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

Academic and Research Staff

Professor Keith A. Nelson

Visiting Scientists and Research Affiliates

Dan E. Leaird,[1] Dr. Andrew M. Weiner[1]

Graduate Student

Gary P. Wiederrecht

5.1 Introduction

The main goal of this project is to measure the strengths of electron-phonon interactions in semiconductors to understand the effect of these interactions on electron mobility at different temperatures. A secondary goal — extremely important for ultrafast electronics — is to determine the responses of electronic materials to input signals which are both fast (i.e., pulses of short duration) and high in repetition rate (i.e., many short pulses in rapid succession).

5.2 High Repetition-rate Signals and Resonant Responses of Crystalline Solids

We have successfully characterized the electronic and vibrational responses of a wide variety of solids to single femtosecond pulses. With earlier JSEP support, we demonstrated that ultrashort optical pulses passing through nearly any material initiate coherent vibrational oscillations that influence the material's transient optical and electrical responses. This finding is important for fast electronics because it indicates the response of materials to fast electrical and optical signals. The material's response to sufficiently short signal duration (i.e., optical or electrical pulse duration) can include transient vibrational oscillations, negatively affecting its usefulness in possible applications such as information processing.

At the same time, the initiation of coherent lattice vibrational oscillations with ultrashort pulses provides us with a unique spectroscopic tool. We can record femtosecond time-resolved "stop-action" spectroscopic observations of coherently vibrating crystal lattices, permitting characterization of well defined, nonequilibirium crystal structures at various stages of vibrational distortion. Characterization of the effects of lattice vibrations (phonons) on electonic energy levels is of particular importance in semiconductors since temperature-dependent carrier mobilities are influenced by electron-phonon interactions.

Figure 1 shows the typical effects of a single ultrashort pulse on a crystalline solid. A femtosecond pulse exerts an "impulse" force on lattice vibrational modes, initiating vibrational oscillations. These oscillations influence the material's optical and electrical properties. The Terahertz-frequency oscillations in the data (shown in figure 1) correspond to individual lattice vibrational oscillation cycles, observed in real-time.

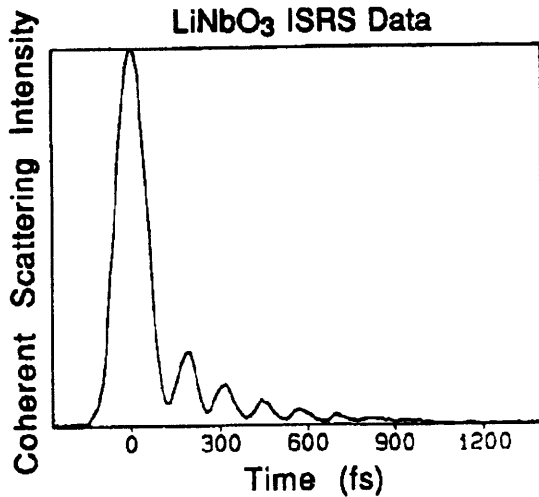[1] Bell Communications Research (Bellcore).

LiNbO₃ ISRS Data

Figure 1. Impulsive stimulated Raman scattering (ISRS) data in which optic phonon oscillations and decay in lithium niobate are monitored. In this noncentrosymmetric crystal, the optic phonon mode is both Raman and IR active, and is strongly coupled to the polariton mode associated with infrared light propagation. The phonon oscillations induced through ISRS are therefore accompanied by IR radiation at the same frequency. Femotosecond pulses have therefore been used to excite phonons and also to produce tunable Terahertz-frequency (far-IR) radiation. The phonon frequency and damping rate depend on the ISRS scattering angle.

We are monitoring the material responses to high-density signals instead of a single signal for two main reasons. First, in any real-world high-speed signal-processing application, it is essential to process signals which are not only short in duration but also frequent, i.e., signals with high information density. It is important, therefore, to understand the differences between a material's response to one isolated pulse and its response to many pulses in rapid succession, the latter being far more relevant to applications.

