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Published in:
Applied Physics Letters

Link to article, DOI:
10.1063/1.4810756

Publication date:
2013

Document Version
Publisher's PDF, also known as Version of record

AIP | Applied Physics Letters

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Citation: Appl. Phys. Lett. **102**, 231120 (2013); doi: 10.1063/1.4810756
View online: http://dx.doi.org/10.1063/1.4810756
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v102/i23
Published by the American Institute of Physics.

Density-dependent electron scattering in photoexcited GaAs in strongly diffusive regime

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(Received 19 March 2013; accepted 22 May 2013; published online 12 June 2013)

In a series of systematic optical pump–terahertz probe experiments, we study the density-dependent electron scattering rate in photoexcited GaAs in the regime of strong carrier diffusion. The terahertz frequency-resolved transient sheet conductivity spectra are perfectly described by the Drude model, directly yielding the electron scattering rates. A diffusion model is applied to determine the spatial extent of the photoexcited electron-hole gas at each moment after photoexcitation, yielding the time-dependent electron density, and hence the density-dependent electron scattering time. We find that the electron scattering time decreases from 320 to 60 fs, as the electron density changes from 10^{15} to 10^{19} cm^{-3}. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4810756]

GaAs is an archetypal direct-gap semiconductor, widely used in a variety of applications ranging from transistors to laser diodes. GaAs is also one of the key materials employed in present-day terahertz (THz) semiconductor optoelectronics, which is largely based on ultrafast photoconductive phenomena.[1–3] A thorough understanding of the photoconductive behavior of this material is, therefore, of both fundamental and technological relevance. The conductivity is defined as $\sigma = eN\mu$, where e is the elementary charge, N is the carrier density, and $\mu = e\tau_s/m^*$ is the mobility dependent on carrier momentum scattering time τ_s and effective mass m^*. The scattering time τ_s can in turn depend on the carrier density,[4] leading to a complex dependency $\sigma = eN\mu = e^2N\tau_s(N)/m^*$. The carrier momentum scattering time is one of the key parameters for the performance of THz optoelectronic systems, e.g., defining the efficiency of traditional THz photoconductive emitters,[5,6] and determining the sign of the THz refractive index nonlinearity of a semiconductor.[7,8]

In most THz applications, the photoconductivity in a semiconductor arises from above-bandgap photoexcitation. Such photoexcitation typically leads to a gradient in the spatial distribution of carriers, which causes carrier diffusion.[10,11] Therefore, the temporal dynamics of the photoexcited carrier density $N = n/V$ depends not only on the excitation parameters and decay of free carriers via recombination or trapping, but also on the expansion of the electron gas volume V via diffusion from the photoexcited region into the bulk of the material. Here n is the number of carriers.

Surprisingly, however, the explicit dependency of electron momentum scattering time on electron density $\tau_s(N)$ in GaAs, especially in photoexcited GaAs where the photocarrier diffusion effects are dominant, is not yet characterized for a wide range of electron densities. Carrier density dependence of the scattering rate has been studied in detail for Si and TiO_2,[4] and has been reported previously over a relatively small density range for GaAs.[9] In this work, we show that the electron momentum scattering time in photoexcited GaAs is a highly dynamic parameter: it decreases by as much as a factor of 4, as the electron density is increased over the range of 10^{15}–10^{19} cm^{-3}.

