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6. AUTHOR(S)
Keith A. Nelson

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Massachusetts Institute of Technology
77 Massachusetts Avenue
Cambrige, MA 02139

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13. ABSTRACT (Maximum 200 words)

Ultrafast time-resolved spectroscopy and coherent control of terahertz phonon-polariton waves in semiconductors are explored. Phonon-polaritons are mixed lattice vibrational/electromagnetic waves that move at light-like speeds through a host crystal, within which they may be used for ultrahigh-bandwidth modulation of optical or electrical properties and as high-speed information carriers. The development of a THz polaritonics platform for such applications is a main objective.

The methods needed for coherent control over phonon-polaritons were refined and exploited for modulation of semiconductor quantum dot and quantum well properties. The work involved spatial and temporal shaping of ultrafast optical fields that were used to generate correspondingly shaped THz fields. Spatiotemporal femtosecond pulse shaping was also extended to include both phase and amplitude profiles, permitting a wide range of important new applications including fully phase-coherent multidimensional spectroscopy, with full pulse shaping capabilities in all of the input light fields, conducted in a fully automated fashion.

Spatiotemporal coherent control over THz phonon-polaritons was demonstrated. In preliminary results, THz field effects on semiconductor quantum dot electronic properties are indicated. This opens the door to new understanding of quantum dot electronic dynamics and to applications involving THz-frequency modulation of optical gain and other properties of quantum dots.

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Principal Investigator:
Keith A. Nelson
Professor of Chemistry
Massachusetts Institute of Technology
Room 6-235
Cambridge, MA 02139
Tel: 617/253-1423
Fax: 617/253-7030
Email: kanelson@mit.edu

Problem Studied and Principle Results

During this grant period, two broad classes of methods for coherent control over semiconductor quantum wells and quantum dots were developed. Both involve spatiotemporal pulse shaping of femtosecond optical waveforms, a technology which was first demonstrated during the prior grant period and refined considerably during the present grant period. In one case the shaped waveforms are used to generate THz waves for coherent control over semiconductor intersubband coherences. In the other they are used for fully coherent nonlinear spectroscopy and control of interband coherences in semiconductor quantum wells and quantum dots. Through this work our capabilities for semiconductor coherent control have been advanced dramatically.

Results on fully automated spatiotemporal femtosecond pulse shaping, in which a single input laser beam with a single femtosecond pulse is transformed into many spatially separated output beams, each with a temporally shaped output waveform, were presented. [1,2] In this work, continued from the first grant period, temporal shaping was executed through specified pixel patterns in one dimension (horizontal, along which the spectral components of the incident pulse are dispersed by a grating) of a 2D liquid crystal spatial light modulator (SLM) and spatial shaping was executed through specified pixel patterns in the perpendicular (vertical) dimension.

In this scheme, each horizontal row of pixels could be used to generate a distinct time-dependent optical waveform, with the different waveforms emerging separated in the vertical dimension. In practice, several contiguous horizontal rows of pixels are used for shaping of a single output waveform. This still permits generation dozens of distinct waveforms.

The use of spatiotemporally shaped optical waveforms for coherent control over THz phonon-polaritons was reported in *Science*. [3] This work is a key enabling development for *THz polaritonics*, a comprehensive methodology for THz wave and complex signal generation, control, guidance, and imaging/readout. Each of the separated beams reaches a different region of a ferroelectric crystal and generates a propagating THz response through impulsive stimulate Raman scattering. In one configuration, analogous to phased array generation of radar (MHz) signals, the THz wavelets emanating outward from each spot are overlapped spatially in the far field, and the superposition THz waveform is controlled through temporal shaping of the incident optical waveforms. In another configuration, the optical waveform is shaped with proportional spatial and temporal shifts such that the THz wave generated at one irradiated region of the crystal arrives at the next irradiated region just as an optical pulse arrives there, is amplified through coherent superposition with the THz response at that region, and continues this way through the array of irradiated regions, finally emerging with a high field amplitude. This remains our primary approach for generation of large-amplitude THz waves, which are important for manipulation of semiconductor intersubband electronic responses. The THz waves may be transmitted out of the ferroelectric crystal and into a semiconductor sample that is in direct contact or through air. Alternately, the ferroelectric crystal may act as a THz waveguide with the semiconductor sample at a surface or interface.

