

# Highly resolved separation of carrier- and thermal-wave contributions to photothermal signals from Cr-doped silicon using rate-window infrared radiometry

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It is shown that the new photothermal technique of lock-in rate-window infrared radiometry is capable of completely separating out photoexcited free-carrier-wave and thermal-wave contributions to the photothermal signal from an *n*-type, Cr-doped Si wafer with a simple experimental procedure, and with superior temporal resolution in the determination of the electronic lifetime and thermal transport time constant.

## I. INTRODUCTION

For some time now several laser-based photothermal techniques have been developed to monitor photoexcited carrier kinetics and transport in semiconductors, the advantage over other, mainly electrical, methods being that electronic effects can thus be monitored in a noncontacting and nondestructive manner, therefore eliminating the need for electrode attachment.<sup>1-5</sup> A distinct disadvantage of those photothermal techniques, however, is the fact that with either frequency-scanned<sup>1,4,6</sup> or with time-resolved<sup>7</sup> detection, both free-carrier (plasma) -wave and thermal-wave responses from semiconductors are strongly coupled together. As a result the interpretation of the convoluted experimental data is usually complicated. The task of deconvoluting the two types of responses becomes cumbersome, and this renders much of the analysis qualitative. As an example, the thermoreflectance technique<sup>1</sup> produces signals  $\Delta R$  which depend on both the ac temperature of the laser-excited semiconductor surface  $\Delta T(\omega)$ , and on the photogenerated electron-hole plasma wave  $\Delta N(\omega)$ ,

$$\Delta R(\omega) = \left( \frac{\partial R}{\partial T} \right) \Delta T(\omega) + \left( \frac{\partial R}{\partial N} \right) \Delta N(\omega). \quad (1)$$

Very tightly focused ( $\sim 1 \mu\text{m}^2$ ) pump beams can, in principle, lead to the domination of  $\Delta R$  by the plasma response,<sup>1</sup> yet this constraint results in the necessity for quite complicated three-dimensional mathematical modeling. Furthermore, it can be detrimental to the study of electronic defects, since the exceedingly high local laser fluences may greatly perturb experiments by saturating trap centers or causing enhanced thermal emission of trapped carriers.

Similarly, pulsed (i.e., impulse response) photothermal radiometry exhibits severe overlap of free-carrier density and thermal effects.<sup>7</sup> In terms of physical interpretation of signals, the latter technique is considered preferable

to the respective frequency-domain (i.e., transfer-function) detection<sup>5,6</sup> due to the inherent ability of transient-response techniques to be interpretable in terms of simple system time-delay constants. The same information can be obtained, in principle, from the frequency-scanned data; however, this method requires the demultiplexing of data over broad frequency ranges, typical of the existing relationship between Fourier transform pairs (i.e., time and frequency domains).

Recently we introduced the lock-in rate-window photothermal instrumentation and detection scheme,<sup>8</sup> which combines the extremely high dynamic reserve of frequency-domain narrow-band detection techniques with the simplicity of interpretation of time-resolved photothermal signals in terms of response delay times. When used with semiconductors, the pulsed radiometric signal may contain both carrier recombination and thermal conduction components.

In Ref. 9 we showed that lock-in rate-window pulsed photothermal radiometry (RW PTR) of deep-impurity-doped Si can yield excess photoexcited carrier bulk lifetimes with optimal signal-to-noise (S/N) ratio, from the simple, unambiguous interpretation of the maximum position of the rate-window signal. We also showed that measured lifetimes are mostly in good agreement with results obtained from the noncontact laser/microwave reflection detection method.<sup>10</sup> In this article we applied the lock-in RW PTR to Cr-doped *n*-type Si, with preoxidized and etched high-quality surface, a sample which exhibits strongly overlapping carrier recombination and thermal conduction transient behavior (Fig. 1). By choosing appropriate pulse durations (i.e., acousto-optically time gating a cw laser beam) for the first time we have been able to separate the two transient contributions completely and measure the respective delay-time constants with superior resolution to both frequency- and time-domain conventional methodologies. This ability of RW PTR enormously simplifies the analytical complexity of conventional

