Bulk Ambipolar Diffusion Measured by Ultrafast X-ray Diffraction

D.A. Reis,[1] P.H. Bucksbaum,[1] A. Cavalieri,[1] R. Clarke,[1] M.F. DeCamp,[1] E.M. Dufresne,[1]
R. Merlin,[1] D.A. Arms,[2] A.M. Lindenberg,[3] A.G. MacPhee,[3] Z. Chang,[4] B. Lings,[5]
J.S. Wark,[5] S. Fahy[6]
[1]FOCUS Center and Department of Physics, University of Michigan, Ann Arbor, MI, U.S.A.
[2]Advanced Photon Source (APS), Argonne National Laboratory, Argonne, IL, U.S.A.
[3]Department of Physics, University of California, Berkeley, CA, U.S.A.
[4]Department of Physics, Kansas State University, Manhattan, KS, U.S.A.
[5]Department of Physics, Clarendon Laboratory, University of Oxford, Oxford, England
[6]Physics Department and National Microelectronic Research Center (NMRC), University College Cork, Ireland

Introduction

Ultrafast strain generation by short-pulse laser absorption has been studied extensively in optical and x-ray pump-probe experiments, for example [1-11]. When the laser pulse duration is short compared to the speed of sound across the penetration depth, then coherent acoustic phonon pulses are developed. Thomsen et al. developed an analytical model of coherent acoustic phonon generation and propagation within the limit of instantaneous stress production [2]. In this case, a static surface strain and a symmetric bipolar acoustic pulse are produced. Typically the stress is thermal; therefore, the model is known as thermoelastic. While this model is an idealization, results have often been in good agreement with the theory. Deviations have been discussed by Thomsen et al., and recently, quantitative differences have been observed in x-ray diffraction [6, 8, 9, 11, 12]. In this work, we report on recent results on strain generation in bulk germanium (Ge) single crystals, where ambipolar diffusion of the electron-hole pairs produces the dominant strain mechanism (through the deformation potential) [13]. In germanium, the diffusion front travels faster than the speed of sound, such that the we can no longer consider the acoustic pulse as arising from instantaneous stresses.

Methods and Materials

Photoexcited carriers are produced in a 280-µm-thick single crystal (001) Ge by a femtosecond near-infrared laser pulse. The Ge is arranged in an asymmetric Laue geometry, with diffraction from the 202 lattice planes (these planes are at a 45° angle between the surface normal) oriented perpendicular to the x-ray polarization (Fig. 1), and the laser is incident on the x-ray transmission side of the crystal. Experiments are performed by using the pump-probe technique. Transient changes to the lattice spacing show up as variations in the anomalous transmission (Borrmann Effect) [14] — as temporal Pendellösung fringes [12]. Deviations from the thermoelastic model manifest themselves in the amplitude

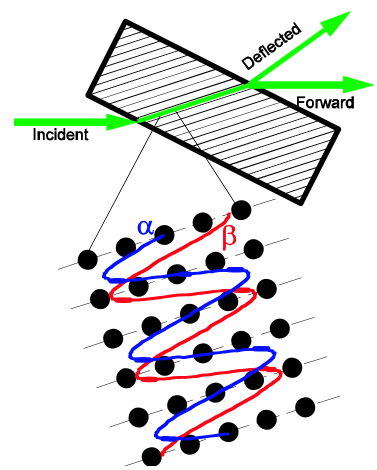

FIG. 1. Diffraction geometry.

and phase of these Pendellösung oscillations. We measure time-resolved diffraction curves for an x-ray energy of 10 keV as a function of laser fluence. The laser fluence is such that the initial carrier concentration varies between approximately a few 10^{19} cm^{-3} and a few 10^{20} cm^{-3}.

