

Chapter 5. Measurement of Electron-phonon Interactions Through Large-amplitude Phonon Excitation

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5.1 Introduction

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The main goal of this project is to determine the responses of electronic materials to input signals which are not only fast (i.e., pulses of short duration) but also high in repetition rate (i.e., many short pulses in rapid succession) as is directly relevant to ultrafast electronics applications. This is accomplished through direct measurement using terahertz-frequency sequences of femtosecond pulses. The responses are heavily dependent on electron-phonon interactions whose strength can be determined.

5.2 High Repetition-rate Signals and Resonant Responses of Electronic Materials

Much of the current work in ultrafast spectroscopy of electronic materials involves irradiation of the sample with a single femtosecond pulse followed by probing of the sample's time-dependent response. This has taught us a great deal about the fundamental properties and ultrafast dynamical responses of semiconductors, metals, and, more

recently, superconductors. What does this type of spectroscopy tell us about the performance of the material in a real ultrafast device?

Most ultrafast signal processing and communications applications involve not just one very fast signal followed by a long waiting period, then another isolated very fast signal, another long waiting period, etc. The main purpose of developing ultrafast devices is to make possible *high-density* signal processing, i.e., processing of very high (terahertz) repetition rate signals. For a material to be appropriate for this type of application, its response to terahertz-frequency signals must be suitable. Usually "suitable" means fast, without long-lived or spurious components.

Ultrafast spectroscopy generally measures material responses to single, isolated ultrafast optical signals. Certainly, if the response to this kind of signal is unsuitable (slow, or a fast initial response with substantial slow "trailing edge" components), then the material can be ruled out for many ultrafast device applications. However, successful performance in this type of test does not ensure successful response to high-density signal inputs of interest in real ultrafast devices. How can we use spectroscopy to provide a better "real-world" test of material performance?

It is now possible to generate complex terahertz frequency signals through femtosecond "pulse-

¹ Bell Communications Research (Bellcore).

shaping" techniques² developed at Bell Communications Research (Bellcore). Recognizing the importance of these techniques for ultrafast electronic materials characterization, we worked in collaboration with Bellcore scientists to conduct the first spectroscopic experiments in which material responses to terahertz-frequency signals were recorded.³ The experiments showed how a crystalline solid could give a very fast response to a single ultrafast input signal, with minimal "trailing edge," and yet give a clearly unacceptable response to a terahertz-frequency sequence of input signals. The sample showed a "spurious" response (due to lattice vibrations) with an amplitude that was very small after one input signal, but which was amplified by successive input signals in the sequence. After the terahertz-frequency sequence was over, the spurious signal dominated the response! This signal was amplified by an "impulsive" driving force on the lattice vibrations exerted by each of the successive input signals. The mechanism for this force, "impulsive stimulated Raman scattering," was discovered and elucidated with earlier RLE support.⁴

Our initial experiments on organic molecular crystals, supported through RLE, received wide publicity through publication (by us and by journalists) in *Science*,³ *Physics News in 1990*,⁵ *Optics News*,⁶ *Science News*,⁷ *Chemistry and Industry*,⁸ *Chemical and Engineering News*,⁹ and other "semi-popular" as well as technical

journals.¹⁰ We have now imported the Bellcore technology into our own lab, as illustrated in figure 1. The figure shows a terahertz repetition-rate sequence of femtosecond pulses generated in our lab using the pulse-shaping method developed at Bellcore.

Our current experiments are focused on semiconductors and high-Tc superconductor materials for which the results will be of immediate practical importance. We expect very substantial "spurious" responses to terahertz-frequency frequency inputs in semiconductor multiple-quantum-well structures due to lattice resonances associated with the layer thicknesses. Similarly, in "quantum dot" structures formed by semiconductor clusters, resonances associated with the cluster size will likely lead to spurious responses to high-density input signals. The size of these responses will probably be large due to strong electron-phonon interactions in semiconductors. High-Tc superconductors have recently been shown to have very strong electron-phonon interactions as well, with specific lattice phonons coupling strongly to the low-energy electronic continuum. We expect that these materials too will show important responses to high-density signals which are not apparent when an isolated, ultrafast signal is used.

In addition to providing a direct test of material performance in ultrafast device applications, the experiments permit characterization of the

² A.M. Weiner, J.P. Heritage, and E.M. Kirschner, "High-resolution Femtosecond Pulse Shaping," *J. Opt. Soc. Am. B* 5(8): 1563-1572 (1988).

³ A.M. Weiner, D.E. Leaird, G.P. Wiederrecht, and K.A. Nelson, "Femtosecond Pulse Sequences Used for Optical Manipulation of Molecular Motion," *Science* 247: 1317-1319 (1990).

⁴ For a recent review see: K.A. Nelson and E.P. Ippen, "Femtosecond Coherent Spectroscopy," *Adv. Chem. Phys.* 75: 1-35 (1989).

