Photoexcited GaAs surfaces studied by transient terahertz time-domain spectroscopy

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The transmission characteristics of an air–GaAs interface and the transient absorption and index spectra of the thin, photoexcited surface layer are investigated subsequent to excitation by a femtosecond laser pulse. We find that the total phase change and transmission of a terahertz (THz) probe pulse are dominated by interface effects. This observation has important implications in the interpretation of THz time-domain spectroscopy data of absorbing media. We also observe that the THz pulse apparently arrives at the detector as much as 60 fs earlier when it is transmitted through an optically excited GaAs wafer. This effect is fully explained in terms of a frequency-dependent transmission and phase shift at the air–GaAs interface and is not associated with superluminal propagation. © 2000 Optical Society of America

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During the past decade, terahertz (THz) time-domain spectroscopy (TDS) has been refined and applied to precise studies of the static optical properties of a wide, and still expanding, range of materials.[1–3] The subpicosecond duration of the THz pulses in a THz TDS system offers the exciting possibility of time-resolved spectroscopy in the THz region in pump–probe experiments. For work done by other groups of researchers in this new field, see Refs. 4–7.

Here we investigate the transient absorption and refractive-index spectra of semi-insulating (SI) GaAs subsequent to excitation by 800-nm photons (excess energy, 120 meV). We use this simple case to illustrate how to analyze data from transient THz TDS of optically excited semiconductors. The typical absorption depth of the pump light is less than 1 μm, and the extraction of the transient dielectric function (i.e., absorption coefficient and refractive index) of the thin, absorbing layer (the surface layer) is complicated by a large, frequency-dependent phase shift at the interface between air and semiconductor. We outline an iterative procedure for extracting the dielectric function from such experimental data and show that it is possible to determine both the absorption and the index of the excited layer and simultaneously to find the frequency-dependent complex transmission function of the air–GaAs interface. We consider only GaAs here, but the data analysis presented is of general validity.

Our experimental setup consists of a THz TDS system driven by the output of a regeneratively amplified Ti:sapphire oscillator (Clark-MXR CPA 1000). The THz pulses are generated by optical rectification at a GaAs (311) surface and are guided over an intermediate focus, where the sample is located, to an electro-optic ZnTe detector crystal. A part of the laser beam is used to excite the sample at a variable time delay with respect to the THz probe beam. The sample is the excited surface layer of a 5.08-cm-diameter, 508-μm thick wafer of semi-insulating GaAs ($\rho > 10^8\ \Omega$ cm). We obtain carrier densities in the 10^{17} to 10^{18} cm^{-3} range in a 1-μm-thick surface layer by femtosecond optical excitation. The spatial profile of the low-frequency components of the THz beam diverges at the sample. For frequencies below 350 GHz, the THz spot size in our experiment is larger than the pump beam. Hence at low frequencies unpumped regions of the sample are probed in addition to the pumped area. The transient transmission of the unpumped regions is unity, with no additional phase shift. This effect is also seen in Fig. 3 of Ref. 4. As a consequence we can determine the complex, transient transmission function of the sample only in the frequency range from 350 GHz to 2.8 THz, with a time resolution of 250 fs.

When we excite SI GaAs with optical intensities of a few microjoules per square centimeter we observe a shift of the THz pulse to earlier times than for its passage through an unexcited sample [see Fig. 1(a)]. The apparent shift is of the order of 50–100 fs. Causality prohibits transit times over the excited region that are faster than the transit time over a corresponding distance in vacuum. The penetration depth of the 800-nm excitation pulse in GaAs is $\sim$1 μm, and, with a group refractive index of $\sim$3.5 for a pulse with center frequency 1 THz, we do not expect to see a time shift toward earlier times of more than 8.3 fs, assuming that the time shift is caused by propagation effects inside the excited layer. We show below that the frequency-dependent transmission coefficient and phase shift at the air–GaAs interface account for the observed negative time shift of the THz pulse.

When a plane wave of cyclic frequency ω is transmitted at normal incidence over an interface between air (refractive index 1) and a medium with refractive index n and field absorption coefficient α_f (related to the power absorption coefficient as $\alpha_p = 2\alpha_f$), the field transmission amplitude t_{12} and the phase shift χ_{12} are[8]

$$t_{12} = \frac{2}{[(1+n)^2 + (\alpha_f c/\omega)^2]^{1/2}}, \tag{1}$$

$$\tan \chi_{12} = -\frac{\alpha_f c}{\omega(1+n)}, \tag{2}$$

where the subscript 12 refers to the interface between air and sample. Hence, if a field is transmitted into

