Pattern Competition in the Photorefractive Semiconductors

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We analytically study the photorefractive Gunn effect in n-GaAs subjected to two external laser beams which form a moving interference pattern (MIP) in the semiconductor. When the intensity of the spatially independent part of the MIP, denoted by $I_0$, is small, the system has a periodic domain train (PDT), consistent with the results of linear stability analysis. When $I_0$ is sufficiently large, the space-charge field induced by the MIP will compete with the PDT and result in complex dynamics, including driven chaos via quasiperiodic route.

KEYWORDS: semiconductors, pattern competition, nonlinear analysis

Spatiotemporal behavior of Gunn domains¹ ² in semiconductors is an interesting and important current research problem.³ ⁹ Gunn diodes are not only good systems for studying nonlinear dynamics, but also potentially important for various applications.⁷ ⁹ In 1996–1997 Segev et al.³ and Subacius et al.⁵ showed, respectively, that optical waves can excite multiple Gunn-domain formation with spatial periodicity, called periodic domain trains (PDT), in deep-impurity doped GaAs and semi-insulating GaAs; both groups used the well known Kroemer’s criterion¹ to determine the number of Gunn domains. In 1998, Bonilla et al.⁵ numerically studied the same dynamical system as Segev et al.; however, their findings are mostly inconsistent with the predictions of Segev et al. Bonilla et al.⁵ found that the number of Gunn domains in the system is not consistent with the prediction of Segev et al. and in many cases the system shows complex or chaotic behavior.

Very recently, Shiu et al. used a linear stability analysis (LSA) to study a shallow-impurity doped GaAs with length $L$ subjected to two external laser beams which form a moving interference pattern (MIP) of intensity $I(x, t) = I_0[1 + m \cos(Kx + \Omega t)]$ and predicted PDT in the Gunn diode.⁷ Here $\Omega$ is the frequency difference of the two optical waves, $K = 2\pi/\Lambda$ is the interference wave number, $\Lambda$ is very small, $L$, $m$ is the modulation depth, and $I_0$ is the average intensity. With considering of time-delay feedback, the diode will emit high-dimensional chaotic microwaves, which is potentially useful for secure microwave communications, memory devices, applications involving photorefractive effects, etc.⁷

The purpose of this manuscript is to clarify different findings of 3, ⁴ and ⁵ using a nonlinear analytic method to study the system of 7, ⁸ as $L \to \infty$. In this approach, the intensity of the spatially independent part of the MIP, $I_0$, plays an important role in determining the simple dynamics (i.e., PDT) or complex dynamics of Gunn domains. When $I_0$ is small enough, the system shows simple dynamics consistent with the result of the LSA, and Kroemer’s criterion is still valid. When $I_0$ is large, the space-charge field induced by the MIP will compete with the PDT and result in complex dynamics. This picture is also useful for understanding the results of other groups³ ⁵ discussed above.

We consider two optical waves incident on a n-GaAs with a biased voltage $V$. The photon energy is just above the bandgap of GaAs, i.e., 1.42 eV, so that electron-hole pairs can be generated by optical excitation. The generation-recombination processes include complete thermal ionization of the donor, generation of electron-hole pairs by the MIP with rate $g$, and recombination of electron-hole pairs with rate $\gamma$. Therefore we can write the dynamical equations for the electric field $E$, the free electron density $n$, and the free hole density $p$ as:

$$\frac{\partial E}{\partial x} = \frac{e}{\epsilon} (n - N_D^+ - p),$$

$$\frac{\partial n}{\partial t} = gI(x, t) - \gamma np + \frac{\partial}{\partial x} \left[ nv(E) - D_n \frac{\partial n}{\partial x} \right],$$

$$\frac{\partial p}{\partial t} = gI(x, t) - \gamma np + \frac{\partial}{\partial x} \left[ pp_f E + D_p \frac{\partial p}{\partial x} \right],$$

$$V = \int_0^L E(x, t) dx.$$  (4)

