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Energy distributions of 2D excitons in the presence of nonequilibrium phonons

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Abstract. For the first time we present the results of an analysis of the energy distribution function of 2D excitons for fixed nonequilibrium phonon spectra. The calculations are based on the kinetic equation for the interacting 2D exciton and 3D acoustic phonon gases. For the low-density exciton gas in GaAs/AlGaAs quantum wells (QW), we find a strong dependence of the exciton energy distribution on the QW width and nonequilibrium phonon spectrum.

1. Introduction

In recent experiments [1] it was shown that the heating of two-dimensional exciton gas (2DExG) by nonequilibrium phonons depends strongly on the width (*d*) of the quantum well (QW) and the exciton density (n_s) . The experimental results evidently show that the energy distribution of the 2DExG for $n_s \ll 10^{10}$ cm⁻² in the presence of nonequilibrium phonons becomes essentially a non-Boltzmann distribution. A qualitative explanation was given in [1], which is based on the selection rules for the exciton–phonon interaction in 2D semiconductor structures.

The nonequilibrium energy distribution f(E) of the 3D excitons in bulk semiconductors was calculated earlier [2] in connection with 'hot-spot' experiments [3]. In the present paper we calculate the energy distribution f(E) for 2D excitons interacting with 3D non-Planckian phonons. The quantum confinement of excitons has an effect on selection rules for the exciton-phonon interaction and makes the analysis quite different from that in the 3D case [2]. We will separately analyse the role of LA and TA phonons taking into account the anisotropy of the coupling matrix element for exciton scattering by phonons. The system of low-density 2DExG differs strongly from the degenerate two-dimensional electron gas, where due to the strong electron-electron scattering the energy distribution is close to Fermian and a certain temperature of 2D electron gas may be introduced. Another specific feature of excitons is their finite lifetime τ_0 . In high-quality GaAs/AlGaAs QWs, $\tau_0 \sim 10^{-9}$ s [4], which is much larger than the characteristic energy exciton–acoustic phonon relaxation time $\tau_{ex-ph} \sim 10^{-11}$ -10⁻¹⁰ s [5–7]. Due to the condition $\tau_0 \gg \tau_{ex-ph}$ one can consider the 2DExG to be in equilibrium with nonequilibrium phonons, neglect generation and recombination processes in the kinetic equation and normalize f(E) to the given exciton density n_s . We will also neglect the exciton-exciton collisions $(\tau_{ex-ex} \gg \tau_{ex-ph})$ and assume that the 2DExG has no effect on the nonequilibrium phonon occupation numbers N_{ω} , which are determined by the phonon generator and the geometry of the experiment [1].

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2. Theory

Under the above assumptions the kinetic equation for the exciton energy distribution $f(E_k)$ is reduced to

$$\sum_{k'} \left[W_{k' \to k} f(E_{k'}) - W_{k \to k'} f(E_k) \right] = 0.$$
(1)

Here k and E_k are the exciton in-plane wave vector and kinetic energy $(E_k = \hbar^2 k^2/2m)$; the transition probability is given by

$$W_{k \to k'} = \frac{2\pi}{\hbar} \sum_{q,\nu} \left| M_{k \to k'}^{q,\nu} \right|^2 \left(N_{\omega_{\nu}(q)} + \frac{1 \pm 1}{2} \right) \delta(E_{k'} - E_k \pm \hbar \omega_{\nu}(q))$$
(2)

where $M_{k \to k'}^{q, \nu}$ is the matrix element for the $k \to k'$ transition with emission (+) or absorption (-) of an acoustic phonon characterized by the polarization $\nu = \text{LA}$, TA, the 3D wave vector q and frequency $\omega_{\nu}(q) = s_{\nu}q$, where s_{ν} is the sound velocity.

The exciton envelope function in the state with the wave vector k can be written as

$$\Psi_{k}(\boldsymbol{r}_{e},\boldsymbol{r}_{h}) = \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_{\parallel}}F(\rho)\,\varphi_{e}(\boldsymbol{z}_{e})\,\varphi_{h}(\boldsymbol{z}_{h}) \tag{3}$$

where r_e , r_h are the electron and hole positions, R is the position of the exciton centre of mass, z is the growth direction, \parallel indicates the vector component in the QW plane, $\varphi_{e(h)}(z)$ is the electron (hole) wave function for the first size-quantized level, $\rho = (r_e - r_h)_{\parallel}$, and $F(\rho)$ is a function describing the relative electron–hole motion. In the simplest variational approach,

$$F(\rho) = \sqrt{\frac{2}{\pi a_0^2}} e^{-\rho/a_0}$$
(4)

with a_0 being the 2D effective exciton radius.