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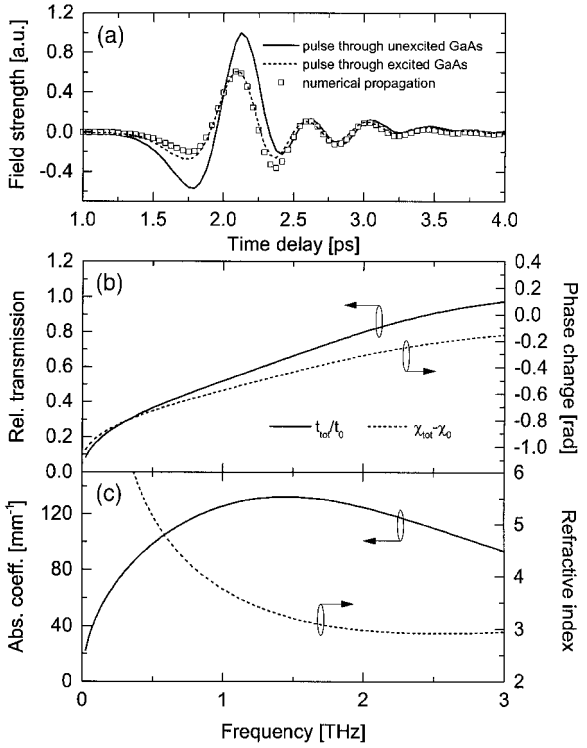

Fig. 1. (a) Experimental and numerical propagation of a THz pulse through photoexcited GaAs. (b) Model transmission coefficient and phase shift. (c) Absorption coefficient and refractive index.

an absorbing medium, there is a negative phase shift of the field caused by the interface. Equations (1) and (2) reduce to the well-known $t_{12} = 2/(1 + n)$ and $\chi_{12} = 0$ for unexcited SI GaAs and for nonabsorbing media in general. In addition to transmission over the air–GaAs interface, propagation through the excited surface layer must also be taken into account. The transmission coefficient through the excited layer is $t_2 = \exp(-\alpha_f L)$, and the corresponding phase shift is $\chi_2 = \omega n L/c$, where L is the thickness of the excited layer, defined by the absorption depth of the excitation pulse. The subscript 2 refers to the excited layer. With the knowledge of the functional form of t_{12}, t_2, χ_{12}, and χ_2 we can model the propagation of a THz pulse through the excited sample, once $\alpha_f(\omega)$ and $n(\omega)$ are known. The Drude theory of conduction gives a simple, yet physically realistic, model of the transient absorption coefficient and refractive index of the excited layer.

In Fig. 1(a) we show two experimental THz pulses, one transmitted through a SI GaAs wafer (the reference pulse, solid curve) and the other (the sample pulse, dashed curve) transmitted through an optically excited GaAs wafer. The main part of the sample pulse arrives earlier than the reference pulse. The data points represented by open squares are the result of a numerical propagation of the reference pulse based on the Drude model, calculated with a carrier density of 10^{18} cm^{-3} in a 1-μm-thick layer and a damping rate $\Gamma = 20$ THz, resulting in absorption and index spectra as shown in Fig. 1(c). These curves are used in the evaluation of t_{12}, t_2, χ_{12}, and χ_2. Figure 1(b) shows the total transmission coefficient and phase change that we applied to the spectrum of the reference pulse to evaluate the numerical pulse profile in Fig. 1(a) (open squares). The open squares in Fig. 1(a) were obtained by Fourier transformation of the reference pulse. Then the transmission coefficients and phase shifts were applied to each frequency component, and the resultant spectrum was transformed back to the time domain by inverse Fourier transformation. The simulated propagation of the reference pulse reproduces the shape of the sample pulse well. The negative delay of the sample pulse with respect to the reference pulse is determined mainly by the interface phase shift χ_{12}, which is negative and within the bandwidth of the THz pulse is approximately linear. In contrast to frequency-independent phase shifts, as observed in total internal reflection,[9] this phase shift does not significantly reshape the THz pulse. Its consequence is mainly a shift of the entire THz pulse to earlier times. In light of Ref. 10 we emphasize that the observed time shift is not associated with superluminal propagation of the THz pulse, even though such might be a first conclusion.

In what follows, we show how to evaluate the transient absorption coefficient and refractive index from actual data. A THz TDS experiment consists of measuring the shapes of THz pulses after propagation through a reference medium and through a sample medium and comparing the spectra of the two pulses. In our experiment the sample–reference pair is recorded with and without excitation of the stationary sample, with all other experimental conditions fixed. The ratio of sample pulse to reference pulse in the frequency domain can be described by

$$\frac{E^{\star}(\omega)}{E(\omega)} = \frac{t_{12}^{\star} t_2^{\star}}{t_{12}} \exp[i(\chi_{12}^{\star} + \chi_2^{\star} - \chi_2)], \tag{3}$$

