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Excitonic parameters of GaN studied by time-of-flight spectroscopy

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We refine excitonic parameters of bulk GaN by means of time-of-flight spectroscopy of light pulses propagating through crystals. The influence of elastic photon scattering is excluded by using the multiple reflections of the pulses from crystal boundaries. The shapes of these reflexes in the time-energy plane depict the variation of the group velocity induced by excitonic resonances. Modeling of the shapes, as well as optical spectra, shows that a homogeneous width of the order of 10 μeV characterizes the exciton-polariton resonances within the crystal. The oscillator strength of A and B exciton-polaritons is determined as 0.0022 and 0.0016, respectively. © *2011 American Institute of Physics*. [doi:10.1063/1.3625431]

The current interest in slow light is mostly related to the fact that any realization of all-optical schemes of quantum communication implies a deterministic retardation of photon pulses.[1] The phenomenon of light delay due to optical dispersion induced by a resonance line[2] may be important for a set of optoelectronic devices, where light should propagate a long distance, or for those operating at a high frequency. The study of this phenomenon can elucidate the mechanism of the light transfer through a medium, which may be either ballistic (polariton-like), with conservation of wave vector,[3] or diffusive, when the wave vector, and possibly frequency, is changed during acts of scattering.[4] Recently, it has been demonstrated that both mechanisms can coexist in GaN, where the light is scattered by neutral donor bound exciton (D^0X) complexes.[5] Weak reflexes, emerging due to the internal reflection of light pulses from the sample boundaries, have been resolved by the time-of-flight spectroscopy in the time-resolved (TR) images recorded in GaN (Ref. 5) and in CdZnTe crystals.[6] It has been shown that the mechanism of their propagation is exclusively ballistic, because otherwise photons lose any memory about the initial direction after a few acts of scattering.[5] Here, we demonstrate that the study of the slow light may bring one more benefit. It allows one to refine excitonic parameters in semiconductors.

For GaN, there is a huge variation in reported values of the exciton oscillator strength (f) and longitudinal-transverse splitting (ω_{LT}). This uncertainty is determined by various factors, such as elastic strain,[7] different quality of samples,[8] and certainly by experimental methods. A high value of ω_{LT} can be obtained, if it is defined via a separation between photoluminescence (PL) peaks, because of a thermalization process.[9] The commonly used modeling of reflectivity spectra[10,11] suffers from uncertainty caused by a large number of the fitting parameters. Besides, both PL and reflectivity techniques probe an area close to the sample surface, rather than its volume, whereas the properties of these regions may differ.[12] A similar uncertainty exists for the width of the resonances, whose reported values vary from 0.2 meV (Ref. 10) up to 2.1 meV (Ref. 13) for A exciton. This parameter is commonly considered as an empirical damping constant (Γ_{eff}). However, the intrinsic linewidth has a certain physical meaning. It characterizes the decay of the coherence for the localized excitation or the dephasing relaxation for exciton-polaritons.[14]

In this paper, we propose to use the data on the ballistic reflexes of the light pulses propagating through GaN crystals to refine the excitonic parameters of bulk GaN. The samples under investigation were free-standing GaN layers with thickness L of 1 and 2 mm grown by hydride vapor phase epitaxy (HVPE). Their carrier concentrations were $5{\cdot}10^{15}$ cm^{-3} and 10^{17} cm^{-3}, respectively. The TR experiments were done at 2 K, using the pulses of a tunable picosecond laser (Mira-HP) and a Hamamatsu streak camera with a resolution about 2 ps. The dynamical range of the detecting system was not less than 10^3. As illustrated by the scheme in Fig. 1, the pulse reflexes can be registered in two experimental configurations. In the back-scattering (reflection) configuration, the even reflexes (2nd and 4th) propagate in the sample a distance of $2L$ and $4L$, respectively, while in the transmission geometry the odd 3rd reflex propagates $3L$, etc. In this study, we focus on the even-order reflexes, because their delay is well-defined with respect to an initial pulse (denoted as $0L$), directly reflected from the sample surface. The complementary continuous wave (cw) transmission and reflectivity measurements were performed at 5 K using a tungsten lamp.