Equation (1) is for the Gauss law. Equation (2) and (3) represent the continuity equations for the electrons and holes, respectively. The circuit condition is given at eq. (4). Here $v(E)$ displays an N-shaped negative differential mobility (NDM). The homogeneous solution of eqs. (1)–(4) is $E_0 = V/L$, $n_0 = (N_D^+ + \sqrt{N_D^2 + 4gI_0/\gamma})/2$, and $p_0 = (-N_D^+ + \sqrt{N_D^2 + 4gI_0/\gamma})/2$, where $N_D^+$ is effective donor concentration. When $I_0$ is very small, $n_0 \approx N_D^+/2 + gI_0/\gamma$, and a natural expansion parameter for high order corrections is $\epsilon = p_0/N_D^+ = gI_0/N_D^2\gamma$. We can rearrange the model equations by the following rescaling process for the dynamical variables and parameters in eqs. (1)–(4):

$$E \to \epsilon E, \quad n \to n, \quad p \to \epsilon p, \quad I \to \epsilon I.$$  (4)

The small parameter $\epsilon$ is
can distinguish the large variables (i.e., $E$ and $n$) from small variable $p$ in our model. Rescaling variables in eqs. (1)–(4), we have \( \frac{\partial E}{\partial x} = \epsilon (n - N_p^{(1)})/\epsilon + O(\delta), \frac{\partial n}{\partial t} = -\partial \nu (w(E) - D_n \partial n/\partial x)/\partial x + O(\delta). \) These results and eq. (3) implies

\[
\frac{\partial E}{\partial t} = -\frac{1}{\epsilon} \epsilon N_p^{(1)} (E - v_0 x) \frac{\partial E}{\partial x} + D_n \frac{\partial^2 E}{\partial x^2} + \frac{1}{\epsilon} J_{\text{rot}}(t), 
\]

(5)

\[
\frac{\partial p}{\partial t} = g I(x, t) - \gamma (N_p^{(1)} + \frac{\epsilon}{\epsilon} \frac{\partial E}{\partial x}) p + \frac{\partial}{\partial x} \left( \rho p q + D_p \frac{\partial p}{\partial x} \right),
\]

(6)

where \( J_{\text{rot}}(t) \) is time-dependent current density. The physical meaning of eq. (5) is quite clear, which represents the Gunn-domain formation and with drift velocity \( v(E_0) \). Then, eq. (5) confirms our LSA results in ref. 7. In the following we want to show the detailed structure of PDT.

In order to get the solution of eq. (5), an ansatz of the electric field, we set \( E(x, t) = E_0 + [E(x, t)] e^{K^{(x-v_0)}} + E^{(x)}(t) e^{i K^{(x-v_0)}} \), where \( v_0 = \nu(E_0) \). \( E_0(x, t) \) is a new variable corresponding to the amplitude of electric-field domains and \( E^{(x)}(t) \) is its complex conjugate; \( K (= 2\pi / L) \) is a bulk parameter of semiconductor. Since \( K \) is small, the spatiotemporal behavior of the dynamical system within the linear dimension \( A \) is dominated by the behavior of \( E_0(x, t) \). Actually, the \( |K| \) is a irrelevant parameter which is verified by the numerical works of ref. 8. When \( E_0 \gg |E(x)| \), we expand \( E(x, t) \) as: \( E(x, t) \approx v_0 + v_0^{(1)} (E - E_0) + v_0^{(2)} (E - E_0)^2 + v_0^{(3)} (E - E_0)^3/6 \), where \( v_0^{(j)} = dJv(E)/dE^{(j)}X = E_0 \). Equation (5) and above expansions for \( E(x, t) \) and \( E^{(j)}(t) \) imply

\[
\frac{\partial E_n}{\partial t} = \left( -\frac{\epsilon}{\epsilon} N_p^{(1)} + D_n K^2 \right) E_n + (i2D_n K - v_0) \frac{\partial E_n}{\partial x} + \frac{1}{2} \frac{\epsilon}{\epsilon} \frac{\partial E_n^{(2)}}{\partial x} + \frac{1}{2} \frac{\epsilon}{\epsilon} \frac{\partial E_n^{(3)}}{\partial x},
\]

(7)

With \( K \to 0 \) (as length \( L \to \infty \)) and \( E_n \sim \Psi(t) e^{i K^{(x-v_0)}} \), eq. (7) implies

\[
\frac{d\Psi}{dt} = -C \frac{\epsilon}{\epsilon} \left( -\frac{\epsilon}{\epsilon} N_p^{(1)} + iK^{(x-v_0)} \right) |\Psi|^2 \Psi.
\]

(8)