Results and Discussion

Figure 2 shows the time-resolved anomalous transmission for a few excitation densities. The frequency of the oscillations corresponds to the Pendellösung length divided by the longitudinal speed of sound in the [001] direction, irrespective of the excitation density. The phases of the oscillations, however, depend strongly on the excitation density, as does an initial transient that is unresolved with the 100-ps resolution afforded by the APS single-bunch duration. This transient, which is not predicted by the thermoelastic model, suggests that there is a supersonic component to the strain generation. However, the frequency of the oscillations at later times are consistent with strain propagation at the speed of sound; this suggests that the supersonic character occurs only at early times. It also suggests that the strain

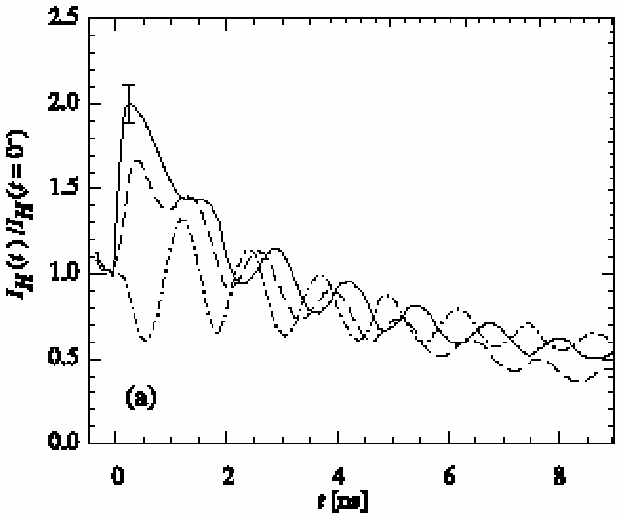

FIG. 2. Time-resolved anomalous transmission (deflected-diffracted) intensity for an incident laser fluence of 35 (solid line), 7 (dashed line), and 2 (dotted-dashed line) mJ/cm^2. The error bar represents the estimated systematic error in the overall scale, adapted from Ref. 13.

generation is, in part, due to rapid ambipolar diffusion of the dense electron-hole plasma and its coupling to the lattice through the deformation potential. Figure 3 shows the amplitude of the transient and the phase of the Pendellösung oscillations, as well as predictions from a full dynamical simulation, including the effects of thermal expansion, the deformation potential, thermal and carrier diffusion, and Auger recombination. The dominant effect is due to the deformation potential and carrier diffusion, without which the data cannot be even qualitatively reproduced. To our knowledge, these results are the first true bulk measure of carrier diffusion in the dense plasma regime. More detailed studies may reveal the density dependence of material properties, such as the carrier diffusion rate and speed of sound.

Acknowledgments

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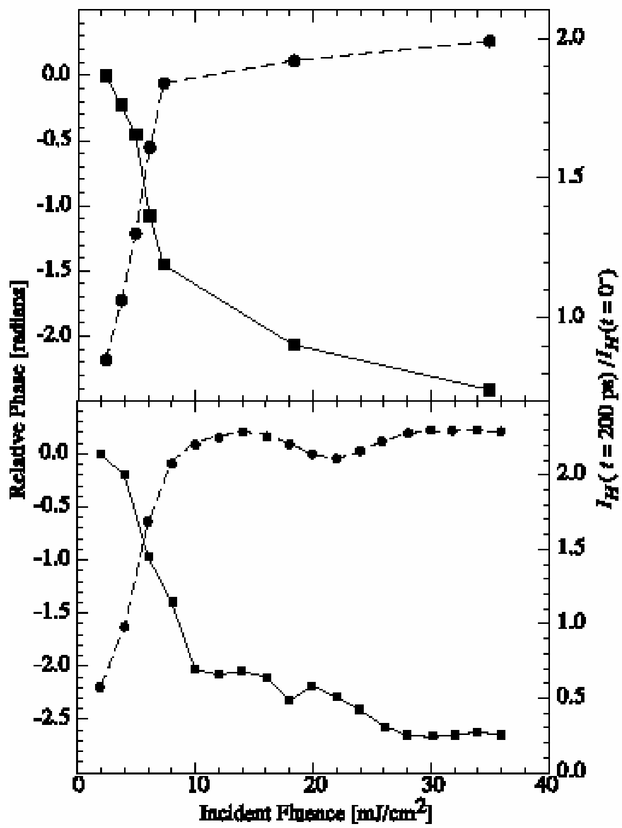

FIG. 3. Upper experiment (lower simulation): The relative phase of the Pendellösung oscillations (squares) and the normalized deflected-diffracted intensity at a time delay of 200ps (circles) as a function of incident optical fluence. From Ref. 13.

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