In GaAs/AlGaAs QWs the following mechanisms contribute to the exciton–acoustic phonon interaction: the deformation potential (DP) and the piezo-acoustic (PA) mechanism. The DP interaction Hamiltonian for creation (annihilation) of one acoustic phonon of wave vector q and mode v can be written as

$$H_{ex-ph}^{DP} = H_{e-ph}^{DP} + H_{h-ph}^{DP} = \sqrt{\frac{\hbar}{2\rho_0 V s_\nu q}} iq \left(\Xi_e e^{\pm iq \cdot r_e} + \Xi_h e^{\pm iq \cdot r_h}\right)$$
(5)

where Ξ_e , Ξ_h are the electron and hole deformation potential constants, ρ_0 is the material density, and V is the whole volume. The PA interaction Hamiltonian is

$$H_{ex-ph}^{PA} = H_{e-ph}^{PA} + H_{h-ph}^{PA} = \sqrt{\frac{\hbar}{2\rho_0 V s_\nu q}} \beta \left(e^{\pm i q \cdot r_e} - e^{\pm i q \cdot r_h} \right)$$
(6)

where β is the exciton average PA constant.

Note that in fact the deformation potential for holes is anisotropic:

$$H_{h-ph}^{DP} = \left(a + \frac{b}{2}\right) \left(u_{xx} + u_{yy}\right) + (a - b) u_{zz}$$
(7)

where the diagonal components of the deformation tensor are

$$u_{\alpha\alpha} = \sqrt{\frac{\hbar}{2\rho_0 V s_\nu q}} \mathbf{i} e_\alpha^{(q,\nu)} q_\alpha \mathbf{e}^{\pm \mathbf{i} \boldsymbol{q} \cdot \boldsymbol{r}_h}$$

and $e^{(q,v)}$ is the polarization unit vector. Therefore Ξ_h in equation (5) is a function of the angle between q and the plane of the QW:

$$\Xi_{h}(\boldsymbol{q}) = \begin{cases} a + \frac{b}{2} - \frac{3}{2}b\left(\frac{q_{z}}{q}\right)^{2} & \text{for LA phonons} \\ -\frac{3}{2}b\left(\frac{q_{z}q_{\parallel}}{q^{2}}\right)^{2} & \text{for TA phonons.} \end{cases}$$
(8)

Since the conduction electrons interact only with LA phonons, Ξ_e vanishes for the exciton–TA phonon interaction.

Calculating the matrix elements

$$M_{k \to k'}^{q,\nu} = \langle \Psi_k | H_{ex-ph} | \Psi_{k'} \rangle$$

and substituting into equation (2) we obtain the expressions for transition probabilities:

$$W_{\boldsymbol{k}\to\boldsymbol{k}'}^{DP} = \frac{\Xi^2(\boldsymbol{q})}{\hbar\rho_0 S_0 s_{\nu}^2} \left(N_{s_{\nu}q} + \frac{1\pm 1}{2} \right) \frac{q^2}{q_z} \Theta\left(\boldsymbol{q} - \boldsymbol{q}_{\parallel}\right)$$
(9)

$$W_{k \to k'}^{PA} = \frac{\beta^2(q)}{\hbar \rho_0 S_0 s_{\nu}^2} \left(N_{s_{\nu}q} + \frac{1 \pm 1}{2} \right) \frac{1}{q_z} \Theta \left(q - q_{\parallel} \right)$$
(10)

where $q = (\hbar/2ms_{\nu})|k^2 - k'^2|$, $q_{\parallel} = |\mathbf{k} - \mathbf{k}'|$, $q_z = \sqrt{q^2 - q_{\parallel}^2}$, S_0 is the sample area, and $\Theta(x)$ is the Heaviside step function:

$$\Theta(x) = \begin{cases} 1 & x \ge 0 \\ 0 & x < 0. \end{cases}$$

The effective exciton DP and PA constants are given by

$$\Xi(\boldsymbol{q}) = \Xi_e Z_e(q_z) \Big/ \left[1 + \left(\frac{m_h}{2m} q_{\parallel} a_0\right)^2 \right]^{3/2} + \Xi_h(\boldsymbol{q}) Z_h(q_z) \Big/ \left[1 + \left(\frac{m_e}{2m} q_{\parallel} a_0\right)^2 \right]^{3/2}$$
(11)

$$\beta(q) = \beta \left\{ Z_e(q_z) / \left[1 + \left(\frac{m_h}{2m} q_{\parallel} a_0\right)^2 \right]^{3/2} - Z_h(q_z) / \left[1 + \left(\frac{m_e}{2m} q_{\parallel} a_0\right)^2 \right]^{3/2} \right\}.$$
 (12)

 $Z_{e,h}$ are the overlap integrals defined as

$$Z_{e,h}(q_z) = \int_{-\infty}^{\infty} \mathrm{d}z \; \varphi_{e,h}^2(z) \mathrm{e}^{\mathrm{i}q_z z}.$$

 m_e, m_h are the electron and hole effective masses.

In the approximation of infinitely high barriers

$$Z_e(q_z) = Z_h(q_z) = \frac{\sin(q_z d/2)}{(q_z d/2) \left[1 - (q_z d/2\pi)^2\right]}$$
(13)

where d is the QW width. This function is equal for 1 at $q_z = 0$ and decreases rapidly for $q_z \sim \pi/d$. Further, for qualitative consideration we shall suppose infinitely high barriers using equation (13) for numerical calculation of f(E), and discuss the effect of finite barriers in the next section.

3. Results and discussion

For numerical calculations of the exciton distribution function f(E) we used the parameters for the heavy-hole free exciton in GaAs/AlGaAs QWs, namely, $m_e = 0.067m_0$, $m_h = 0.15m_0$ (m_0 is the free-electron mass), $a_0 = 100$ Å, $\Xi_e = 7.3$ eV, a = -6.7 eV, b = -2 eV, $s_{LA} = 5 \times 10^5$ cm s⁻¹, $s_{TA} = 3 \times 10^5$ cm s⁻¹.

We calculated the exciton energy distribution only for the DP interaction since the PA interaction is known to be important only for very low energies $E \ll 1 \text{ meV}$ [7].

For calculation of f(E) we have chosen the nonequilibrium phonon spectrum N_{ω} consisting of only low-frequency phonons $\omega < \omega_0$, where $\hbar \omega_0$ is the high-energy cut-off which has a value of several meV. This is typical for the experiments with nonequilibrium phonons when the phonon generator and the region with 2DExG are separated by the semiinsulating GaAs substrate in which effective scattering of high-frequency phonons takes place (see [1] and references therein). The value of the cut-off ω_0 may be varied by changing the material of the substrate and the distance between the phonon generator and 2DExG. In principle the arbitrary nonequilibrium phonon spectrum N_{ω} may be used for the calculation. For example, it is known [3] that in the 'hot-spot' regime a deficit of low-energy phonons exists, which is opposite to what is the case for the high-energy ω_0 cut-off. In the present paper, to show the sensitivity of the 2DExG energy distribution f(E) to the nonequilibrium features of the phonon spectrum N_{ω} , we take

$$N_{\omega} = \left\{ \exp\left[\frac{\hbar\omega}{k_B T(\omega)}\right] - 1 \right\}^{-1}$$
(14)

where $T(\omega) = 12$ K for $\omega < \omega_0$ and $T(\omega) = 4.5$ K for $\omega > \omega_0$ (k_B is the Boltzmann constant).

The results of the calculation show that f(E) depends on the width of QW, the value of ω_0 , phonon polarization and the angular distribution of nonequilibrium phonons.



Figure 1. The energy distribution f(E) in the presence of LA nonequilibrium phonons with a high-frequency cut-off ($\hbar\omega_0 = 2$ meV) for the QWs with different widths (d). The arrow shows $E = \hbar\omega_0$.

