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All optical method for investigation of spin and charge transport in semiconductors: Combination of spatially and time-resolved luminescence

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A new approach is demonstrated for investigating charge and spin diffusion as well as surface and bulk recombination in unpassivated doped semiconductors. This approach consists in using two complementary, conceptually related, techniques, which are time-resolved photoluminescence (TRPL) and spatially resolved microphotoluminescence (μPL) and is applied here to p⁺ GaAs. Analysis of the sole TRPL signal is limited by the finite risetime. On the other hand, it is shown that joint TRPL and μPL can be used to determine the diffusion constant, the bulk recombination time, and the spin relaxation time. As an illustration, the temperature variation of these quantities is investigated for p⁺ GaAs.

I. INTRODUCTION

Determination of diffusion constants and characterization of surface and bulk recombination of semiconductors is of importance for the designing of devices, such as photovoltaic, microelectronic, and spintronics systems. The diffusion constants are usually obtained from the values of minority carrier mobilities.1–3 The charge and spin diffusion constants can also be determined using spin grating experiments.4 In spite of their strong interest, these experiments are relatively elaborate since they involve time-resolved four-wave mixing. For determination of the recombination dynamics, time-resolved photoluminescence (TRPL),5 cathodoluminescence,6,7 or conductivity8 have been used. Independent theoretical analysis9,10 has shown the strong potential of time-resolved techniques for investigating bulk and surface recombination, and for the determination of diffusion constants using analysis of the various exponential modes in the transient, for each of which the characteristic time and amplitude can, in principle, be determined independently. Such analysis has only been performed for silicon.8 Experiments on p⁺ GaAs have investigated bulk11 and to some extent surface12 recombination kinetics, as well as spin and charge lifetimes.13–15

Both time and spatially resolved luminescence has also been used for investigating minority carrier and transport in surface free n-type GaAs.16

In the present work, it is first shown, in the case of doped unpassivated GaAs, that analysis of the charge and spin transients can be performed using an inverse Laplace transform method, which gives well-defined temporal modes. However, the finite TRPL rise time only enables to observe the first two modes, while their amplitudes are difficult to determine. On the other hand, the joint use of TRPL and of spatially resolved microphotoluminescence (μPL) enables to determine the charge and spin lifetimes as well as the charge and spin diffusion constants. The μPL technique, which consists in imaging the photoluminescence profile using a local CW laser excitation,17 is formerly very similar to TRPL since the spatial concentration profile also exhibits modes. This method is applied to investigation of the temperature dependences of diffusion constant, lifetime, spin relaxation time for p⁺ GaAs, for which the interest for spintronics arises from the strong electronic polarization under circularly polarized excitation. Section II presents a description of the method with an illustration at low temperature and shows the conceptual analogies of the two techniques. Section III presents a discussion of the limits of the sole TRPL and an interpretation of the joint TRPL and μPL results. In Sec. IV, the method is applied to investigation of temperature effects and spectrally dependent dynamic characteristics.

II. DESCRIPTION OF THE METHOD

A. Experimental

We have investigated a thin GaAs film (acceptor concentration \( N_A = 10^{18} \text{ cm}^{-3} \), thickness \( d = 3 \mu\text{m} \)). This structure is grown on a GaInP layer, which is itself grown on a semi-insulating substrate which does not contribute to the luminescence. The thin GaInP layer confines the photogenerated carriers and also strongly reduces the recombination at the back film interface. The present section is devoted to results obtained at a temperature near 15 K, while the results as a function of \( T \) will be explained in Sec. IV.

For TRPL, as described in Ref. 18, the excitation source was a circularly polarized mode-locked frequency-doubled Ti:Sa laser (1.5 ps pulse width, 80 MHz repetition frequency, wavelength 780 nm) and the emitted light was dispersed by a spectrometer (resolution 0.12 nm) and detected by a streak camera. The average power was adjusted between 5 and 500 μW and the diameter of the excitation spot was 50 μm that is, much larger than the diffusion length, so that light excitation was spatially homogeneous. The spin-related

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difference signal $I_d(t) = I_+(t) - I_-(t)$, defined as the difference between the $\sigma^+$- and $\sigma^-$-polarized components of the transient luminescence signal was also monitored. Figure 1 shows the sum and difference transients obtained at 20 K normalized to unity at their maximum as well as, for comparison, the time response of the streak camera. These transients reveal a slow increase, in a time of the order of 70 ps, an exponential signal between 100 ps and 300 ps, and will be interpreted in Sec. III B.