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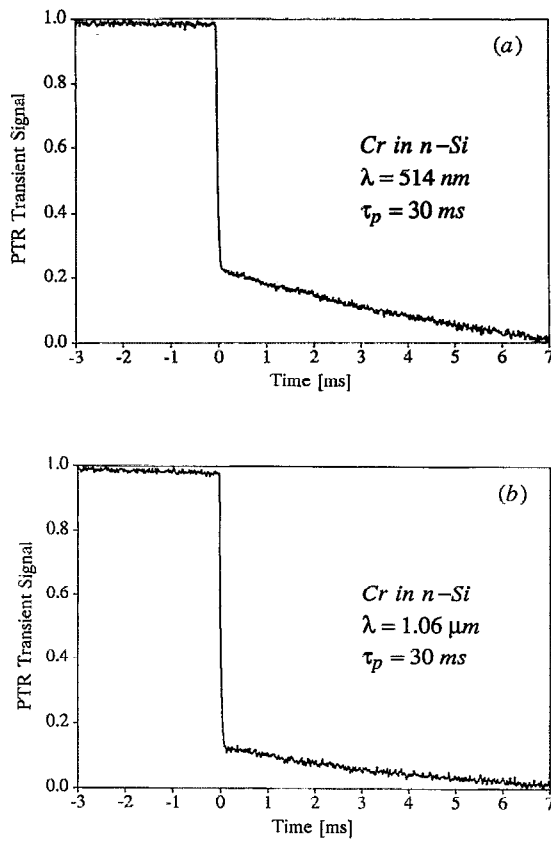


FIG. 1. PTR transient response of a Cr-doped  $n$ -type Si wafer ( $1.7 \times 10^{13}$  Cr atoms/cm<sup>2</sup>; 5–13  $\Omega$  cm) to (a) an Ar-ion laser "pulse;" and (b) a Nd-YAG laser "pulse." Pulse duration  $\tau_p = 30$  ms.

photothermal-based semiconductor diagnostic techniques, such as thermoreflectance<sup>1</sup> and frequency-domain PTR.<sup>6</sup>

## II. EXPERIMENTAL RESULTS AND DISCUSSION

The experimental apparatus has been described previously.<sup>9,11</sup> Computer software was generated which enabled the automatic scanning of the repetition period  $T_0$  of the optical pulse, while keeping the pulse duration  $\tau_p$  fixed at a predetermined value which, in turn, depended on the photothermal decay process being monitored. The two laser wavelengths chosen, 514 nm and 1.06  $\mu$ m, were used to probe the surface ( $\sim 0.5$   $\mu$ m) and the bulk ( $\sim 1$  mm) of the Si wafer, respectively.<sup>12</sup>

The infrared radiation from the sample was collected by two off-axis paraboloidal mirrors and detected using a liquid-N<sub>2</sub>-cooled mercury-cadmium telluride detector. Two experimental runs were performed at each wavelength, one with  $\tau_p = 30$   $\mu$ s and a second run with  $\tau_p = 1$  ms. The temperature rises were estimated to be less than 1 K in both cases. Figure 2 shows the lock-in RW-PTR in-phase signal as a function of the laser time-gated square pulse repetition period  $T_0$ , resulting from a pulse of duration  $\tau_p = 30$   $\mu$ s. The solid lines are the theoretical simulations using the expression for the fundamental Fourier component  $b_1$  of the repetitive transient PTR signal  $S_{IR}(t; \tau)$  over the period  $T_0$ , using the fact that the fast decay in Fig. 1 is a purely exponential function:<sup>9</sup>