⁵ A.M. Weiner, D.E. Leaird, G.P. Wiederrecht, and K.A. Nelson, "Femtosecond Optical Control Over Molecular Motion," in *Physics News in 1990*, ed. P.F. Schewe (New York: American Institute of Physics, 1990), pp. 25-26.

⁶ A.M. Weiner, D.E. Leaird, G.P. Wiederrecht, and K.A. Nelson, "Femtosecond Multiple Pulse Impulsive Stimulated Raman Scattering," *Opt. News* 15(12): 29-31 ("Optics in 1989" section).

⁷ "Sculpting Light to Maneuver Molecules," *Sci. News* 137(14): 223 (1990).

⁸ "Molecule Motions," *Chem. Ind. News* 7: 202 (1990).

⁹ "Lasers Manipulate Molecular Motion," *Chem. Eng. News* 68(12): 21 (1990).

¹⁰ K.A. Nelson, "Impulsive Stimulated Raman Scattering with Single-pulse and Multiple-pulse Excitation," in *Proceedings of the 12th International Conference on Raman Spectroscopy*, ed. J.R. Durig and J.F. Sullivan (Chichester: Wiley, 1990), pp. 19-22; A.M. Weiner, D.E. Leaird, G.P. Wiederrecht, M.J. Banet, and K.A. Nelson, "Spectroscopy with Shaped Femtosecond Pulses: Styles for the 1990s," in *Picosecond and Femtosecond Spectroscopy from Laboratory to Real World*, ed. K.A. Nelson (Bellingham, Washington: SPIE, 1990), *SPIE Proc. Ser.* 1209: 185-197 (1990); A.M. Weiner, D.E. Leaird, G.P. Wiederrecht, and K.A. Nelson, "Femtosecond Multiple-pulse Impulsive Stimulated Raman Spectroscopy," *J. Opt. Soc. Am. B*, forthcoming.

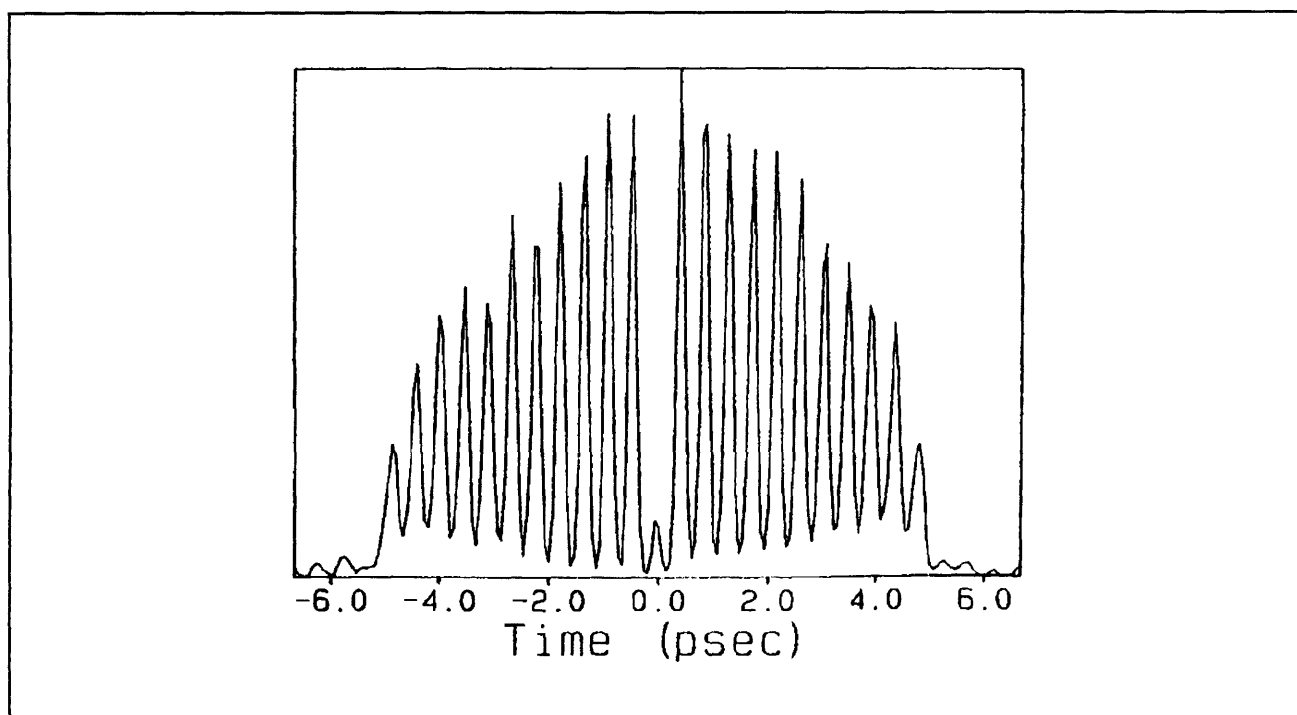


Figure 1. A 2.1-terahertz repetition rate pulse sequence produced through pulse-shaping techniques. The responses of electronic materials to high-density signals of this type are examined to provide a realistic test of their performance characteristics in ultrafast device applications.

electron-phonon interactions of the samples. These are important in mediating carrier mobility at different temperatures (i.e., different levels of phonon excitation). In high- T_c superconductors, the mechanism for strong electron-phonon coupling and its importance in superconductivity are not well understood. Our experiments may resolve these issues since they provide a unique opportunity to examine the effects of specific phonon modes on electronic energies.