For µPL, as described in Ref. 17 and illustrated in the left panel of Fig. 2, the sample was excited by a CW tightly focused light (Gaussian radius $\sigma \approx 0.6 \mu m$, energy 1.59 eV) in a modified commercial optical microscope. An image of the luminescence was taken using a CCD camera and a small light excitation power (several $\mu W$) in order to keep a photoelectron concentration smaller than $10^{16} \text{cm}^{-3}$ and to avoid effects of ambipolar diffusion19 and of Pauli blockade.20 Such image, at a temperature of 15 K, is shown in the top right panel of Fig. 2. The bottom right panel shows an angular-integrated section of this image in the same conditions as Fig. 1, together with a fit and the cross section of the laser profile.

B. Theory

For the two techniques, the luminescence intensity is given by

$$I = C \int_0^d e^{-z \gamma} n(r, z, t) dz,$$

(1)

where $C$ is a constant. Here, $t$ is the time and, as shown in the left panel of Fig. 2, $r$ is the distance to the excitation spot and $z$ is the depth. This equation allows for the possibility of reabsorption of the luminescence with a coefficient $\gamma$. The photoelectron concentration $n(r, z, t)$ is a solution of the following diffusion equation:

$$\frac{\partial n(r, z, t)}{\partial t} = g(r, z, t) - \frac{n(r, z, t)}{\tau} + D \Delta n(r, z, t),$$

(2)

where $D$ is the charge diffusion constant, $\tau$ is the bulk lifetime, and $\Delta$ is the Laplacian operator. Here, $g(r, z, t)$ is the number of thermalized electrons created in the conduction band per unit volume and per unit time and takes into account the possible spatial diffusion of hot electrons during thermalization. For µPL, one has $g(r, z, t) = g_\mu(r, z)$ so that, in Eq. (1), $\partial n(r, z, t)/\partial t = 0$. For TRPL, $g(r, z, t) = g_{TR}(z, t)$ so that $n(r, z, t)$ does not depend on $r$. Equation (2) is solved in Appendix by imposing the Robin boundary conditions $\partial n/\partial z|_{z=0} = \gamma n/d$ and $\partial n/\partial z|_{z=d} = -\gamma' n/d$, where the dimensionless parameters $\gamma$ and $\gamma'$ are related to the recombination velocities $S$ and $S'$ of the front and back surfaces, respectively, by

$$\gamma = \frac{Sd}{D}, \quad \gamma' = \frac{S'd}{D}.$$  

(3)
For TRPL, the difference signal \( I_{\text{TRPL}}(t) \) is also given by Eq. (1) provided the charge concentration \( n \) is replaced by the spin concentration \( s = n_+ - n_- \), where \( n_\pm \) is the concentration of electrons of spin \( \pm 1/2 \), with a quantization axis along the direction of light excitation. The quantity \( C \) is replaced by \( C|P| \) where, for cubic semiconductors, the initial polarization \( P_i \) is equal to \( \pm 0.5 \) for \( \sigma^\pm \)-polarized light excitation. The time evolution of \( s \) is given by an equation similar to Eq. (2) where \( g_{\text{TRPL}}(z, t) \) is replaced by \( g_{\text{TRPL}}(z, t)P \), and \( \tau \) is replaced by the spin lifetime \( \tau_s = (1/\tau + 1/T_1)^{-1} \), where \( T_1 \) is the spin relaxation time. The spin diffusion constant \( D_s \) can be smaller than \( D \) if spin drag is present.\(^{21} \) Within these changes, Eqs. (7), (A6), and (A3) also give the expression for \( I_T(t) \), as a function of the characteristic times \( \tau_m \) of the various modes for the decay of \( s \).

