

Resonant Light Delay in GaN with Ballistic and Diffusive Propagation

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We report on a strong delay in light propagation through bulk GaN, detected by time-of-flight spectroscopy. The delay increases resonantly as the photon energy approaches the energy of a neutral-donor bound exciton (*BX*), resulting in a velocity of light as low as 2100 km/s. In the close vicinity of the *BX* resonance, the transmitted light contains both ballistic and diffusive components. This phenomenon is quantitatively explained in terms of optical dispersion in a medium where resonant light scattering by the *BX* resonance takes place in addition to the polariton propagation.

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Production of “slow light” is a subject of extensive studies nowadays [1]. Propagation of an electromagnetic wave through a dielectric medium can be considerably slowed down via ballistic or diffusive propagation, due to either peculiarities of dispersion or multiple scattering acts, respectively. The ballistic light propagation through the dispersive medium exhibits strong retardation of the energy transport in the vicinity of an absorption resonance [2], so that the group velocity v_g can be formally zero or even negative [3]. In semiconductors, this effect is closely related to the exciton-polariton formation [4]. The pulse propagation time agrees with that calculated from the polariton group velocity in different semiconductors, namely, in GaP:N around an isolated bound exciton (*BX*) line [5], CuCl near a free exciton resonance [6], and ZnO in the *BX* vicinity [7].

Light diffusion involving multiple photon scattering has been observed in various disordered media, including dielectric microspheres, polymers, and ZnS nanocrystals [8]. It is accompanied by intriguing phenomena, such as weak localization and coherent backscattering of light [9]. A strong slowdown of light transfer due to diffusive motion of photons was discussed in, e.g., [10]. In conventional bulk semiconductors, no experimental evidence for slow-light formation due to multiple scattering has been reported so far, to our knowledge.

In this Letter we report the first observation of a strong delay of light propagation in a high quality bulk GaN crystal, which amounts to 470 ps for a 1 mm long way, corresponding to a light velocity of about 2100 km/s. We demonstrate a resonant nature of this phenomenon and show that both light diffusion and retarded ballistic propagation of exciton polaritons can contribute to the delay. Because of the wide use of GaN in modern optoelectronics, this effect is promising for practical applications.

The studies have been done by time-of-flight spectroscopy using freestanding GaN epilayers grown by hy-

dride vapor phase epitaxy, which are among the best available. They exhibit photoluminescence (PL) spectra with the width of neutral-donor *BX* lines about 0.5 meV. We focused on three samples of different thicknesses, L : 1, 2, and 0.3 mm. The first sample has the carrier density of $8 \times 10^{15} \text{ cm}^{-3}$, two others around $5 \times 10^{16} \text{ cm}^{-3}$. In these experiments, a Hamamatsu streak camera with a ~ 20 ps temporal resolution was exploited to record the time-resolved (TR) images. The third and second harmonics from a tuned Ti:sapphire femtosecond pulsed laser were used as a light source. The temporal width of the detected laser pulse (20–50 ps) was determined by the instrumental accuracy. The transmitted light was registered in the forward direction along the normal to the surface. Conventional backward PL was measured for reference.

With excitation in a highly absorbing region by a 4.66 eV laser line, the backward TR PL images [Fig. 1(a)] show a fine excitonic structure with the pronounced *A* and *B* free exciton lines and the *BX* doublet at 3.472 eV, which is due to the exciton localization at Si and O donors. The transmitted PL images turn to be absolutely different. They exhibit a sharp quenching of the signal at a boundary located just below this doublet [Fig. 1(b)]. This boundary at ~ 3.47 eV is rather sharp in the best 1-mm sample; in other samples it is more diffuse and redshifted. The other remarkable feature is the bending of the PL streak towards this boundary. It means that the emission leaving the sample appears with a delay in time, which increases with approaching the *BX* resonances.

