Near-field investigation of exciton dynamics under semiconductor surface

V. Vasilenko\textsuperscript{a,}\textsuperscript{*}, V. Lozovski\textsuperscript{a,b}

\textsuperscript{a} Department of Semiconductor Electronics, Kyiv National Taras Shevchenko University, Glushkov Avenue 2, Building 5, Kyiv-22, 03022, Ukraine

\textsuperscript{b} V. Lashkariov Institute of Semiconductor Physics, National Academy of Sciences of Ukraine, Nauki Avenue 45, Kyiv-28, 03028, Ukraine

Abstract

The near-field images calculation method for the semiconductor surface with the excitons generated by strong focused laser pulse was proposed. Calculation was performed using Green function method in the frame of concept of local field. The main characteristic of the proposed approach is maximal usage of the analytical calculations. The near-field images for the Si surface were studied. Developed approach is universal and could be able to find with experimental data on time-resolved near-field microscopy some parameters of exciton such as diffusion constant, relaxation time, and surface state density.

1. Introduction

Due to rapid development of micro- and nano-technologies in modern electronics the studies of semiconductor surface is the actual problem for the last 30–40 years. These studies need the non-destructive methods, which can provide obtaining the important characteristics of the surface. One of these methods developed last decades is the scanning near-field optical microscopy (SNOM) [1–6]. Unfortunately, SNOM becomes useless for the researching fast dynamical processes such as relaxation, transporting, dephasing of the electrons and the excitons in the subsurface domains of semiconductor, because this researching needs as the high spatial resolution as the high temporal resolution. Ultra-fast scanning near-field optical microscopy i.e. UF-SNOM with combined spatial and time high resolution is able to satisfy all of these conditions [7–9]. The modulating detection in the combined SNOM supplies us with the consideration of the measured local-field distribution as static. This favorable circumstance allows us to use the near-field images calculations method developed earlier for other problems [10–12]. Then, the near-field images calculations method was proposed in this paper for the semiconductor surface with the inhomogeneous exciton distribution. The method is based on the formally exact solution of Lippman–Schwinger equation. The near-field images computation for Si surface with Gauss-like exciton distribution was proposed in this work. Then, the developed approach could be able for the analysis of the experimental data on the ultra-fast near-field microscopy when the power fast pulses of the strong-focused Gauss-like laser beam radiates the semiconductor surface.

2. The linear response of the inhomogeneous exciton distribution

Let us consider the semiconductor surface, which was acted by the strong focused Gauss-like light pulse with the energy of quantum equal to the energy of the exciton generation. The near-field images computation for Si surface with Gauss-like exciton distribution was proposed in this work. Then, the developed approach could be able for the analysis of the experimental data on the ultra-fast near-field microscopy when the power fast pulses of the strong-focused Gauss-like laser beam radiates the semiconductor surface.
the shape of intensity profile of the generating light and it is Gaussian-like in XY plane (parallel to the semiconductor’s surface). However, it is determined by the Burger–Lambert law of the absorption along Z-axis (normally to the semiconductor surface):

$$\delta n(x, y, z, t = 0) = n_m e^{-\frac{x^2 + y^2}{\sigma^2}},$$

(1)

with \(n_m\) is the maximum of exciton concentration which is achieving after action of light pulse in the center of the light spot at the surface of semiconductor, \(\sigma_{xy}\) is the half-width of the Gauss-like distribution in plane XY and \(\sigma_z\) is the characteristic length of the light penetration inside the sample. The \(\sigma_{xy}\) parameter is determined by the width of the light pulse which generates excitons and the \(\sigma_z\) parameter determines the absorption properties of the semiconductor and it can be expressing via absorption factor \(\alpha\) as \(\alpha = 1/2\sigma_z\).

After stopping of light pulse action the distribution of the excitons at the subsurface layer will be changed due to processes of diffusion and relaxation. In order to describe the evolution of nonequilibrium exciton distribution (1), one should write the continuity equation

$$\frac{\partial \delta n}{\partial t} = -\frac{1}{c} \text{div}(\delta n) - R,$$

(2)

where \(\delta n\) is excess exciton density,

$$R = -\frac{\delta n}{\tau},$$

(3)

is the relaxation velocity, where \(\tau\) is exciton lifetime. The current \(\delta j\) consists of only diffusion component

$$\delta j_D = -D \nabla \delta n,$$

(4)

where \(D\) is diffusion constant.