Here \( C = (\epsilon N_p^{(1)} / \epsilon + D_n K^2 + i K \nu_0) \). With \( \Psi(t) = R(t) e^{i \Theta(t)} \), eq. (8) implies \( R(t) = -(a/b)^{1/2} \) \( r_0^{(1)} e^{i K^{(x-v_0)}} \), \( \Theta(t) = -K \nu_0 \left( -1 + a_1 \right)^{1/2} \), and eq. (7) is related to initial value of \( R(t) \). When \( a > 0 \) (i.e., the operating point is within the regime of positive differential mobility PDM), \( b > 0 \), and \( t \to \infty \), \( R(t) \) and \( \Theta(t) \) tend to 0 and \( -K \nu_0 \), respectively. These results mean that the origin on the complex plane of \( \Psi(t) \) is a stable focus, which also implies electric-field profile in material is homegeneous, i.e., \( E(x, t) = E_0 \). When \( a < 0 \) (i.e., the NDM regime), \( b > 0 \), and \( t \to \infty \), \( R(t) = -(a/b)^{1/2} \) and \( \Theta(t) = -K \nu_0 + a_1^{(2)}(8b) t \). This solution describes a circular trajectory with counterclockwise angular frequency \( K \nu_0 + a_1^{(2)}(8b) \), radius \( -(a/b)^{1/2} \), and transient response time \( |a|^{-1} \). The transition from stable focus to stable limit cycle is a supercritical Hopf bifurcation. Since \( K \to 0 \), \( E_0 \sim \Psi(t) e^{i K \nu_0} \) and \( \Psi(t) = R(t) e^{i \Theta(t)} \) for \( a < 0, b > 0 \), and \( t \to \infty \):

\[
E(x, t) = E_0 + (-a/b)^{1/2} \cos[K(x - v_0 t - a_1^{(2)}(8b) t)],
\]

(9)

which means that the PDT has a spatial period \( \Lambda \) and a travelling velocity \( v_0 + a_1^{(2)}(8b) \). Since \( v_0 \sim 10^7 \text{ cm/s} \) is much larger than \( a_1^{(2)}(8b) \), we neglect the term \( a_1^{(2)}(8b) \) below.

In general, \( E_n \) may be written as

\[
E_n = \sum_{q=1}^{\infty} \Psi_n(t) e^{i q K^{(x-v_0)}} = \sum_{q=1}^{\infty} -\frac{a_n}{b} \left[ \left( \frac{r_1(1 - e^{2a_1 t})}{2 \gamma} + a_n \right) \right]^{1/2},
\]

(10)

where \( a_n = e N_p^{(1)} / \epsilon + D_n K^2 q^2 \). However, eq. (10) needs to be corrected when we consider the coupling between spatial modes. This is very important for self-organizing systems. For example, consider two spatial modes \( \Psi_1 e^{i K \nu_0} \) and \( \Psi_2 e^{i K \nu_0} \) with \( a_2 > 0 > a_1 \) and \( b > 0 \). From equations for \( R(t) \) and \( \Theta(t) \), we know that \( |\Psi_1| \to (a_1/b)^{1/2} \) and \( |\Psi_2| \to 0 \) when \( t \to \infty \). Then \( |\Psi_2|/|\Psi_1| \) will become zero. This result implies that only unstable spatial modes (i.e., \( a_n < 0 \)) have contributions for pattern-forming systems. However, if we combine these two modes, i.e., \( E(x, t) = E_0 + \sum_{q=1}^{\infty} \Psi_n(t) e^{i q K^{(x-v_0)}} \), and substitute it into eq. (5), we have

\[
\frac{d\Psi}{dt} = -(a_1 + i K \nu_0) \Psi_1 - \sum_{m_1 + m_2 + m_3 + m_4 = 1} \frac{\beta_1}{s^4} \Psi_{m_1} \Psi_{m_2} \cdots \Psi_{m_4},
\]

(11)

\[
\frac{d\Psi_2}{dt} = -(a_2 + i K \nu_0) \Psi_2 - \sum_{m_1 + m_2 + m_3 + m_4 = 2} \frac{\beta_2}{s^4} \Psi_{m_1} \Psi_{m_2} \cdots \Psi_{m_4},
\]