When the laser pulse was tuned to 3.486 eV to be resonant with the free excitons, the transmitted signal includes the excited PL along with the nonabsorbed part of the pulse. Both components are cut and bent in a similar way [Fig. 2(b)]. The excited PL was negligible, when the excitation energy was tuned to 3.464 eV, i.e., below the exciton resonances. This permits us to study the propagation of the laser pulse solely. Again, we observe the similar

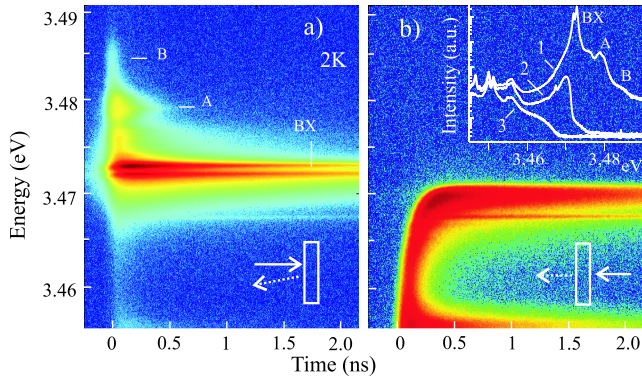


FIG. 1 (color online). (a) Backward and (b) forward TR PL images recorded in the 1-mm sample with excitation by a 4.66 eV laser line. In the colored images, the PL intensity (logarithm scale) increases with the color variation from dark blue to red. The inset presents time-integrated spectra of backward PL in the 1-mm sample (1) and forward PL in the 1-mm (2) and 2-mm (3) samples.

bending towards *BX*. The images recorded at different temperatures show a shift of the cutoff with the increase of temperature. This shift is ~ 10 meV in the 2–70 K range, which is consistent with the variation of the band gap in GaN and, hence, of the *BX* energy.

There is one more important feature—an additional curved streak (“reflex”), which is clearly seen in the images without PL taken with high resolution [Fig. 2(d)–2(f)]. Its bending corresponds to the threefold delay time of the first transmitted pulse. We assume that this feature appears due to the triple coherent passage of a wave packet (forward-back-forward) provided by the light reflection at the sample boundaries. This seems to indicate that the pulse propagation in our samples is predominantly ballistic, otherwise a photon should lose the direction memory after a few acts of scattering. The diffusion model predicts an extension of the pulse in the time domain but not an extra peak, as it is illustrated by Fig. 2(c), calculated using the models described below.

On the other hand, the reflex is clearly seen only in the best 1-mm sample. In other samples with higher impurity concentration, usually only a smooth cloud is recorded. Besides, the reflex is not visible close to *BX* and disappears with the temperature rise, while the delay still exists. All this shows that a nonballistic contribution to the delay of light in the *BX* vicinity cannot be excluded.

In an attempt to gain further information, we have measured the far-field angular dependence of the transmitted light. We expect the ballistic propagation to yield a narrow angular diagram of the output intensity, while the diffusion is likely to provide a much broader distribution. For this experiment, we used a Xe lamp, whose light passed through a monochromator impinges normally to the sample surface. The output intensity was registered at different angles by means of rotation of a stage, holding the lamp and a cryostat, with respect to a detecting system. As one can see from Fig. 3, the angular distribution of the light intensity clearly indicates a superposition of two components—relatively narrow and broad ones. They are ascribed to the ballistic and diffusive parts of the light, respectively. We found that the intensity ratio between these components depends on the energy. The ballistic part dominates in the region of relative transparency, while the diffusive mechanism plays the major role in the vicinity of the *BX* resonances.

The coexistence of two regimes of light propagation is a surprising observation. It is well known that the ballistic propagation time $T_B(\omega)$ depends linearly on the sample thickness $T_B(\omega) = L/v_g(\omega)$, while the delay time caused by a conventional diffusion process would increase quadratically with the sample thickness. Thus the presence of the diffusive component could result in a marked extension of the streaks in the time-resolved images, which is not observed. The question arises why so different processes yield similar propagation times. To clarify that we model both processes separately.