In our experiments, we determine the ultrafast photoconductivity response using optical pump–THz probe (OPTP) spectroscopy.[2,3,9,12,13] A semi-insulating (SI) bulk GaAs wafer is photoexcited using sub-50 fs laser pulses, centered at 400 nm, with variable pump fluence in the range of $1 - 200\,\mu$J/cm^2. The optical penetration depth determines the initial thickness of the photoexcited electron-hole gas, and amounts to $d_0 = 15$ nm at 400 nm wavelength.[14] The correspondingly large charge carrier density gradient efficiently drives the diffusion of photoexcited electrons and holes into the bulk of the sample, which will be analyzed below. At a variable time delay t_p after the photoexcitation, the complex-valued *sheet* photoconductivity spectra of GaAs were resolved by ultrashort THz probe pulses with useful spectrum covering the frequency range of 0.5–2.5 THz. The experiment was carried out at room temperature and in transmission configuration. The pump-probe delay time t_p was varied in the range of 10-1000 ps. A minimum pump-probe delay of 10 ps was chosen in order to ensure the complete cooling of the carriers after 400 nm excitation, which occurs via intervalley $L - \Gamma$ transfer with a time constant of 3.5 ps.[8,15] We note that in GaAs the photoconductivity is almost fully dominated by the contribution of electrons rather than holes, owing to a much lighter electron effective mass as compared to the holes, $m_e = 0.067m_0$ vs $m_h = 0.45m_0$ (m_0 is the electron rest mass).[16]

The direct product of an OPTP measurement is the time- and frequency-resolved complex-valued THz field transmission spectrum $\hat{t}(\omega, t_p)$, which can be directly converted into the *sheet* complex-valued conductivity spectrum $\hat{\sigma}_s(\omega, t_p)$, e.g., using a fairly accurate thin-film approximation known as the Tinkham equation.[3,17] The sheet conductivity $\hat{\sigma}_s$ is related to (bulk) conductivity as $\hat{\sigma} = \hat{\sigma}_s/d$, where d is the thickness of the conductive layer, in our case the spatial extent of the electron gas. Under conditions of diffusion, d is naturally a

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dynamic parameter, which needs to be determined *before* the (bulk) conductivity spectrum $\hat{\sigma}(\omega, t_p)$ can be reliably inferred from the THz transmission measurements.

Exemplary measurements of the complex-valued sheet conductivity spectra of GaAs excited at 400 nm with pump fluence of 13 μJ/cm^2 are shown in Fig. 1. These spectra were measured at respective pump-probe delays of $t_p = 10$ ps, 100 ps, and 300 ps. Like all other recorded spectra in this work, these data are described very well by the Drude model (solid lines in Fig. 1)

$$\hat{\sigma_s}(\omega) = \hat{\sigma}(\omega)d = \frac{N_s e^2 \tau_s}{m^*} \frac{1}{1 - i\omega\tau_s}. \quad (1)$$

The values for the sheet electron density N_s, obtained from the Drude fits, are shown in Fig. 1. Since the time-dependent effective thickness $d(t_p)$ of the electron gas, defined by the diffusion, is not yet determined, the free electron density $N(t_p) = N_s(t_p)/d(t_p)$ cannot be quantified at this stage. However, the electron momentum scattering rate $\gamma(t_p) = \tau_s^{-1}$ can be readily obtained from the complex sheet conductivity spectra: in the Drude model it is given by the crossing frequency of the real and imaginary parts, multiplied by 2π (note that the THz probe frequency is $\omega/2\pi$).

It is apparent from Fig. 1 that the electron momentum scattering rate $\gamma = \tau_s^{-1}$ is strongly dependent on the pump-probe delay, and hence on the free carrier density N: it shows a monotonous decrease with decreasing N. We observed the same trend in all our measurements, i.e. over the whole range of pump fluences 1-200 μJ/cm^2 and pump–probe delays of 10–1000 ps.

In order to establish the dependency of electron momentum scattering time on the carrier density $\tau_s(N)$, one needs to correlate the above determined values for scattering times $\tau_s(t_p)$ with the carrier density $N(t_p) = N_s(t_p)/d(t_p)$. To this end, we determine the effective thickness of the electron gas $d(t_p)$ as a function of pump-probe delay numerically. In previous studies, the diffusion of an electron-hole gas in GaAs at the delays $t_p > 3$ ps was found to be ambipolar:[9,10] that is, electron and hole populations diffuse together, and the expansion of the electron-hole plasma is governed by the diffusivity of its least mobile constituent—the holes. This effect is demonstrated in Fig. 2, where we present the numerical solution of a drift-diffusion equation,[18] describing co-diffusion of electrons and holes.

