Controlling the photoluminescence of gallium arsenide with trains of ultrashort laser pulses

Zhan Hu,[1,2] Sima Singha,[1] and Robert J. Gordon[1,*]
[1]*Department of Chemistry, University of Illinois at Chicago, Chicago, Illinois 60607, USA*
[2]*Institute of Atomic and Molecular Physics, Jilin University, Changchun 130021, People's Republic of China*
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A train of three equally spaced femtosecond laser pulses is used to induce electron recombination in GaAs(100). Photoluminescence from the E_1 transition is observed when the spacing between the pulses is an integer multiple of the longitudinal-optical phonon period. This effect is also controlled by varying the relative optical phase and the polarization plane of the pulses. These observations are explained by a mechanism in which hot electrons excited in a plasma collide with the underlying lattice to produce coherently excited phonons, which scatter carriers into the L valley, where they recombine.

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I. INTRODUCTION

Controlling the behavior of matter with light has challenged chemists and physicists for decades.[1,2] A limiting factor in any coherent control experiment is the loss of phase information through interaction with the environment.[3] To control reactions at high temperatures and/or densities, it is necessary either to complete the process before decoherence sets in or to design control schemes that suppress decoherence.[4,5] The effect of decoherence is especially severe in the solid state, where dephasing occurs typically in a few picoseconds. Previous experiments with solid materials used quantum interference to control carrier populations,[6] electron flow,[7] and coherent phonon oscillation[8] in semiconductors, as well as photoemission of electrons from metal surfaces.[9] All such studies used lasers with pulse intensities well below the damage threshold of the material so that the dephasing time was longer than the pulse width of the laser. Normally, one would not expect to observe coherent effects at intensities high enough to melt the lattice. Here we show that, to the contrary, it is possible to control electronic behavior of a solid at fluences well above the melting threshold.

One strategy for controlling the dynamics of a solid is to use trains of pulses to induce large-amplitude oscillation of the nuclei and to harness this nuclear motion to alter the subsequent dynamics of the material. It is well known that an ultrashort laser pulse focused on a crystal produces coherent phonon oscillations by imposing concerted motion on a thermally excited lattice.[10] By using pairs of pulses with appropriate delays, it is possible to enhance or completely suppress the amplitude of the oscillations[11] as well as to increase the relative amplitudes of specific modes.[12] With a longer train of pulses it is possible further to enhance the phonon amplitude.[13]

The present study was motivated by the idea that coherently driven lattice vibrations may be used to control ablation of a material. Several reports have appeared on the use of shaped laser pulses to control the ablation of semiconductors[14–17] and metals[18–20] and to modify the optical properties of glasses.[21] Our own work on the ablation of Si with pairs of ultrashort laser pulses revealed qualitative changes in the surface damage and an order of magnitude increase in spectral emission with increasing delay between the pulses.[22,23] The effects in our previous studies were clearly incoherent inasmuch as the pulse spacing was as long as 100 ps. In the present study, pulse spacings comparable to the phonon period were used in the hope of inducing coherent electronic and nuclear effects.

II. METHODS

The apparatus employed is similar to one used previously to study the ablation of materials by femtosecond laser pulses.[22,23] Single pulses are generated by a Ti:sapphire laser (Spectra Physics Tsunami oscillator and Spitfire amplifier) with a peak intensity at 805 nm and a bandwidth of 24 nm (full width at half maximum), falling to 1% at 772 and 834 nm. A half-wave plate and polarizer are used to reduce the pulse energy to within the range of 0.7–3.4 μJ, with a shot-to-shot variation of less than 3%. The multiphonon intrapulse interference phase scan (MIIPS) technique[24] is used to assure that the pulses are transform limited. These pulses are shaped by a 640 pixel dual-mask spatial light modulator (CRI Inc., SLM-640-D-VN), using a standard 4-f focusing configuration to disperse the laser beam and image it onto the SLM and then to recompose and recollimate the shaped beam. A pair of cylindrical lenses (f=300 mm) and gratings (1800 grooves/mm) are used for this purpose.