1. TRPL signal

For a time-dependent, spatially homogeneous, light excitation, the concentration is expressed as

\[
n(z, t) = \sum_m a_m u_m(z) e^{-t/\tau_m},
\]

where \( a_m \) are functions of time and \( u_m(z) \) are the orthogonal spatial eigenfunctions of the Laplacian operator, satisfying the above boundary conditions, and given by Eq. (A1).\(^5 \)\(^{22} \) The characteristic times \( \tau_m \) of the various modes are given by

\[
1/\tau_m = 1/\tau + D_0^2 \mu_m^2 / d^2,
\]

where the angle \( \theta_m \) is given by the nonlinear equation

\[
\theta_m = \arctan(\gamma / \theta_m) + \arctan(\gamma' / \theta_m) + (m - 1)\pi.
\]

The final expression of the luminescence intensity, found using Eq. (1), is

\[
I_{\text{TRPL}}(t) = C \sum_m c_m e^{-t/\tau_m},
\]

where \( c_m \) are functions of time, given by Eq. (A3). In the simplified case where the buildup of the thermalized electron population is instantaneous, one has \( g_{\text{TRPL}}(z, t) \propto \delta(t) e^{-xz} \), where \( x \) is the absorption coefficient. The functions \( c_m \) are constants, \( c_m = n_0 c^0_m \) where \( n_0 \) is proportional to the excitation power and \( c^0_m \), given by Eq. (A4), only depends on \( \gamma \) and \( \gamma' \). Assuming that \( \gamma' = 0 \), the dependences of \( c^0_m \) on \( \gamma \) are illustrated in Fig. 3, neglecting photoluminescence reabsorption (\( \gamma_0 = 0 \)). For a passivated surface (\( \gamma = 0 \) which gives \( c^0_m = 1 \)), only one mode is present in the transient, of time equal to \( \tau \). If on the other hand \( S \) is not negligible, determination of \( c^0_m \) gives \( \gamma \) and \( \theta_m \) using Eq. (6). Provided more than one mode is observed, these angles, together with the measured values of \( \tau_m \), give \( D \) using Eq. (5) from which \( \tau \) and \( S \) are obtained.\(^{23} \)

2. \( \muPL \) profile

For \( \muPL \), the spatially inhomogeneous rate of creation of thermalized electrons is approximated by \( g_r(r, z) \propto x^* e^{-x^* z}; e^{-(r/r^*)^2} \), where \( \sigma^* \) and \( x^* \) can differ from the Gaussian radius \( \sigma \) of the laser spot and from the absorption coefficient \( x^* \), respectively, because of hot electron diffusion. In analogy with Eq. (7) and, as shown in Appendix, the luminescence profile can be expressed as the sum of spatial modes as given by

\[
I_{\muPL}(r) \propto \sum_m c^0_m \int_0^\infty K_0 \left( \frac{r'}{\tau_m} \right) e^{-(r-r')^2/\sigma^2} dr'.
\]

The spatial mode of order \( m \) is proportional to the modified Bessel function of the second kind \( K_0 \) convoluted with the effective excitation profile and multiplied by the same amplitude \( c^0_m \) as for TRPL. The mode of order \( m \) decays as a function of \( r \) with a characteristic distance

\[
L_m = \sqrt{D \tau_m},
\]

which decreases with \( m \). Modal analysis of the experimental spatial profile in principle gives the values of \( c^0_m \) and \( L_m \). However, such analysis requires a precise knowledge of \( \sigma^* \) and is generally not possible as soon as the kinetic energy of electrons at creation becomes significant.

As shown in the Appendix, the diffusion equation also has an analytical solution given by Eq. (A8), obtained using a treatment similar to Ref. 24. This solution is expressed as a function of \( S, S' \), and of the bulk diffusion length \( L = \sqrt{D \tau} \). For the calculation of the profile, it is possible to take \( S = S' = 0 \) and \( L = L_1 \), which is equivalent to replacing in Eq. (8) all \( L_m \) by \( L_1 \). This implies that the profile essentially depends on one parameter, \( L_1 \).

III. INTERPRETATION

A. Analysis of the sole TRPL transient

A convenient method for the analysis of the decays in Fig. 1 is to choose as a time origin the time \( t_0 \) of the signal maximum and to perform an inverse Laplace transform, using a numerical resolution of the following 1D inverse Fredholm integral of the first kind
where $J(t)$ is the searched distribution of time constants, $e(t)$ represents the noise. This algorithm is based on a compression of the data obtained with the singular value decomposition of the exponential kernel and a robust optimization of the data obtained with the singular value decomposition.