The substitution of Eqs. (3) and (6) into Eq. (2) gives the next equation

$$\frac{\partial \delta n}{\partial t} = D \Delta (\delta n) - \frac{\delta n}{\tau},$$

(5)

The boundary condition, which takes into account the influence of the semiconductor surface, consists in that the surface relaxation current is equal to the diffusion current at the surface

$$J_S = J_D|_{z=0}.$$

(6)

Obviously, this boundary condition can be rewritten in the form

$$\frac{\partial \delta n}{\partial z}|_{z=0} = -\frac{s}{D} \delta n,$$

(7)

where \(s\) is surface relaxation rate, which depends on surface state density of the semiconductor under consideration.

In order to find the expression which describes an evolution of nonequilibrium exciton concentration one needs to solve Eq. (5) with boundary conditions [Eqs. (1) and (7)]. For simplifying Eq. (5) the next replacement is used

$$\delta n(x, y, z, t) = e^{-i\omega t} u(x, y, z, t).$$

(8)

The substitution of Eq. (8) into Eq. (5) gives the simple form of the equation

$$\frac{\partial u}{\partial t} = D \Delta u,$$

(9)

with boundary conditions

$$u(x, y, z, t = 0) = n_m e^{-\frac{x^2 + y^2}{\sigma^2}}, \quad \frac{\partial u}{\partial z}|_{z=0} = -\frac{s}{D} u.$$

(10)

The solution of Eq. (9) is

$$u(x, y, z, t) = \int_{-\infty}^{\infty} dx' \int_{-\infty}^{\infty} dy' \int_{-\infty}^{\infty} dz' u(x', y', z', t = 0) g(x, y, z, x', y', z', t),$$

(11)

where \(g(x, y, z, x', y', z', t)\) is the Green function of Eq. (9) with boundary conditions (10). The expression for the Green function \(g(x, y, z, x', y', z', t)\) was found in Ref. [13] and equals to

$$g(x, y, z, x', y', z', t) = \frac{1}{4\pi Dt} e^{\frac{x^2 + y^2}{4Dt \sigma^2}} \left( \frac{1}{4\pi Dt} \right)^\frac{1}{2} e^{\frac{(x-x')^2}{4Dt \sigma^2}} + \frac{1}{4\pi Dt} e^{\frac{(x-x')^2}{4Dt \sigma^2}} \frac{s}{D} e^{\frac{(z+z')^2}{4Dt \sigma^2}} \text{Erfc} \left( s \sqrt{\frac{1}{4Dt} + z + z'} \right).$$

(12)

Using the next standard integral

$$\int_0^\infty e^{-x} \text{Erfc}(b + cx) dx = \begin{cases} c < 0, & \text{Erfc}(b) + e^{b^2 - c^2} (\text{Erfc} \left( b + \frac{1}{2} \sqrt{c^2} \right) + 1) \\ c = 0, & \text{Erfc}(b) \\ c > 0, & \text{Erfc}(b) - e^{b^2 - c^2} \text{Erfc} \left( b + \frac{1}{2} \sqrt{c^2} \right) \end{cases}$$

(13)

the integration in Eq. (11) can be performed. Then, with taking into account Eqs. (8) and (11), the nonequilibrium concentration of excitons can be obtained in the form

$$\delta n(x, y, z, t) = n_m e^{-i\omega t} e^{-\frac{x^2 + y^2}{\sigma^2}} \sigma_{xy} + 2Dt \left( \frac{1}{2} e^{\frac{x^2 + y^2}{4Dt \sigma^2}} \text{Erfc} \left( \sqrt{\frac{Dt}{2\sigma_x}} \frac{z}{2\sigma_z} \right) + e^{\frac{x^2 + y^2}{4Dt \sigma^2}} e^{\frac{(z-z')^2}{4Dt \sigma^2}} \text{Erfc} \left( \sqrt{\frac{Dt}{2\sigma_x}} \frac{z + z'}{2\sigma_z} \right) - 2\sigma_z \sigma_{xy} \frac{s}{2\sigma_x - D} \left( e^{\frac{x^2 + y^2}{4Dt \sigma^2}} e^{\frac{2s\sigma_z}{2\sigma_x - D}} \text{Erfc} \left( \frac{z}{2\sigma_x} + s \sqrt{\frac{1}{4Dt}} \right) + e^{\frac{x^2 + y^2}{4Dt \sigma^2}} e^{\frac{(z-z')^2}{4Dt \sigma^2}} e^{\frac{2s\sigma_z}{2\sigma_x - D}} \text{Erfc} \left( \frac{z + z'}{2\sigma_x} + s \sqrt{\frac{1}{4Dt}} \right) \right) \right).$$