(12)

where \( m_n = \pm 1 \) or \( \pm 2 \), \( \beta_{m_n} = c N_p^{(1)} / \epsilon + \text{imaginary} \). Based on the slaving principle, we know that the stable mode \( \Psi_2 \) will be determined by the unstable mode \( \Psi_1 \), and that \( \Psi_2 \), and hence the ratio \( |\Psi_2|/|\Psi_1| \equiv r_2(t, K, N_p) \), will be nonzero. For simplicity, in the following we assume that \( r_2 = 1 \) (i.e., \( |\Psi_2|/|\Psi_1| \)) is independent of \( r \) and denote the number by \( r \), which can be determined by experiments (see below). From \( E(x, t) = E_0 + [E(x, t) + E^{(x)}(t)]/2 \) and eq. (10), for large \( t \) we get

\[
E(x, t) = E_0 + \left( -a_1 / b \right)^{1/2} \left( -r + \cos[K(x - v_0 t)] \right),
\]

(13)

Therefore, the maximum and minimum fields are equal to \( E_{\text{max}} = E_0 + (-a_1/b)^{1/2}(1 - r)^{-1} \) and \( E_{\text{min}} = E_0 - (-a_1/b)^{1/2}(1 + r)^{-1} \), respectively. According to Pockel's
effect, the difference in refractive index between $E_{\text{max}}$ and $E_{\text{min}}$ is $-n^2 \gamma_{41}(-a_1/b)^{1/2}(1-r^2)^{-1}$, where $n$ is refractive index of pure material and $\gamma_{41}$ is the electro-optic coefficient ($n = 3.3$ and $\gamma_{41} = 1.43 \times 10^{-10}$ cm/V for GaAs when the wavelength of the incident light is 1 $\mu$m). The profile of eq. (13), shown in Fig. 1, is strongly $r$ dependent. If $r$ is near 0, $E(x,t)$ is approximately equal to $\cos(K(x-v_0 t))$. If $r$ is near 1, eq. (13) shows an obvious shock-wave structure. In some senses, $r$ is related to the product $N^2_2 \Lambda$ in Kroemer’s criterion for domain formation. To see this, we first consider $r \to 1$. In this case, $(\Psi_s - [\Psi_{s+1}]/[\Psi_s])$, and hence $(|a_2| - |a_1|)/|a_1|$, is much smaller than 1 and there are many excited unstable Fourier modes. Thus, $-eN^2_2 \varphi_s^{(1)}/\gamma$ will be much larger than $D_n K^2$ and it follows from eq. (10) that Kroemer’s criterion for sharp domain formation is $a_1 \Lambda/v_0 \approx eN^2_2 \varphi_s^{(1)} \Lambda/(v_0 e) \gg 1$, i.e. $N^2_2 \Lambda \approx -v_0 e^{-1/\varphi_s^{(1)}}(\approx 10^{-2} \text{cm}^{-2})$. On the other hand, in the case $r \to 0$, $-eN^2_2 \varphi_s^{(1)}/\gamma$ is of the same order as $D_n K^2$ and Kroemer’s criterion for the appearance of sinusoidal electric-field domain is $a_1 \Lambda/v_0 \approx eN^2_2 \varphi_s^{(1)} \Lambda/(v_0 e) \approx 1$, i.e. $N^2_2 \Lambda \approx -v_0 e^{-1/\varphi_s^{(1)}}(\approx 10^{-2} \text{cm}^{-2})$. Therefore, Kroemer’s criterion appears in eq. (13) via the representation of $r$ parameter. To determine the relation between $r$ and $N^2_2 \Lambda$, we can measure $(E_{\text{max}} - E_0)/(E_0 - E_{\text{min}}) = (1 + r)/(1-r)$ at different values of $N^2_2 \Lambda$. Then we will find that $r$ is a function of $N^2_2 \Lambda$. In other words, $r$ can be determined by experiments.

