Optical measurement of the ambipolar diffusion length in a ZnCdSe–ZnSe single quantum well

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We describe a straightforward technique for the measurement of carrier diffusion in semiconductors. Using an optical microscope we can spatially image luminescence with a resolution of ~500 nm. We measured the ambipolar diffusion length in a $Zn_{0.75}Cd_{0.25}Se$–ZnSe single quantum well by fitting the spatially resolved luminescence profile with the solution of the two-dimensional diffusion equation. The ambipolar diffusion length was found to be 498 nm at a carrier density of $\sim 1\times10^{18}$ cm^{-3} and we deduce an ambipolar diffusion constant of 1.7 $cm^2\,s^{-1}$. © *1997 American Institute of Physics.* [S0021-8979(97)04201-1]

The recent demonstration[1] of a ZnCdSe–ZnSe laser with a continuous, room-temperature operating lifetime of over 100 h has led to renewed interest in wide band-gap II–VI quantum well materials for blue-green optoelectronic applications. To fully develop their potential it is essential to understand the basic physical processes of light emission and carrier dynamics in these novel materials.

Carrier transport properties in two-dimensional heterostructures are of great interest due to the importance of these structures for optoelectronic devices. Carrier diffusion lengths are critical parameters in the calculation of the *p-n* junction injection ratio and laser diode threshold current[2] as well as providing vital information on carrier lifetime and mobility. Although there have been several studies of diffusion in bulk ZnSe,[3,4] little is known about the transport properties of carriers in ZnCdSe–ZnSe quantum wells at room temperature. Carrier transport has been extensively studied during the development of III–V optical and electronic devices and several techniques have been developed to measure the diffusion length, including the original Haynes–Shockley experiment.[5] More recent techniques include short circuit photocurrent measurements,[6] electron beam induced currents,[7] cathodoluminescence (CL),[8] time-of-flight (TOF) measurements[9] and transient grating techniques.[10]

In this communication we present the first measurement of the ambipolar diffusion length in a ZnCdSe–ZnSe single quantum well at room temperature. Using an optical microscope we spatially image the luminescence profile created by an exciting laser pulse focused to a spot size of ~500 nm. The diffusion length is obtained by fitting the solution of the two-dimensional diffusion equation to the experimentally measured luminescence profile. This method has the advantage that no further sample processing is needed as is the case for CL and TOF measurements. Furthermore, since no electrical contacts are involved, the technique is equally accessible to the measurement of either the minority carrier diffusion length in doped samples or the ambipolar diffusion length in undoped materials.

The recent improvements in material growth which have led to increased II–VI laser operating lifetimes are reflected in long luminescence lifetimes and high photoluminescence efficiency at room temperature. In our samples the carrier lifetime was found to be greater than 1 ns for carrier densities up to $\sim10^{19}$ cm^{-3}.[11] These carrier lifetimes are comparable with those measured in III–V laser diodes[12] indicating the good quality of our sample. This long lifetime combined with the high room-temperature PL efficiency permits the straightforward optical measurement of diffusion length.

The sample studied was a 6 nm $Zn_{0.75}Cd_{0.25}Se$ single quantum well grown by molecular beam epitaxy (MBE) on a (100) GaAs substrate. The sample consisted of a GaAs buffer layer, a 30 nm ZnSe layer, a 150 nm $ZnS_{0.07}Se_{0.93}$ buffer layer, a 500 nm ZnMgSSe cladding layer, a 100 nm ZnSSe barrier layer, the 6 nm $Zn_{0.75}Cd_{0.25}Se$ quantum well, followed by a further 100 nm $ZnS_{0.07}Se_{0.93}$ barrier layer and a 200 nm ZnMgSSe capping layer. This separate confinement heterostructure, although undoped, is a typical laser structure,[13] the wide band-gap ZnMgSSe layers providing good optical confinement in the active region.