(14)

Then, the linear response of the system can be written as

$$\chi(\vec{r}) = \chi(\omega, n_m) e^{-i\omega t} e^{-\frac{x^2 + y^2}{\sigma^2}} \sigma_{xy} + 2Dt \left( \frac{1}{2} e^{\frac{x^2 + y^2}{4Dt \sigma^2}} \text{Erfc} \left( \sqrt{\frac{Dt}{2\sigma_x}} \frac{z}{2\sigma_z} \right) + e^{\frac{x^2 + y^2}{4Dt \sigma^2}} e^{\frac{(z-z')^2}{4Dt \sigma^2}} \text{Erfc} \left( \sqrt{\frac{Dt}{2\sigma_x}} \frac{z + z'}{2\sigma_z} \right) - 2\sigma_z \sigma_{xy} \frac{s}{2\sigma_x - D} \left( e^{\frac{x^2 + y^2}{4Dt \sigma^2}} e^{\frac{2s\sigma_z}{2\sigma_x - D}} \text{Erfc} \left( \frac{z}{2\sigma_x} + s \sqrt{\frac{1}{4Dt}} \right) + e^{\frac{x^2 + y^2}{4Dt \sigma^2}} e^{\frac{(z-z')^2}{4Dt \sigma^2}} e^{\frac{2s\sigma_z}{2\sigma_x - D}} \text{Erfc} \left( \frac{z + z'}{2\sigma_x} + s \sqrt{\frac{1}{4Dt}} \right) \right) \right).$$

(15)

where \(\chi(\omega, n_m)\) is maximal susceptibility of exciton cloud, which equals to [14,15]

$$\chi(\omega, n_m) = \frac{\epsilon_0 \omega \sigma_{xy}}{\epsilon_0 \omega - \omega^2 + i\alpha n_m n_m},$$

(16)

where \(\omega\) is frequency of the incident light, \(\varepsilon\) is dielectric constant of the semiconductor, \(\omega_0\) is resonance frequency of the excitons, \(a_0\) is Bohr radius, \(\gamma\) is homogeneous broadening and \(\omega_{LT}\) is exciton longitudinal-transverse splitting.

Then, the problem reduces to calculation of the near-field image of the “object” which is the undersurface domain of inhomogeneous distribution of excitons, which is characterized by linear response [Eq. (15)].

3. Modeling of SNOM illumination

The semiconductor surface with the nonequilibrium exciton distribution is scanning by the illuminating mode of SNOM. The phase-detecting scheme of the experiment [16–18] allows consider that the probe detects the local-field distribution as static in any point of time (because it allows run experiment with time
resolution ~100 fs). The experimental setup of calculation is shown in Fig. 1. The probe, which linear dimension is much less than the characteristic linear dimensions of the object, is moved along the scanning plane. The field radiated by the probe generates the currents inside the object. Those currents generate the field, which is detected by the detector in far-zone. This field is strongly dependent on the distribution of the local field inside the object. The intensity of the field at the as function of coordinates of the probe, which usually named as near-field image (NFI) can be written as [10–12]

\[ N_{\mu}(\vec{R}_p, \omega) = \left| \int_{V_s} d\vec{R}_q \chi_{\mu}(\vec{R}, \vec{R}_p, \omega) \right|^2, \]  

(17)

Eq. (17) determines the dependence of intensity of i-component of the field at the detector on the probe coordinates when the j-component of external field, \( \chi(\vec{R}, \vec{R}_p, \omega) \) is an effective susceptibility tensor which connects local currents inside the object with the external field \( \vec{E}(\vec{R}, \omega) \).

\[ f(\vec{R}, \vec{R}_p, \omega) = \chi(\vec{R}, \vec{R}_p, \omega) \vec{E}(0)(\vec{R}, \omega). \]  

(18)

The effective susceptibility takes into account near-field interaction in the system. The integration in Eq. (17) is over volume localization of the excitons \( V_s \). \( \vec{R}_p \) is the probe coordinate. Eq. (17) means that computation of near-field images needs to calculate the effective susceptibility of the considered system.

