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Measurement of Ambipolar Diffusion Coefficient of Photoexcited Carriers with Ultrafast Reflective Grating-Imaging Technique

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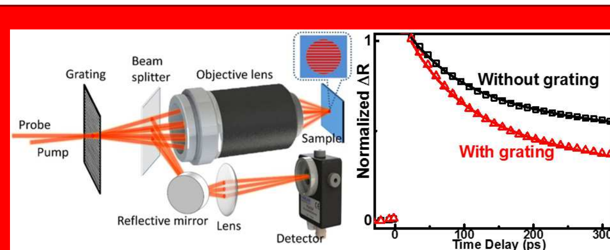
Supporting Information



A novel ultrafast reflective grating-imaging technique has been developed to measure ambipolar carrier diffusion in GaAs/AlAs quantum wells and bulk GaAs. By integrating a transmission grating and an imaging system into the traditional pump−probe setup, this technique can acquire carrier diffusion properties conveniently and accurately. The fitted results of the diffusion coefficient and diffusion length in bulk GaAs agree well with the literature values obtained by other techniques. The diffusion coefficient and diffusion length of GaAs/AlAs quantum wells are found to increase with the well layer thickness, which suggests that interface roughness scattering dominates carrier diffusion in GaAs/AlAs quantum wells. With the advantages of simple operation, sensitive detection, rapid and nondestructive measurement, and extensive applicability, the ultrafast reflective grating-imaging technique has great potential in experimental study of carrier diffusion in various materials.

carrier diffusion coefficient, carrier diffusion length, GaAs/AlAs quantum wells, bulk GaAs

Carrier diffusion, one fundamental form of electron transport in semiconductors, is important for semiconductor physics and device applications. Carrier diffusion properties, such as diffusivity and diffusion length, are associated with carrier scattering time, carrier mean free path, and carrier lifetime.[1] Studying carrier diffusion in semiconductors can help understand the interactions between electrons and other elementary excitations, reveal the dominant scattering mechanism, and facilitate the design of electronics and optoelectronics.

Semiconductor quantum wells (QWs) have attracted extensive research interest for several decades, since this structure not only provides a platform to study many quantum physics effects such as quantum confinement,[2] quantum Hall effect,[3] and quantum tunneling,[4] but also has important applications in semiconductor lasers,[5] high electron mobility transistors,[6] and photodetectors.[7] Study of photoexcited carrier diffusion in quantum wells is necessary to gain fundamental knowledge for optoelectronic applications. For example, in photodetectors[8] and semiconductor lasers,[9] how fast and how far the generated carrier can diffuse away from the active region determines the response rate and efficiency of the devices, respectively. The most important carrier diffusion parameters include the carrier diffusivity, D, and the diffusion length, L, which have been measured by several optical techniques including transient grating diffraction,[10] mask photoluminescence (PL),[11] mask transmission,[12] and spatial scanning pump−probe.[13] However, limitations are found in these existing techniques. For example, transient grating diffraction usually has weak diffraction signal. Mask PL and mask transmission methods require sizable PL intensity and optical transparency of the sample, respectively. Furthermore, mask−sample contact in mask transmission technique can cause surface contamination or undesired strain effect to the sample. Spatial scanning pump−probe is time-consuming due to the multiple scanning processes.

Here, we present a novel optical technique, ultrafast reflective grating-imaging (URGI), to measure carrier diffusivity and diffusion length in several different GaAs/AlAs quantum well structures, along with bulk GaAs. URGI is a significant modification of traditional ultrafast pump−probe system, as it incorporates a transmission grating (photomask) and an optical imaging setup to create a density gradient of excited carriers along the sample surface and enables the detection of in-plane carrier diffusion. Carrier dynamics purely from diffusion can be extracted by considering the difference in transient reflectivity change between the grating-modulated and no grating cases. The URGI technique has advantages of large signal, simple setup, wide applications, and rapid measurement, which