Second, the data in figure 1 illustrate our ability to use short light pulse to control solid-state structure and behavior in a very precise way. We can use pulses to literally move ions or molecules in crystals toward or away from each other. In addition, we can monitor the motions and their effects in real time. Unfortunately, the extent of our optical control is limited because, with a single pulse, we can only move material very small distances — less than 10^{-3} Å. These small dis-

tances have dramatic effects in signal processing, as figure 1 illustrates. However, if we could drive lattice vibrations much harder — for example, to amplitudes exceeding 0.1 Å — we could induce permanent structural changes in solids, fabricate new materials, etc. We could also characterize electron-phonon interactions over a wide range of phonon displacements.

To measure material responses to high repetition-rate sequences of femtosecond pulses, we first needed to generate the pulse sequences. We could have approached this problem by splitting a single femtosecond pulse many times with partial reflectors and aiming all the resulting beams into a sample with desired timing. This extremely cumbersome approach would have involved dozens of partial and high reflectors, lenses, and precision delay lines. Instead, we chose to collaborate with scientists at Bellcore who have
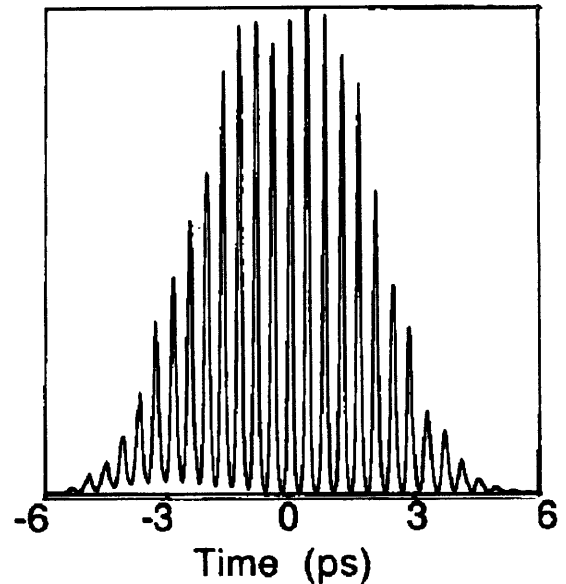
Femtosecond Pulse Sequence