Calculation of the effective susceptibility \( \chi(\vec{R}, \vec{R}_p, \omega) \) needs to use the Green function method which consists in solution of Lippmann–Schwinger equation [10–12] (here and below variable \( \omega \) will be omitted)

\[ E_i(\vec{R}) = E_i^{(0)}(\vec{R}) - \int_{V_s} d\vec{R}_q \chi_{ij}(\vec{R}, \vec{R}_p) \chi_{ji}(\vec{R}) E_j(\vec{R}_q), \]  

(19)

with

\[ \chi_{ij}(\vec{R}, \vec{R}_p) = G_{ij}^{(+)}(\vec{R}, \vec{R}_p) - \nu P \delta_{ij} G_{ij}^{(-)}(\vec{R}, \vec{R}_p) G_{ij}^{(+)}(\vec{R}, \vec{R}_p), \]  

(20)

generalized photon propagator [10] which is taking into account the probe. In this equation \( P \) is the electrical susceptibility of the probe, \( \nu \) is its volume, \( G_{ij}^{(+)}, G_{ij}^{(-)}, G_{ij}^{(+)} \) are Green functions which describe propagation of electromagnetic field in the system with a flat interface [23].

The solution of Eq. (19) can be written via the effective susceptibility, which has a sense of a self-consistent response on the external field [10–12]

\[ E_i(\vec{R}) = E_i^{(0)}(\vec{R}) - \int_{V_s} d\vec{R}_q \chi_{ij}(\vec{R}, \vec{R}_p) \chi_{ji}(\vec{R}) E_j(\vec{R}_q), \]  

(21)

where the effective susceptibility is equal to

\[ \chi_{ij}(\vec{R}, \vec{R}_p) = \left[ \delta_{ij} + \int_{V_s} d\vec{R}_q \chi_{ij}(\vec{R}, \vec{R}_p) \chi_{ji}(\vec{R}) \right]^{-1}. \]  

(22)

Then the near-field imaging reduces to calculation the integral in Eq. (17).

Let us consider such SNOM configuration that the frequency of the external field microscopy \( \omega \) is not equal to resonance frequency of the exciton \( \omega_0 \). In this case, contribution of the excitons in optical constant is small [see Eq. (16)] and the exciton distribution satisfy the next inequality

\[ \chi(\omega, n_m) \chi(\vec{R}) \ll 1. \]  

(23)

It means that the system (which is described by Eq. (21)) is not resonance, therefore the integral term in the brackets of Eq. (22) can be considered as small parameter. Therefore, the effective susceptibility could be rewritten in the form of Born expansion

\[ \chi_{ij}(\vec{R}, \vec{R}_p) = \chi(\vec{R}) \left[ \delta_{ij} - \int_{V_s} d\vec{R}_q \chi_{ij}(\vec{R}, \vec{R}_p) \chi(\vec{R}) \right]. \]  

(24)

This equation can be base of numerical calculations.

4. Numerical calculations

As it was noted above, the near-field image of the inhomogeneous exciton distribution is defined by the effective susceptibility of the system under consideration (see Eq. (17)). The analysis of Eq. (24) shows that the inequality (23) allows us to consider the initial susceptibility (15) as first approximation (\( \chi(\vec{R}) \) is small parameter) of the effective susceptibility (24). Then the intensity at SNOM detector in first approximation is equal to

\[ N_{\mu}^{(0)} = \left| \int_{V_s} d\vec{R}_q \chi(\vec{R}) \right|^2. \]  

(25)