After leaving the SLM optics, the pulse train is focused with a microscope objective (0.25 NA) onto a GaAs(100) crystal. The beam profile, measured with a scanning knife edge, has a spot diameter of 3.6 μm. The pulse width after the objective, determined from measurement of the electric field (see below), is 62 fs. The GaAs wafer is mounted in air on a computer-controlled xyz translation stage (Newport, 426), which rasters the sample so that a fresh surface is exposed to each laser shot. Fluorescence from the sample is collected with a convex lens (f=50.8 mm) aligned perpendicular to the laser direction, focused onto the 500 μm entrance slit of a 0.25 m monochromator (Jarrell Ash 82/462), and detected by a photomultiplier tube (PMT, Hamamatsu R928). Typically 20 shots are averaged to obtain a single data point. In some experiments the monochromator is replaced with a spectrograph (Princeton Instruments, Spectrapro 2300i) having a 500 μm entrance slit and equipped with

a nongated thermoelectrically cooled charge coupled device (CCD) camera (Pixis 400). All of the data reported at a fixed fluorescence wavelength were obtained with the monochromator/PMT combination. The laser trigger, translation stage, pulse-shape generator, and CCD camera are all controlled with a laboratory computer, which also retrieves the data from a digital oscilloscope, allowing the experiment to be fully automated.

A variety of phase functions are applied to the SLM to produce different shaped laser pulses. These include a sequence of binary phases,[25] multiple independent combs,[26] and a variable π step function [27] and a sine phase function. The last of these produces a sequence of evenly spaced pulses, generated by the phase function [28]

$$\psi(\omega) = A \sin[(\omega - \omega_{ref})T + \phi]. \quad (1)$$

The amplitude A determines the number of pulses in the train, T determines the spacing between them, and ϕ is a constant phase offset. The phase lag of an adjacent prepulse is given by $\Phi = (\omega_0 - \omega_{ref})T + \phi$, and the phase lag of an adjacent postpulse is given by $\pi - \Phi$, where ω_0 is the carrier frequency of the laser and ω_{ref} is a reference frequency.

The optical field is measured by using an interferometric field cross-correlation method[29] in which one mask of the SLM generates the pulse train to be measured, and the other mask generates a single moving pulse to scan across the pulse train. The interferometric output is measured by a photodiode. Figure 1 shows the measured amplitude and phase of the electric field for a three-pulse train, generated by the phase function

$$\psi(\omega) = 1.2566 \sin[756 \text{ fs}(\omega - 2.34 \text{ rad/fs}) + \phi] \quad (2)$$

with ϕ equal to either 0 or π.

III. RESULTS

The fluorescence spectrum produced by a p-polarized laser is shown in Fig. 2(a). The spectrum obtained with a single transform-limited pulse is dominated by two atomic transitions of neutral Ga atoms riding on a broad continuum. Various pulse shapes were tested for their effectiveness in altering this spectrum. The only one that produced a marked change is a train of three equally spaced pulses, which were generated using a sine phase function.[28] Very little difference is observed when the pulse separation, Δt, is a half-integer multiple of the period, τ, of the longitudinal-optical (LO) phonon. A striking change, however, is produced at integer multiples of τ. Under that condition an additional continuum peaked near 430 nm appears. This photoluminescence (PL) band is in the region of the $E_1(L_{1c} \rightarrow L_{3v})$ transition of GaAs,[30,31] previously observed in the reverse direction by electroreflectance and ellipsometry.[32] Figure 2(b) shows that this band diminishes in intensity as the laser polarization is rotated out of the plane of incidence and vanishes entirely for s polarization.