Figure 2. Cross-correlation trace of a sequence of femtosecond pulses at Terahertz repetition rates produced through pulse-shaping techniques. A single femtosecond pulse was passed through two gratings, two lenses, and a spatially varying mask to produce the output shown. The output can be used to exert repetitive "impulsive" driving forces on selected vibrational modes to achieve larger amplitudes than can be driven with a single pulse.
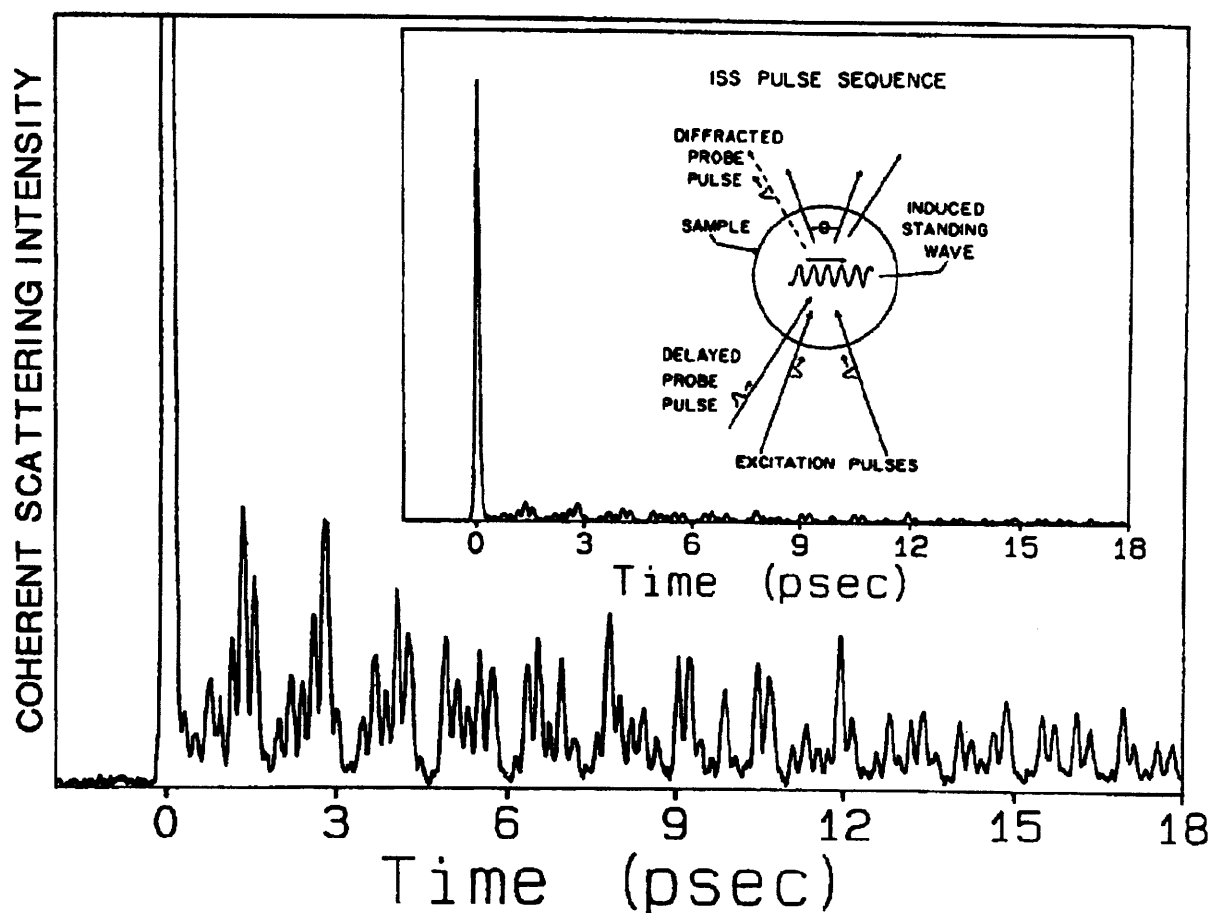
Figure 3. ISRS data from the α-perylene organic molecular crystal. Several phonon modes are excited by the femtosecond excitation pulses. This gives rise to the characteristic "beating" pattern which contains sum and difference frequencies. The "spike" at t = 0 is due to the essentially instantaneous electronic response of the crystal to the excitation pulses. The inset shows that the vibrational signal intensity is only about 4% that of the spike. In some signal processing applications (especially those involving threshhold devices), the comparatively slow response due to vibrations could be disregarded.

recently developed femtosecond optical "pulse-shaping" techniques through which a single femtosecond pulse can be converted into a sequence with the desired timing. In this technique, a single pulse, passed through a simple optical network consisting of only two lenses, two gratings, and a spatially varying mask, yields a pulse sequence similar to the one shown in figure 2. Sequences with different temporal profiles are selected by changing the mask.

