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Damage threshold prediction of crystal materials irradiated by femtosecond lasers based on ionization model and two-temperature model

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ABSTRACT

Crystal materials may be damaged irradiated by high-power femtosecond laser pulse during terahertz radiation generation in optical rectification, limiting the further increase of the intensity and the conversion efficiency of the generated terahertz radiation. It remains a big challenge to predict the damage threshold of crystal materials by femtosecond lasers. In this paper, the interaction mechanism between the femtosecond laser pulse and the crystal material has been analyzed and the related theoretical model has been built up. Consequently, two types of models for the damage threshold prediction of crystal materials based on the ionization model and the two-temperature model have been proposed. On the basis, the evolution of free electron in crystals and the temperature variation of electron and lattice have been discussed in detail. The influence of the major parameters of femtosecond laser pulses on the damage threshold has also been analyzed quantitatively. The results show that, the actual damage threshold of the crystal material may be between the calculated values based on the ionization model and the two-temperature model. For the incident femtosecond laser pulse with larger central wavelength, the damage threshold of crystal materials becomes relatively larger.

1. Introduction

As a new branch of science, terahertz (THz) science has been increasingly attracted widespread attention, which is focused on the range of the electromagnetic spectrum (0.1–10 THz), and earlier referred to as the far-IR range [1–3]. In recent years, optical rectification (OR) by femtosecond laser pulses becomes a widely used method for the generation of intense THz pulses [4], and nonlinear crystal materials are much more promising for THz generation by OR [5,6]. It has been shown from the previous studies that the THz radiation intensity and generation efficiency commonly increases with the intensity of the pump femtosecond laser pulse [7]. Consequently, high-power femtosecond laser pulse has been used to generate THz radiation with higher energy to meet the requirement of many promising applications. However, the further increase of the intensity and the conversion efficiency of generated THz radiation are inevitably limited by the damage to nonlinear crystal materials due to the intense femtosecond laser pulse. Therefore, it is of great importance to predict the damage threshold of crystal materials by femtosecond lasers. By far, although the experimental and numerical study of the damage threshold of fused silica and silicon is widely performed [8–10], the damage threshold of lithium niobate ($LiNbO_3$) crystal material commonly used as nonlinear crystal in optical rectification is lack of effective theoretical and experimental research.

In this paper, the prediction model for the damage threshold of optical rectification crystals based on the ionization model and the two-temperature model have been proposed. The damage threshold of the $LiNbO_3$ crystal has further been predicted and discussed. Firstly, the interaction mechanism between the femtosecond laser pulse and the crystal material has been analyzed and the corresponding theoretical model has been built up. Then, the model of the evolution of the electron and lattice temperatures determined by the two-temperature model has also been established. Finally, the variations of the damage threshold of $LiNbO_3$ crystal with the pulse duration and the central wavelength of the femtosecond laser pulse have also been analyzed.

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2. Theoretical models

The main purpose of this study is to predict the damage threshold of $LiNbO_3$ crystals using the following two types of assumptions based on: (1) free electron density in the ionization model, (2) the lattice temperature with the two-temperature model.

2.1. The ionization model

It is usually assumed that the damage of optical materials occurs when the density of free electron in the optical materials reaches the critical density. Consequently, the damage threshold could be considered as the minimal fluence that exactly creates the critical density. Contrary to metals, the conduction band of crystal materials is initially empty and the crystal is transparent to the femtosecond laser pulse. The free electrons from the valence band must be first transferred in the conduction band by nonlinear ionization processes to start the femtosecond laser pulse heating [11], a large amount of free electrons generated by avalanche ionization and photo-ionization due to the absorption of the laser pulse energy by crystal materials transit from the valence band to the conduction band. In this process, photo-ionization provides the initial electrons needed for the avalanche ionization, that is, the free electrons are activated by the photo-ionization early, and then by both avalanche ionization and photo-ionization simultaneously during the interaction between the femtosecond laser pulse and the crystal material. When the critical density is achieved, the free electrons are excited into the conduction band and begin to absorb the femtosecond laser pulse energy rapidly.