Figure 2(c) shows that the PL is restricted to a very narrow range of fluences, with a sharp threshold at 12 J/cm^2 and falling to zero above 25 J/cm^2, corresponding to intensities of $0.64-1.34 \times 10^{14}$ W/cm^2 for individual pulses in the train. These fluences are two orders of magnitude greater than the melting threshold[33] of 0.15 J/cm^2. In principle it is possible to determine the temperature, T_L, of the underlying lattice from the PL spectrum. Such a determination is complicated, however, by the fact that we measure a time-integrated spectrum; transient measurements of the E_0 under much milder conditions showed that the carrier temperature changes over a period of up to 10 ps.[34,35] An upper bound to the local temperature may be obtained from the red edge of the spectrum, which is determined by the band gap. Using experimental values for the temperature variation in the E_1 band gap,[36] we obtain temperatures clustered around 550 K [Fig. 2(d)]. The apparent decline of T_L with fluence is at first surprising and is likely the result of competition between ejection of energetic particles from the surface and propagation of thermal and mechanical energies into the crystal during the recombination time.

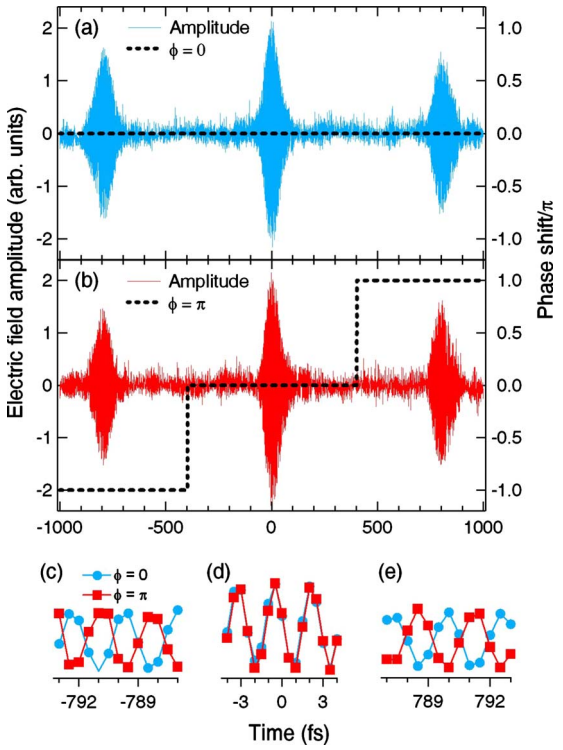

FIG. 1. (Color online) Amplitude and phase of a three-pulse train. The amplitude and phase of the entire pulse train is shown in panels (a) and (b) for ϕ equal to 0 or π, respectively. The dashed lines show the phase offset of the first and third subpulses from the middle one. Panels (c)–(e) show an expanded view of a few cycles of the electric field in each of the three pulses for both phase offsets.

Next we investigated the effect of pulse separation and fluence on the intensity of the emission at a fixed wavelength of 450.8 nm. Figure 3(a) displays fluorescence maxima at integer multiples of the phonon period out to $\Delta t = 11\tau$. In some runs we observed fewer peaks but with as much as 50% modulation. In assigning the peaks in this figure, we find that a phonon period of 124.6 fs gives the best fit to the data. The shift from the room temperature value[8] of 114 fs is indicative of the elevated temperature of the lattice.[37] Figure 3(b) plots the ratio of the fluorescence for a triple pulse to that of a single pulse of the same total energy as a function of