where the superscript * refers to the excited sample. For simplicity, we assume that the unexcited sample is without dispersion and absorption. The transient dielectric function changes smoothly into the bulk dielectric function over several absorption lengths of the visible beam. Therefore we ignore interface effects between the excited surface layer and the bulk material of the sample. By using Eqs. (1) and (2) we can express the ratio of the sample pulse spectrum to the reference pulse spectrum as a transmission amplitude t_m and a phase ϕ_m, so $E^{\star}/E = t_m \exp(i\phi_m)$:

$$t_m(\omega) = \frac{(1 + n_0)\exp(-\alpha_f L)}{[(1 + n_0 + \Delta n)^2 + (\alpha_f c/\omega)^2]^{1/2}}, \tag{4}$$

$$\phi_m(\omega) = \frac{\omega \Delta n L}{c} + \arctan\left(-\frac{\alpha_f c/\omega}{1 + n_0 + \Delta n}\right), \tag{5}$$

where n_0 is the static refractive index and $\alpha_f = \alpha_f(\omega)$ and $\Delta n = \Delta n(\omega)$ are the frequency-dependent, transient absorption coefficient and refractive index, respectively. These coupled, nonlinear equations can be

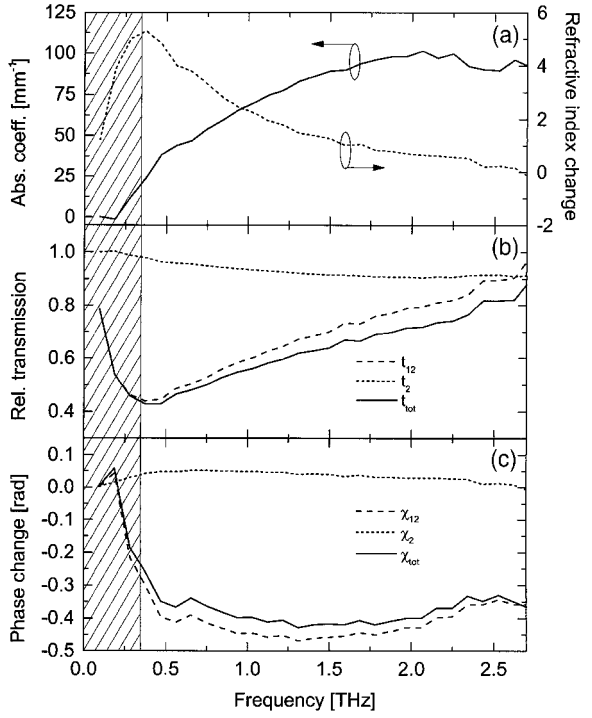

Fig. 2. Experimental results. (a) Transient absorption coefficient and refractive index. (b) Transmission coefficient. (c) Phase shift with respect to an unexcited sample.

solved by iteration. The Newton–Raphson method for nonlinear systems of equations[11] can be applied to yield improved values of α_f and Δn from initial values $\alpha_f^{(0)}$ and $\Delta n^{(0)}$. We can obtain good initial values by assuming that t_m and ϕ_m originate from the interface only. Figure 2(a) shows the transient absorption coefficient α_f and the refractive index Δn of SI GaAs, recorded 4.2 ps after excitation with a 100-fs, 800-nm, 2.4-μJ/cm^2 laser pulse. The data are extracted by use of the Newton–Raphson algorithm. Above 350 GHz there is qualitative agreement between the absorption and index spectra in Fig. 2(a) and the numerical data in Fig. 1(c), indicating that, after thermalization of the carriers (typical relaxation time in GaAs after 800-nm excitation is less than 1 ps), the Drude model gives a reasonable description of the experimental data. Note that the vertical scale in Fig. 1(c) gives the absolute refractive index of the excited layer, whereas the transient index, relative to the static value, is shown in Fig. 2(a).

Figure 2(b) shows transmission amplitude t_m and the contributions from the interface and the surface layer. Figure 2(c) shows total phase change ϕ_m, together with the contributions from the interface and the surface layer. The contribution from the interface to both the transmission amplitude and the phase change is substantial. The low-frequency (<350-GHz) features of the experimental data, marked by the hatched area in Fig. 2 ($\Delta n \rightarrow 0$, $t_m \rightarrow 0$, and $\phi_m \rightarrow 0$), are artifacts of the experiment, as explained above.

In conclusion, we have demonstrated an iterative method for extracting the transient dielectric functions of semiconductors in the THz range subsequent to excitation with femtosecond pulses. The analysis of transient THz TDS data of optically dense, excited surface layers, measured in transmission, is relatively complicated, which suggests that it is better to perform experiments in a reflection geometry.[3] The carrier densities of the excited surface layer studied in our research are comparable with carrier densities in highly doped semiconductors. The reflection geometry offers a straightforward analytical approach to the extraction of the dielectric function.[8]

We have explained the apparent shift to earlier times of the transmitted THz pulse after excitation of the sample in terms of the frequency-dependent transmission function over the air–GaAs interface and shown that it is not caused by superluminal propagation effects in the sample.

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