley, C. L. Felix, J. J. Plombon,M. S. Sherwin, M. Sundaram, P. F. Hopkins, and A. C. Gossard, Phys. Rev. B **48** 2376 (1993)
2. *"Far-Infrared Nonlinear Response of Electrons in Semiconductor Nanostructures,"* by M. S. Sherwin, N. Asmar, W. W. Bewley, K. Craig, C. Felix, B. Galdrikian, E. G. Gwinn, A. G. Markelz, A. C. Gossard, P. F. Hopkins, M. Sundaram, and B. Birnir, in **Free-Electron Laser Spectroscopy in Biology, Medicine and Materials Science**, Proceedings of the SPIE **1854**, p. 36 (1993)
3. *"Resonant Harmonic Generation and Dynamic Screening in a Double Quantum Well, "* J. N. Heyman, K. Craig, B. Galdrikian, M. S. Sherwin, K. Campman, P. F. Hopkins, S. Fafard, and A. C. Gossard, Phys. Rev. Lett. **72**, 2183 (1994)
4. *"Far-Infrared Saturation Spectroscopy of a Single Square Well,"* K. Craig, C. L. Felix, J. N. Heyman, A. G. Markelz, M. S. Sherwin, K. L. Campman, P. F. Hopkins and A. C. Gossard, Semicond. Sci. Technol. **9**, 627 (1994)
5. *"Probing Terahertz Dynamics in Semiconductor Nanostructures with the UCSB Free-electron Lasers, "* S. J. Allen, K. Craig, P. Guimaraes, J. Heyman, J. Kaminski, B.J. Keay, J.S. Scott, M. Sherwin, P.F. Hopkins, A.C. Gossard, D. Chow, M. Lui,and T-K. Liu, J. of Luminescence, Vol. 60-61
6. *"Resonant Harmonic Generation Near 100 μm in an Asymmetric Double Quantum Well,"* J. N. Heyman, K. Craig, M. S. Sherwin, K. Campman, P. F. Hopkins, S. Fafard and A. C. Gossard, in "Quantum Well Intersubband Transition Physics and Devices," ed. H. C. Liu, B. F. Levine, J. Y. Andersson NATO ASI series, Kluwer Academic Publishers, Dordrecht, p. 467
7. *"Probing Terahertz Electron Dynamics in Semiconductor Nanostructures with the UC Santa Barbara FELs,"* J. P. Kaminski, S. J. Allen, M. Sherwin, B. Keay, J. S. Scott, K. Craig, J. Heyman, P. Guimaraes, K. L. Campman, P. F. Hopkins, A. C. Gossard, D. Chow, M. Lui and T. Y. Liu, Nuclear Instruments and Methods in Physics Research **A341**, 169 (1994)
8. *"Temperature and intensity dependence of intersubband relaxation rates from photovoltage and absorption,"* by J. N. Heyman, K. Unterrainer, K. Craig, B. Galdrikian, M. S. Sherwin, K. Campman, P. F. Hopkins, and A. C. Gossard, Physical Review Letters **74** (14), 2682-5 (1995).
9. *"Nonlinear quantum dynamics in semiconductor quantum wells,"* by M. S. Sherwin, K. Craig, B. Galdrikian, J. Heyman, A. Markelz, K. Campman, S. Fafard, P. F. Hopkins, and A. Gossard, Physica D **83** (1-3), 229-42 (1995).
10. *"Optical rectification as a probe of quantum dynamics in a heterostructure,"* by K. Unterrainer, J. N. Heyman, K. Craig, B. Galdrikian, and M. S. Sherwin, Superlattices and Microstructures **17** (2), 159-62 (1995).

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"Undressing a Collective Intersubband Excitation in a Quantum Well," by K. Craig, B. Galdrikian, J. N. Heyman, A. G. Markelz, J. B. Williams, M. S. Sherwin, K. Campman, P. F. Hopkins, and A. C. Gossard, Phys. Rev. Lett., submitted (1995).

8. SCIENTIFIC PERSONNEL SUPPORTED BY THIS PROJECT AND DEGREES AWARDED DURING THIS REPORTING PERIOD:
Dr. James Heyman (Post-doctoral fellow, currently Assistant Professor of Physics at Macalaster College in St. Paul, MN)
Jon Williams, Graduate Research Assistant.