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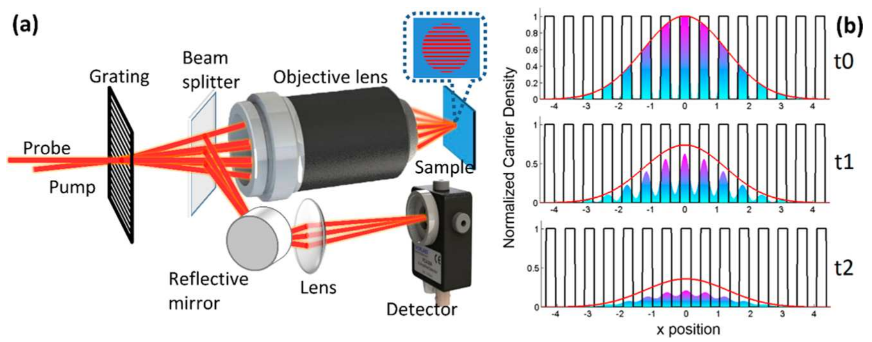

Figure 1. (a) Experimental setup of ultrafast reflective grating-imaging technique (URGI); (b) Schematic diagram of spatial and temporal evolutions of carrier density profiles generated by a regular Gaussian pump (the red curves) and a grating-modulated pump (the blue-purple shades), corresponding to the carrier density profiles without grating and with grating measurements, respectively.

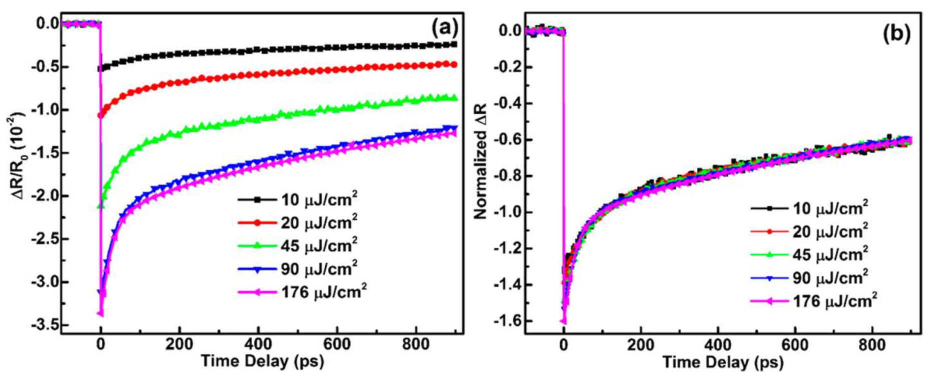

Figure 2. (a) Transient differential reflection signals $\Delta R/R_0$ in 8 nm/8 nm QWs measured in the absence of the grating with regular Gaussian pump at different excitation fluences; (b) $\Delta R/R_0$ signals normalized with its value at 80 ps, showing density-independent carrier recombination time.

overcome the limitations of current methods. In this paper, the validity of our technique is first confirmed by comparing our measured diffusion coefficients in bulk GaAs with the reported values. Then the dependence of carrier diffusivity and diffusion length on well thickness at room temperature is studied. Our results support that the interface roughness scattering is the dominant mechanism that limits photoexcited carrier diffusion in GaAs/AlAs quantum wells.

■ RESULTS AND DISCUSSION

Experimental Results. Figure 1a shows the experimental setup of URGI. Pump and probe beams are focused to the same point on a transmission grating with a 4° relative angle. The pump spot radius on the grating is about two times of that of the probe, with their centers overlapping. The diffracted beams from the grating pass through an objective lens and converge at the sample surface. A clear shrunken grating image from the focused laser spot can be generated at the sample surface by precise adjustment of the distance between the grating and the objective lens, as shown in the inset of Figure 1a. The reflected probe beams first pass through the same imaging objective lens, are then directed by a beam splitter and a mirror toward a lens, and then are loosely focused into a photodetector. The grating-modulated pump generates a carrier density grating along the sample surface, as shown in Figure 1b. The generated carrier grating decays and finally disappears due to both local carrier recombination as well as carrier diffusion from the illuminated area to the dark area. Because the pump and probe beams irradiate on the same spot at the grating whose conjugate image is formed at the sample surface, both the pump and probe spots on the sample surface will have the same grating-modulated intensity pattern (bright and dark slits) and the illuminated area of probe (bright slits) should overlap with the pump-illuminated area (see Figure S1 in Supporting Information for images of pump and probe spots on the sample surface). That is, the probe beam can only sense carriers in the illuminated area, and the probe reflection will only monitor the carrier density change in these regions. Therefore, the temporal reflection change of the probe beams reveals the decay process of the carrier grating generated by the pump beams, which contains information on both carrier recombination and diffusion. In order to obtain accurate information about carrier diffusion, it is necessary to subtract the contribution of carrier recombination. By removing the grating, a separate measurement equivalent to traditional pump−probe is performed to acquire the carrier lifetime. By contrast, in traditional pump−probe measurements, the laser spots at the sample surface are usually much larger than the carrier diffusion length, which means carriers cannot effectively diffuse out of the probe region (the red curve in Figure 1b) before recombination. So, the in-plane diffusion effect can safely be neglected when using a large laser spot,[14,15] and the transient reflection change in this case purely reflects the carrier recombination process and can yield carrier lifetime.