Figure 1(a) shows the spectral dependencies of the delay times measured along the pulses of different energies for the same length of propagation, namely, through the 2-mm-long sample and for $2L$ replicas in the 1-mm-long sample. The delay time of the basic transmitted pulse in the 1-mm sample, multiplied by two in order to simulate the 2-mm length, is shown for comparison as well. Obviously, the delay times are smallest for the $2L$ reflexes, when the photon scattering, enhancing the light path, is excluded. The difference in the

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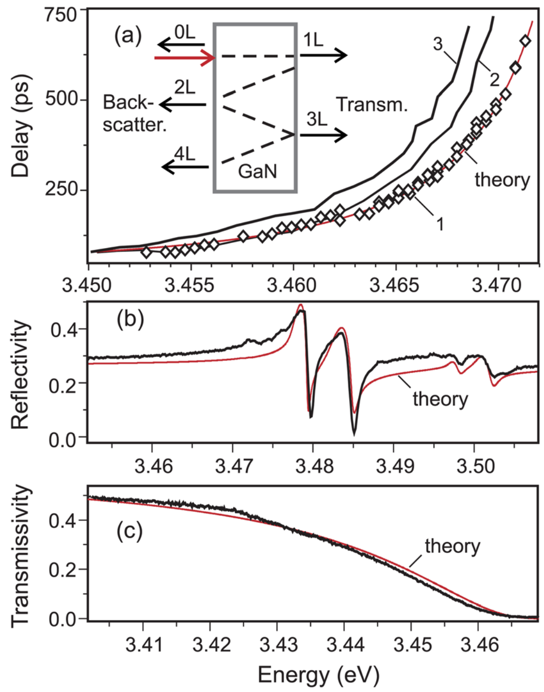

FIG. 1. (Color online) (a) Delay time dependencies obtained using (1) 2*L* pulse reflexes in the 1-mm sample; (2) basic pulses transmitted through the same sample, multiplied by two; and (3) basic pulses in the 2-mm sample. The inset shows the scheme of the back-scattering and transmission measurements, where n·*L* denotes the series of replicas. (b), (c) Experimental spectra of reflectivity (b) and transmission (c) measured in the 1-mm sample. The thin red lines are simulated spectra calculated as described in the text.

dependences for the 1-mm and 2-mm samples is related to different donor concentrations, which influence the oscillator strength of the D^0X resonances and, hence, the scattering cross-sections and polariton group velocity.[5] Figures 1(b) and 1(c) present reflectivity and transmission spectra, respectively, measured in the 1-mm sample. Figure 2 shows the sequence of the appearance of the $2L$ and $4L$ reflexes arising with the shift of the pulse from the D^0X lines towards a transparency region. The delay and shape of these reflexes reproduce the variation of the group velocity $v_g(\omega)$, which, in turn, is controlled by the parameters of the excitonic resonances. Near the D^0X lines, these reflexes are strongly curved and their higher-energy edge is extended along the time axis until full disappearance.

To analyze the experimental data, we use the model of the ballistic light propagation in a medium with several resonances, which, in general, can be inhomogenously broadened.[5] The delay $T(\omega) = L/v_g(\omega)$ is given by the group velocity $v_g(\omega) = d\omega/dk$, where the wave vector $k(\omega) = (\omega/c)\sqrt{\varepsilon(\omega)}$. The complex dielectric function $\varepsilon(\omega)$ is written as