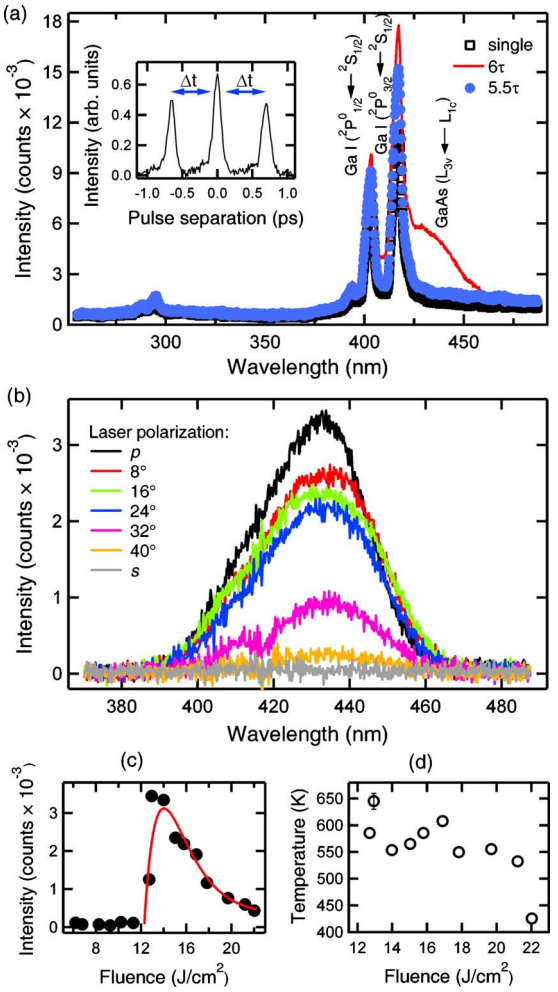

FIG. 2. (Color online) Fluorescence from an irradiated GaAs crystal. (a) Laser-induced breakdown spectrum of GaAs produced by a single pulse, a triple pulse with $\Delta t = 5.5\tau$, and a triple pulse with $\Delta t = 6\tau$, at a fluence of 13.5 J/cm^2. The inset shows the cross correlation of the pulse train. (b) Difference spectrum obtained by subtracting the 5.5τ spectrum from the 6τ spectrum, revealing the E_1 band. Angles refer to the direction of the laser polarization vector with respect to the incident plane. (c) Peak height of the difference spectrum as a function of laser fluence with p-polarization. The curve is drawn to guide the eye. (d) Temperature of the local lattice estimated from the red edge of the spectra used to construct panel (c). The error bar is the standard deviation calculated from the spectra in panel (b).

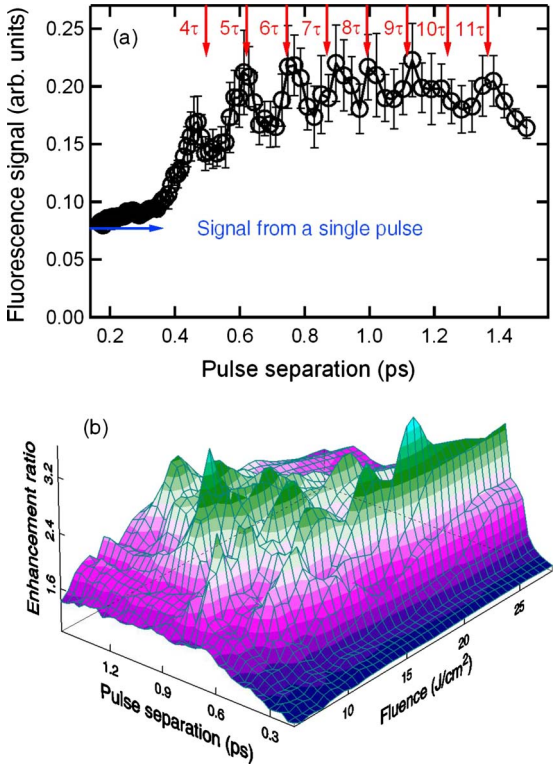

FIG. 3. (Color online) Emission at 450.8 nm as a function of pulse separation and laser fluence. (a) Emission intensity vs pulse separation for a fixed fluence of 12.8 J/cm^2. The arrows indicate multiples of 124.6 fs. (b) Ratio of the fluorescence intensity produced by a triple pulse to that produced by a single pulse of the same total intensity.

pulse separation and fluence, again illustrating the narrow fluence range compatible with coherent phonon excitation.

