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Electronic diffusivity measurement in silicon by photothermal microscopy

Benoît C. Forget,^{a)} Isabelle Barbereau, and Danièle Fournier
*Laboratoire d'Instrumentation, Université Pierre and Marie Curie, ESPCI, 10 rue Vauquelin,
75231 Paris Cedex 05, France*

Suneet Tuli and Amalendu B. Battacharyya
C.A.R.E., Indian Institute of Technology, Hauz Khas, New Delhi 110016, India

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In this letter we demonstrate that a photothermal microscopy experiment can be used to determine the electronic diffusivity (or carrier mobility) in the same way it is now widely used to measure locally thermal diffusivity of various nonsemiconductor materials. The main difficulty lies in the fact that in order to separate thermal and carrier diffusion, the experiment must be performed for a relatively large distance between the pump and probe beams. Photothermal signals are therefore rather weak and great experimental care must be taken. We present and discuss experimental results on Si. © 1996 American Institute of Physics. [S0003-6951(96)02034-7]

Photothermal microscopy is now a widely used technique for thermal diffusivity measurement on various materials.^{1,2} It has also been successfully used for investigations on semiconductor materials³⁻⁶ and devices.⁷⁻⁹ In the case of semiconductors, one of the main difficulties in concurrent determination of all the electronic transport properties (diffusivity, lifetime, and surface recombination velocity) is their correlated effect on photothermal signal.¹⁰ It is thus important to find experimental conditions for which one, or more, of these parameters can be determined unambiguously. In this letter we show that studying the phase of photoreflection signal far from the injection point allows such a measure for the electronic diffusivity.

After discussing the experimental setup, we will discuss briefly the free carrier diffusion equation. We will show that for our experimental conditions, this equation is simplified and its solution permits an easy and unambiguous determination of the electronic diffusivity. Our experimental results clearly exhibit a passage from a regime in which the interpretation of the photothermal signal is rather complex (meaning that one must consider heat diffusion as well as free carrier diffusion) to one where the electronic diffusivity measurement is unambiguous for a Si sample.

There are many techniques to measure the diffusivity (or mobility) of free carriers, the most widely used being: conductivity, Hall effect, magnetoresistance, and time of flight techniques.^{11,12} The main difference, and great advantage, of photothermal microscopy is that it requires no contact, neither electrical nor of any other type, with the sample.

The principle of the photothermal experiments is to heat a sample by the absorption of an intensity modulated light source (in our case an Ar⁺ laser beam) and to measure the temperature rise. In the case of photoreflection this is done *in situ* and without contact by reflecting a second laser (the probe laser is a 670 nm laser diode) on the surface and measuring through a lock-in amplifier (EG&G 5206 lock-in and HMD Electronik Dynatrac 511/531 heterodyne detector for high frequency measurements), the part of the reflected beam which is modulated at the same frequency as the excitation

beam. The Ar⁺ laser (or pump laser) is modulated by an acousto-optic modulator at frequencies ranging from 100 Hz to 10 MHz. The experiments presented in this letter were performed at modulation frequencies ranging from 500 kHz to 2 MHz. Both laser beams are focused through a microscope and thus the heating and the temperature measurement are done at the micron scale. In our setup the dichroic mirror is mounted on an orientable holder which can be positioned precisely with step motors.¹³ We will measure the photothermal signal as a function of the distance between pump and probe beams.

In the case of semiconductors, one must consider two important aspects. First, if the photon energy $h\nu$ of the pump laser is greater than the band gap E_g , electron-hole pairs will be generated by optical absorption. These free carriers will then diffuse in the bulk of the sample until they recombine, generally through a nonradiative process. Consequently, carrier dynamics play an important role in the conversion of photon energy into heat and therefore the surface temperature rise T , which modifies the reflectivity R , is dependent upon the electronic transport properties of the sample. Second, in a photoreflection experiment the free carrier density n , contributes directly to the reflectivity modulation through Drude effect. The relative variation of reflectivity is thus expressed as follows:

$$\frac{\Delta R}{R} = \frac{1}{R} \frac{\partial R}{\partial T} \Delta T + \frac{1}{R} \frac{\partial R}{\partial n} \Delta n. \quad (1)$$

In the case of silicon, the two contributions are easily distinguishable as they are of opposite sign. Therefore, under the modulated excitation used in our experiments the two contributions are 180° phase shifted. Studying the phase of the signal clearly indicates which of the two contributions is dominant.

