Directed Hypersonic Strain Waves Imaged with Ultrafast Electron Microscopy

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Intense, ultrafast photoexcitation of semiconducting materials produces several discrete but inter-related energy relaxation processes that span large spatial and temporal scales [1]. Initial charge-carrier excitation, and generation of a dense electron-hole plasma, produces hypersonic plasma waves that propagate outward from the excitation zone. Related to this, myriad scattering processes (electron-electron, electron-phonon, *etc.*) occur within the first few picoseconds following photoexcitation, ultimately leading to the launch of coherent propagating strain waves [2]. In thin membranes of semiconducting materials, propagating strain waves display Lamb-type symmetries and hypersonic velocities (depending on mode type and membrane thickness) [3]. In addition to often-used ultrafast spectroscopic methods, femtosecond X-ray and electron scattering has been used to study both hypersonic plasma waves and acoustic-phonon wavetrains [4,5]. Observations from these experiments, combined with knowledge of the associated spatiotemporal scales, lead to questions about preferred nucleation points/regions, propagation directions, and time-varying phase-velocity dispersion behaviors.

Here, we discuss the results of real-space imaging, with ultrafast electron microscopy (UEM) [6], of hypersonic coherent strain-wave dynamics in Ge [7]. While much is known about ultrafast charge-carrier dynamics in semiconductors like Ge and GaAs, several open questions remain pertaining to local acoustic-phonon behaviors and possible connections to hypersonic plasma waves [8]. Specimens consisted of 2° wedge-polished Ge single crystals. The polishing process employed produces linear nanoscale striations on the polished surface having a repeating ridge-like morphology [Figure 1(a,b)]. Intense, *in situ* femtosecond photoexcitation of these specimens produces coherent acoustic-phonon wavetrains that propagate parallel to the striations and with initial phase velocities well in excess of the bulk speed of sound [*e.g.*, 35 km/s vs. 5 km/s, respectively; Figure 1(c,d)]. Further, while the phase velocity of each wavefront is fixed across the micrometer imaging field of view, each subsequent wavefront has a slightly reduced velocity, ultimately decaying to the approximate bulk speed of sound over one nanosecond [Figure 1(e)]. Comparison of this imaged time-varying phase-velocity dispersion behavior to calculated guided acoustic modes suggests a single, first-order symmetric Lamb mode is responsible for the observed contrast dynamics [Figure 1(f)]. By using a plasma lensing method to determine time zero (moment of *in situ* specimen photoexcitation) [9], a significant delay (100 ps) was observed to occur before emergence of the first coherent contrast wave (a total of 200 ps passed before quantifiable waves were observed). This suggests a connection between Auger recombination dynamics and strain-wave nucleation and launch [10].

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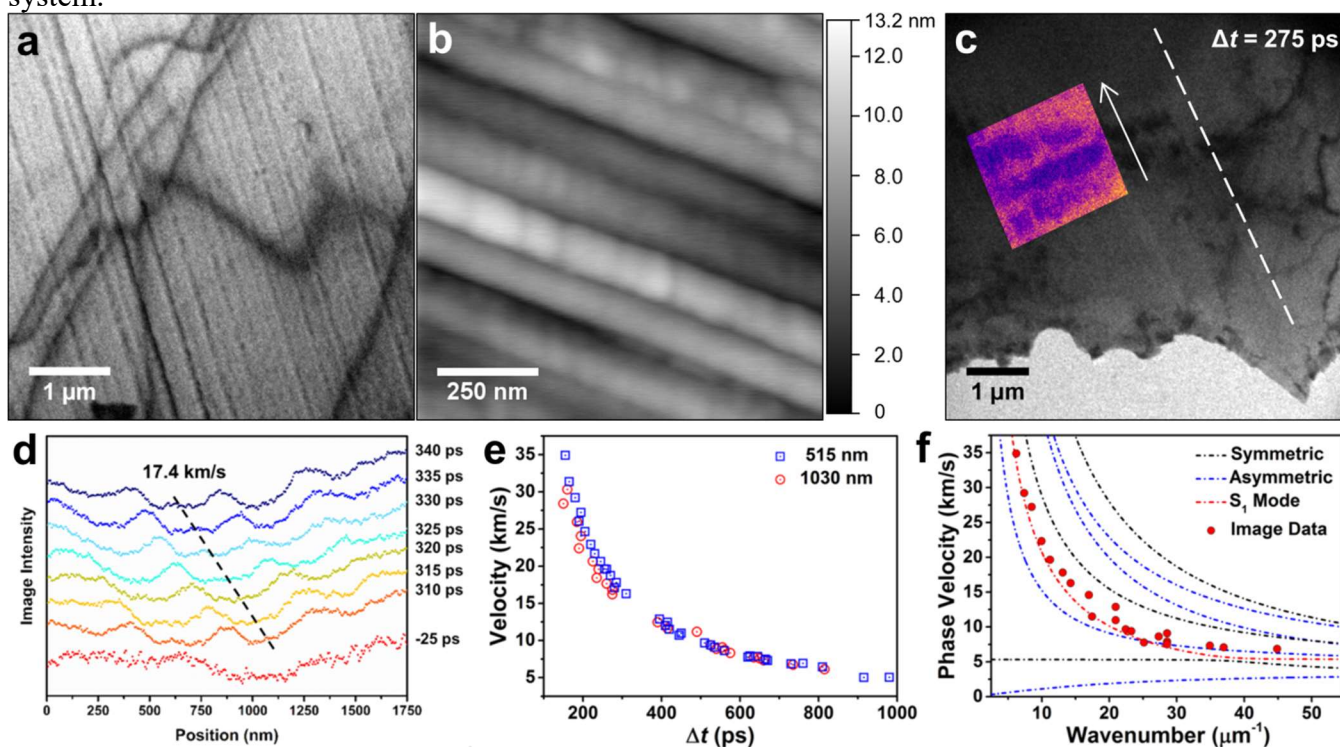[5] M. Harb *et al*, Phys. Rev. Lett. **100** (2008), p. 155504.

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Figure 1. Directed hypersonic acoustic-phonon wavefronts in Ge. (a) Bright-field TEM image of linear polishing striations in a 2° Ge wedge specimen. (b) AFM image (tapping mode) of the polished Ge surface (average ridge width = 110 ± 30 nm; n = 19 ridges; error is one standard deviation). (c) Bright-field UEM image obtained 275 ps after *in situ* femtosecond photoexcitation (Δt). The false-colored region is meant to highlight contrast features arising from a coherent wavetrain, and the arrow indicates the wavefront propagation direction. The dashed line marks the general linear striation orientation. (d) Line scans from select UEM images (*e.g.*, 310 ps, 315 ps, 320 ps, *etc.*) from the false-colored region shown in (c). One wavefront propagating at 17.4 km/s is highlighted with a dashed black line. A pre-excitation line scan (-25 ps) is shown for comparison. (e) Time-varying phase-velocity dispersion of propagating acoustic phonons generated with two different photoexcitation wavelengths (515 and 1,030 nm, blue squares and red circles, respectively). Time zero was determined with a plasma lensing method [9], and the first quantifiable wavefronts occurred 200 ps after photoexcitation. (f) Experimental phase-velocity dispersion (red dots) compared to calculated guided acoustic modes (dot-dashed lines). Symmetric modes are shown in black, and asymmetric modes are shown in blue. The first-order symmetric mode is shown in red and most closely matches the experiment.