Relativistic Doppler Frequency Up-conversion and Probing the Initial Relaxation of a Non-Equilibrium Electron-Hole Plasma in Silicon

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***Abstract*—we demonstrate experimentally the relativistic Doppler frequency up-conversion of the THz pulses from the counter-propagating ionized plasma front in silicon. The observed frequency up-conversion can be well modeled by the 1D FDTD simulations if significant short scattering time (well below 10 fs) in the plasma is assumed. To further elucidate the scattering rate in the electro-hole plasma, we performed pump probe experiment employing ultra-broadband (150 THz) THz-Mid-Infrared pulse. The results show the scattering time decreases from ~200 fs down to ~20 fs when the carrier density increases up to 10^{19}-cm^{-3}, and then saturates for higher densities. Such scattering time dependence on plasma carrier density can be very well fitted by the Drude model for thermalized electron-holes, and the saturation behavior is attributed to electron-hole phase-space restriction as the plasma becomes degenerate. The resultant much shorter scattering time measured with non-thermalized plasma is in good accordance with the Doppler experiment, which demonstrates Doppler geometry an effective method for probing non-equilibrium plasma dynamics.**

I. INTRODUCTION

When an electromagnetic wave reflects off a counter-propagating plasma front, it will be frequency-upshifted by the relativistic Doppler effect [1]. The frequency upshift factor $\Gamma_{\text{theo}} = \omega_{\text{out}} / \omega_{\text{in}}$ of a lossless (collision-free) plasma depends on the velocities of the plasma front and of the wave. Indeed, the charge carriers themselves do not have to move, only a fast-propagating charge-density gradient is required. Such a situation can be obtained by the optical interband excitation of a semiconductor material, in which the propagating optical pulse generates a fast moving plasma front. The semiconductor has to be a volume material in order to enable the propagation effect, while the carriers themselves can be spatially confined (e.g., in quantum structures) or unconfined.

II. RESULTS

Here, we study the relativistic interaction between THz pulses and counter-propagating carrier plasma fronts in undoped bulk silicon excited by 30-fs optical pulses [2]. The THz pulses with a bandwidth of 20 THz are derived from laser-ionized air. A strong frequency up-shift would be expected if the plasma in Si were collision-free, as Γ_{theo} amounts to 8.8. The upper panel of Fig. 1 displays experimental data as a function of the delay time between the optical excitation pulse and the THz pulse. When the THz pulse encounters a propagating excitation front (around delay time zero), a frequency up-shift is observed. It is much weaker, however, than expected from Γ_{theo}. A reduction of the up-shift factor can be explained by the fast collisions of the charge carriers.

In order to assess the influence of charge-carrier collisions in a quantitative manner at least in an effective relaxation picture, knowledge of the momentum scattering time as a function of carrier density is required. It turns out that the literature does not provide reliable information for Si, which led us to perform conventional pump/probe measurements of the dynamical conductivity of the charge carriers in Si from which to derive Drude scattering times [3]. In the experiment, we employ a white-light continuum as a probe pulse, containing a very broad frequency up until 150 THz [4-5]. The lower panel of Fig. 1 displays the resultant Drude scattering time over a large range of carrier densities for a pump-probe delay of 1 ps. The scattering time decreases from the low-density value of 200 fs down to 20 fs, if the density is raised to the 10^{19}-cm^{-3} range. The data are very well explained by theory as shown by the full line in the graph of Fig. 1 [3].

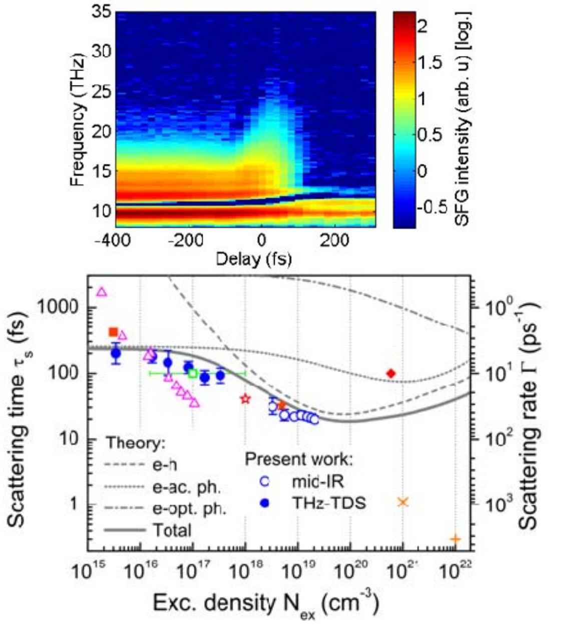

Fig. 1. Measured frequency upshift by Doppler reflection (top); Measured and calculated scattering time in a thermalized electron-hole plasma as a function of carrier density (bottom). The symbols in other colors than blue are from the literatures [6-13].

One has to note, however, that the THz pulse does not interact with a thermalized electron-hole plasma in the Doppler experiment, but that the carrier distributions encountered by the THz pulse at the different locations in the sample are far from Fermi-Dirac distributions. In order to account for this situation, we also measured the conductivity response if the optical-pump THz-probe delay is reduced, and a non-thermal electron-hole plasma is probed. One finds, that the Drude scattering time decreases significantly below 20 fs in the 10^{19}-cm^{-3} range.

Employing 1D-FDTD simulations [1], we model the Doppler measurements and succeed in reproducing the measured spectra of the top panel of Fig. 1 fairly well, if we assume the Drude scattering time to be well below 5 fs. Doppler reflection experiments apparently are extremely sensitive probes of the initial scattering processes of non-equilibrium photo-excited charge carriers, because any up-shifted THz radiation arises due to the interaction with the moving plasma front, and hence probes the initial charge-carrier dynamics quite specifically. The approach should also be applicable to other material systems.

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