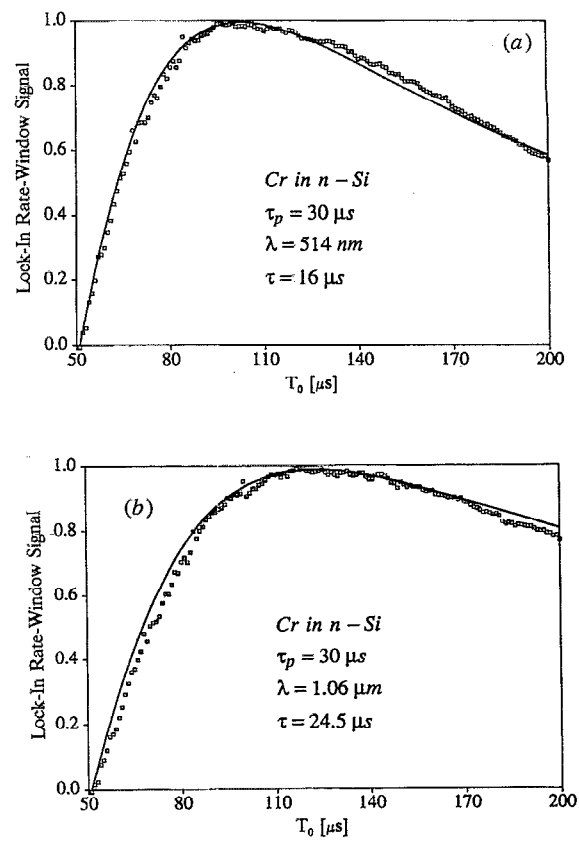


FIG. 2. Lock-in rate-window photothermal radiometric response to a repetitive square laser pulse of duration  $\tau_p = 30$   $\mu$ s. Squares: data; lines: theory, Eq. (2). (a) 514 nm photon excitation of Cr-doped  $n$ -type Si wafer; (b) 1.06  $\mu$ m photon excitation.

$$S_{IR}(t; \tau) = C_1 \tau \begin{cases} 1 - e^{-t/\tau}, & t \leq \tau_p \\ (e^{\tau_p/\tau} - 1) e^{-t/\tau}, & t > \tau_p \end{cases} \quad (2)$$

and

$$b_1(\tau; T_0) = C_2 \tau \left( 1 - \cos(\omega_0 \tau_p) + \frac{\omega_0}{\sqrt{\tau^{-2} + \omega_0^2}} [\sin(\omega_0 \tau_p + \theta) - (1 + e^{-(T_0 - \tau_p)/\tau} - e^{-T_0/\tau}) \sin \theta] \right), \quad (3)$$

where

$$\theta = \tan^{-1}(\omega_0 \tau), \quad \omega_0 = 2\pi/T_0, \quad (4)$$

$C_1$  and  $C_2$  are constants independent of any photoexcited carrier characteristic time constants, and  $\tau$  is the carrier bulk recombination lifetime.

The PTR response, Eq. (2), assumes complete domination of the PTR signal by the photoexcited free-carrier density kinetics, when the carrier surface recombination velocity is very low. The excellent fits of the theory to the data, normalized to unity for convenience, prove the free-carrier interband recombination origin of the signal. No actual transient traces could be obtained on an oscilloscope for this short  $\tau_p$ , because the extremely small PTR signal could not be made to rise above the instrumental noise level, even after averaging several thousand transients.<sup>11</sup> It

is important to note that the high sensitivity of the theoretical curve to  $\tau$  in Figs. 2(a) and 2(b) renders the fit to the experimental data shown such that only true single-exponential recombination decays can yield the detailed agreement shown in Fig. 2, provided the PTR response is linear with the incident radiation intensity (which was the case with all our data). Any deviations from exponential decay manifest themselves as an inability to fit the entire data curve to a single exponential function, Eqs. (2)–(4). The analytical advantage of scanned RW PTR over conventional PTR impulse responses (even when the latter can be of a high enough S/N ratio to give reliable slopes) is the pronounced intolerance of the former measurement technique to deviations from the assumed decay law, compared to a relatively forgiving mild nonconstancy of the transient slope. Thus, Figs. 2(a) and 2(b) may also be viewed as testing criteria for pure exponential decays in semiconductors. A slight deviation of 4%–6% in slope constancy, well within the averaged transient S/N ratio in high-quality *n*-type Si substrates where the transient was directly measurable,<sup>9</sup> produced RW-PTR scans of the type shown in Fig. 2. These transients when fitted theoretically using Eqs. (2)–(4) exhibited significant deviations either before or after the maximum, in the range of 13%–20%. The most likely deviations from semiconductor recombination purely exponential decay behavior are known to be due to high-surface-recombination velocities.<sup>13,14</sup> Such a surface recombination velocity contribution to our RW-PTR signal decay was highly unlikely in our case, due to the high quality of the preoxidized and etched surface. Furthermore, in the case of nonexponential decays, adjusting  $\tau$  in the theoretical curve of Fig. 2 cannot produce an acceptable fit throughout the entire  $T_0$  range shown, in which case a more complete model involving surface recombination effects must be used,<sup>9</sup> and the experimenter is thus most easily alerted to the nonexponential character of the decay. On the other hand, if the RW time scale chosen for an experiment does not include the recombination time constant, no extremum in the RW-PTR response will be observed. Usually the lock-in RW  $T_0$  is set at about one order of magnitude greater than the range of characteristic recombination time constants in a given type of semiconductor ( $\tau \lesssim 20 \mu\text{s}$  for deep-level doped silicon<sup>9</sup>). In earlier work<sup>11</sup> we demonstrated theoretically and in practice that the S/N ratio of the lock-in rate-window filter is higher than that obtained by a boxcar rate-window averager at the extremum typically by a factor of 2. The S/N ratio of the boxcar averager approaches monotonically from below that of the lock-in rate window as the gate width of the boxcar increases toward  $T_0/2$ .