Carrier transport dynamics have been measured with URGI in three MBE grown GaAs/AlAs quantum well samples on GaAs substrates and one GaAs bulk wafer. All quantum well samples have 30 periods, with each period consisting of 6 nm/6 nm, 8 nm/8 nm, and 14 nm/2 nm of GaAs/AlAs (the determination of sample thicknesses can be found in Supporting Information). Pump and probe pulses are generated from a Ti:sapphire oscillator, with 80 MHz repetition rate, 800

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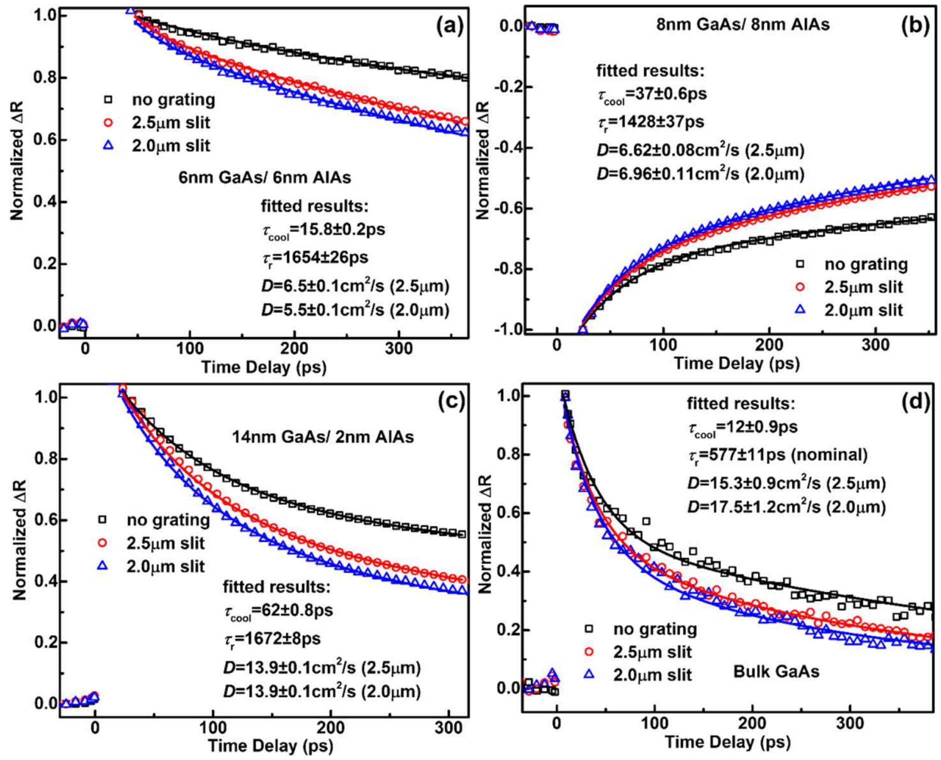

Figure 3. Normalized transient $\Delta R/R_0$ signals measured with and without grating modulations in (a) 6 nm/6 nm QWs; (b) 8 nm/8 nm QWs; (c) 14 nm/2 nm QWs; and (d) bulk GaAs, respectively. Pump fluence is 57 μJ/cm^2 in all measurements. The solid curves are the fittings with eq 5 derived in the text.

