Time resolved luminescence of photoexcited p-type gallium arsenide by population mixing

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A novel technique in time resolved luminescence spectroscopy called population mixing using a subpicosecond cw mode-locked dye laser has been developed and applied to p-type GaAs at low temperatures. Using this technique the relaxation lifetime for electron recombination was measured to be 39 ± 7 ps for p-type GaAs with Zn at 6×10^{18} cm^{-3} hole concentration. This is comparable to the relaxation time measured by a streak camera.

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Time resolved measurements of photoexcited semiconductors are important and of great interest in the study of semiconductors.[1] The advent of picosecond lasers and techniques made ultrafast time resolved measurements possible.[2-3] Luminescence relaxation decay times of materials have been measured using either a streak camera or a gating method on a picosecond time scale.[4-8] However, subpicosecond technology has been limited to time resolved absorption and reflectivity studies.[9-12] The purpose of this letter is to describe a novel technique for the measurement of the luminescence relaxation and to report the direct fast luminescence kinetics of photogenerated electrons in heavily doped p-GaAs. The new method, called population mixing, has subpicosecond resolution and is limited only by the pulse duration of the laser. This method is of general interest to the laser and semiconductor communities, and has the advantage of being able to isolate fast relaxation components of carriers in heavily doped semiconductors.

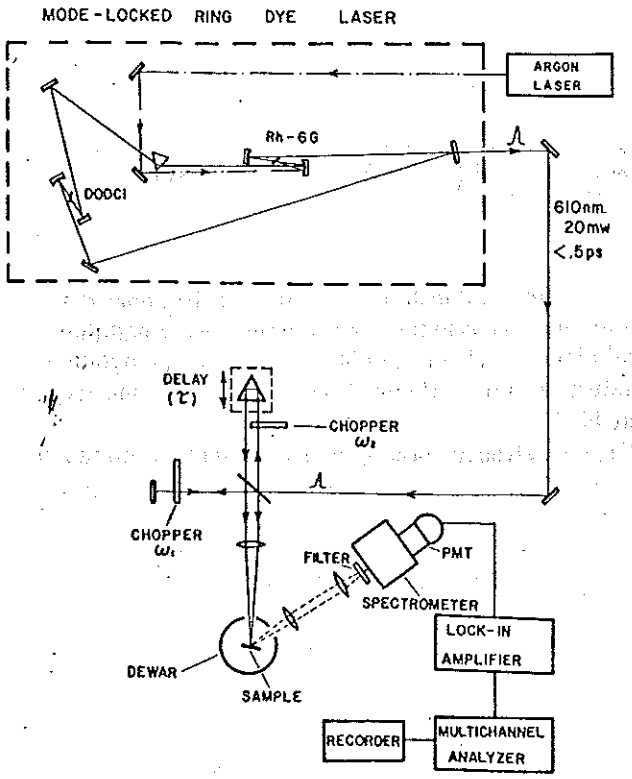

FIG. 1 Subpicosecond photoluminescence time resolved apparatus.

The intensity of luminescence from photogenerated carrier recombination in a semiconductor is proportional to the product of the population of conduction band electron density and the valence band hole density.[13,14] In this new technique, two identical excitation pulses are used to pump a semiconductor with a time delay τ between them. The electron and hole densities generated by the first pulse at time t are $n(t)$ and $p(t)$, and the photogenerated densities generated by the second pulse are $n(t+\tau)$ and $p(t+\tau)$. The photoluminescence produced by radiative recombination of these carriers is given by

$$I_L(t,\tau) \propto [n_0 + n(t) + n(t+\tau)]\,[p_0 + p(t) + p(t+\tau)], \quad (1)$$

where n_0 and p_0 are background electron and hole densities. The two excitation pulses are repeated at rate N ($N \ll 1/T$ where T is the carrier lifetime). The total average signal measured by a slow photodetection system is given by the time integral of $I_L(t,\tau)$

$$\langle I_L(t,\tau)\rangle \propto I_s(\tau) + I_B, \quad (2)$$

where

$$I_s(\tau) \propto N\int_{-\infty}^{\infty} n(t+\tau)p(t)\,dt + N\int_{-\infty}^{\infty} n(t)p(t+\tau)\,dt. \quad (3)$$

In the population mixing technique, the time-delay dependence remains only in $I_s(\tau)$. The free carriers from the first pulse mix and recombine with the free carriers from the second pulse generating the luminescence $I_s(\tau)$. The shape of $I_s(\tau)$ will depend on the forms of $n(t)$ and $p(t)$, which are determined by the type of recombination processes present. The other techniques (streak camera and Kerr gate) measure the dominant emission component. For p-type semiconductors $I_L(t) = n(t)p_0$, and for n-type semiconductors $I_L(t) = p(t)p_0$ since the $n(t)p(t)$ term is usually a much smaller term. The background signal I_B is usually much larger than the signal $I_s(\tau)$. I_B is subtracted from $\langle I_L(t,\tau)\rangle$ by a heterodyne technique. The signal from the photodetector is passed to a lock-in amplifier. The first pulse train is modulated at a frequency W_1 and the second pulse train is modulated at a frequency

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W_2. By setting the lock-in at frequency $\Omega = |W_1 - W_2|$ the background contribution I_B is subtracted from $\langle I_L(t,\tau)\rangle$ leaving $I_s(\tau)$. In this experiment, the excitation pulses are much shorter in duration than the lifetimes of photogenerated electrons, T_e, and holes, T_h. Therefore, the carriers form instantly and decay exponentially with a characteristic decay time. The majority-carrier decay was neglected because of the extremely slow recombination time in comparison to the decay of the minority carriers.[13,15]

The experimental setup is shown in Fig. 1. The 0.5-ps pulses at 610 nm were produced by a ring mode-locked dye laser at repetition rate of 125 MHz with an energy of 40 pJ per pulse. The pulse train was divided by a beam splitter into two pulse trains of equal intensity. The two pulse trains were modulated at 1430 and 2000 Hz, respectively. Both beams, whose pulses were ~ 10 pJ, were focused by a lens onto a sample. The spot size was about 25 μm. The sample was p-type GaAs doped at $6\times 10^{18}/\text{cm}^3$ Zn concentration at 115 K. The luminescence was collected and imaged onto the slit of a spectrometer. An RCA7265 photomultiplier and a PAR lock-in amplifier were used to display the signal which was fed into a Tracor Northern signal averager.