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

Here, $\varepsilon_b = 9.5$ is the background dielectric constant. Each j-term within the sum is the convolution of the resonance line, characterized by the homogeneous width Γ_j, with the Gaussian centered on the same frequency $\omega_{0,j}$. The parameter Δ_j describes the inhomogeneous width of the broadened resonances. Spatial dispersion is taken into account by the term $\beta k^2 = (\hbar^2/2M_j)k^2$, where the effective masses M_j equal to $0.5m_0$, $0.6m_0$, and $0.8m_0$ (m_0 is free electron mass) for the A, B, and C excitons, respectively,[10] being considered as infinite for D^0X. Such a representation is allowed for frequencies not too close to ω_0 of the free exciton resonance which is sufficient for the consideration of our data on cw and TR light transmission. The estimations showed that the changes made by the spatial dispersion are not essential, being within the 20% limit. Therefore, Eq. (1) with the omitted βk^2 term can be used to simulate the reflectivity spectra using the equation

$$R(\omega) = |[(n-1)^2 + \kappa^2]/[(n+1)^2 + \kappa^2]|, \tag{2}$$

where $n(\omega) = \mathrm{Re}\sqrt{\varepsilon(\omega)}$ and $\kappa(\omega) = \mathrm{Im}\sqrt{\varepsilon(\omega)}$. The intensity of the transmitted signal is estimated as

$$I(\omega) = (1-R)^2 \exp(-D)/[1 - R^2 \exp(-2D)], \tag{3}$$

where $D = L\alpha$ is the optical density and $\alpha = 2\omega\kappa(\omega)/c$ is the absorption coefficient.

The fitting procedure includes the simulation of reflectivity spectra [Fig. 1(b)] to determine approximately the energies $E_j = \hbar\omega_{0,j}$ and f values of the exciton-polariton resonances. At this stage, the inhomogeneous width Δ_j plays the role of an effective damping parameter, used with the conventional reflectivity modeling. We underline that the determined parameters characterize rather a region close to the surface, than the bulk GaN. Next step is the simulation of the cw transmission spectra [Fig. 1(c)], which gives us the Γ values of the exciton-polaritons. These refer to the GaN volume, where the pulses propagate. The final correction of the excitonic parameters is made by means of the simulation of the delay, curvature, and attenuation of the transmitted reflexes. The different delay of the components of the pulse, which provides its curvature along the time axis, depends predominantly on the oscillator strength. Note that such a dominant influence of each of the parameters on a certain process simplifies the numerical simulation.

This simulation shows that the influence of the D^0X resonances is important at a distance less than 1-2 meV from the PL peaks [see Fig. 2(d)]. Consideration of the reflexes outside this range gives us the parameters of the exciton polaritons. In this modeling, we assumed that the parameters of the D^0X resonances are similar for both lines O^0X_A (3.4714 eV) and Si^0X_A (3.4723 eV), related to the oxygen and silicon donors. Their oscillator strength and homogeneous width for the 1-mm sample are obtained by fitting the experimental data as $5 \cdot 10^{-6}$ and 1 μeV, respectively (these values should vary with the donor concentration). A deviation from these values exceeding 10% does not allow one to reproduce the shape of the reflexes shown in Fig. 2. The inhomogeneous width of the D^0X resonances is determined by the consideration of the light attenuation near these lines. A width over 35 μeV causes quenching of the higher-energy tail of the light reflexes at the energies, where the signal is observed in the experimental images. It provides a shift of the apparent pulse center towards lower energy, as it is illustrated by the inset in Fig. 2(a). The obtained values for the D^0X resonances are substituted into Eq. (1) to determine