In order to model the photoreflection signal, we must obtain the spatiotemporal distribution of the free carrier density and of the temperature rise. The diffusion equation for free carriers considering harmonic excitation of frequency $f = \omega/2\pi$ is the following:

$$\frac{\partial n}{\partial t} = j\omega n = D\nabla^2 n - \frac{n}{\tau} + \Phi, \quad (2)$$

^{a)}Electronic mail: forget@optique.espci.fr

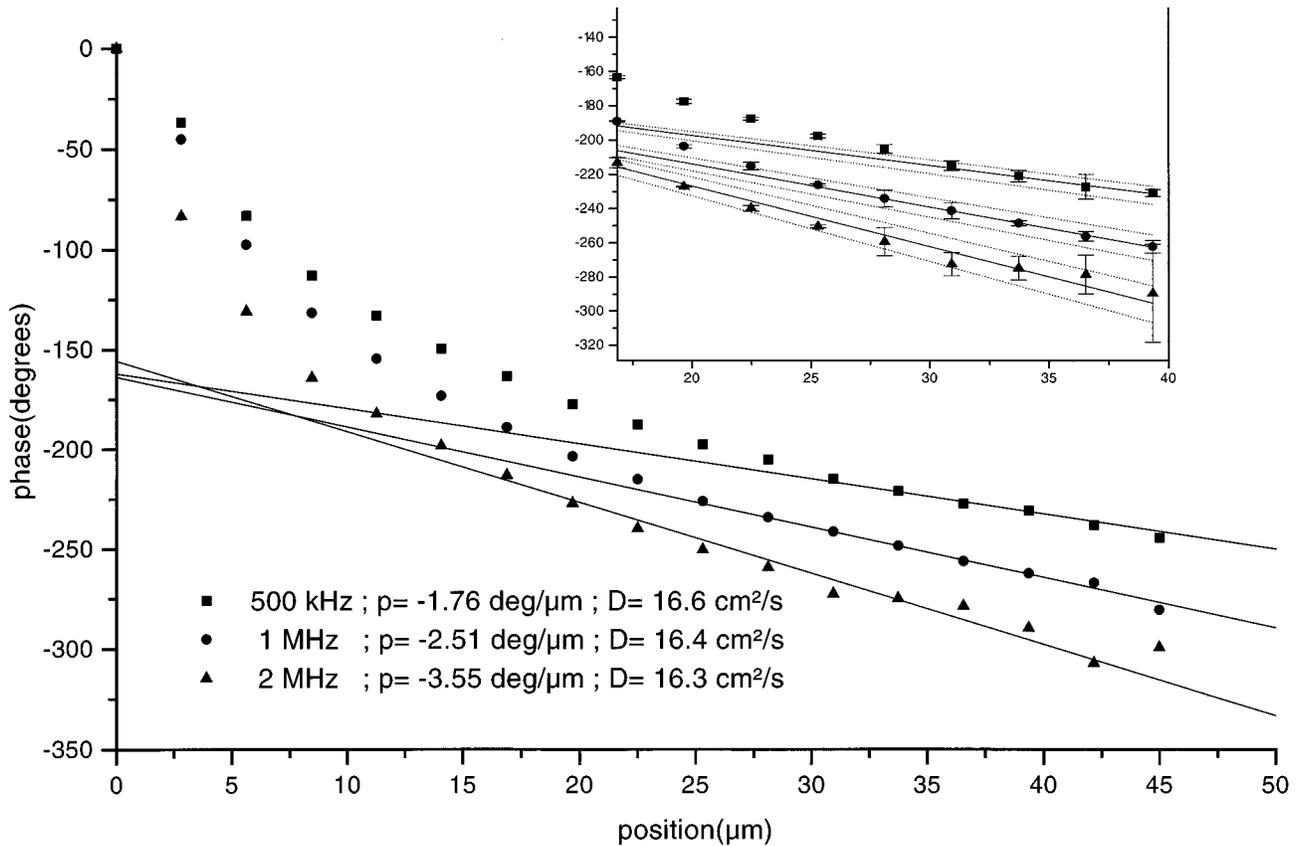


FIG. 1. Phase of the photoreflexion signal on Si as a function of the pump-probe distance for three modulation frequencies : 500 kHz, 1 MHz, and 2 MHz. The solid lines are the linear dependencies predicted by Eq. (6) and slopes, p , are fitted to calculate the diffusivity. The inset shows the same data with error bars. We have also plotted the linear dependencies for variations of the diffusivity of $\pm 15\%$ (dashed lines) at each frequency.

where D is the electronic diffusivity (related to mobility μ by Einstein's relation $D = kT_0/q\mu$ with k the Boltzmann constant, T_0 the ambient temperature, and q the electron charge), τ is the lifetime and Φ is a modulated source term which depends on the photon flux, the optical absorption, and the spatial distribution of the pump beam on the surface of the sample.

We must also consider the thermal diffusion equation, which depends on the free carrier density since nonradiative recombination is a heat source

$$\frac{\partial T}{\partial t} = D_T \nabla^2 T + \frac{E_g}{\rho C} \frac{n}{\tau} + \frac{h\nu - E_g}{\rho C} \Phi, \quad (3)$$

where D_T is the thermal diffusivity, ρ is the density, and C the specific heat. These equations can be solved (using Hankel transformation scheme to consider 3D diffusion in the sample) to obtain a complete solution which considers the finite size of the pump and probe beams, bulk and surface recombination, and optical absorption.¹⁴