nm central wavelength, and 100 fs duration. Pump and probe spot sizes at the sample surface were measured to be 70 and 35 μm, respectively (see Figure S1 in Supporting Information for images of pump and probe spots on the sample surface). The polarizations of the pump and probe were set perpendicular to each other to eliminate the coherence artifacts. All measurements were performed at room temperature. Figure 2a shows the transient differential reflection signals measured in the 8 nm/8 nm QW sample in the absence of the grating with regular Gaussian pump. The photoexcited carriers generated by the pump pulse modulated the refractive index of GaAs layers, n, and induced a negative probe reflection change $\Delta R = R - R_0$ in the sample, where R and R_0 are the reflection with and without pump, respectively. The magnitude of differential reflection $\Delta R/R_0$ increases with pump fluences and saturates at high fluences due to the fact that the injected carrier density reaches the maximum available electron density of state. Using the transfer matrix method,[16] we found that 1 μJ/cm^2 pump fluence corresponds to an excited carrier density of 3.4×10^{16}/cm^3 (see Supporting Information for details of carrier density estimation). From the experimental data shown in Figure 2a, it can be seen that the signals show saturation features at pump fluences between 45 and 90 μJ/cm^2, suggesting a saturation density between 1.5×10^{18}/cm^3 and 3.1×10^{18}/cm^3. Considering the density of states for GaAs quantum wells, $g = \frac{4\pi m^*}{h^2}$ (where m^* is the effective mass of electron),[17] the thickness of the well, and the bandwidth of the laser pulse, about 0.107 eV (from 770 to 825 nm), a theoretical value of saturation density is estimated to be 2.75×10^{18}/cm^3 for the detected energy region, falling within the experiment-suggested range (1.5×10^{18}/cm^3 to 3.1×10^{18}/cm^3). This good agreement confirms both the high quality of our sample and the accuracy of our pump−probe experiments. The decay of $\Delta R/R_0$ consisted of two exponential components, one fast decay within several tens of picoseconds and another slow decay on nanosecond time scales. While the fast decay comes from hot carrier cooling in GaAs quantum wells,[18,19] the slower one reflects the carrier recombination process. As shown in Figure 2b, if normalized with its value at 80 ps, $\Delta R/R_0$ signals after 80 ps totally overlap, indicating that the carrier recombination time is independent of carrier density. A similar feature has been observed in all QW samples, which confirms the existence of density-independent carrier recombination time in GaAs/AlAs QWs within our measured carrier density range.

Shown in Figure 3 are the transient $\Delta R/R_0$ signals measured in QW samples with grating-modulated pump as well as with conventional Gaussian pump at the same fluence for comparison. The two gratings used in this study have transparent slit widths of 2 and 2.5 μm on the sample surface, respectively. The transparent slit width is half of the grating image period. The signals have been normalized at certain delay times within 20−50 ps in order to eliminate or weaken the nonlinear effects from carrier cooling and to facilitate comparison between the slow decay processes, with and without grating modulation. Specifically, the time delay of normalization is 46 ps for 6 nm/6 nm QW, 24 ps for 8 nm/8 nm QW, 24 ps for 14 nm/2 nm QW, and 7 ps for GaAs substrate. It should be noted that the sign of $\Delta R/R_0$ signal of GaAs/AlAs quantum well structures depends on the sign of dR/dn, which is a structure-dependent parameter that varies with layer thickness and composition, due to the interference of the reflections from different interfaces in the quantum wells (see Supporting Information for more details). The key feature observed for all signals is that the decay rates with grating

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modulations are obviously faster than without the grating. The smaller the grating slit, the faster the decay rate. This observation is expected from the measurement principle. Without grating modulation, the slow decay process only reflects the carrier recombination; with grating modulation, the slow decay process is affected by not only the local carrier recombination, but also the carrier diffusion from the illuminated (probed) area to the dark (unprobed) area. Such carrier diffusion accelerates the carrier density decay in the probed area and results in faster decay rates observed in cases with grating modulation. The smaller the grating slit, the larger the contribution to carrier density decay from carrier diffusion, hence, an even faster decay.