Figure 2. The energy distribution f(E) for d = 25 Å and different cut-offs ($\hbar\omega_0$) of the LA nonequilibrium phonon spectrum. The arrows show $E = \hbar\omega_0$.



Figure 3. The energy distribution f(E) for d = 25 Å and $\hbar\omega_0 = 2$ meV, when interaction with only LA or TA phonons is taken into consideration. The arrow shows $E = \hbar\omega_0$.

Figure 4. The energy distribution f(E) for anisotropic (1) and isotropic (2) TA phonon spectra (see the text); d = 25 Å, $\hbar\omega_0 = 2$ meV. The arrow shows $E = \hbar\omega_0$.

First we present results for the isotropic distribution of nonequilibrium phonons. Figure 1 shows f(E) for different d. From comparison with the Boltzmann distribution (the dashed line) it is clearly seen that f(E) is nonequilibrium and decreases rapidly for $E > \hbar\omega_0$. The difference from the Boltzmann distribution becomes larger for narrow QWs and correspondingly the decrease of f(E) is more rapid while d decreases. Figure 2 shows f(E) for different ω_0 (d = 25 Å). It is seen that while ω_0 increases the rapid decrease for f(E) starts at higher $E \sim \hbar\omega_0$. The effect of the phonon polarization (LA, TA) on f(E) is shown in figure 3. For the parameters which were used in this calculation (see the caption to figure 3), f(E) differs strongly for LA and TA modes. For the interaction with TA modes, f(E) is almost of Boltzmann form, while f(E) is essentially nonequilibrium if only LA phonons are taken into consideration. The analysis shows that the effect of phonon polarization on f(E) is mainly due not to the difference in the sound velocities but to the strong anisotropy of the deformation potential for TA phonons—see equation (8).

The general feature in the establishing of f(E) in the presence of nonequilibrium phonons is the imbalance between emission and absorption of phonons with $\omega > \omega_0$ by 2DExG. Due to the low number of phonons with $\omega > \omega_0$ there are only few absorption transitions $\mathbf{k} \to \mathbf{k}'$ for $E_{\mathbf{k}'} - E_{\mathbf{k}} > \hbar\omega_0$, but the emission process $\mathbf{k}' \to \mathbf{k}$ may be very effective. This qualitatively should lead to the fast decay of f(E) for $E \ge \hbar\omega_0$. Moreover the calculations show the dependence of f(E) on the width, d, of the QW. This fact reflects the specific features of the exciton-phonon interaction in 2DExG.

The selection rules for the exciton-phonon transition give the maximum phonon momentum and hence the maximum phonon energy, $\hbar\omega^{\max}$, which is active in the interaction. This energy is determined by the restrictions of the *q*-projections parallel, q_{\parallel} , and perpendicular, q_z , to the plane of the QW. The momentum conservation which is included in equations (9) and (10) in the form of a Heaviside step function gives restrictions for q_{\parallel} . The maximum in-plane phonon wave vector, q_{\parallel}^{\max} , depends on *k* and the type of

the transition. For the exciton with k = 0 only an absorption process is possible and

$$q_{\parallel}^{\max}(k=0) = 2ms_{\nu}/\hbar.$$

For excitons with $k \ge 2ms_{\nu}/\hbar$, $q_{\parallel}^{\text{max}}$ differs for emission and absorption of phonons:

$$q_{\parallel}^{\max} = 2k \pm 2ms_{\nu}/\hbar.$$

For the perpendicular direction q_z is limited by the overlap integral (13), and the major contribution in the exciton-phonon transitions for LA polarization is given by phonons with $q_z^{\max} \sim \pi/d$. Hence

$$\omega_{LA}^{\max} \sim s_{LA} \sqrt{\left(q_{\parallel}^{\max}\right)^2 + \left(q_{z}^{\max}\right)^2}.$$