A number of additional measurements were made to elucidate the PL mechanism. Figure 4(a) shows that the phonon structure vanishes for s polarization [as expected from Fig. 2(b)] and is also absent for discrete line emission regardless of the laser polarization. Figure 4(b) shows that the phonon structure is present only for oblique angles of incidence and vanishes entirely for angles $\leq 25°$. In another experiment, we shifted the optical phase of the first pulse by an amount $-\phi$ and that of the last pulse by $+\phi$ with respect to the middle pulse for a fixed Δt. Figure 4(c) shows that the 6τ phonon oscillation increases by 45% for a phase shift of 1 rad while the trough at 5.5τ diminishes slightly. At 1.8 rad the phonon oscillation is suppressed.

IV. DISCUSSION

It is well known that the interaction of ultrashort (subpicosecond) pulses with semiconductors produces structural changes on a time scale much shorter than the thermalization time. Time-resolved x-ray diffraction studies of InSb irradiated with 50 fs pulses probed the transition from solid to liquid.[38,39] Inertial displacement of the lattice atoms was observed for at least 250 fs, with the intensity of the diffracted light dropping to 20% within 500 fs, revealing dissolution of the lattice structure down to a depth of at least 50 nm. Similarly, optical microscopy measurements revealed that melting of GaAs is completed in several hundred femtoseconds.[33] Experiments[23] and simulations[40] show that the melt front produced by such pulses propagates inward at subsonic speeds for tens of picoseconds. It is likely that the phonon oscillations observed here occur in the intact lattice beneath this moving melt front.

A mechanism that is consistent with all of our observations is phonon scattering of electrons and holes into the L valley of the Brillouin zone followed by radiative recombination. (See Fig. 5.) During the early part of the first pulse, electrons are promoted from the valence to the conduction

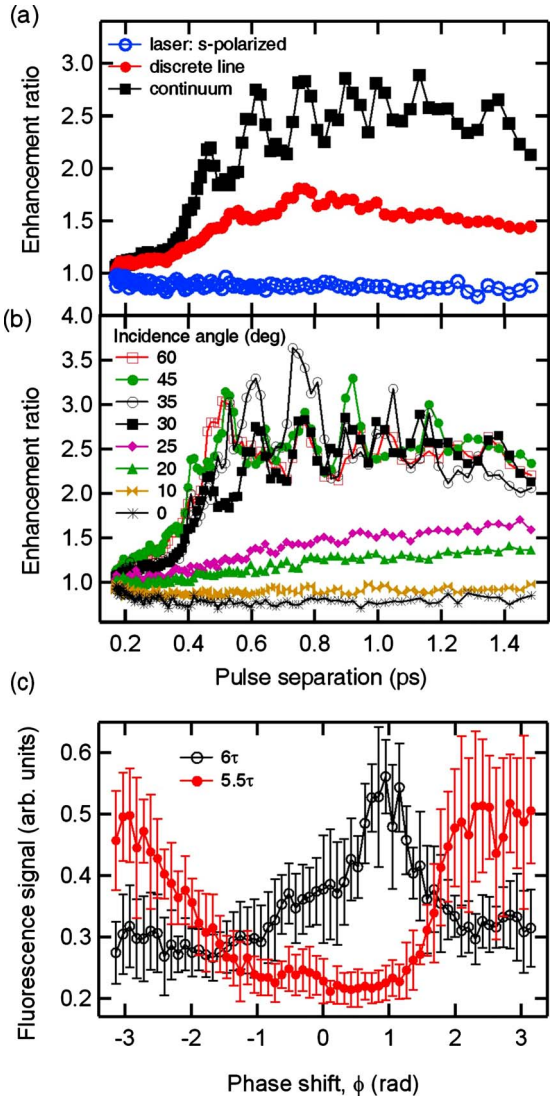

FIG. 4. (Color online) Dependence of the fluorescence intensity on properties of the three-pulse train. (a) Comparison of continuum emission at 450.8 nm produced with *p*-polarized (top trace) and *s*-polarized (bottom trace) radiation and discrete emission at 417.2 nm (middle trace) produced with *p*-polarized light, with a total fluence of 12.8 J/cm^2. (b) Effect of incidence angle on the continuum emission at 450.8 nm. A fixed pulse energy of 1.5 μJ was used in all the traces. (c) Effect of the optical phase shift on the 450.8 nm emission for pulse separations of 5.5τ (filled symbols) and 6τ (open symbols).