In order to get hole distribution, we assume $p(x,t) = p_0 + [p_s(x,t)e^{iK(x+v_0 t)} + p_s^*(x,t)e^{-iK(x+v_0 t)}]/2$, and substitute such $p(x,t)$ into eq. (6) to find

$$\frac{\partial p_s}{\partial t} = -(D_p K^2 + \gamma N^2_2) p_s + D_p \frac{\partial^2 p_s}{\partial x^2} + (2i K D_p + \mu_p E_0) \frac{\partial p_s}{\partial x} + S(x,t),$$

and

$$\frac{\partial p_s^*}{\partial t} = -\gamma N^2_2 p_s^* + D_p \frac{\partial^2 p_s^*}{\partial x^2} + \mu_p E_0 \frac{\partial p_s^*}{\partial x} + 2g I_0 \lambda \cos(K x + \Omega t) + p_0 \left(\mu_p - \gamma/e\right) \frac{\partial E}{\partial x}.$$  

(14)

Here $S(x,t) = 2g I_0 e^{-iK(x+v_0 t)}m \cos(K x + \Omega t) + p_0 (\mu_p - \gamma/e)(\frac{D_p}{2} + i KE_0) e^{-iK(x+v_0 t)}$. Equation (14) is a linear partial differential equation with external driving function $S(x,t)$ including the MIP and multiple Gunn domains. Thus $p_s$ can be considered as a linear combination of a homogeneous solution $p_s^h(x,t)$ and an inhomogeneous solution $p_s^i(x,t)$. Since the characteristic length of $p_s$ is equal to $\Lambda (<< L)$, we may treat $K$ as zero in eq. (14) and write $\frac{\partial^2 p_s}{\partial t^2} = -(\gamma N^2_2 + D_p \frac{\partial^2 p_s}{\partial x^2} + \mu_p E_0 \frac{\partial p_s}{\partial x}) + 2g I_0 \lambda \cos(K x + \Omega t) + p_0 \left(\mu_p - \gamma/e\right) \frac{\partial E}{\partial x}.$

(15)

It is easy to find solution of $p_s^h$ and show that $p_s^h = 0$ as $t \to \infty$. When $E_s = \sum_{q=1}^\infty q^r \exp(-a_1/b)^{1/2} \exp[iqK(x-v_0 t)]$ is substituted into eq. (15), we can find solution for $p_s^i$. Therefore, the hole distribution $p(x,t) = p_0 + Re(p_s^i)$ is

$$p(x,t) = \frac{g I_0}{\gamma N^2_2} + \frac{2g I_0 \lambda}{A} \left(|D_p K^2 + \gamma N^2_2| \sin(K x + \Omega t) - (K \mu_p E_0 - \Omega) \cos(K x + \Omega t)\right) - D \sum_{q=1}^\infty \frac{q^r \exp\left(-a_1/b\right)}{\gamma N^2_2 + q^2 K^2 D_p^2} \cos[qK(x-v_0 t)] + (\gamma N^2_2 + q^2 K^2 D_p^2) \sin[qK(x-v_0 t)] \right].$$

(16)

Here $A = (D_p K^2 + \gamma N^2_2)^2 + (K \mu_p E_0 - \Omega)^2$ and $D = g I_0 \lambda \left(\mu_p - \gamma/e\right) - (a_1/b)^{1/2} / \gamma N^2_2$. The pattern profiles of eqs. (13) and (16) correspond to simple dynamics.

When $I_0$ is large and the higher order corrections in eqs. (1)–(4) need to be included, then we have

$$\frac{\partial E}{\partial t} = -\epsilon e (n - N^2_2) / e = -\epsilon E / \epsilon \varphi_s^{(0)},$$

$$\frac{\partial n}{\partial t} + \frac{\partial}{\partial x} \left[n(eE) - D_n \frac{\partial n}{\partial x}\right] = \epsilon \left[g I(x,t) - \gamma \left(N^2_2 + \frac{\epsilon}{e} \frac{\partial E^{(0)}}{\partial x} \right) p^{(0)} \right],$$

where $p^{(0)}$ and $E^{(0)}$ are solutions of eqs. (16) and (13), respectively. These results imply that up to order $O(\epsilon)$ we have

$$\frac{\partial E}{\partial t} + \frac{1}{\epsilon} e N_2 \varphi_s^{(0)} e(E) = \epsilon \frac{\partial E}{\partial x}$$

$$\frac{\partial}{\partial x} \left[n(eE) - D_n \frac{\partial n}{\partial x}\right] = \epsilon \left[g I(x,t) - \gamma \left(N^2_2 + \frac{\epsilon}{e} \frac{\partial E^{(0)}}{\partial x} \right) p^{(0)} \right],$$