Theoretical Model to Extract Carrier Diffusion Properties from URGI. In order to extract the diffusion coefficient from the $\Delta R/R_0$ signals and study the physics of carrier diffusion, we have developed a theoretical model that considers both carrier recombination and diffusion excited by the grating-modulated pump. An amplitude transmission function of the grating is defined to take the grating-modulated pump into account: $t(x') = \frac{1}{2}[1 + \sum_{n=\text{odd}} \frac{4}{n\pi} \sin\frac{n\pi}{2}\cos(nfx')]$, where x' is the coordinate of the grating plane, and $f = \frac{\pi}{w}$ is the spatial modulation frequency with grating slit width w. Due to the finite aperture of the objective lens, only the first 3 orders (0 and ±1) of the diffractive beams are collected and generate the grating image with the amplitude right after the grating expressed as $\tilde{U}(x') = U_0 t(x') = \frac{1}{2}U_0\left(1 + \frac{4}{\pi}\cos(fx')\right) = U_0\left(\frac{1}{2} + \frac{1}{\pi}e^{ifx'} + \frac{1}{\pi}e^{-ifx'}\right)$, where the three terms in the last bracket () stand for the zeroth and $\pm$ first orders, respectively. After passing through the objective lens, the amplitude formed at the imaging plane is[20] $\tilde{U}(x, y) = U_0 A_0\left(\frac{1}{2} + \frac{1}{\pi}e^{ifVx} + \frac{1}{\pi}e^{-ifVx}\right)e^{i[OO']}e^{ikx^2+y^2/2z}$, where A_0 is the damping factor due to reflection, $[OO']$ is the optical length between the spot center at the grating O′ and the laser spot center on the sample O, $k = 2\pi/\lambda$ is the wave vector, z is spatial distance between the focal point of objective lens and the laser spot center on the sample O, and V is the magnification factor of objective lens (20× in our case). The intensity of the optical image at the sample surface can be expressed as $I = \tilde{U}\tilde{U}^* = U_0^2 A_0^2\left(\frac{1}{2} + \frac{2}{\pi}\cos(fVx)\right)^2$.

In our experiments, the probe spot is much smaller than the pump and focuses on the center of the pump. Thus, in the model, the pump intensity sensed by probe can be approximated as uniform, but the probe intensity is still treated as Gaussian. The initial carrier density grating generated by the pump pulse takes the form:

$$N(x, 0) = N_0\left(\frac{1}{2} + \frac{2}{\pi}\cos(Fx)\right)^2 = N_0\left[\left(\frac{1}{4} + \frac{2}{\pi^2}\right) + \frac{2}{\pi}\cos(Fx) + \frac{2}{\pi^2}\cos(2Fx)\right] \quad (1)$$

where N_0 is the carrier density at laser spot center, and $F = fV = \frac{\pi}{W}$ is the spatial frequency of the grating image at the sample surface with W as the grating slit width ($W = w/V$).

After the initial carrier cooling stage at a short time delay, the evolution of carrier density is governed by both carrier recombination and carrier diffusion, written as

$$\frac{\partial N(x, t)}{\partial t} = D\frac{\partial^2 N(x, t)}{\partial x^2} - \frac{N(x, t)}{\tau_r} \quad (2)$$

where D is the diffusion coefficient and τ_r is the carrier lifetime or, more accurately, the carrier density decay time (see later discussion for the case of bulk GaAs substrate). Under the initial condition of eq 1, the solution of the diffusion equation, eq 2, is

$$N(x, t) = N_0 e^{-t/\tau_r}\left[\left(\frac{1}{4} + \frac{2}{\pi^2}\right) + \frac{2}{\pi}\cos(Fx)e^{-F^2Dt} + \frac{2}{\pi^2}\cos(2Fx)e^{-4F^2Dt}\right] \quad (3)$$

The relative change in transmission or reflection intensity, $\Delta I_t/I_{t0}$ or $\Delta I_r/I_{r0}$, is proportional to carrier density $N(x, t)$. Thus, the intensity change of reflected probe is

$$\Delta I_r(x, y, t) = CI_{r0}(x, y)N(x, t) = CI_{00}G(x, y)\left(\frac{1}{2} + \frac{2}{\pi}\cos(Fx)\right)^2 N(x, t) \quad (4)$$

where C is a scaling factor, I_{00} is the intensity at the laser spot center, and $G(x, y) = e^{-4\ln 2(x^2+y^2)/r^2}$ is the Gaussian envelope of the probe intensity, with r as the full width at half maximum (fwhm). By integrating the probe intensity over the whole sample surface, the final analytical expression of the detected $\Delta R/R_0$ signal with URGI takes the following form:

$$\Delta R(t) = \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} \Delta I_r(x, y, t)\mathrm{d}x\mathrm{d}y \cong Ae^{-t/\tau_r}\left[\left(\frac{1}{4} + \frac{2}{\pi^2}\right)^2 + \frac{2}{\pi^2}e^{-F^2Dt} + \frac{2}{\pi^4}e^{-4F^2Dt}\right] \quad (5)$$

where $A = CI_{00}N_0\frac{\pi r^2}{4\ln 2}$. The detailed derivation of eq 5 can be found in the Appendix of the Supporting Information. While τ_r can be acquired from the measurement without grating modulation, there are only two fitting parameters in eq 5: the scaling factor A and the carrier diffusion coefficient D. The three terms of eq 5 describe the contribution from the three frequency components that make up the grating: carrier recombination of the zero frequency term, recombination, and diffusion of the F frequency term that originates from the interference of 0 order and +1 (or −1) order, and recombination and diffusion of the $2F$ frequency term from interference of +1 order and −1 order. When there is no grating ($F = 0$), those interference terms will disappear and eq 5 will reduce to a single exponential decay that only describes the nominal carrier recombination dynamics.

Eq 5 is derived under the condition that the carrier cooling effect can be neglected. For the case of a relatively long τ_{cool}, a modified theoretical model is used for fitting (see Supporting Information for details):

$$\Delta R(t) = (Ae^{-t/\tau_r} + Be^{-t/\tau_{cool}})\left[\left(\frac{1}{4} + \frac{2}{\pi^2}\right)^2 + \frac{2}{\pi^2}e^{-F^2Dt} + \frac{2}{\pi^4}e^{-4F^2Dt}\right] \quad (6)$$

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When there is no grating ($F = 0$), eq 6 reduces to a form of two exponential decays that describe the nominal carrier recombination and the carrier cooling dynamics.

■ FITTED RESULTS AND DISCUSSION

The solid curves in Figure 3 show the fitting results of the measured $\Delta R/R_0$ signals. The signals for the no grating cases were first fitted with two exponential decays to obtain the carrier cooling time τ_{cool} and the carrier lifetime τ_r. Then with τ_{cool} and τ_r known, the signals with gratings were fitted using eq 5 or eq 6 to acquire the carrier diffusion coefficient (see Supporting Information for details of fitting process). All the experimental data can be fitted well, suggesting that our model captures the relevant carrier diffusion dynamics. Moreover, the fitted diffusion coefficients obtained with two different gratings on the same sample are very close to each other. Such self-consistence of the fitted results further confirms the effectiveness of our URGI technique and theoretical model. The acquired ambipolar diffusion coefficients for all samples are plotted in Figure 4 as solid symbols. The open symbols represent previously reported values in bulk GaAs measured at room temperature under high excitation ($>10^{17}/cm^3$), which vary in the range of 15−20 cm^2/s.[12,21,22] Our bulk GaAs result, which is obtained at a carrier density around $2 \times 10^{18}/cm^3$, agrees well with the published data, validating the utilization of URGI to measure the ambipolar diffusion coefficient. The reported values of the carrier diffusion coefficient in MBE grown GaAs/$Al_xGa_{1-x}As$ quantum wells can vary over a relatively large range (10−50 cm^2/s) at room-temperature due to the variation in growth condition and composition.[10,11,23−25]

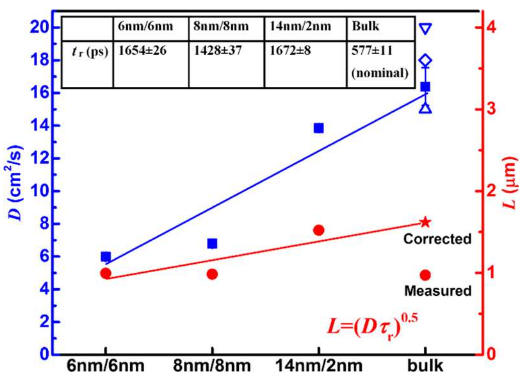

Figure 4. Fitted ambipolar diffusion coefficients D and the derived diffusion lengths L in GaAs/AlAs QWs and bulk GaAs. Inset: carrier lifetime in all samples. The down triangle, diamond, and up triangle data are from references13, 14, and9, respectively. The solid star data is estimated from an equivalent carrier lifetime. Solid lines are guides for eyes.