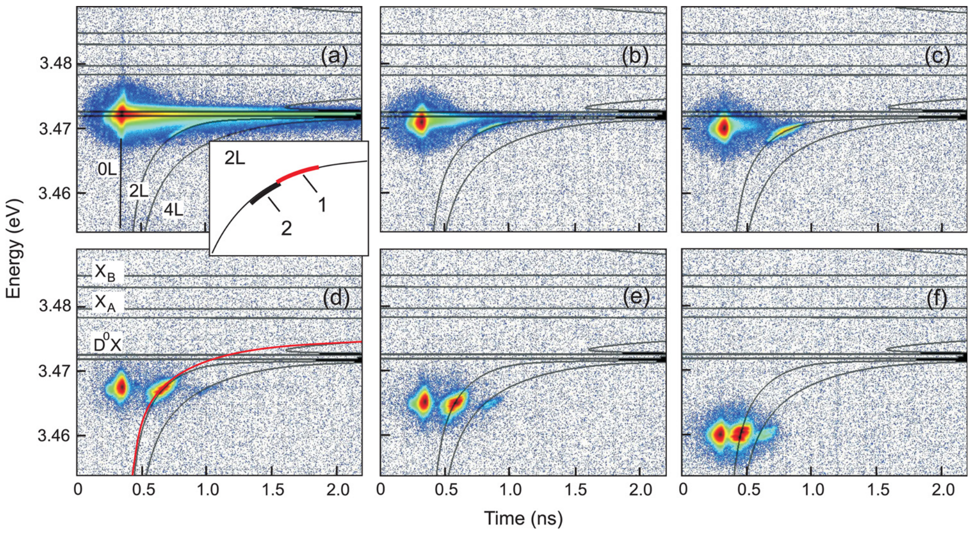

FIG. 2. (Color online) Selected TR images of replicas of impinging pulses at different energies, recorded in the back-scattering configuration. The delay time dependencies calculated for the 2L and 4L reflexes are plotted over these images. The extra curve in (d) is the delay time dependence calculated neglecting the D^0X resonances. The inset shows the contour of the transmitted pulse with Δ_{D^0X} assumed to be (1) 0.035 meV and (2) 0.1 meV.

finally the low-temperature parameters of the exciton-polaritons, inherent for the bulk GaN (Table I).

The oscillator strength values, being 0.0022 and 0.0016 for the A and B excitons, respectively, are smaller than the values determined using the surface-probing reflectivity data.[10,11] In the region below $\omega_{0,A}$, the A exciton resonance affects the light propagation stronger than the remote B and, especially, C excitons. Therefore, the accuracy of its parameter determination is higher. Note that further increase of the f value for the B exciton would unrealistically diminish the A-exciton f value. The longitudinal-transverse splitting for the A excitonic series is 0.8 ± 0.1 meV, estimated as $\omega_{LT} = f_j \omega_{0,j}/\varepsilon_b$. This value is somewhat less than the splitting ~1 meV between the PL peaks, ascribed to transverse and longitudinal A exciton emission using polarization-dependent PL measurements in the same 1-mm sample.[15] The homogeneous width of 13 ± 1 μeV, assumed to be the same for all exciton-polariton resonances, is similar to the value recorded for polaritons in other direct gap semiconductor CuCl (3.4 eV room-temperature bandgap).[14] At low temperatures, this parameter has to be small in perfect crystals, being determined mostly by the acoustic phonon scattering.[16] The inhomogeneous widths of all exciton-polariton resonances do not influence markedly the light reflexes. As a result, their values cannot be derived from these experiments. Thus, we cannot exclude that the exciton-polaritons propagate at low temperatures through the GaN crystal as coherent excitations with narrow homogeneous linewidth.

TABLE I. Summary of exciton-polariton parameters of bulk hexagonal GaN derived from cw and TR transmission data, shown together with reported data obtained by a reflectivity technique. The accuracy is ±0.3 meV for the energies, being within ±10% limits for other parameters.

Sample	Parameter	A	B	C	Ref.
HVPE free standing crystals	E (meV)	3478.4	3483.4	3501.6	This work
	f	0.0022	0.0016	0.0004	
	Δ (meV)	0.85	1.0	1.0	
	Γ (μeV)	13	13	13	
LEO method grown layers	E (meV)	3479.1	3484.4	3502.7	Ref. 10
	f	0.0033	0.0029	0.0007	
	Γ_{eff} (meV)	0.2	0.7	1.2	
Homoepitaxial MOCVD layers	E (meV)	3476.7	3481.5	3498.6	Ref. 11
	f	0.0027	0.0031	0.0011	
	Γ_{eff} (meV)	0.7	1.5	3.1	

In conclusions, the simultaneous fitting of both time-resolved images and cw optical spectra gives us an opportunity of accurate determination of the exciton-polariton parameters. Our data demonstrate that the exciton-polaritons propagating within a GaN crystal at low temperature have homogeneous width of the order of 10 μeV and the oscillator strength which is 20%–30% lower than the currently accepted values. Besides, we show that the study of the light propagation is a unique way to estimate the parameters of D^0X resonances.

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