where $S(x,t) = \epsilon \left|p^{(0)}(E^{(0)}) - \mu_p E^{(0)} p^{(0)} - (D_p + D_h) (p^{(0)} / \partial x) / \epsilon \right.$ and represents the electron-hole coupling. To understand the underlying physics of eq. (19), we follow the same procedure as the case of simple dynamics, to get
\[ \frac{d\Psi}{dt} = -C\Psi - \frac{1}{\varepsilon} \left( \frac{e}{\varepsilon} N_{D}^{2} v_{0}^{(3)} + iKv_{0}^{(2)} \right) |\Psi|^{2}\Psi \]

\[ + \int_{0}^{\Lambda} \bar{S}(x,t)e^{-iKx}dx, \quad (20) \]

which is a generalization of eq. (8) and represents a self-sustained oscillator (i.e., PDT) under the external driving force \( F(t) \) and the external driving force \( F(t) \) under the external driving force \( F(t) \). The dynamical variables can be analytically expanded to Fourier series based on the form of \( I(x,t) \). Therefore they can get multiple Gunn-domain order by order. Actually the physical idea of Segev et al. is the same as Subacˇius et al. The spatial period of \( I(x,t) \) determines the spacing of Gunn domains and \( I_{0} \) (or \( I_{0}f(t) \)) generates background electrons. However, the work of Segev et al. didn’t discuss the stability of multiple Gunn-domain, which will lead to the travelling domains with the same moving velocity (i.e., \( \Omega/K \)) of \( I(x,t) \). Furthermore, according to the prediction of Segev et al. the moving velocity of electric-field profile in PDM or NDM regime has no any difference. This unsuitable prediction can be validated and verified by eq. (20). As for the numerical work of Bonilla et al., they showed complicated spatiotemporal behaviors of Gunn domains, e.g., quasiperiodic route to chaos, are mostly observed in their simulations. In other words, PDT didn’t appear in their system. Therefore, they concluded that Kroemer’s criterion can’t give the correct numbers of Gunn domains. We think that \( m = 0.1 \) used in their study is large enough to generate driven chaos due to the competition between SCF and PDT. Therefore both considerations of Kroemer’s criterion and pattern competition may give correct physics in light-triggered Gunn-domain systems.

Finally we want to discuss the validity of our approach to the light-triggered Gunn-domain in semi-insulating semiconductors. In this case all of free electrons are excited from valence band, i.e., \( N_{D}^{2} = 0 \) and \( n_{0} = (g_{l0}/\gamma)^{1/2} \). Then, there are no small variables or parameters among \( E, n, p, \) and \( I \). Therefore, we can’t get the solutions order by order via the present approach. However, the physics we studied in the present manuscript still can address both PDT and pattern competition which can be observed in semi-insulating semiconductors. The large \( I_{0} \) is the key point to produce PDT, where \( I_{0} \) can generate sufficient \( v_{0} \) to satisfy the Kroemer’s criterion, i.e., \( n_{0} \lambda \gg 10^{12} \text{ cm}^{-2} \). And the role of small \( m \) is to determine the spatial periodicity, i.e., \( \lambda \), of multiple Gunn-domain and weak SCF. When \( m \) is sufficiently large, the competition between stronger SCF and PDT will appear and results in complex dynamics. In order to find these phenomena in semi-insulating semiconductors, we suggest that people shall keep \( I_{0} \) and \( \Lambda \) fixed, and tune \( m \) in experiments.

In conclusion, using a nonlinear analytic method we confirm the LSA results of ref. 7 and find that the competition between the SCF and the PDT could be a dominant factor to determine the spatiotemporal pattern in semiconductor. We hope that the formulation of the present manuscript can be further verified by numerical calculations or experiments.

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11) In the present manuscript we consider \( m \) is a fixed values and much small than 1. Therefore, SCF shall be proportional only to \( m \).
12) When \( \gamma / C_{22} \) is much small than 1, eq. (20) can be reduced to eq. (8). In other words, \( I_0 \) shall be much small than \( N_{D}^{2} \gamma / g \). For GaAs parameters \( g = 0.44 \times 10^{-15} \text{cm}^{-3}\text{sec}^{-1}\text{W}^{-1} \), \( \gamma = 10^{-10} \text{sec}^{-1}\text{cm}^{2} \), and \( N_{D} = 10^{16} \text{cm}^{-3} \), then the intensity requirement for PDT formation is \( I_0 \ll 2.27 \text{kW/cm}^{2} \).