The difference in lifetime  $\tau$  between the results obtained with the Ar and Nd-YAG laser, Figs. 2(a) and 2(b), can be explained by the widely different optical penetration depths of the two excitation pulses. The 514 nm pulse probes a region very close to the surface, in which near-surface defects can provide an additional free-carrier recombination channel, thus shortening the effective lifetime.<sup>14</sup> On the other hand, the deeply penetrating 1.06  $\mu\text{m}$  pulse is expected to give a better measurement of the

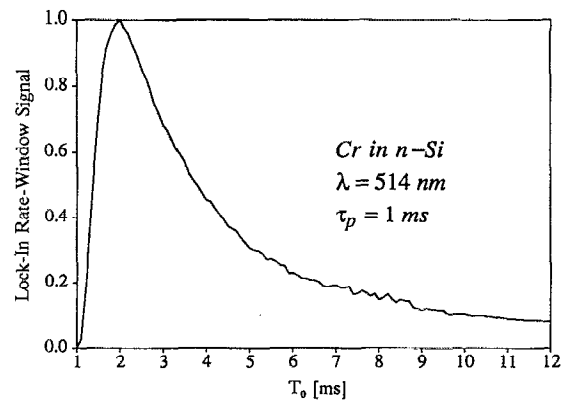


FIG. 3. Lock-in rate-window photothermal radiometric response of Cr-doped Si wafer to a repetitive square Ar-laser pulse of duration  $\tau_p = 1$  ms.

true value of the bulk recombination lifetime. In this case near-surface recombination is much less significant in its contribution to the effective lifetime, and therefore the effective lifetime is longer and characteristic of bulk processes.

When the pulse duration was increased to  $\tau_p = 1$  ms, the lock-in RW-PTR response of the Cr-doped silicon wafer to the Ar laser excitation is shown in Fig. 3. This response, too, is normalized to unity for convenience. A very similar curve was obtained with Nd-YAG laser excitation. The maximum of the in-phase signal at about 2 ms from the onset of the pulse is in good agreement with domination of the infrared radiometric signal by a thermal transient, which in the 500- $\mu\text{m}$ -thick free-standing Si wafer requires a round-trip time<sup>11</sup>

$$t_{rt} \approx L^2 \pi / 4 \alpha_s \approx 1.96 \text{ ms.} \quad (5)$$

The fact that signals generated with Nd-YAG laser excitation exhibit similar in-phase rate-window maxima is indicative of the much higher carrier-diffusion coefficient<sup>15</sup>  $D$  than the thermal diffusivity  $\alpha_s$ . Photoexcited carrier recombination and thermal energy release follows rapid electronic transport through the bulk of the wafer, so that for both laser pumps similar distributions of free carriers recombining across the thickness  $L$  may be expected by the time the 1 ms pulse is switched off, irrespective of the widely different original optical penetration depths. A complete theoretical description of the combined optical and thermal transport effects in time-gated laser-pulse photoexcited semiconductors is currently under investigation.

### III. SUMMARY

The comparison between Figs. 2 and 3 shows the ability of RW photothermal detection to completely separate out electronic and thermal transport contributions to overlapping PTR signals from a Cr-doped Si wafer. This can be done with excellent temporal resolution, by a simple change in  $\tau_p$ . Similar results have been observed with Fe-doped Si wafers as well. It is worth mentioning that the

completely separate, highly resolved results shown in Figs. 2 and 3 cannot be obtained either with frequency-scanned PTR detection, because of the time-multiplexed nature of this methodology, or with pulsed laser excitation, due to the fixed (i.e., noncontinuously variable) repetition rate and pulse duration of today's laser technologies.

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