The new Laplace transform is known as an ill-posed problem, the very good signal-to-noise ratio of the experimental data gives two well-defined peaks such that

$$I_{TR}(t-t_0) = c_1^* e^{-(t-t_0)/\tau_1} + (1 - c_1^*) e^{-(t-t_0)/\tau_2},$$

and

where $\tau_1 = 256$ ps, $\tau_2 = 66$ ps, and $c_1^* = 0.82$. In the same way, the difference signal is described by $I_{TR}(t-t_0) = c_s^* e^{-(t-t_0)/\tau_s} + (1 - c_s^*) e^{-(t-t_0)/\tau_s}$, where $\tau_s = 215$ ps, $c_s^* = 0.725$, and $t_0 = 140$ ps. Curves d and e of Fig. 1 show the individual exponential modes. The data are very well approximated by their sums (curves c).

The two successive exponential decays of Eq. (11) are attributed to the first two modes defined in Eq. (7). Other interpretations implying ambipolar diffusion, Aufer recombination at high density, change of recombination time caused by temperature dependence of the bimolecular recombination, or stimulated emission by the photoexcited carriers, can be ruled out since the transient stays exactly the same if the excitation power is decreased by two orders of magnitude. The result is shown as a dotted line in the bottom right panel of Fig. 2 and very well approximates the data. It is concluded that combination of TRPL and $\mu$PL allows us to determine the key parameters for recombination and transport.

Investigation of the spin dynamics is performed by calculating the transient of the spin polarization $P = (n_+ - n_-)/(n_+ + n_-)$, defined as the ratio of the difference and sum transients. This transient is shown in Fig. 4, taking the time origin at $t_0 = 140$ ps in order to avoid the effects of the slow signal increase. As seen in the inset, the fast tail has almost completely disappeared and the transient is essentially described by a single exponential of characteristic time $T_1 \approx 1150$ ps. Using Eq. (7) and the corresponding equation for spin orientation, one can show that this result implies that $D_s \approx D$ so that, in Eq. (6), $\theta_1 = \theta_1s$, and $\theta_2 = \theta_2s$. The transients at short time calculated using Eq. (7) for several values of the ratio $D_s/D$ are shown in the main figure along with the data. From the good agreement obtained for $D_s/D$ near unity, we conclude that $D_s/D = 1 \pm 0.1$, in agreement with Ref. 21 for a similar doping. It is concluded that, if present, spin drag only marginally affects spin transport.

**B. Combined TRPL and $\mu$PL Investigation**

As shown in the Appendix, the higher order spatial modes do not affect the $\mu$PL signal for distances larger than $2 \mu$m. Thus, analysis of the profile at longer distances can be restricted to the lowest order mode of Eq. (8). This gives $L_1 = 1.33 \mu$m from which we find $D = 80 \pm 15$ cm$^2$/s using Eq. (9). Taking $S' = 0$ for simplicity, Eq. (6) becomes $\theta_1 \tan \theta_1 = \theta_2 \tan \theta_2$ and gives using Eq. (5) a nonlinear equation whose resolution gives $\tau = 270 \pm 50$ ps, $L = 1.46$ nm, and $S = 4.6 \times 10^4$ cm/s. However, since $\tau$ is very close to $\tau_1$, the uncertainty on the determination of $S$ is very large, of at least one order of magnitude. The profile is finally calculated using Eq. (A8), using the above determined parameter values and $\sigma = 0.6 \mu$m. The result is shown as a dotted line in the bottom right panel of Fig. 2 and very well approximates the data. It is concluded that combination of TRPL and $\mu$PL allows us to determine the key parameters for recombination and transport.

**FIG. 4.** Ratio of the spin to charge diffusion constants. The inset shows the light polarization transient, calculated from curves b of Fig. 5 and taking the time of the maximum $t_0$ as the time origin. This transient mostly exhibits a single exponential. The dots show the same transient at short times. The curves are calculated using Eq. (7) taking a ratio $D_s/D$ of the spin to charge diffusion constants equal to 0.3 (a), 0.9 (b), 1 (c), 1.1 (d), 1.3 (e), and 1.5 (f). From the almost ideal exponential behavior of the transient, it is concluded that $D_s/D = 1 \pm 0.1$. 