In GaAs the main contribution in ω_{LA}^{\max} is determined by $s_{LA}q_z^{\max}$. Apparently the value of $\hbar s_{LA}q_{\parallel}^{\max}$ is equal to 0.03 meV for the excitons at the bottom of the band and increases up to 0.3 meV for $E_k \sim 1$ meV. The value of $\hbar s_{LA}q_z^{\max}$ is much larger and has a value of 1 meV for d = 100 Å. The fact that $q_{\parallel}^{\max} \ll q_z^{\max}$ leads to the small angles θ_{LA}^{\max} between the normal to the plane of QW and q of the phonons which are absorbed or emitted by 2DExG:

$$\tan \theta_{LA}^{\max} = q_{\parallel}^{\max} / q_z^{\max} \ll 1.$$

However, this is not true for TA phonons, when $\Xi_h(q)$ is strongly anisotropic and $\Xi_e = 0$ see equation (8). This anisotropy makes phonons with small θ inactive in the interaction with excitons, and the maximum probability of the transition is found for larger angles $\theta_{TA}^{\max} > \theta_{LA}^{\max}$. As a result we have a decrease of the maximum energy and

$$\hbar\omega_{TA}^{\max} < \hbar s_{TA} \sqrt{\left(q_{\parallel}^{\max}\right)^2 + \left(q_{z}^{\max}\right)^2}$$

The restrictions in q and correspondingly in $\hbar\omega$ obviously do not effect f(E) if the phonon spectrum N_{ω} is Planckian. In that case the exciton distribution is always a Boltzmann one. A nonequilibrium f(E) appears only if N_{ω} is non-Planckian and the frequency interval $0 < \omega < \omega^{max}$ includes nonequilibrium features of N_{ω} . For the interaction with LA phonons in GaAs/AlGaAs QWs, $\hbar\omega_{LA}^{\text{max}} \sim 1 \text{ meV}$ for d = 100 Åand ~4 meV for d = 25 Å. As a result, for our model N_{ω} —see equation (14)—f(E)should be nonequilibrium if $\omega_0 < \omega^{\text{max}}$. Therefore f(E) is sensitive to the values of d and ω_0 , and to the phonon polarization. If $\omega_0 < \omega^{max}$ excitons cannot be effectively 'heated' to the energies $E > \hbar \omega_0$. This fact is clearly seen for narrow QWs and small ω_0 —see curves 1 in figure 1 and figure 2. Qualitatively, excitons with $E > \hbar \omega_0$ effectively relax, emitting phonons with $\omega_0 < \omega < \omega^{\text{max}}$, and this emission process cannot be compensated by the phonon absorption due to the deficit of phonons with $\omega > \omega_0$. In the case of wide QWs or large ω_0 we have $\omega_0 > \omega^{\text{max}}$, and excitons do not feel nonequilibrium features of N_{ω} . Hence f(E) does not differ strongly from the Boltzmann distribution—see curves 3 on figure 1 and figure 2. The dependence of f(E) on a phonon polarization (LA, TA) comes from the difference in ω^{max} ($\omega_{TA}^{\text{max}} < \omega_{LA}^{\text{max}}$). To show this we choose d and ω_0 so that f(E)differs strongly for LA and TA phonons—see figure 3, where f(E) is almost of Boltzmann form for TA phonons, while for LA phonons f(E) decreases rapidly for $E > \hbar \omega_0$.

The effect of finite barriers modifies the dependence of f(E) on d. In particular, it may be important for narrow QWs ($d \ll 100$ Å) when the exciton wave function essentially penetrates into the barriers. Then the rapid decrease of overlap integral starts at smaller q_z in comparison to the case of infinite barriers. Our calculations show that for Al_{0.33}Ga_{0.67}As barriers and QW with d = 25 Å, q_z^{max} decreases by 40% while for d = 100 Å the changes in q_z^{max} are negligible in comparison with what is found for infinite barriers. So in the case of finite barriers the well width dependence will be slightly reduced.

We analysed the case of an anisotropic phonon distribution, which is more realistic in the experiments with nonequilibrium phonons when the phonon generator and 2DExG are located on opposite sides of the GaAs substrate [1]. We consider nonequilibrium TA phonons incident and specularly reflected at the surface with 2DExG in the confined cone. To show the effect of the anisotropy of the phonon distribution we solved the kinetic equation (1) using equation (9) with phonon occupation numbers

$$N(q) = \left\{ \exp\left[\frac{\hbar s_{TA}q}{k_B T(q)}\right] - 1 \right\}^{-1}$$
(15)

where

$$T(q) = \begin{cases} 12 \text{ K} & \text{for } s_{TA}q < \omega_0 \text{ and } q_{\parallel} < q_z \\ 4.5 \text{ K} & \text{otherwise.} \end{cases}$$