9. REPORT OF INVENTIONS : Voltage-tunable narrow-band detector for terahertz radiation. (UC Case No. 95-326-1).

10. BRIEF OUTLINE OF RESEARCH FINDINGS:

The problem we proposed to study is harmonic generation from semiconductor heterostructures at Terahertz frequencies. During the course of the proposal, we expanded the scope of our research to examining intersubband relaxation processes in quantum wells. We have observed extremely large, voltage-tunable, resonant optical nonlinearities in quantum wells, which can be successfully modeled by self-consistent theory which includes the electron-electron interactions. We have performed what I believe will be a classic measurement of the intersubband relaxation time for quantum wells with energy level spacings below the LO phonon energy. Finally, we have observed the collective intersubband absorption in a quantum well to shift from its "dressed" energy towards its undressed energy on excitation with intense Terahertz radiation. More detailed summaries are given below.

1. *"Far-Infrared Second-Harmonic Generation in GaAs/AlGaAs Heterostructures: Perturbative and Nonperturbative Response."* (Ref. 1)
We reported measurements of far-infrared (FIR) harmonic generation from $GaAs/Al_xGa_{1-x}As$ heterostructures. The samples studied were a modulation-doped $Al_{0.3}Ga_{0.7}As/GaAs$ heterojunction and a sample with ten modulation-doped half-parabolic quantum wells. The samples were driven with intense far-infrared radiation from a molecular gas laser at 29.5 cm^{-1} and the University of California-Santa Barbara free-electron laser at 51.3 cm^{-1}. The FIR radiation was polarized parallel to the growth direction. Second harmonics of the FIR were detected from both the semi-insulating GaAs substrate and from the confined electrons. For the heterojunction sample, the second-harmonic power generated by the electrons depended quadratically on fundamental power at low power, as expected from time-dependent perturbation theory. However, this dependence became subquadratic at higher powers, indicating a nonperturbative response. At high FIR powers, electrons were also ionized from the heterojunction and half-parabolic wells. For the heterojunction at f=29.5 cm^{-1} in the perturbative regime, the surface second-order susceptibility was computed to be $\chi_S^{(2)}=(1.0\pm0.75)*10^{-8} esu^{-1} cm^3$. This value agrees, within experimental error, with a simple model of the heterojunction as a triangular quantum well.. The second-order polarizability of a conduction electron in the heterojunction is nine orders of magnitude larger than that of a valence electron in pure GaAs.

2. *"Resonant Harmonic Generation and Dynamic Screening in a Double Quantum Well, "* (Ref. 3)
Second- and third-harmonic generation were observed in a semiconductor heterostructure which approximates a two-state system with an 11-meV level spacing. A resonance in the second- (third-) harmonic generation is found when the depolarization-shifted infrared absorption peak is Stark tuned through 2 (3) times the pump frequency. These resonances are thus associated with the depolarization-shifted, and not the bare, intersubband energy. Data are analyzed with a theory of second-harmonic generation including dynamic screening. Saturation is observed at pump intensities >10 kW/cm^2.

3. *"Temperature and intensity dependence of intersubband relaxation rates from photovoltage and absorption,"* (Ref. 8)
We reported intersubband-scattering times (T_1) in a semiconductor heterostructure with intersubband spacing below the LO phonon energy. T_1 was determined by simultaneous measurements of the intersubband absorption and the photovoltage induced by far-infrared radiation (FIR) near the intersubband transition frequency. At the lowest temperature (T=10 K) and FIR intensity (I=10 mW/cm^2), $T_1=1.2\pm0.4$ ns, several times longer than predicted

theoretically. T_1 decreases strongly with increasing temperature and FIR intensity, to 20 ps at T=50 K in the linear regime, and to 15 ps at T=10 K and I=2 kW/cm^2.

4. *"Undressing a Collective Intersubband Excitation in a Quantum Well,"* (submitted)
We have experimentally measured the 1-2 intersubband absorption in a single 40 nm wide modulation-doped $Al_{0.3}Ga_{0.7}As$/GaAs square quantum well as a function of frequency, intensity, and charge density. The low-intensity depolarization-shifted absorption occurs near 80 cm^{-1}(10 meV or 2.4 THz), nearly 30% higher than the intersubband spacing. At higher intensities, the absorption peak shifts to lower frequencies. Our data are in good agreement with a theory proposed by Zaluzny, which attributes the red-shift to a reduction in the depolarization shift as the excited subband becomes populated.

Technical information on all but #4 has been included in previous reports. The latter observation is significant because the observed red-shift is a new kind of nonlinear effect associated with the electron-electron interaction. The observed nonlinearity may give rise to optical bistability, period-doubling and chaos in quantum wells excited with intense Terahertz fields.