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IV. DISCUSSION

A. Temperature effects

The analysis of Sec. III has also been performed as a function of temperature. As seen in the bottom panel of Fig. 5, which shows the sum TRPL signals, the decay time $\tau_1$ at the long time increases with temperature. This shows that $\tau_1$ is dominated by the decrease with temperature of the bulk recombination efficiency $S$ rather than by the increase of surface recombination velocity. In agreement with Eq. (A4) and Fig. 3, the increase of $S$ is responsible for the observed temperature increase of the amplitude of the fast exponential decrease before 400 ps. The top panel of Fig. 5 shows the temperature dependence of the difference signal. Up to about 80 K, the time $\tau_{11}$ of the long time transient increases with temperature, in the same way as for the sum transient. Further temperature increase induces a decrease of $\tau_{11}$, which reveals a temperature decrease of the spin relaxation time $T_1$. The difference transient also exhibits a temperature increase of the amplitude of the short time transient. Such effect is caused by the increase with temperature of the surface recombination velocity, which will increase $\tau_{11}$ as shown in Fig. 3. Note also that, in agreement with independent observations, the characteristic time for the increase of the two signals weakly depends on temperature. Also visible is the slow tail at very long times observed for the sum transient and to some extent for the difference transient at the lowest temperature of 10 K. This finding will be discussed in Sec. IV B.

The sum $\mu$PL profiles are also shown in Fig. 6 for selected temperatures. The spatial decay is slightly faster when the temperature is decreased, thus revealing a limited decrease of $L_1$. Also shown in the figure are the best fits of the experimental data using Eq. (A8) using $S = S' = 0$. The agreement with the experimental data is satisfactory. Table I gives the values of $\tau_1$ and of $\tau_2$, as obtained by inverse Laplace analysis, of $L_1$ as well as the resulting values of $D$ and $\tau$.

At room temperature, $D = \mu kT/q$, as defined by the Einstein relation, is close to the value of 50 cm$^2$/s, calculated using a typical minority carrier mobility of 2000 cm$^2$/Vs. The value of $D$ only slightly increases with decreasing temperature, because of the approximate $1/T$ dependence of the mobility $\mu$. The difference between $\tau$ and $\tau_1$ becomes visible at temperatures larger than about 100 K because in this temperature range, the surface recombination velocity starts to play a role. However, the value of $\tau$ at 300 K is smaller by a factor of 3 than the bulk radiative lifetime for the present acceptor concentration and the temperature increase is characterized by an exponent of the order of 0.5 as a function of $T$ and closer to 1 as a function of the temperature of the electron gas. These exponents are smaller than the one of $3/2$ found for bulk radiative recombination. These two differences are attributed to nonradiative recombination at the rear surface, for which the time decreases with increasing temperature. At room temperature, the characteristic time for the back surface recombination, of the order of $\langle S'/d \rangle/(\tan(\theta_1))$ provided $y' \ll y$, can be estimated using the value of $S' = 10^5$ cm/s found on a sample with a doping higher by one order of magnitude. One finds a value slightly smaller than 1 ns, showing that back surface recombination could affect the value of $\tau$. Finally, the fact that $\tau$ is not larger than its expected value for radiative recombination suggests that photon recycling effects are weak. Such recycling would result in an increase of $\tau$ and is indeed expected to be small in the presence of a substrate.
Analysis of the dependence of $T_1$ as a function of lattice temperature as well as of the photoelectron temperature $T_e$, as determined from the shape of the PL spectrum, is shown in Fig. 7. A power law is found for the two curves with exponents of 1.16 and of 1.41 as a function of $T$ and $T_e$, respectively. The latter exponent is very close to that found using Hanle effect measurements for a similar doping, which shows the relevance of the exchange interaction of the electronic spins with the hole spins (known as the Bir Aronov Pikus mechanism).\(^5\)

### B. Energy-resolved analysis at $T = 9$ K

In the present section, we apply analysis of the TRPL signal to spectroscopic investigation as a function of luminescence energy. Unlike higher temperatures, the transient at $T = 9$ K depends on energy. The luminescence spectra at 9 K for selected times after the laser pulse are shown in the bottom panel of Fig. 8. While the spectrum shape does not strongly change during the signal increase, the spectrum after the onset of the decrease exhibits a stronger high energy signal (above 1.5 eV) and a relatively weak signal at an energy lower than 1.49 eV. Conversely, at long times, the luminescence peaks at a lower energy of 1.492 eV, implying an energy-dependent recombination time $\tau_2$. The value of $\tau_2$, determined using inverse Laplace analysis [Eq. (10)], is shown in the inset of Fig. 8 as a function of energy and is larger at low energy. Such behavior is well-known in disordered systems at low temperatures and indicates photoelectron localization in potential wells caused by the random distribution of acceptors.\(^3\) Although at this acceptor concentration the holes remain delocalized,\(^3\) the electron localization decreases the overlap between electron and hole wave functions, which explains the lifetime increase.