In quantum well samples, D shows an increasing trend with increasing GaAs layer thickness at room temperature. Similar trends have also been observed before in the low temperature range of 10−230 K.[26] With an excitation photon energy of 1.55 eV, carriers can only be excited in the GaAs layer because AlAs has a band gap larger than the photon energy. The inhomogeneity of the GaAs/AlAs interface can serve as an effective scattering potential for 2D electron motion,[27] which originates from the roughness-induced local fluctuation in quantization energy. With larger GaAs layer thickness, the roughness/thickness ratio is smaller, hence, smaller carrier-interface scattering. Interface roughness scattering is well accepted in literature as the main limiting factor of carrier mobility in quantum wells.[28−30] As a complement, our experimental result further indicates that carrier-interface scattering is the dominant mechanism in carrier diffusion of GaAs/AlAs quantum wells at room temperature.

The inset of Figure 4 lists the carrier lifetime in each sample. All quantum wells possess very close carrier lifetime values, while that in bulk GaAs is much smaller. It is necessary to point out that the fitted nominal carrier lifetime in bulk GaAs is not the actual recombination lifetime, but rather the carrier density decay time which includes both effects from the carrier recombination and carrier diffusion along the cross-plane direction. In quantum wells, carrier movement along the growth direction is restricted by the AlAs barrier layers. So, the excited carriers can only diffuse along in-plane directions in the GaAs wells. However, in bulk GaAs, the excited carrier can diffuse deeper into all directions, which will decrease the carrier density at the probed-surface and, hence, results in a smaller apparent carrier lifetime. Despite the fact that the nominal lifetime measured in bulk GaAs substrate without grating modulation includes the cross-plane diffusion effect, our approach to acquire the in-plane diffusion effect is still valid because with grating, the signal decay rate is also determined by carrier recombination, cross-plane diffusion, and in-plane diffusion. It is the difference in the decay rate between the without and with grating cases that reflects the in-plane diffusion effect, as can be seen in our model (eq 5). The carrier diffusion length L can be estimated using the following relation: $L = \sqrt{D\tau_r}$ (here τ_r is the real carrier recombination lifetime); the results are plotted in Figure 4 as filled red circles. The diffusion lengths in all samples are around 1−2 μm, on the same order as literature values.[12,24,31] The measured diffusion length is much smaller than probe spot size (35 μm), which supports our assumption that diffusion effects can be neglected for the case without grating modulation. Assuming that the bulk GaAs should possess an equivalent carrier lifetime to that of quantum wells (∼1600 ps), a corrected diffusion length for bulk GaAs is obtained and displayed in Figure 4 by a star point. Considering the correction, the diffusion length L also increases with GaAs layer thickness. This increasing trend also indicates that the interface roughness scattering governs the carrier diffusion motion in QWs.

■ CONCLUSION

An ultrafast reflective grating-imaging technique has been developed and employed to measure ambipolar carrier diffusion in GaAs/AlAs quantum wells and bulk GaAs. The measured diffusion coefficients in bulk GaAs were determined self-consistently and agree well with literature values obtained from other techniques. The diffusion coefficient and the diffusion length in GaAs/AlAs quantum wells were found to increase with GaAs well layer thickness, which supports that the interface roughness scattering is the dominant mechanism for carrier diffusion in GaAs/AlAs quantum wells. This URGI technique is simple in operation since it only takes two measurements to determine the diffusion properties; it is sensitive in detection since it takes the relatively large reflection change as signal; and it is widely applicable to both transparent and nontransparent samples. URGI is expected to find extensive applications in the experimental study of diffusion

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of all kinds of photoexcitations such as carriers, excitons, and phonons in various materials.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphotonics.7b00187.

(1) CCD images of pump and probe spots on surface, (2) MEB growth of supperlattice and determination of sample thicknesses, (3) Calculation of the excited carrier density in GaAs/AlAs quantum wells (QWs), (4) Carrier cooling time and lifetime from the fitting of the non-grating curve, (5) Sign change of ΔR in different samples, (6) Influence of GaAs substrate on the overall reflection of GaAs/AlAs SL, (7) Sensitivity analysis, (8) Appendix, and (9) Modified model for grating-curve fitting when carrier cooling needs to be considered (PDF).

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Notes

The authors declare no competing financial interest.

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