For the ballistic propagation, the group velocity $v_g(\omega) = d\omega/dk$, where the wave vector $k(\omega) = (\omega/c)\sqrt{\epsilon(\omega)}$. The

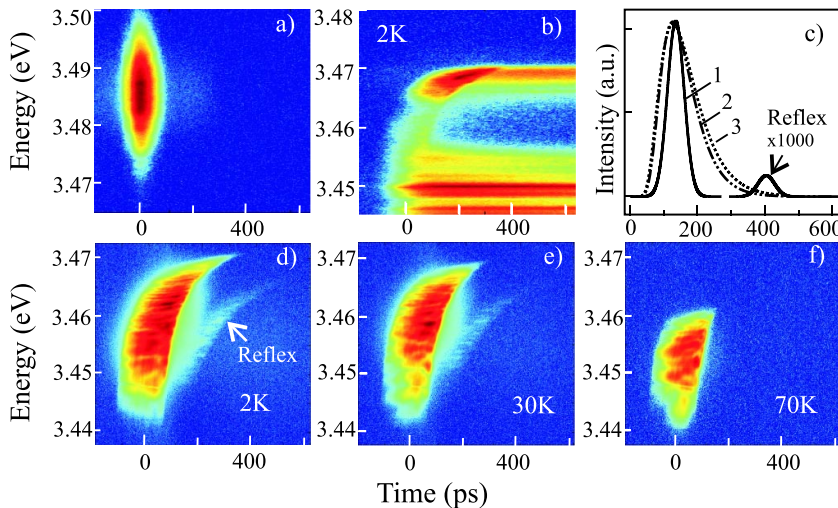


FIG. 2 (color online). TR images (logarithm scale) of 3.486 eV pulses recorded (a) without a sample and (b) through the 1-mm sample. (c) The pulse shape at 3.465 eV calculated for ballistic (1) and diffusive propagations in the sample neglecting (2) and taking into account (3) the 97% light reflection at boundaries. (d)–(f) TR images of 3.464 eV pulses passing through the sample at different temperatures.

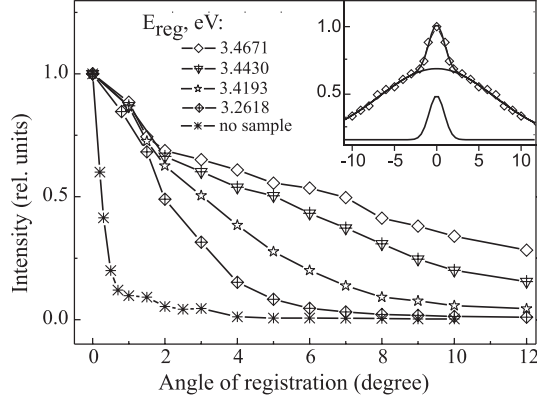


FIG. 3. Normalized angular diagrams of the light intensity measured at different photon energies in the 1-mm sample (and without it). The inset presents deconvolution of a diagram into narrow and broad components. The width of the ballistic part is determined by the light scattering at the sample surface. Some distortion of the broad component at large angles is due to the shading by cryostat windows.

frequency dependent dielectric constant in the vicinity of exciton resonances can be written neglecting spatial dispersion as

$$\varepsilon(\omega) = \varepsilon_b + \sum_j \int \frac{f_j \omega_{0,j}}{\omega_{0,j} + \xi - i\Gamma_j - \omega} \frac{1}{\sqrt{\pi}\Delta_j} \exp\left(\frac{-\xi^2}{\Delta_j^2}\right) d\xi. \quad (1)$$

Here, ε_b is the background dielectric constant; $j = A, B, C, BX$ denotes an exciton resonance. For simplicity we consider a single bound exciton state. Each resonance is described by a frequency $\omega_{0,j}$, an oscillator strength f_j , and a damping term Γ_j . The inhomogeneous broadening of the exciton line is taken into account by means of convolution with the Gaussian with the width Δ_j . We underline that