The light polarization spectra at selected times after the laser pulse, shown in the top panel of Fig. 8, exhibit the progressive build up of a polarization dip near 1.49 eV. This implies that, both for electrons of higher and lower energy, the lifetime $\tau_1$ is decreased or the spin relaxation time $T_1$ is increased with respect to their values at 1.49 eV. At high energy, as seen from the inset which shows the spectral dependences of $\tau_2$ and of $T_1$, the polarization increase is caused by an increase of $T_1$ from 1.5 ns at 1.48 eV to about 3 ns at 1.50 eV, while $\tau_1$ weakly depends on energy. This increase is caused by the reduced Coulomb interaction of energetic electrons with holes, which reduces the efficiency of spin relaxation. Below 1.49 eV, the increased polarization must be caused by the increase of $T_1$, which dominates the opposite effect of the increase of $\tau_1$. In spite of the relatively large difference, the polarization at 1.49 eV is substantially smaller than the value 1 at 1.48 eV, which shows the importance of the polarization decrease caused by the reduced Coulomb interaction of energetic electrons with holes.

### TABLE I. Values of effective diffusion length of the characteristic times of the first two TRPL modes and of the calculated charge diffusion constant and bulk recombination time for selected temperatures. Values in parentheses give uncertainties.\(^32\)

<table>
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<tr>
<th>$T$ (K)</th>
<th>$L_1$ (µm)</th>
<th>$\tau_1$ (ps)</th>
<th>$\tau_2$ (ps)</th>
<th>$D$ (cm²/s)</th>
<th>$\tau$ (ps)</th>
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<td>51(10)</td>
<td>111(15)</td>
<td>261(50)</td>
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<td>268</td>
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</tr>
</tbody>
</table>

FIG. 7. Dependence of the spin relaxation time $T_1$ as a function of the lattice temperature $T$ and of the temperature of the photoelectron gas $T_e$. At high temperature, these dependences follow a power law, of exponents 1.16 and of 1.41, respectively. The latter exponent is close to the one predicted by the Bir Aronov Pikus model for relaxation by exchange with holes.

FIG. 8. Energy-dependent TRPL at 10 K. The bottom panel shows the luminescence spectra at 10 K taken after a delay of 50 ps, corresponding to the maximum of the signal (a), 150 ps (b), 400 ps (c), 550 ps (d), 700 ps (e), 900 ps (f), 1300 ps (g), and 2000 ps (h) after the laser pulse. The top panel shows the corresponding light polarization spectra, where curves g and h have been omitted, as well as regions of poor signal-to-noise in the polarization spectra. The inset shows the energy dependences of $\tau_1$, and $T_1$, extracted from the sum and difference transients according to Eq. (7) and of the resulting spin relaxation time $T_1$.\(^5\)
poor accuracy of the determination of \( T_1 \) in conditions of weak signal conditions, the increased value of \( T_1 \) near 1.48 eV is visible in the inset of Fig. 8. This decrease of the spin relaxation efficiency of localized electrons by exchange with holes is viewed as a consequence of the smaller overlap of their wavefunctions, which decreases the electron-hole collision probability.

V. CONCLUSION

The charge and spin dynamics of \( p^+ \) GaAs have been investigated as a function of temperature using a novel method. This technique consists in combining spatially and time-resolved luminescence investigations. These two complementary, all optical, techniques, although quite different from an experimental point of view, have close conceptual analogies since corresponding signals are the solutions of very similar diffusion equations. The time decay of the signal exhibits well-defined exponential modes which are isolated using an inverse Laplace transform method. The spatial luminescence profile for a tightly focussed light excitation, given by an analytical resolution of the diffusion equation, is also predicted to exhibit spatial modes. The combination of these two techniques gives the charge and spin diffusion constants as well as bulk recombination time and spin relaxation time.