We will make two remarks. First, the expression of the temperature rise is composed of two thermal waves: one due to the thermalization of the photon energy in excess of the band gap ($h\nu - E_g$) and one due the thermalization of the remaining energy (E_g) through the nonradiative recombination of free carriers. Second, since the thermal diffusivity is smaller than the electronic diffusivity, the thermal waves are attenuated faster than the plasma waves. Therefore even if the thermal contribution to the photoreflexion signal is

dominant for superimposed pump and probe beams ($r=0$), for sufficiently large pump-probe distances the thermal wave will be completely attenuated and the photoreflexion signal will be uniquely due to the free carriers, as our experimental results clearly show (see Fig. 1).

We have measured the photoreflexion signal on a 300 μm thick p -type Si wafer. We have scanned the position of the pump beam from side to side of the probe beam for a total distance of 90 μm and plotted the phase of the signal as a function of this distance for three modulation frequencies: 500 kHz, 1 MHz, and 2 MHz. The reciprocity theorem indicates that the signal obtained this way is the same as the one obtained if we had scanned the probe beam.¹⁵

For sufficiently large distances (roughly 30 μm at 500 kHz, 25 μm at 1 MHz, and 20 μm at 2 MHz) the phase is a linear function of the distance for all three experimental curves. As we move towards the center ($r=0$ μm) the phase departs from this linear behavior and tends towards 0° . This is due to the fact that we find ourselves in the situation discussed above, where the thermal contribution dominates the signal for superimposed beams. If the free carrier contribution had dominated the signal at $r=0$ μm the phase would have been near -180° as one can see if the linear behavior is prolonged to $r=0$. This 180° phase shift between the two contributions is due to the opposite signs of the plasma and thermal factors in Eq. (1).