The work above permits highly refined control over THz waves, with wide-ranging applications in THz signal generation [4], spectroscopy, and coherent control. A second set of developments has permitted comparable progress in nonlinear spectroscopy and coherent control at optical wavelengths, with direct application to semiconductor interband coherences. The first refinement is based on overlapping the spatially separated (and spatially coherent) outputs of the spatiotemporal pulse shaper. In this case interferences among the outputs arise, with wavevectors given by the pixel patterns in the vertical dimension of the 2D SLM. Essentially, the vertical dimension is used for wavevector shaping, complementary to the spatial shaping described above and similar to the Fourier shaping in the horizontal dimension. [5] The second development is a more fundamental step, aimed at shaping of both phase and amplitude profiles of the output waveform rather than phase-only shaping. [6] This is achieved by forming sawtooth grating patterns with successive pixels along the vertical dimension, and using the first-order diffracted field that is thereby produced. In this case the phase of the diffracted field is determined by the spatial phase of the grating pattern that produced it, and the amplitude of the diffracted field is determined by the grating amplitude. Combined phase and amplitude control over a single output field requires many contiguous rows of pixels for formation of the specified grating patterns, so it comes at the cost of a substantial reduction in the number of independently shaped output fields. Even in this case, many (perhaps 10) such fields can be produced, so this cost is not significant in practice for most of the applications of interest. In particular, fully phase-coherent four-wave mixing spectroscopy can be conducted through generation of four distinct outputs. [7] The spatiotemporal pulse shaper is the only active element required, eliminating multiple delay lines, interferometric feedback systems, and great complexity that is ordinarily associated with measurements in which the relative phases of multiple non-collinear, non-time-coincident beams must be controlled. In addition to enormous simplification, the new approach is far more powerful since the waveform in each of the incident beams can be shaped temporally – the equivalent of a femtosecond pulse shaper in each beam. This development enables extensive control over interband electronic coherences. MIT has applied for a patent (listed below) based on the advances in pulse shaping.

In ongoing work, THz waves are generated in ferroelectric-semiconductor hybrid structures that permit coherent spectroscopy and control of the semiconductor intersubband response. Fully phase-coherent four-wave mixing measurements on quantum well exciton and biexciton transitions are also under way. These two sets of measurements will ultimately be combined, so that THz control over coupling between different optical-frequency coherences may be examined. The objectives are fundamental understanding of intersubband and interband dephasing dynamics, coupling between intersubband and interband coherences, and coupling between different interband coherences, and demonstration of practical applications in THz-frequency modulation of semiconductor optical properties including optical gain.

Publications

(a) peer-reviewed journals (numbers are references for preceding text)

1. “Multidimensional control of femtosecond pulses using a programmable liquid crystal matrix,” T. Feurer, J.C. Vaughan, R.M. Koehl, and K.A. Nelson, *Optics Letters* **27,** 652-654 (2002).

2. “Automated two-dimensional femtosecond pulse shaping,” J.C. Vaughan, T. Feurer, and K.A. Nelson, *Journal of the Optical Society of America B* **19**, 2489-2495 (2002).

3. “Spatiotemporal coherent control of lattice vibrational waves,” T. Feurer, J.C. Vaughan, and K.A. Nelson, *Science* **299**, 374-377 (2003).

4. “Typesetting of THz waveforms,” T. Feurer, J. Vaughan, T. Hornung, and K.A. Nelson, *Optics Letters* **29**, 1802-1804 (2004).

5. “Automated spatiotemporal diffraction of ultrashort laser pulses,” J.C. Vaughan, T. Feurer, and K.A. Nelson, *Optics Letters* **28,** 2408-2410 (2003).

6. “Diffraction-based femtosecond pulse shaping with a two dimensional spatial light modulator,” J.C. Vaughan, T. Hornung, T. Feurer, and K.A. Nelson, *Optics Letters* **30**, 323-325 (2005).

7. “Degenerate four-wave mixing based on two-dimensional pulse shaping,” T. Hornung, J.C. Vaughan, T. Feurer, and K.A. Nelson, *Optics Letters* **29**, 2052-2054 (2004).

(b) conference proceedings

“Phonon-polaritons: Controlled propagation and amplification,” T. Feurer, N.S. Stoyanov, J.C. Vaughan. D.W. Ward, and K.A. Nelson, *Femtochemistry,* A. Douhal and J. Santamaria, eds. (World Scientific, 2002), pp. 377-389.

“Automated two-dimensional femtosecond pulse shaping and phased-array THz generation,” J.C. Vaughan, T. Feurer, and K.A. Nelson, *Ultrafast Phenomena XIII*, R.D. Miller, M.M. Murnane, N.F. Scherer, and A.M. Weiner, eds. (Springer-Verlag, 2002), pp. 214-216.