band. As the intensity grows, an avalanche generates a plasma on the surface of the crystal. This electron-hole plasma rapidly expands above the surface and absorbs additional photons during the first pulse and continues to absorb strongly during subsequent pulses in the train.

The dominant absorption mechanisms at our intensities are collisional and resonance absorption.[41] The former, also known as inverse bremsstrahlung, results from collisional damping of the electric field of the laser as it propagates through the plasma.[42] This effect is insensitive to the laser polarization, and, while undoubtedly present under our conditions, it cannot explain the strong polarization dependence

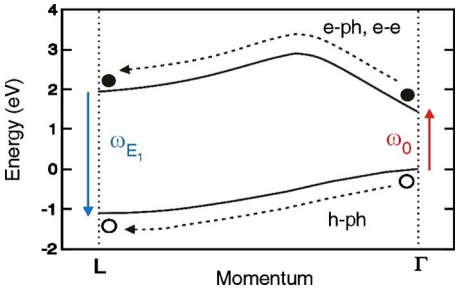

FIG. 5. (Color online) Mechanism for phonon-assisted recombination. A laser photon of frequency ω_0 creates an electron-hole pair. Electron-electron and electron-phonon scattering transport electrons, and hole-phonon scattering transports holes into the L valley. Recombination produces photons with energy peaked near 3 eV.

of the PL. In the latter process, obliquely incident *p*-polarized radiation, having an electric field component along the carrier density gradient, drives a resonant electrostatic wave in the plasma. This resonance is caused by the quivering of electrons in the plasma above the surface. The electron oscillation frequency in the plasma is given by $\omega_{pe} = 5.64 \times 10^4 n_e^{1/2}$, where n_e is the number density of electrons in units of cm^{-3}.[42] A resonance between electromagnetic oscillation and plasma oscillation occurs at a density such that ω_{pe} equals the laser frequency. The region where this condition is satisfied is referred to as the critical surface, which separates the underdense and overdense regions of the plasma. Under this condition, a *p*-polarized electromagnetic wave induces an electrostatic wave in the plasma. Resonance absorption of light occurs at the critical surface of the plasma, where an evanescent electromagnetic wave penetrates up to the skin depth of the plasma, and the rest of the incident wave is reflected. This resonance is a property of the plasma formed above the crystal and is unrelated to the properties of the crystal itself.

The critical plasma density at 800 nm equals 1.7×10^{21} cm^{-3}. Reflectance measurements showed that carrier densities on the order of 10^{22} cm^{-3} are generated near the melting threshold of GaAs.[33,43] Still greater densities are produced in our experiment, where the laser intensity is two orders of magnitude greater than the melting threshold. The complete absence of phonon-enhanced luminescence for *s*-polarized laser radiation [Figs. 2(b) and 4(a)] and the weak enhancement for *p*-polarized radiation at small incident angles [Fig. 4(b)] are consistent with of a resonant excitation mechanism.