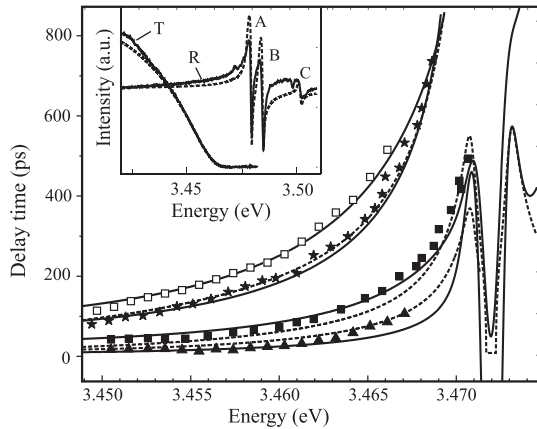


FIG. 4. Delay times measured in the 1-mm (squares), 2-mm (stars), and 0.3-mm (triangles) samples. Open squares are the data on the reflex in the 1-mm sample. The fitting curves are presented for ballistic (solid lines) and diffusive (dotted lines) propagation. The inset presents reflection (R) and transmission (T) spectra, measured using a lamp in the 1-mm sample, with their simulations (dotted lines).

only this formalism has allowed us to fit the reflection and transmission spectra simultaneously (see inset in Fig. 4). Note that the conventional approach [11] yields a reasonable fitting of reflection spectra but strongly overestimates the absorption of light near the exciton resonances.

We have performed a modeling assuming $\varepsilon_b = 9.5$ and $\hbar\omega_{0,j}$ equal to 3.4720, 3.4785, 3.4837, and 3.5016 eV for $BX, A, B,$ and C excitons, respectively. For either of the resonances, $\hbar\Gamma_j$ is taken as 0.012 meV, which is of the order of the homogeneous line width of an exciton polariton in other semiconductors [12]. The inhomogeneous width $\hbar\Delta_j$ is taken as 0.75 meV, close to the average PL linewidth in our spectra. For the A, B and C excitons, f equals 0.0026, 0.0017, and 0.00037, respectively, which is comparable with the previously reported values for GaN [11].

The oscillator strength for the bound exciton with the donor concentration N can be estimated as [13]

$$f = \frac{4\pi N}{\hbar\omega_{0,BX}} \left(\frac{e|p_{cv}|}{m_0\omega_{0,BX}} \right)^2 \left[\int \Psi(\mathbf{r}, 0) d\mathbf{r} \right]^2, \quad (2)$$

where m_0 is the free electron mass, e is the elementary charge, p_{cv} is the interband matrix element, and $\Psi(\mathbf{r}, \boldsymbol{\rho})$ is the envelope of the electron-hole pair function depending on the center of mass position \mathbf{r} and the relative electron-hole vector $\boldsymbol{\rho}$. Under the assumption that the exciton is localized as a whole it has the form

$$\Psi(\mathbf{r}, \boldsymbol{\rho}) = \frac{1}{\pi R^{3/2} a_B^{3/2}} \exp(-\mathbf{r}/R) \exp(-\boldsymbol{\rho}/a_B). \quad (3)$$

Here we took the exciton localization radius $R = a_B$, where $a_B = 2.8$ nm is the Bohr radius, which allows us to reproduce the observed value of the BX binding energy [14]. N is taken equal to the carrier density; $p_{cv} = 10^{-19}$ g cm/s. The best fit is obtained with $f \approx 2 \times 10^{-5}$ for the 1-mm sample, what is somewhat smaller than an estimation according to Eq. (2). For other samples f is taken an order of magnitude higher following the difference in the carrier densities. Figure 4 demonstrates a reasonable fit with these parameters and clarifies the origin of the sharp cutoff. The calculated dependencies have a strong dip near the BX resonance. It appears because the phase and group velocity of light strongly decrease here, while the absorption is enhanced [3].