A straightforward optical microscope setup, as shown in Fig. 1, was used to measure the ambipolar diffusion length. A stilbene-3 dye laser, synchronously pumped using the third harmonic of a mode locked Nd:YAG laser, producing 5 ps pulses tunable between 420 and 460 nm at a repetition rate of 76 MHz was used to excite carriers in the sample. To spatially filter the collimated beam it was focused through a 10 μm pinhole using a ×20, 0.4 NA microscope objective (MO1). A 20 cm focal length lens (L1) was used to recollimate the beam before it was focused onto the sample using a ×50, 0.6 NA, infinity tube-length microscope objective (MO2). The sample was mounted with the quantum well in the focal plane of MO2. The luminescence was collected by MO2 and focused using a second 20 cm focal length lens (L2) via the beamsplitter (BS) onto a 1317×1035 pixel air-

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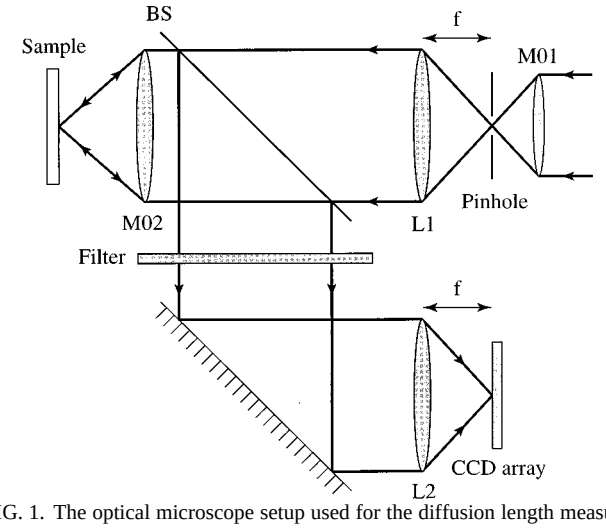

FIG. 1. The optical microscope setup used for the diffusion length measurement. M01=×20, 0.4 NA microscope objective, M02=×50, 0.6 NA microscope objective, L1,L2 $f=20$ cm focal length lenses, BS=beamsplitter.

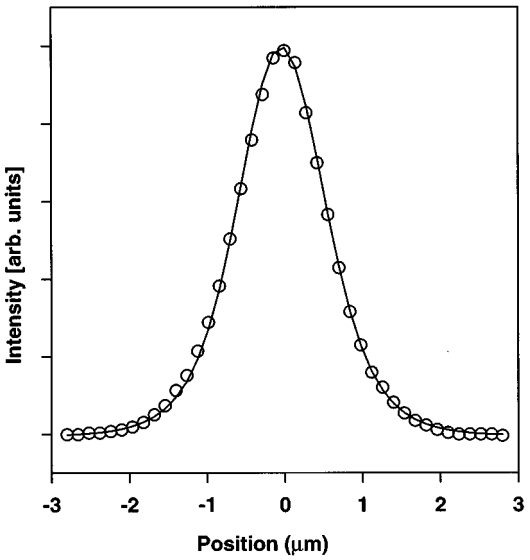

FIG. 2. Experimentally measured luminescence profile (open circles) and best fit to the data (solid line).

cooled CCD array for data collection. A colored glass filter having optical density >5 at the excitation wavelength was used to filter the laser line from the collected luminescence ensuring that laser light was not detected by the CCD. The choice of a 20 cm focal length lens to focus the collected luminescence onto the CCD provides an overall system magnification of ×50 between the object and detector planes. Thus, the CCD pixel size of 6.8 μm gives a detector resolution of 136 nm.