Figures 3 and 4, respectively, show crystalline responses to single- and multiple-pulse excitation. For the initial demonstration, we used an organic molecular crystal since its

vibrational and electronic properties were well understood. Figure 3 shows the response to a single femtosecond pulse. The "impulse" force exerted by the pulse drives several lattice vibrational modes whose simultaneous oscillations yield the "beating" pattern in the data. The "spike" in the data at t = 0 is caused by the instantanous electronic response of the crystals to the excitation pulses. The signal due to lattice vibrations is only about 4 percent as strong as that due to the electronic response. In some signal processing applications, this "slow" signal could be disregarded, using instead the ultrafast electronic response as the basis for a rapid switch.
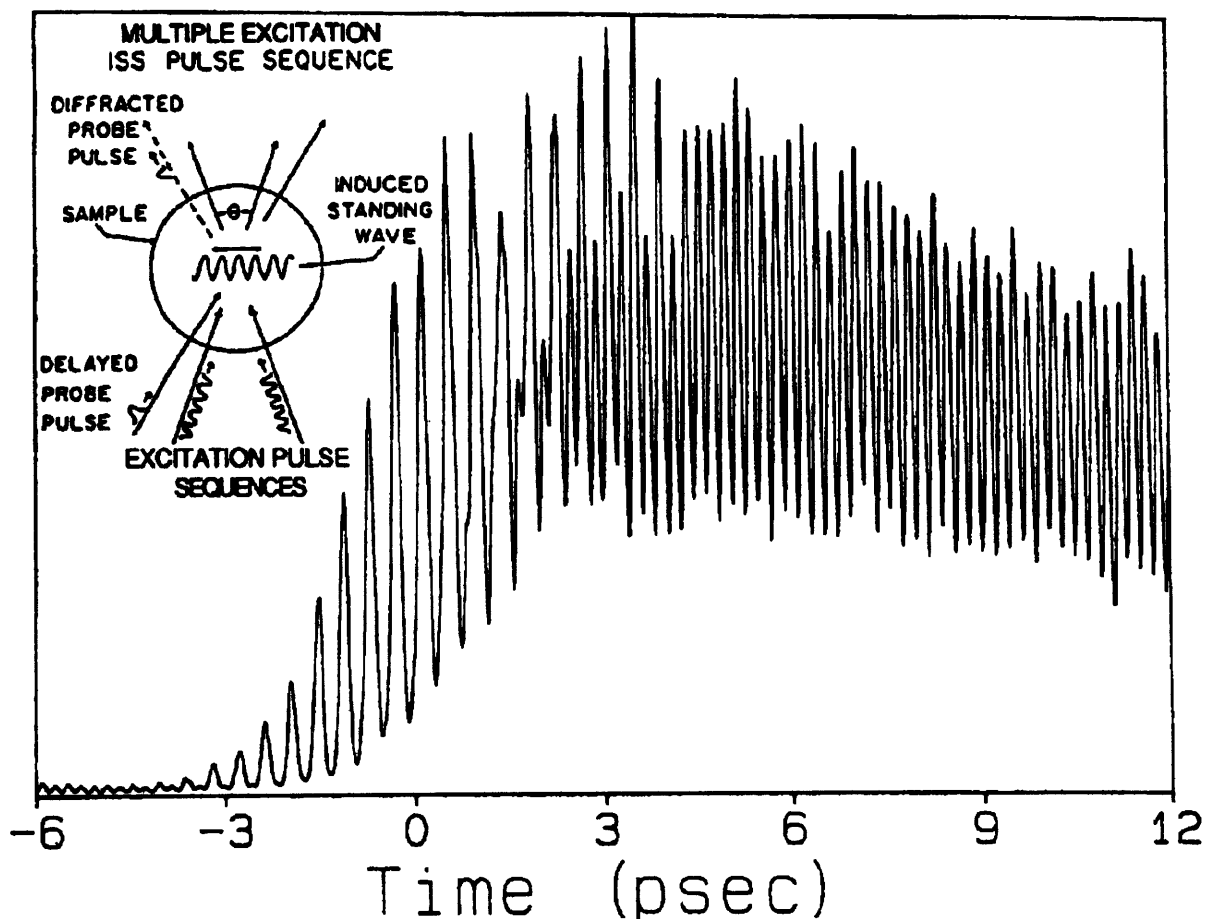
Figure 4. ISRS data from the perylene crystal driven by the pulse sequence shown in figure 2, which is tuned to drive the 80 cm^{-1} lattice vibrational mode. At first the signal is dominated by the instantaneous electronic response of the sample to each of the pulses in the sequence. After about 10 pulses, signal due to repetitively driven lattice vibrations is apparent. Signal from the vibrational mode grows stronger with each successive pulse, eventually reaching intensity levels comparable to the strongest electronic response. Selective amplification of the 80 cm^{-1} mode has been demonstrated. The strong response due to the amplified vibrational mode could not be ignored in any signal processing application.