A luminescence signal at 840 nm was observed at 570, 1430, 2000, and 3430 Hz. The signal at 570 Hz was found to vary linearly with the pump intensity of either beam. When either pump beam was blocked, the signal at 570 Hz vanished. It is estimated that under the conditions of the experiment, the density of the photogenerated carriers was $\sim 5\times 10^{16}$ carriers/cm^3, at least two orders of magnitude less than the background density.

The photoluminescence intensity detected at 840 nm is plotted as function of delay time τ in Fig. 2(a). The signal is symmetric. The data of Fig. 2(a) are plotted on a semilog scale in Fig. 2(b). The decay time for the exponential slope is found to be 39 ps. The average decay time obtained from five separate runs at 115 K is 39 ± 7 ps. The decay time was comparable with the photoluminescence lifetime measured by a streak camera on the same sample.

The recombination decay time of the photogenerated carrier measured by the population mixing technique in heavily doped samples of p-type GaAs is found to be 39 ± 7 ps at 115 K. It should be kept in mind that the streak camera technique measures the time dependence of the total luminescence. The dominant component, in the total luminescence, is the relaxation of the minority carrier $[p_0 n(t)]$ since the term $n(t)\,p(t)$ is insignificant at the intensity levels commonly used. On the other hand, the population mixing technique measures a lifetime which depends on the relaxation of both minority and majority carriers. In the case of GaAs the relaxation time of the majority carriers is much longer than minority carriers,[15] for p-type $T_h/T_e > 10^3$. Since the maximum delay that can be measured in the present apparatus is 150 ps, the component of majority-carrier relaxation cannot be accurately time resolved. This slow component contributes to the background. The measured decay time 39 ± 7 ps is assigned to the relaxation of the electrons (minority carriers). The relaxation time is the time for the excess electrons to be trapped at recombination centers. Population mixing is a direct time resolved optical technique capable of subpicosecond resolution. This technique will prove to be a powerful method for measuring time resolved luminescence and the time evolution of electron and hole distribution functions of semiconductors, now that reliable cw subpicosecond lasers are available.[16]

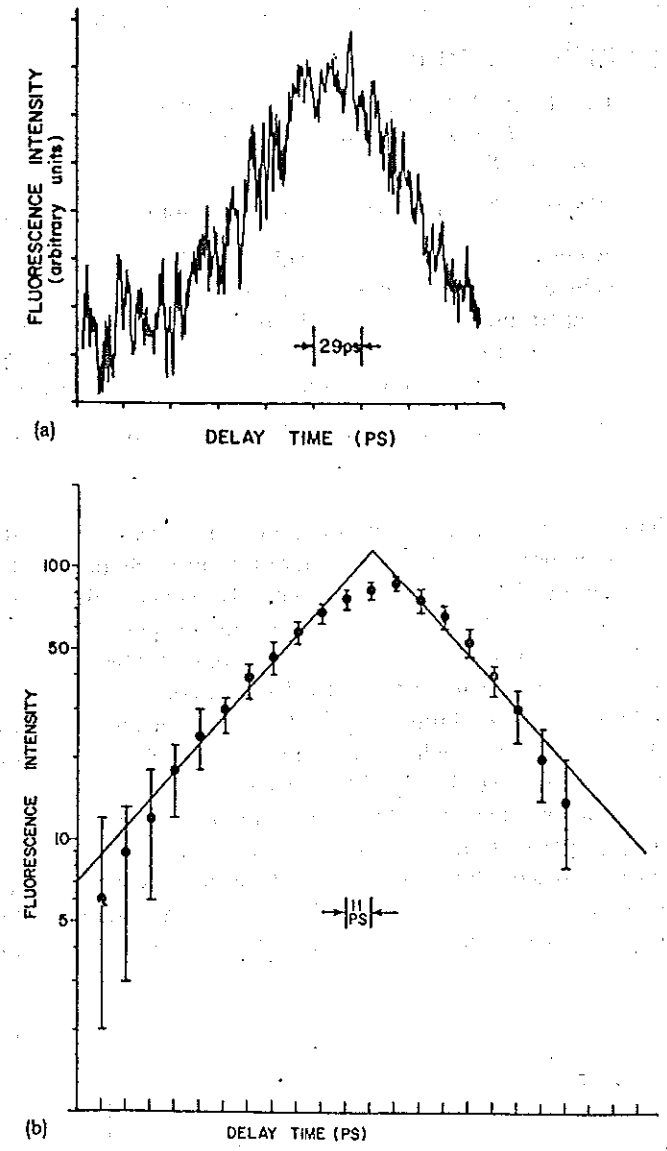

FIG. 2. (a) Correlation of the photoluminescence measured at wavelength 840 nm. (b) data of (a) is plotted on a semilog scale. The decay time is measured to be 39 ± 7 ps.

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