The overall spatial resolution of the system was estimated to be ~500 nm by removing the filter and measuring the full width at half-maximum of the image of the laser profile reflected from the surface of the sample. Using the theory of Török *et al.*[14] we calculate that the effect of the ZnMgSSe capping layer is to reduce the spot size by less than 10% at the quantum well. Therefore, the value of 500 nm is a good estimate of the overall system resolution. For all measurements carriers were excited in the ZnSSe barrier layers with the laser wavelength set at 430 nm; luminescence at a wavelength of 500 nm is emitted after carriers relax into the well. Carrier capture times in quantum wells have been theoretically predicted to be of the order of 10 ps;[15] this time is short compared to the carrier lifetime (~1 ns) so carriers do not diffuse significantly in the lateral direction before capture in the quantum well. A comparison of the diffusion length obtained by exciting carriers directly in the well and in the ZnSSe barrier layers verified that there was no difference in the values of diffusion length measured for the same carrier density in the well. However, to achieve the high densities which occur in laser diodes it was necessary to generate carriers in the barrier layer rather than directly in the quantum well.

Since carriers are confined in the plane of the quantum well, carrier transport can be well described using the two-dimensional diffusion equation:

$$\frac{\partial N(r,t)}{\partial t} = D\nabla^2 N(r,t) - \frac{N(r,t)}{\tau}, \tag{1}$$

D is the ambipolar diffusion constant, $N(r,t)$ is the carrier density as a function of the radial coordinate, r, and time, t, and τ is the carrier lifetime. We have assumed an exponential carrier decay rate and diffusive transport in the plane parallel to the plane of growth. Given an initial Gaussian[16] distribution of $1/e$ width, σ_0, Eq. (1) may be solved explicitly to give

$$N(r,t) = \frac{\sigma_0^2 N_0}{4Dt+\sigma_0^2} \exp\left(\frac{-t}{\tau}\right) \exp\left(\frac{-r^2}{4Dt+\sigma_0^2}\right), \tag{2}$$

for the carrier density as a function of position and time where N_0 is the initial carrier density. The signal, $I(r)$, recorded by the experimental setup shown in Fig. 1 is proportional to the time integral of (2), i.e.,

$$I(r) \propto \int_0^\infty N(r,t)dt. \tag{3}$$

The integral was found to depend only on the diffusion length, $L_D = \sqrt{D\tau}$, and not to vary independently with D and τ. Therefore, it was not possible to independently and uniquely determine D and τ, but by fitting the experimentally measured luminescence profile with L_D as a variable parameter using (3) it was possible to determine the diffusion length unambiguously. Since the initial distribution of carriers created in the well cannot be measured directly it was also necessary to include σ_0 as a fitting parameter. Values of σ_0 obtained in the fitting process were ~500 nm, consistent with the measurement of the system resolution. The quality of the fits obtained indicates that the assumption of an initial Gaussian distribution is indeed valid.

Figure 2 shows a typical luminescence profile (open circles) obtained at a carrier density of $\sim 1\times10^{18}$ cm^{-3} using the experimental setup shown in Fig. 1. In all cases the measured spatial distribution was found to be circularly symmetric to within experimental error and so the calculation of the diffusion length was independent of the section of the profile used. The best fit to the measured profile using Eq. (3) is also

shown (solid line); an ambipolar diffusion length of $L_D=498$ nm was obtained from this fit. From separate measurements, a carrier lifetime of 1.5 ns was obtained at this carrier density, giving an approximate value for the ambipolar diffusion constant, D, of 1.7 $cm^2\ s^{-1}$. Despite the recent improvements in II–VI technology[1] this value of D is still less than typical values of D reported for GaAs[16,17] at room temperature, indicating that there is still room for further improvements in II–VI material quality leading to potentially longer II–VI laser diode operating lifetimes.

In conclusion we have described a new, high-resolution, technique for the measurement of the carrier diffusion length in semiconductors. The method involves spatially imaging the luminescence profile created using a laser pulse focused to a spot size of ~500 nm. The diffusion length is obtained by fitting the measured profile with the solution of the two-dimensional diffusion equation. The high resolution of our system enables us to measure small diffusion lengths on the order of 500 nm. We have used this method to measure the ambipolar diffusion length in an undoped $Zn_{0.75}Cd_{0.25}Se$–ZnSe single quantum well. At a carrier density of $\sim 1\times 10^{18}$ cm^{-3} the diffusion length was found to be $L_D=498$ nm and the diffusion constant $D=1.7$ $cm^2\ s^{-1}$.

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