Now we can use a simple diffusion model to simulate the expansion of the electron-hole gas. The ambipolar diffusion constant is

$$D_{ab} = \frac{(n+p)D_n D_p}{D_n p + D_p n} = \frac{2\mu_n \mu_p}{\mu_n + \mu_p} \frac{k_B T}{e} \approx \frac{2k_B T \mu_p}{e}, \quad (2)$$

where p and n are the concentrations of holes and electrons, respectively, and D_p and D_n are the corresponding diffusion constants. Here, we have used the Einstein relation $D = k_B T \mu / e$, and assumed $n = p$.

In our simple model we ignore the possible effect of bulk or surface recombination on the spatial profile of the electron and hole densities, and we use the literature value for the hole mobility $\mu_h = 360\,\mathrm{cm^2 V^{-1} s^{-1}}$.[19] The skin depth of GaAs at 400 nm $d_0 = 15$ nm is much smaller than both the optical and THz wavelengths in our experiments, so the initial density distribution $n(x, t_p)$ of the electrons and holes can be considered a delta function, and the diffusion profile can be modeled by the simplest form of the diffusion equation,[18] yielding the straightforward solution

$$n(x, t_p) = \frac{N_{s0}}{d_0 \sqrt{4\pi D_{ab} t_p}} \exp\left(-\frac{x^2}{4 D_{ab} t_p}\right), \quad (3)$$

where x is the depth coordinate and N_{s0} is the sheet density of electrons right after the photoexcitation (see inset of Fig. 2). At each pump-probe delay t_p, we estimate the effective thickness d of the electron layer as the ratio of the total number of carriers and the maximum value of the density $d = \int_x n(x)dx / \max[n(x)]$, as shown in the inset of Fig. 2. We thus obtain the effective thickness of the layer at each pump delay $t_p > 0$ as $d(t_p) = \sqrt{\pi D_{ab} t_p}$.

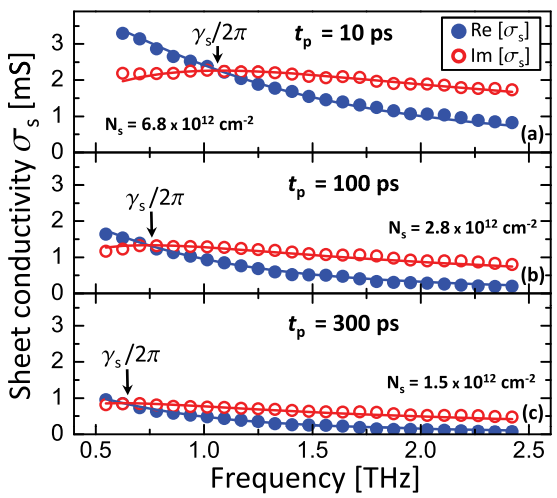

FIG. 1. Symbols—complex sheet conductivity spectra of photoexcited GaAs at pump fluence 13 μJ/cm^2 measured at selected pump-probe delays t_p of (a) 10 ps, (b) 100 ps, and (c) 300 ps. Lines—Drude fits. The arrows indicate the scattering rates γ_s, and the extracted electron sheet densities N_s (see Eq. (1)) are indicated in the figure.