To consider the diffusive propagation we assume that elastic neutral-donor scattering dominates the delay, neglecting any inelastic scattering by impurities or acoustic phonons [15]. We write the diffusion equation for the concentration of photons $n(\omega, x, t)$ at frequency ω , coordinate x , and time t

$$\frac{\partial n(\omega, x, t)}{\partial t} = D(\omega) \frac{\partial^2 n(\omega, x, t)}{\partial x^2} - \frac{n(\omega, x, t)}{\tau(\omega)}. \quad (4)$$

Here frequency dependent $D(\omega)$ and $\tau(\omega)$ are a diffusion coefficient and photon lifetime, respectively. The lifetime is defined as $\tau(\omega) = \tau_0[l_A(\omega)/l_0]$, where l_0 and τ_0 are the

photon mean free path and time between the two acts of scattering in the region of relative transparency; $l_A(\omega)$ is the absorption length. All the photons are assumed to be initially created in a thin layer near the sample surface: $n(\omega, x, 0) = n(\omega, 0)\delta(x)$. We have performed calculations of the pulse propagation in the diffusive model neglecting and taking into account the light reflection at boundaries and found no qualitative difference [Fig. 2(c)]. Below the light reflection is neglected. The delay time due to diffusive photon motion, $T_D(\omega)$, can be found from the condition $\partial n(\omega, L, t)/\partial t = 0$ which corresponds to the intensity maximum. In the general case it is written as

$$T_D(\omega) = \sqrt{\frac{\tau(\omega)^2}{16} + \frac{L^2\tau(\omega)}{4D(\omega)}} - \frac{\tau(\omega)}{4}. \quad (5)$$

We have the two following limiting cases: (i) small absorption, $\tau(\omega) \gg L^2/D(\omega)$ where the delay scales as a second power of the sample thickness, $T_D(\omega) = L^2/[2D(\omega)]$; (ii) strong absorption, $\tau(\omega) \ll L^2/D(\omega)$, where the delay is proportional to the sample thickness as with the ballistic propagation, $T_D(\omega) = (L/2) \times \sqrt{\tau(\omega)/D(\omega)}$. The latter result is a consequence of the competition between the “fast” absorption and “slow” diffusion processes.

The diffusion constant $D(\omega) = v_g(\omega)l_{tr}(\omega)/3$, where $l_{tr}(\omega) = [N\sigma(\omega)]^{-1}$ is the photon mean free path determined by the light scattering cross section $\sigma(\omega)$ and the concentration of scattering centers N . Assuming that the light scattering is caused by the bound exciton resonances one may write $\sigma(\omega)$ in a general form [16,17]

$$\sigma(\omega) = \frac{f^2}{4\pi N^2 c^4} \frac{\omega_{0,BX}^6}{(\omega_{0,BX} - \omega)^2 + \gamma^2}. \quad (6)$$

Here, γ is an effective broadening parameter. The main contribution to the delay time comes from the strong frequency dependence of the cross section. Strictly speaking, Eq. (6) describes scattering caused by individual BX . This is valid only for the diluted donor concentration satisfying the condition $N^{1/3}\lambda \ll 1$, where λ is a wavelength inside the crystal. Near BX , this is fulfilled for $N \sim 10^{15} \text{ cm}^{-3}$, while the donor density is higher in our samples. On the other hand, the spatial distribution of donors is inevitably inhomogeneous and clusters of closely lying BX act as virtual scattering centers with effective concentration and localization radius. For the sake of demonstration, we present here $T_D(\omega)$ calculated as for the individual scatterers. We use $\gamma = 0.75 \text{ meV}$, $\tau_0/l_0 = 2 \times 10^{-9} \text{ s/cm}$, and f calculated according to Eqs. (2) and (3). While for the 1-mm sample some agreement is achievable with $N = 8 \times 10^{15} \text{ cm}^{-3}$, for other samples we are impelled to take the effective $N = 2 \times 10^{16} \text{ cm}^{-3}$ to obtain a reasonable fitting (Fig. 4).

Our simulations are done in the limiting cases of either ballistic or diffusive light propagation. We abstain from the fitting in a combined model, because the ratio between

these components is too dependent on the sample quality. Our main goal is to demonstrate that the diffusive mechanism can give a delay of the same order of magnitude as the ballistic one and that its dependence of the delay on the thickness can be linear as well.

In conclusion, we reported here a strong delay in light transfer through high quality bulk GaN samples and present a quantitative description of this phenomenon in the frameworks of optical dispersion and resonant elastic scattering models. We believe that our findings have a particular importance for the development of a new generation of smart optoelectronic devices, which rely on a controlled delay of light propagation.

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