As one can see from Eq. (25), the \( N_{\mu}^{(0)}(t) \) does not depend on the probe position, therefore this is a background level (zero level) of the near-field images which does not change when the probe is scanning along the surface. From the other hand \( N_{\mu}^{(1)}(t) \) is changing with time, because susceptibility [Eq. (15)] depends on a time. The value \( N_{\mu}^{(1)}(t) \) can be measured with UF-SNOM experiments. The similar results can be obtained with using so-called far-field spectroscopy. Taking into account Eq. (13), one can calculate \( N_{\mu}^{(1)}(t) \) and obtain

\[ N_{\mu}^{(1)}(t) = 4\pi \sigma^2_s \sigma_{\mu} \chi(\sigma, n_m) e^{i \omega t} \left( \frac{D_0^2}{D - 2s^2} \right) \text{Erfc} \left( \sqrt{\frac{2}{4\sigma^2_s}} t \right)^2. \]  

(26)

The system under consideration is characterized by two recombination processes: the bulk recombination and the surface recombination. It means that one should introduce two relaxation times \( \tau_S = D s^2 \) – the time, which characterizes the influence of surface relaxation; \( \tau_D = 4\sigma^2_s / D \) – the time, which characterizes the relaxation by diffusion processes in the bulk. Then, using the designations for \( \tau_S \) and \( \tau_D \), one can rewrite Eq. (26) in the form

\[ N_{\mu}^{(1)}(t) = e^{\frac{-t}{\tau_S}} \left( \frac{\sqrt{\tau_S} e^{\frac{D}{2\tau_D}} \text{Erfc} \left( \sqrt{\frac{t}{\tau_S}} \right)}{\sqrt{\tau_S} - \sqrt{\tau_D} e^{\frac{D}{2\tau_D}} \text{Erfc} \left( \sqrt{\frac{t}{\tau_D}} \right)} \right)^2. \]  

(27)

where the renormalization on the maximal value of \( N_{\mu}^{(1)}(t) \) was performed.

One should note that Eq. (27) is valid in the case of the near-field approximation (when the linear dimension of the nonequilibrium excitons location area is less than wavelength of the probing beam: \( \sqrt{\sigma^2_s + 2D\tau} < \lambda \)). This inequality leads to limitation on measuring time.
\[
N_{ij}^{(0)}(t) = e^{-\frac{t}{\tau_{SD}}}.
\] (28)

Let us analyze now the dependence of the NFI level background [Eq. (27)] on the surface relaxation rate. In the case of low influence of the surface relaxation \(\tau_S \gg \tau_{SD}\), the expression for the level background is equal to
\[
N_{ij}^{(0)}(t) = e^{-\frac{2t}{\tau_{SD}}}.
\] (29)

Therefore, in this case the diffusion processes do not influence on the zero level, only bulk relaxation processes leads to change of the near-field image background. In the case of strong surface relaxation influence (\(\tau_S \ll \tau_{SD}\)), the expression for background level has the next form
\[
N_{ij}^{(0)}(t) = e^{-\frac{t}{\tau_{SD}}} \left[ \left( \frac{t}{\tau_{SD}} \right) \right]^2.
\] (30)

Then, the curves which describe the time dependence of the NFI level (with different surface relaxation rate) should be situated between the curves defined by Eqs. (29) and (30) as it shown in Fig. 2.

When the relaxation in the system is slow \(\tau \gg \tau_S, \tau_{SD}\), the expression for background level is equal to
\[
N_{ij}^{(0)}(t) = \left( \frac{t}{\tau_{SD}} \right) \left[ \left( \frac{t}{\tau_{SD}} \right) \right]^2.
\] (31)

For short time between the generation of currents and the act of observation, namely, when measurement time \(t < \tau_S, \tau_{SD}\), the zero level has the next form
\[
N_{ij}^{(0)}(t) = e^{-\frac{3t}{\tau_{SD}}}.
\] (32)

Because of the time dependence of \(N_{ij}^{(0)}(t)\) can be measured with UFSNOM and far-field spectroscopy experiments, one could suppose, that expressions [Eq. (27), (29)–(32)] can be used for computation of the experimental data of time-resolved near-field microscopy such parameters of semiconductor surface as diffusion constant \(D\), relaxation time \(\tau\), and surface relaxation rate \(s\). For example, using Eq. (32) one can calculate the parameters \(\tau\) and \(\sqrt{\tau_{SD}D} = s/\sigma_z\).