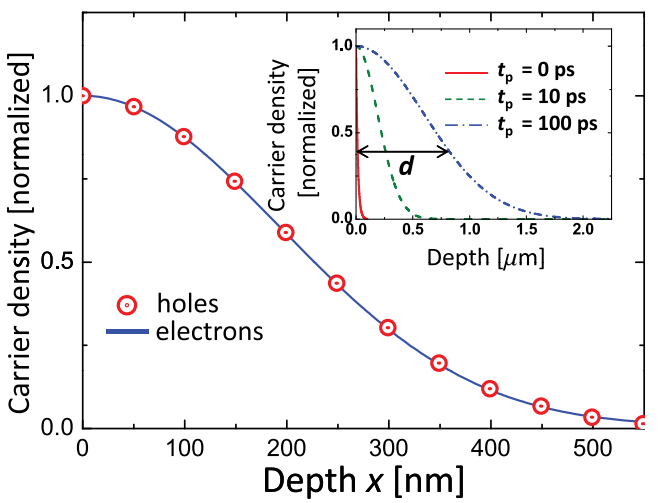

FIG. 2. Numerical solution (at $t_p = 10$ ps) of the drift-diffusion equation illustrating the ambipolar diffusion, independent of pump fluence. Inset: The profile of electron concentration at selected pump-probe delays, and the effective electron gas width d.

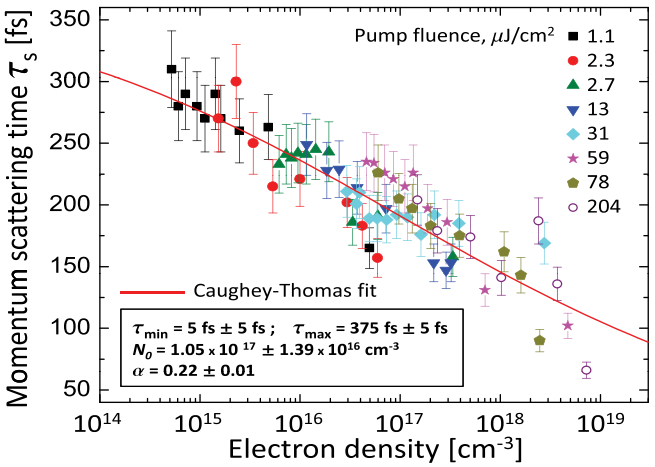

FIG. 3. Symbols—electron momentum scattering time as a function of electron density. The different colors correspond to the data from OPTP measurements at different pump fluences, as indicated in the figure. Solid line—Caughey-Thomas fit, with the parameters shown in the figure.

Now the (bulk) electron density at all times can be evaluated using the expression

$$N(t_p) = \frac{\sigma_s(t_p)}{d(t_p)} \frac{m^*}{e^2 \tau_s(t_p)}, \tag{4}$$

where $\sigma_s(t_p) = e^2 N_s(t_p)\tau_s(t_p)/m^*$ and $\tau_s(t_p)$ can be extracted from the Drude fits (see Eq. (1) and Fig. 1).

With the knowledge of the density $N(t_p)$, we are able to correlate the values of electron momentum scattering time $\tau_s(t_p)$ with the corresponding values of electron density $N(t_p)$ for *every* pump-probe delay t_p, and for *all* pump fluences used in the experiments. This yields the dependency of electron momentum scattering time in photoexcited GaAs on carrier density, shown in Fig. 3, which is the main result of our work. As one can see, τ_s decreases by a factor of 4, approximately from 320 fs down to 60 fs, as the electron density increases in the range of $5 \times 10^{14} - 8 \times 10^{18}\,\mathrm{cm}^{-3}$, i.e., over 4 orders of magnitude.

We emphasize the significant overlap between the datasets originating from different OPTP measurements made at different pump fluences, which are shown with different color-coding in Fig. 3. This overlap confirms the reliability of our analysis, in particular, in estimating the effective width of the electron-hole plasma as a function of pump-probe delay, and in excluding the effect of recombination on the carrier spatial profile. Further, it demonstrates that the electron momentum scattering rate in photoexcited GaAs solely depends on the carrier density, but not directly on the excitation fluence. Hence, one can exclude the influence of the phonons generated via electron cooling after 400 nm excitation, since this phonon population would be proportional to the excitation fluence. The observed increase in electron scattering rate with carrier density is most likely caused by electron-hole and/or defect and (equilibrium) phonon scattering, enhanced by the density-dependent phase-space filling effect.[4]

In order to simplify the use of our data in applications, e.g., in modeling of GaAs-based devices, we parameterize the measured dependency of electron momentum scattering time of the electron density $\tau_s(N)$ using an empirical Caughey-Thomas relation, originally formulated for the density-dependent mobility[20]

$$\tau_s = \frac{\tau_{max} - \tau_{min}}{1 + (N/N_0)^{\alpha}} + \tau_{min}, \tag{5}$$

where $\tau_{max,min}$, N_0, and α are the fit parameters. The Caughey-Thomas fit to the measured data and the best fit parameters are shown in Fig. 3.