As of November 1, 1990, this work is no longer sponsored through the RLE Joint Services Electronics Program. Support for our work on semiconductor and superconductor responses to terahertz frequency signals is currently being sought.

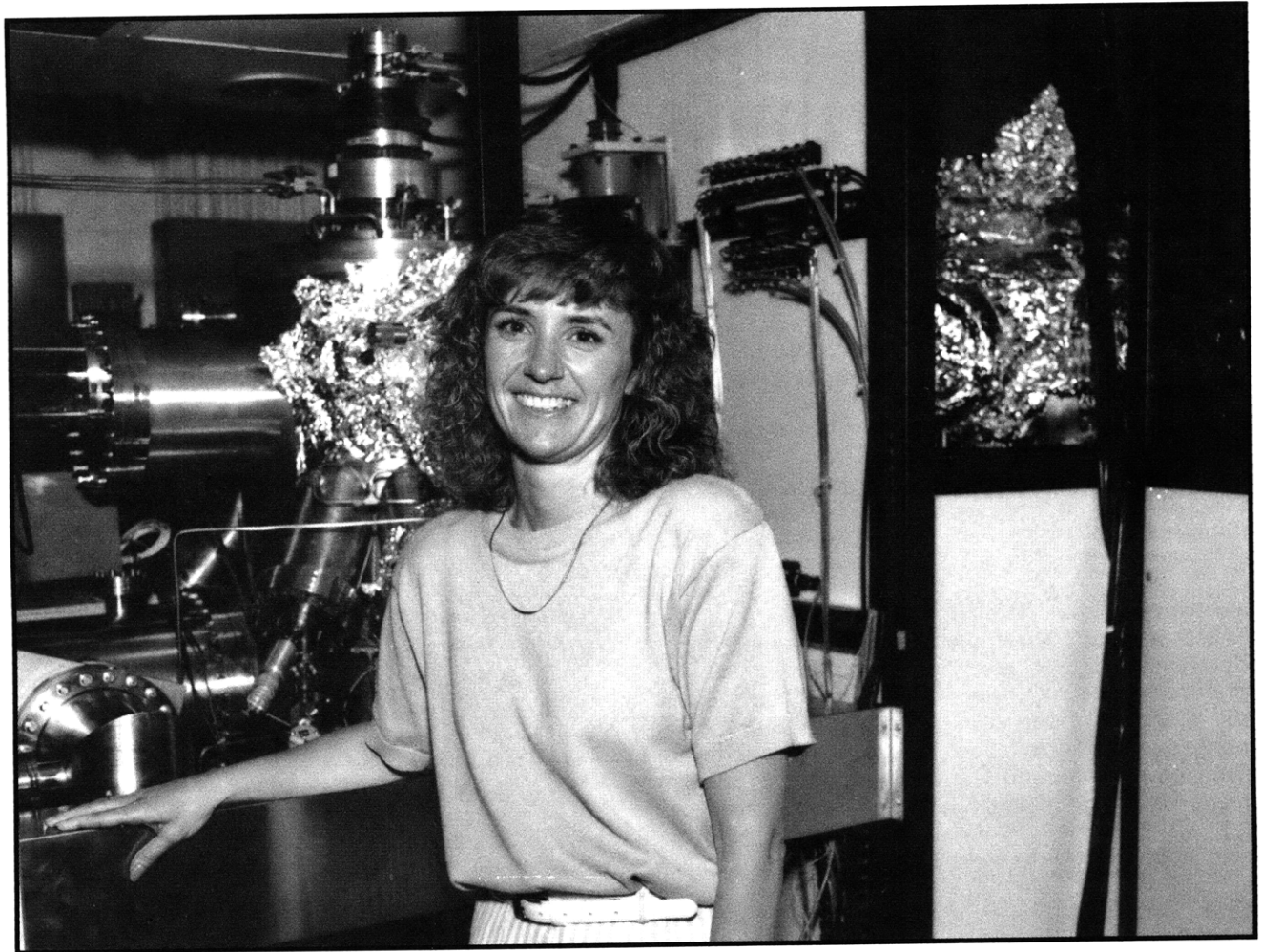
Publications

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Professor Leslie A. Kolodziejki's research is focused on the fabrication of semiconductor lasers based on II-VI compounds. (Photo by Paul McGrath)