Using Eqs. (24) and (17), let us find of the electromagnetic field intensity detected by the SNOM detector as the second approximation on the small parameter \(z(\hat{R})\)
\[
N_{ij}(\hat{R}, \sigma) = \int_{\hat{V}_S} d\vec{R} \chi(\hat{R}) \left[ \delta_{jk} \int_{\hat{V}_S} d\vec{R} \chi(\hat{R}) \right] \left[ \delta_{ij} \int_{\hat{V}_S} d\vec{R} \chi(\hat{R}) \right] \chi(\hat{R})^2.
\] (33)

Eq. (33) can be rewritten by using the expression for the generalized photon propagator [Eq. (20)]

\[
N_{ij}(\hat{R}, \sigma) = \sum_{\kappa} \int_{\hat{V}_S} d\vec{R} \chi(\hat{R}) \left[ \delta_{jk} \int_{\hat{V}_S} d\vec{R} \chi(\hat{R}) \right] \left[ \delta_{ij} \int_{\hat{V}_S} d\vec{R} \chi(\hat{R}) \right] \chi(\hat{R})^2.
\] (34)

One should note that the first and second terms in the right part of Eq. (34) do not depend on the probe position. Therefore, these terms describe the background level of NFI. The third term in the right part of Eq. (34) determines the NFI.

To calculate the near-field images one needs to evaluate the integrals in Eq. (34). To evaluate these integrals one needs to know the explicit form of Green functions \(G_{ij}^{(0)}(\hat{R}, \hat{R}')\), \(G_{ij}^{(1)}(\hat{R}, \hat{R}')\) and \(G_{ij}^{(2)}(\hat{R}, \hat{R}')\). Calculation of these Green functions was made in the framework of approach reported in [19]. Namely, one performed the inverse Fourier transformation of the Green function of two semi-spaces with flat plane interface written in \(k, z\)-representation [20,21]. Using the near-field approximation, one obtains
\[
\tilde{G}^{(0)}(\vec{r}, \vec{r}') = \frac{2k_0^2}{\varepsilon + 1} \tilde{D}^{(0)}(\vec{r}, \vec{r}'),
\] (35)
\[
\tilde{G}^{(1)}(\vec{r}, \vec{r}') = \frac{2k_0^2}{\varepsilon + 1} \tilde{D}^{(0)}(\vec{r}, \vec{r}') + \frac{k_0^2}{\varepsilon} \tilde{D}^{(0)}(\vec{r}, \vec{r}') \tilde{M}.
\] (36)
\[
\tilde{G}^{(2)}(\vec{r}, \vec{r}') = \frac{2k_0^2}{\varepsilon + 1} \tilde{D}^{(0)}(\vec{r}, \vec{r}') + \frac{k_0^2}{\varepsilon} \tilde{D}^{(0)}(\vec{r}, \vec{r}') \tilde{M}.
\] (37)

The next designations were used in Eqs. (35)–(37): the photon propagator of free space written in the near-field approximation [22]
\[
\tilde{D}^{(0)}(\vec{r}, \vec{r}') = \frac{1}{4\pi} \left[ \frac{1}{k_0^2 \vec{R} \cdot \vec{R} - 3} \tilde{\mathbf{U}} - \frac{3}{k_0^2 \vec{R} \cdot \vec{R}} \tilde{\mathbf{e}}_z \tilde{\mathbf{e}}_z \right].
\] (38)

where \(\tilde{\mathbf{U}}\) is unit dyadic, \(\tilde{\mathbf{R}} = \vec{r} - \vec{r}'\), \(\mathbf{R} = |\vec{r} - \vec{r}'|\), \(\tilde{\mathbf{e}}_z = \vec{R}/R\).
\[
\tilde{M} = \frac{1 - \varepsilon}{1 + \varepsilon}
\] (39)

\(\tilde{\mathbf{r}}_S = (x, y, z)\), and \(\varepsilon\) is the dielectric constant of the semiconductor, \(k_0 = \omega/c\), \(c\) is the light velocity. One should note that in the near-field approximation, the photon propagators [Eqs. (35)–(37)] consist only of a real part in the case when the semiconductor dielectric constant is real. This circumstance very simplifies the numerical calculations.