In summary, we have demonstrated that the electron momentum scattering time in photoexcited GaAs decreases by a factor of 4, in the range of 320 – 60 fs, as the carrier density increases over 4 orders of magnitude, in the range of $5 \times 10^{14} - 8 \times 10^{18}\,\mathrm{cm}^{-3}$, independent of the excitation fluence. Accurate parameterization of the measured dependency with the Caughey-Thomas relation will allow for easy implementation of our findings in applications, such as transport simulations.

We acknowledge financial support from the International Max Planck Research School, Max Planck Society, EU Marie Curie Program (Career Integration Grant 334324 "LIGHTER"), Danish Council for Independent Research (FTP project ALFIE), and Danish Proof-of-Concept Foundation (Grant 7.7 "Ultrahigh-speed wireless data communications").

[1]M. Tonouchi, Nat. Photonics **1**, 97 (2007).
[2]R. Ulbricht, E. Hendry, J. Shan, T. F. Heinz, and M. Bonn, Rev. Mod. Phys. **83**, 543 (2011).
[3]P. U. Jepsen, D. G. Cooke, and M. Koch, Laser Photon. Rev. **5**, 124 (2011).
[4]E. Hendry, M. Koeberg, J. Pijpers, and M. Bonn, Phys. Rev. B **75**, 233202 (2007).
[5]P. U. Jepsen, R. H. Jacobsen, and S. R. Keiding, J. Opt. Soc. Am. B **13**, 2424 (1996).
[6]E. Castro-Camus, J. Lloyd-Hughes, and M. B. Johnston, Phys. Rev. B **71**, 195301 (2005).
[7]M. C. Hoffmann and D. Turchinovich, Appl. Phys. Lett. **96**, 151110 (2010).
[8]D. Turchinovich, J. M. Hvam, and M. C. Hoffmann, Phys. Rev. B **85**, 201304 (2012).
[9]M. C. Beard, G. M. Turner, and C. A. Schmuttenmaer, Phys. Rev. B **62**, 15764 (2000).
[10]T. Dekorsy, T. Pfeifer, W. Kutt, and H. Kurz, Phys. Rev. B **47**, 3842 (1993).
[11]G. Klatt, F. Hilser, W. Qiao, M. Beck, R. Gebs, A. Bartels, K. Huska, U. Lemmer, G. Bastian, M. B. Johnston, M. Fischer, J. Faist, and T. Dekorsy, Opt. Express **18**, 4939 (2010).
[12]K. P. H. Lui and F. A. Hegmann, Appl. Phys. Lett. **78**, 3478 (2001).
[13]F. Kadlec, H. Nemec, and P. Kuzel, Phys. Rev. B **70**, 125205 (2004).
[14]S. Zollner, J. Appl. Phys. **90**, 515 (2001).
[15]N. A. van Dantzig and P. C. M. Planken, Phys. Rev. B **59**, 1586 (1999).
[16]A. L. Mears and R. A. Stradling, J. Phys. C: Solid State Phys. **4**, L22 (1971).
[17]R. E. Glover and M. Tinkham, Phys. Rev. **108**, 243 (1957).
[18]S. M. Sze, *Semiconductor Devices, Physics and Technology* (Wiley, New York, 1985).
[19]B. A. Ruzicka, L. K. Werake, H. Samassekou, and H. Zhao, Appl. Phys. Lett. **97**, 262119 (2010).
[20]D. M. Caughey and R. E. Thomas, Proc. IEEE **55**, 2192 (1967).