The corresponding f(E) is shown in figure 4 (curve 1) for $\hbar\omega_0 = 2$ meV and d = 25 Å. The difference from the case of the isotropic phonon distribution (curve 2) is clearly seen. While for the isotropic spectrum N_{ω} , f(E) does not differ strongly from the equilibrium Boltzmann distribution, the anisotropic contribution (15) makes f(E) essentially nonequilibrium. The important feature is that the decrease of f(E) for $E > \hbar\omega_0$ becomes more rapid in the case of anisotropic N(q). The reason for this is the increasing role of the exciton transitions with absorption of phonons incident on the plane of the QW with angles $\theta < \pi/4$ ($q_{\parallel} < q_z$). This fact effectively increases $\hbar\omega^{\text{max}}$ and hence the role of high-energy phonons becomes important in the heating of 2DExG.

We also present the results of the numerical calculation of f(E) when the nonequilibrium phonon spectrum possesses a monochromatic isotropic contribution to the equilibrium (Planckian) background spectrum with T = 4.5 K:

$$N_{\omega} = P \,\delta(\hbar\omega - \hbar\omega_1) + \left[\exp\left(\hbar\omega/k_B T\right) - 1\right]^{-1}.$$
(16)

At the present time the monochromatic high-frequency spectrum (16) is not reliable experimentally because of the difficulties in the generation of monochromatic highfrequency $(10^{11}-10^{12} \text{ Hz})$ phonons. However, we expect the theoretical consideration for monochromatic phonons to be helpful for using a 2DExG as a potential phonon spectrometer. Figure 5 shows f(E) for $\hbar\omega_1 = 2$ meV and two values of P. It is seen that f(E) has resonant peaks at $E = n\hbar\omega_1$ (n = 1, 2, 3). The first peak ($E_1 = \hbar\omega_1$) is due to the absorption of a phonon by the 2D exciton with k = 0. Other peaks correspond to absorption processes involving more than one phonon (see the inset in figure 5). It is seen that resonant peaks broaden while the phonon power P increases.

4. Conclusions

Theoretical analysis and corresponding numerical calculations show that the energy distribution of 2D excitons in the presence of nonequilibrium phonons is a non-Boltzmann distribution and depends both on the width of the QW and the phonon spectrum. Our results show that f(E) reflects the features of the nonequilibrium phonon spectrum N_{ω} , and that 2DExG may be used for obtaining information about the spectrum of nonequilibrium phonons.

Experimentally, f(E) may be studied in several ways. In principle, the annihilation of excitons with simultaneous emission of optical phonons results in a luminescence spectrum



Figure 5. The energy distribution f(E) for monochromatic phonon spectrum (16); $\hbar\omega_1 = 2 \text{ meV}$, d = 25 Å. 1: P = 2.25 meV; 2: P = 0.45 meV. The inset shows the exciton transitions with absorption of a phonon of energy $\hbar\omega_1$.

which allows one to measure directly f(E) for the wide range of E (see [3] and references therein). Information about the high-energy tail of f(E) may be also obtained by studying the population of light-hole excitons whose energy is several meV higher than the bottom of the heavy-hole exciton band. This method was used in [1] to study the heating of 2DExG in wide QWs (d = 200 Å). In narrow QWs where the nonradiative processes become important the effect of population of high-energy exciton states and correspondingly the qualitative features of the high-energy tail of f(E) may be studied from the comparison of the exciton luminescence quenching in the presence of nonequilibrium phonons with equilibrium data. Such a technique was used in [1] for QWs with d = 25 Å.

Our results are in agreement with the experiment [1] where the authors obtained effective heating of the 2DExG in wide QWs (d = 200 Å) and an absence of heating by nonequilibrium phonons in narrow QWs (d = 25 Å). In [1] the authors used qualitative estimation of the maximum frequency $\omega^{\text{max}} \sim \pi s/d$ of phonons which are active in the exciton-phonon interaction. This is appropriate for LA phonons; however, care must be taken for TA phonons when the DP constant for interaction with 2DExG is strongly anisotropic. This makes f(E) sensitive to the polarization and angular distribution, N(q), of nonequilibrium phonons.

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