As an illustration, we consider a surface of Si (under temperature about 10 K) with Gaussian distribution of excitons (1). The distribution of excitons was characterized by maximum concentration \(n_m = 10^{17} \text{ cm}^{-3}\), and dimensions of the inhomogeneity domain \(\sigma_x = \sigma_z = 0.2 \lambda = 1 \mu\text{m}\) is the wavelength of the external probing field). The excitons were characterized by the next parameters: diffusion constant \(D = 12 \text{ cm}^2/\text{sec}\), lifetime \(\tau = 10^{-11}\text{cs}\), dielectric constant of the semiconductor \(\varepsilon = 11.6\), resonance frequency of the exciton \(\omega_B = 1.2 \text{eV}\), Bohr radius \(a_B = 4.3 \text{nm}\), homogeneous broadening of the exciton resonance caused by acoustic phonons \(\gamma = 0.01 \text{meV}\), exciton longitudinal-transverse splitting \(\omega_{lt} = 0.07 \text{meV}\). The probe radius was chosen much less than the wave of long-range monochromatic external field \(\tau_p \ll \lambda\), namely \(\tau_0 = 5 \text{nm}\) at the wavelength \(\lambda = 1 \mu\text{m}\). The dielectric constant of the probe was supposed as \(\varepsilon_{pro} = 2.25\) (glass). Time relaxation was chosen as \(\tau = \sigma_z^2/2D\).

The curves of \(N_{ij}^{(0)}(t)\) are shown in Fig. 2 for infinite and zero surface relaxation rates. The background levels of NFI are shown in arbitrary units. The results of numerical calculations of the near-field images are shown in Figs. 3 and 4. Coordinates in the figures.

![Fig. 2. Dependence level of NFI on time.](image-url)
are given in arbitrary units – \( a/\lambda \), with \( a \) any parameter of dimension. The intensity of the near-field images are represented as normalized. The distance between the scanning plane and semiconductor surface is equal to 0.2 \( \lambda \) for all near-field images calculated in this work. Every figure contains the graphics of NFI sections along the line \( y = 0 \), the polarization the both illumination and detected field are parallel to \( X \)-axis.

The NFI are calculated for three points of time \( t = 0, 0.2 \tau, 0.7 \tau \), and for two surface relaxation rates \( s = 0 \) (Fig. 3) and \( s = \infty \) (Fig. 4). Evidently, that near-field images intensity is decreasing

**Fig. 3.** Near-field images of the exciton distribution at three point of a time for zero surface relaxation rate \( s = 0 \).

**Fig. 4.** Near-field images of the exciton distribution at three point of a time for infinite surface relaxation rate \( s = \infty \).
with time. The decreasing in Fig. 3. is less than the corresponding decreasing in Fig. 4. This fact can be explained by influence of the surface relaxation. The shape of NFI for zero surface relaxation rate is very similar (but not equal) to the shape of the corresponding NFI for infinite surface relaxation rate. It can be explained by the fact that the exciton distribution shape in the plane XOY (see Eq. (14)) does not depend on surface relaxation rate \( s \) (The exciton distribution shape depends on surface relaxation only along OZ-axis). The broadening of peaks of field intensity with time one can see from Figs. 3 and 4. The reason of the broadening is diffusion process.

All of those theoretical results mean that the studying of evolution process of NFI and zero level allows to determine with sub-micron resolution and in noncontact manner such properties as exciton diffusion constant \( D \), relaxation time \( \tau \), surface relaxation rate \( s \), half-width \( \sigma_z \) and accordingly absorption factor \( \alpha \).

5. Conclusions

The calculation method for ultra-fast near-field microscopy of the semiconductor surface with the inhomogeneous exciton distribution is proposed. The calculations were performed using Green function method and based on solving the equation for self-consistent field (Lippmann–Schwinger equation). The solution of Lippmann–Schwinger equation was obtained in analytical form in the frame of local-field concept. Moreover, this solution allows to obtain the analytical expression for the intensity of the scattered far-field at the SNOM detector. This intensity can be found in the ultra-fast scanning optical near-field microscopy experiments. Therefore, the usage the obtained analytical expressions for description the experimental data allows to determine such semiconductor parameters as diffusion constant, relaxation time, and surface recombination rate. One should note that those semiconductor parameters could be found with submicron resolution. Moreover, the proposed method for determination of parameters, which describe the properties semiconductor surface, is nondestructive because the ultra-fast scanning near-field optical microscopy is noncontact.

References