“Direct visualization of the Guoy phase shift,” N.S. Stoyanov, T. Feurer, D.W. Ward, and K.A. Nelson, *Ultrafast Phenomena XIII*, R.D. Miller, M.M. Murnane, N.F. Scherer, and A.M. Weiner, eds. (Springer-Verlag, 2002), pp. 401-403.

“Polaritonics in complex structures: Confinement, bandgap materials, and coherent control,” D.W. Ward, E. Statz, J.D. Beers, T. Feurer, J.D. Joannopoulos, K.A. Nelson, R.M. Roth, R.M. Osgood, and K.J. Webb, *Ultrafast Phenomena XIV*, T. Kobayashi, T. Okada, T. Kobayashi, K.A. Nelson, and S. DeSilvestri, eds. (Springer, 2004), pp. 298-300.

“Degenerate four-wave mixing spectroscopy based on two dimensional pulse shaping,” T. Hornung, J.C. Vaughan, T. Feurer, and K.A. Nelson, *Ultrafast Phenomena XIV*, T. Kobayashi, T. Okada, T. Kobayashi, K.A. Nelson, and S. DeSilvestri, eds. (Springer, 2004), pp. 569-571.

"Typesetting THz waveforms," J.C. Vaughan, T. Feurer, T. Hornung, and K.A. Nelson, *Ultrafast Phenomena XIV*, T. Kobayashi, T. Okada, T. Kobayashi, K.A. Nelson, and S. DeSilvestri, eds. (Springer, 2004), pp. 717-719.

(c) conference presentations (no proceedings)

Note: this is a partial list of invited talks only

"Terahertz polaritonics" (presented by Dr. Thomas Feurer), *27th Infrared and Millimeter waves Conference*, September 24, 2002, San Diego, California.

"Coherent control over collective modes in condensed phases," *Optical Femtosecond Laser Control of Microscopic Dynamics*, September 26, 2002, Bad Honnef, Germany.

"Introduction to pulse shaping techniques," (tutorial), *Coherent Control in Atoms and Molecules Summer School*, September 30, 2002, Cargese, Corsica, France.

"Terahertz polaritonics: Ultrafast dynamics, ultrafast signals, and materials fabrication and processing," *NSF Materials Chemistry Workshop*, October 18, 2002, Newark, Delaware.

"Terahertz polaritonics: An integrated multifunctional platform," (awarded best presentation) *International Workshop on Multifunctional Materials*, October 29, 2002, Pucon, Chile.

"Terahertz polaritonics," *Quantum Electronics & Laser Science Conference*, June 4, 2003, Baltimore, Maryland.

"Terahertz polaritonics: Spatial, temporal, and spatiotemporal coherent control," *Optical Society of America Annual Meeting*, October 8, 2003, Tucson, Arizona.

"Coherent control over collective polariton excitations" (presented by Dr. Thomas Feurer), *3rd International Conference on Optimal Control of Quantum Dynamics*, Ringberg Castle, Tegernsee, Germany, December 10, 2003.

"Terahertz nonlinear spectroscopy and coherent control," *DOE-NSF-NIH Workshop on Opportunities in Terahertz Science*," February 13, 2004, Arlington, Virginia.

Imaging applications: Coherent waveform generation in optics, polaritonics, acoustics, x-rays," *ARO Workshop on Buried Nanostructure Imaging*, December 13, 2004, Arlington, Virginia.

"Terahertz polaritonics: Spatial, temporal, and spatiotemporal coherent control," *7th Symposium on Chemical Reaction Dynamics in Condensed Matter*, March 4, 2004, Laguna Beach, California.

"Fully phase coherent, phase matched multidimensional spectroscopy," *Frontiers in Optics 2004/Optical Society of America Annual Meeting and Laser Science XX*, October 13, 2004, Rochester, New York.

"The future of coherent optical spectroscopy: Multidimensional, automated, powerful, robust," *Symposium on Multidimensional Spectroscopy*, January 19, 2005, Massachusetts Institute of Technology, Cambridge, Massachusetts.

"Terahertz polaritonics," *2nd Center for Integrated Photonic Systems Annual Meeting: Challenges for 21st Century Photonics*," Massachusetts Institute of Technology, May 20, 2005, Cambridge, Massachusetts.

"MHz-THz acoustic and dielectric responses of complex materials," *5th International Discussion Meeting on Relaxations in Complex Systems*, July 11, 2005, Lille, France.

<u>Participating Scientific Personnel</u>

Dr. Thomas Feurer, Research Associate

Dr. Thomas Hornung, Postdoctoral Fellow

David Ward, Ph.D. 2005

Joshua Vaughan, Ph.D. 2005

Eric Statz, current graduate student

<u>Inventions</u>

Diffraction-based pulse shaping with a 2D spatial light modulator