This method is applied between 9 K and 300 K to \( p^+ \) GaAs thin films, covered by a natural oxide layer. The diffusion constant is found to decrease slightly upon increasing temperature. The characteristic times \( T_1 \) and \( T_2 \) of the TRPL modes increase with temperature, implying that the surface recombination velocity increase is masked by the decrease of the efficiency of bulk recombination. The spin relaxation time \( T_1 \) decreases with increasing temperature and its dependence as a function of the temperature

\[
\frac{2}{d\beta_m^2} = 1 - \frac{\sin(2\theta_m)}{2\theta_m} + \gamma^{-1}[1 - \cos(2\theta_m)] + (\theta_m/\gamma)^2 \left[ 1 + \frac{\cos(2\theta_m)}{2\theta_m} \right]. \tag{A2}
\]

The corresponding eigenvalue is given by \( \lambda_m = (\theta_m/d)^2 \) and the eigenfunctions are such that \( \int_0^1 u_m(z)u_m(z')dz = \delta_{mn} \), where \( \delta_{mn} \) is the Kronecker symbol.

For TRPL, we search for a solution of the type \( \sum_m u_m(z)c_m(t) \), where \( c_m(t) \) can be a function of time. Inserting this expression into Eq. (2), multiplying by \( u_m(z) \) and integrating over \( z \), we find that the general solution of the time-resolved diffusion equation is given by Eq. (7) where the characteristic times \( \tau_m \) of the various modes are given by Eq. (5), and

\[
c_m = \int_0^t e^{t'/z_0} \left[ \int_0^t g_{TR}(z,t')u_m(z)dz \right] dt'. \tag{A3}
\]

For \( t' \) larger than the time for thermalization, \( \int_0^t g_{TR}(z,t')u_m(z)dz \) is zero so that for \( t \) larger than this same time, \( c_m \) converges to a value which depends on the details of thermalization. In the case, considered in Sec. II B, where thermalization effects are neglected, one has \( g_{TR}(z,t) = \alpha \delta(t)e^{-z^2} \) in Eq. (2). The amplitudes \( c_m \), found by imposing that, at \( t = 0 \), \( n(z,0) = n_0e^{-z^2} \), are given by \( c_m = n_0c_m^0 \) where

\[
c_m^0 = \beta_m^0A_m(x)A_m(\theta_m) \tag{A4}
\]

and

\[
A_m(x) = \left[ x^2 + (\theta_m/d)^2 \right]^{-1} \times \left\{ \theta_m(1/d + x/\gamma) - e^{ad} \left[ \cos(\theta_m) \frac{\theta_m}{d} + \frac{x}{\gamma} \right] \right. \\
\left. + \sin(\theta_m) \left[ x - \frac{\theta_m^2}{\gamma d} \right] \right\} \tag{A5}
\]

is the amplitude of the projection of \( e^{-z^2} \) on \( u_m(z) \). Note that the functions \( c_m \) defined by Eq. (A3) differ from their values of Eq. (A5) even at long times, provided the time \( \tau_m \) is not very long as compared with the characteristic time \( \tau_{th} \) of thermalization. This can be shown in the simple case where thermalization is characterized by a single exponential of time \( \tau_{th} \) and assuming that the photogenerated concentration profile as a function of depth does not change during thermalization. Taking \( g_{TR}(z,t) = x\tau_{th}^{-1}e^{-z^2/t_{th}} \), one finds

\[
I_{TR}(t) = C \sum_m \frac{c_m^0}{1 - \tau_{th}/\tau_m} \left[ e^{-t/\tau_m} - e^{-t/\tau_{th}} \right]. \tag{A6}
\]

The effective amplitude \( c_m^0/(1 - \tau_{th}/\tau_m) \) of mode \( m \) can differ significantly from \( c_m^0 \) for high order modes for which \( \tau_m \) is comparable with \( \tau_{th} \).

The same method can be applied to find the solution of the diffusion equation for \( \mu PL \) and gives Eq. (8). This solution also uses the amplitude of the projections of \( e^{-z^2} \) on \( u_m(z) \) so that the same coefficient \( c_m^0 \) is used.
for spatial and time modes. Fig. 9 shows the luminescence profiles corresponding to the first three spatial modes, calculated using $S'$ = 0. For a low value of $S = 10^3$ cm/s (bottom panel), the spatial mode of order 1 is strongly dominant at all distances. This is expected from Fig. 3, which shows that the higher order modes are negligible at low $\gamma$. In the opposite case where $S = 10^5$ cm/s (top panel of Fig. 9), one finds $c_0^2/\sigma_1^2 = 0.4$ and $v^0_3/\sigma_1^2 = 0.17$ so that the second order mode is only negligible with respect to the first order one at distances larger than about 2 $\mu$m. It is concluded that higher order modes do not prevent the determination of $L_1$.