A question we address next is how the laser energy is coupled to the lattice. An established mechanism for coherent phonon excitation in polar semiconductors is coupling of polarization and nuclear displacement by screening of the surface space charge.[8] This mechanism by itself, however, is insufficient to explain our observations because the overdense plasma does not transmit electromagnetic radiation beyond the skin depth. A more complete description of the laser-matter interaction includes impulsive excitation of the lattice by energetic electrons extracted by the laser field and redriven into the plasma. Like resonance absorption, such ponderomotive excitation of the electrons in the plasma, also

known as Brunel or vacuum heating,[44] requires *p*-polarized light incident at an oblique angle. The polarization and incidence angle thresholds shown in Figs. 2(b) and 4(b) indicate that the electric field must have at least a 40% component along the direction of the plasma gradient to produce these energetic electrons. The fraction of energy transferred by an electron of mass m_e to an atom of mass m_a in a head-on collision is determined by the mass ratio, such that $\Delta E/E = 4m_e/m_a$.[45] The minimum energy required to excite a single LO phonon is therefore 1.06 keV. There is ample experimental[46] and theoretical[47] evidence that at the intensities of our experiment Brunel absorption on the order of 10% produces hot electrons with energies of several keV. Also, Ref. 46 shows that Brunel excitation turns on at 3×10^{13} W/cm^2, which is close to our threshold intensity. The sensitivity of the phonon oscillations to the optical phase of the laser is consistent with a ponderomotive mechanism, with the phase of the field determining whether the electron is pulled into the vacuum or returned to the plasma.[48]

We consider next the mechanism for scattering of carriers into the L valley. Hot electrons, which may be produced by Brunel excitation, are injected by the first pulse into the conduction band of the L valley. Holes, which have much a greater mass ($0.57m_e$ vs $0.067m_e$) (Ref. 49) and lower mobility, are not injected rapidly into the L valley without appropriate pulse shaping. Instead, electrons slowly rescatter back into the Γ valley, where they recombine to emit near IR radiation.[35,50–52] Only when we use *p*-polarized pulse trains do we see the E_1 emission.

The enhancement observed with integer-τ spacing is evidence for phonon-mediated injection of holes into the L valley. It is known that one of the decay paths of the LO(Γ) phonon produces an acoustic phonon with momentum along the Γ-L axis.[53] Also, the temperature dependence of the decay of the LO(Γ) phonon has been attributed entirely to decay into an LO(L) phonon.[54] It is plausible that the 400 fs delay before the first phonon peak reflects the time required for the scattering of holes by LO(L) phonons.

We comment lastly on the termination of the phonon oscillations after $\Delta t = 11\tau$. The dephasing time of the LO(Γ) phonon is 5.7 ps at 215 K.[54] The 2.8 ps length of the pulse train at which the last phonon oscillation is observed [Fig. 2(a)] is consistent with a lattice temperature ≥470 K, depending on how much dephasing occurs between pulses in the train. Another possible explanation of the termination of the phonon peaks is expansion of the plasma. Brunel excitation occurs for a plasma scale length (defined as[42] $L^{-1} = \partial \ln n_e/\partial z$) on the order of 10% of the laser wavelength.[47] The scale length is initially very short because of the ultrashort duration of the laser pulse. As the plasma produced by the first pulse expands, this condition eventually cannot be sustained.

A number of reports have appeared describing the use of shaped ultrafast laser pulses to control the ablation of solid materials.[14,15] The mechanisms in such cases might be better described as "thermodynamic control." In such studies, the early part of the optimum pulse produces nonequilibrium states of matter that are interact more strongly with later parts of the pulse. This effect is operative here as well, inasmuch as the plasma generated by the first pulse shields underlying layers from further ablation. Nevertheless, the sensitivity of the phonon oscillations to the optical phase of the laser shows that coherence plays a role in generating the PL. The size and shape of the ablation craters, however, are insensitive to the pulse shape, indicating that coherence effects did not play a role in the ablation.

In summary, we have shown that it is possible to use a train of femtosecond laser pulses to control the recombination of electrons and holes in GaAs. The mechanism consists of laser generation and excitation of a plasma, excitation of phonons by ballistic electrons produced in the plasma, phonon scattering of carriers into the L valley of the Brillouin zone, and radiative recombination of the carriers. We have shown that it is possible control this effect by varying the delay between the pulses, their relative optical phases, and their polarization.

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*rjgordon@uic.edu

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