Figure 4 shows the crystal's response to the sequence of pulses shown in figure 2. We have crafted this sequence so that the time between pulses (419 fs) is precisely equal to the oscillation period of one of the lattice vibrational modes. The impulse forces exerted by the pulses resonantly drive this mode in a manner analogous to repetitively pushing a child on a swing so that larger oscillation amplitudes are reached in time. The data show that for the first several pulses, the intense "spike" due to the electronic response dominates the signal. After about 10 pulses, however, the amplified vibrational response is apparent. By the end of the pulse sequence (t > 3ps), the signal due to

vibrations is as intense as the strongest electronic "spike." This illustrates one important difference between the response of the material to high-density input signals versus the response to a single pulse. *We could not ignore the long-lived vibrational response in any real-life signal-processing application.*

The results also illustrate the increased optical control over the sample afforded by judicious multiple-pulse excitation. In this case, through repetitive excitation, we increased the vibrational amplitude by almost a factor of 10. Amplification is also highly mode-selective, since the pulse sequence is timed to drive only one of the lattice

vibrations. This is clear from the single-frequency oscillations in the data, as compared to the beating pattern in figure 3 that results from excitation of several vibrational modes.

At present, we are beginning experiments in which these techniques are applied to semiconductors. We expect to drive large-amplitude vibrational responses and to observe their effects on electronic energy levels. This will permit characterization of the electron-phonon interactions. Another very interesting application will be to multiple quantum well structures. Repetitive excitation should lead to large-amplitude responses of acoustic-like phonons which traverse a single well in the time between pulses of the sequence.

It is worth noting that our primary outside collaborator in this project, Dr. Andrew M. Weiner, performed research for his Ph.D. under the supervision of Professor Erich P. Ippen. Under earlier JSEP support, we collaborated with Dr. Weiner and Professor Ippen to conduct some of the first femtosecond time-resolved spectroscopy experiments on molecular crystals.

Our work in this area is widely recognized within the ultrafast optics and electronics community. *Optics News* described our research in a summary of 1989 highlights, and an article in *Science* is forthcoming. At present, we are preparing more detailed reports.

Publications

Nelson, K.A. "Impulsive Stimulated Raman Scattering with Single-pulse and Multiple-pulse Excitation." *Proceedings of the 12th International Conference on Raman Spectroscopy.* Forthcoming.

Nelson K.A., and E.P. Ippen. "Femtosecond coherent spectroscopy." *Adv. Chem. Phys.* 75: 1-35 (1989).

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

Weiner, A.M., D.E. Leaird, G.P. Wiederrecht, and K.A. Nelson. "Femtosecond Pulse Sequences Used for Optical Manipulation of Molecular Motion." *Science.* Forthcoming.

Weiner, A.M., D.E. Leaird, G.P. Wiederrecht, M.J. Banet, and K.A. Nelson. "Spectroscopy with Shaped Femtosecond Pulses: Styles for the 1990s." *Proceedings of the SPIE Conference on Optoelectronics and Lasers.* Forthcoming.

The system shown is a modular chemical beam epitaxy facility for the epitaxial growth of compound semi-conductors. Research Specialist Steven C. Shepard places a sample into the ultra high vacuum introduction chamber, while Visiting Scientist Dr. Hidehito Nanto considers the layout of the growth reactor contained in the largest enclosure. The main circular chamber seen in the photo is the ultrahigh vacuum transfer chamber.