We will now obtain a simplified expression to account for the linear dependence of the phase for large pump-probe

distances. As discussed above, we need to solve only the carrier diffusion Eq. (2). We will also take into account two more experimental conditions. First we will neglect the convolution effects due to the finite size of the pump and probe beams (which in our case have a diameter of roughly 1 μm thus much smaller than the pump-probe distance) and consider a spherical plasma wave generated by a point source. We will thus rewrite expression (2) as

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial n}{\partial r} \right) = \left(\frac{1 + j\omega\tau}{D\tau} \right) n. \quad (4)$$

We will furthermore, consider high modulation frequencies such as $\omega\tau \gg 1$, thus the plasma wave vector defined as $\zeta \equiv \sqrt{1 + j\omega\tau/D\tau}$ becomes $\zeta \equiv \sqrt{j\omega/D}$. Considering a lifetime of the order of magnitude of 50 μs ,¹⁶ the range of modulation frequencies used (500 kHz–2 MHz) satisfies this condition. The outgoing spherical wave is expressed as¹⁷

$$n(r) = \frac{A}{r} e^{-\zeta r} = \frac{A}{r} e^{-r\sqrt{\frac{\omega}{2D}} - jr\sqrt{\frac{\omega}{2D}}}, \quad (5)$$

where A is determined by boundary conditions, and since our measurement method will use the phase of the signal as a function of r , we will not detail its expression here. The phase of the free carrier density is a linear function of r

$$\arg[n(r)] = -\sqrt{\frac{\omega}{2D}} r. \quad (6)$$

Therefore, in our experimental conditions, measuring the slope of this phase at a fixed modulation frequency permits the unambiguous determination of the electronic diffusivity as it depends solely on this diffusivity. The experimental data validate this simplified model. All three measurements are consistent with the value for ambipolar diffusivity in Si found in literature.^{18,19}

The inset in Fig. 1 shows the phase of experimental data with error bars. The phase is plotted against theoretical curves for three values of the electronic diffusivity at each modulation frequency, in order to evaluate the precision of the estimated value of D . We conclude that the electronic diffusivity is $16.5 \pm 2.5 \text{ cm}^2/\text{s}$ ($\pm 15\%$).

We must make two remarks concerning the precision. First one should remember that the diffusivity is proportional to the square of the slope of the phase as a function of the pump-probe distance. Therefore, the relative error on the diffusivity is twice that of the slope measurement. Second, this slope must be measured for large pump-probe distances in order to ensure that the photo reflection consists of only the free carriers' contribution. Increasing modulation frequency favors the free carrier contribution over the thermal one¹⁰ and our experimental results confirm this as the free carrier contribution is reached for a shorter distance at 2 MHz than at 500 kHz (20 μm compared to 30 μm). On the other hand, the amplitude of the signal decays exponentially with the square root of the modulation frequency, therefore high frequency measurements are more subject to noise. In any case, for such distances the signal to noise ratio is rather weak and thus averaging must be used to improve this precision, which

is already comparable to that obtained for thermal diffusivity measurement using other microscopic photothermal techniques.^{20,21}

In conclusion, we have shown that the photothermal microscopy can be used to determine unambiguously and without contact the electronic diffusivity of semiconductor materials. The main advantage of our experimental scheme is that the slope of the phase depends solely on the electronic diffusivity: the concurrent determination of all electronic transport properties (diffusivity, lifetime, and surface recombination velocity) using complementary photothermal methods (mirage detection and photoreflexion) is therefore facilitated. Furthermore, our measurement is done at a microscopic scale, permitting diffusivity measurement in electronic structures or devices of small dimensions. The measurement through the slope of the photoreflexion signal's phase must be used carefully. One must insure first that the slope measurement is done at pump-probe distances for which it is certain that the signal is dominated by the free carrier contribution; second that the point source approximation can be used; and finally that the modulation period ($1/f$) is much shorter than the free carrier lifetime. All these considerations result in the need to perform experiments for relatively large distances between the pump and probe beams and thus for rather weak photoreflexion signals.

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