Note finally that, for $\mu$PL, the diffusion equation also has an analytical solution, obtained by using a similar approach as Ref. 24. The in-plane Fourier transform $G(\xi, z)$ of the Green’s function $G(r, z)$, which is the response to a Dirac excitation at the point $r = 0, z = z'$, satisfies an ordinary differential equation with respect to $z$. One obtains

$$G(\xi, z - z') = \frac{\tau}{4\pi\lambda k} \left[ e^{-\kappa|z - z'|} + \frac{\kappa - \gamma}{\kappa + \gamma} e^{-\kappa(|z' + z|)} + \frac{2(\kappa - \gamma')}{\kappa + \gamma} \frac{\sinh \kappa c^* z + \gamma \sinh \kappa^* c z + \gamma \sinh \kappa^* z + \gamma \sinh \kappa c z}{\cosh \kappa c z + \gamma \cosh \kappa^* c z + \gamma \cosh \kappa^* z + \gamma \cosh \kappa c z} \right], \tag{A7}$$

where $\kappa = \sqrt{1 + \xi^2 L^2}$, $z^* = z/L$, and $z'^* = z'/L$. The photo-electron concentration is then given by

$$n(r, z) = 2\pi \int_0^\infty \xi \phi(\xi) J_0(\xi \lambda) d\xi \int_0^d e^{-x\xi} G(\xi, z - z') dz', \tag{A8}$$

where $\phi(\xi) \propto e^{-\xi^2/4}$ is the Fourier transform of the excitation profile and $J_0(\xi \lambda)$ is a Bessel function of the first kind.

Because of its integral form, this solution does not allow us to physically discuss the shape of the luminescence profile in the same way as Eq. (8) and requires the knowledge of three parameters which are $S$, $S'$, and $L$. However, its main interest is that it is possible to take $S = S' = 0$ and $L = L_1$, which is equivalent to take in Eq. (8) all $L_m$ equal to $L_1$. Shown by open triangles in Fig. 9 is, for both values of $S$, the profile calculated using Eq. (A8). In the bottom panel, the result closely corresponds to the first spatial mode. This is expected because higher order modes are negligible. In the top panel, the result only slightly differs from the sum of the first three modes near the center and closely corresponds to this sum for $r > 1$ $\mu$m. It is concluded that Eq. (A8) is in all cases a good approximation to the profile using the only parameter $L_1$.

![FIG. 9. The solid lines show the concentration profiles corresponding to the first 3 spatial modes, calculated using Eq. (8) for $D = 100$ cm$^2$/s, $\tau = 300$ ps, $\sigma = 0.5$ $\mu$m, $\gamma = 0$, and $S' = 0$. $S$ is taken equal to $10^5$ cm/s in the bottom panel and $10^3$ cm/s in the top panel. The sums of these modes are shown by full triangles and are in good agreement with the profiles, shown by open triangles, calculated using Eq. (A8) and using $S = S' = 0$ and $L = L_1$. The effect of modes of $m > 1$ is negligible for $S = 10^5$ cm/s, and only slightly affects the profile for $S = 10^3$ cm/s for $r < 1$ $\mu$m.](image-url)
The same calculation performed for a realistic value of $\alpha_l = 0.3$ shows that, for $c > 1$, reabsorption is equivalent, as expected, to increase the importance of surface recombination. Conversely, at very low $c$, photons originating from the recombination of secondary electrons are not included in the PL intensity, which leads to an artificially high $\gamma$. Thus, care should be taken in using the usual equation Eq. (1) for calculating the luminescence intensity at low $c$.

A similar approach in principle enables to determine the spin diffusion constant using the spin-polarized TRPL and µPL. However, such method is relatively imprecise and will not be discussed here.

Since the TRPL results corresponding to the µPL ones at 15 K were obtained at a slightly higher temperature of 20 K, the value of $D$ was modified, by a value smaller than the uncertainty, using an interpolation with the data obtained at 40 K.