The rate equation approach commonly used to predict the evolution of the conduction electron density N_e is given [12,13], *i.e.*,

$$\frac{\partial N_e(t)}{\partial t} = \eta(E)N_e(t) + w_{\mathrm{pi}}(E) - \sigma N_e(t) - gN_e^2(t) \tag{1}$$

where E is the electric field of the femtosecond laser pulse; $\eta(E)$ denotes the electron avalanche rate; $w_{\mathrm{pi}}(E)$ represents the photo-ionization rate; σ and g are electron diffusion and electron–ion recombination, respectively. For simplicity, the electron diffusion and recombination term can be neglected in Eq. (1) for the pump laser pulse duration $\tau < 10$ ps, *i.e.*

$$\frac{\partial N_e(t)}{\partial t} = \eta(E)N_e(t) + w_{\mathrm{pi}}(E) \tag{2}$$

Based on Thornber's model, the avalanche rate $\eta(E)$ of the conduction electron density can be expressed as [14]

$$\eta(E) = \frac{v_s eE}{E_g}\exp\left\{-\frac{E_i}{E(1+E/E_p)+E_{kT}}\right\} \tag{3}$$

where v_s is the saturation drift velocity; e is the electron charge; E_g is the band gap energy. E_i, E_p, and $E_{kT} = E_i kT/E_g$ are the fields for electron overcome the decelerating effects of ionization scattering, optical phonon scattering and thermal scattering in one mean free path, respectively.

Photo-ionization can be further divided into multiphoton ionization and tunnel ionization. In the process of multiphoton ionization in crystal materials irradiated by high-power femtosecond laser pulses, photons are absorbed simultaneously to produce free electrons. Further increasing the intensity of pump laser pulse provides the condition for tunnel ionization.

The photo-ionization rate used in this work is approximately by the Keldysh's expression [15]

$$w_{\mathrm{pi}}(E) = \frac{2\omega}{9\pi}\left(\frac{\omega m_{eff}}{\hbar\sqrt{\gamma_1}}\right)^{3/2} Q(\gamma,x) \times \exp\left\{-\pi < x+1 > \frac{\kappa(\gamma_1)-\xi(\gamma_1)}{\xi(\gamma_2)}\right\} \tag{4}$$

where ω is the lasers frequency; m_{eff} is the effective electron mass; $\hbar$ is the reduced Planck's constant; $\gamma = \omega(m_{eff}E_g)^{1/2}/(eE)$ denotes Keldysh's parameter in crystal; $\gamma_1 = \gamma^2/(1+\gamma^2)$, $\gamma_2 = 1/(1+\gamma^2)$; and

$$\begin{cases} Q(\gamma,\chi) = \sqrt{\frac{\pi}{2\kappa(\gamma_2)}} \times \sum_{n=0}^{\infty} \exp\left\{-n\pi\frac{\kappa(\gamma_2)-\xi(\gamma_2)}{\xi(\gamma_1)}\right\} \times \Phi\left\{\frac{\pi}{2}\sqrt{\frac{2\langle\chi+1\rangle-2\chi+n}{\kappa(\gamma_2)\xi(\gamma_2)}}\right\} \\ \chi = \frac{2}{\pi}\frac{E_g}{\hbar\omega}\frac{\sqrt{1+\gamma^2}}{\gamma}\xi\left(\frac{1}{1+\gamma^2}\right), \quad \Phi(z) = \int_0^z \exp(y^2-z^2)dy \end{cases} \tag{5}$$

where $\Phi(z)$ is the Dawson integral; $\langle z\rangle$ is the integer part of the number z; The complete elliptic integrals of the first and second kinds can be expressed by

$$\begin{cases} \kappa(x) = \int_0^{\frac{\pi}{2}} (1-x^2\sin^2\theta)^{-\frac{1}{2}}d\theta \\ \xi(x) = \int_0^{\frac{\pi}{2}} (1-x^2\sin^2\theta)^{\frac{1}{2}}d\theta \end{cases} \tag{6}$$

In this assumption, a small volume of crystal material can be damaged if its free electron density is equal to or above the critical electron density. For femtosecond laser pulse, the critical electron density n_{cr} is defined as the free electron density at which the plasma frequency $\omega_p = eN_e^{1/2}(\varepsilon_0 m_e)^{-1/2}$ is equal to the laser frequency. Therefore, the critical density n_{cr} can be expressed by

$$n_{cr} = \frac{4\pi^2 c^2 m_e \varepsilon_0}{\lambda^2 e^2} \tag{7}$$

where ε_0 is the electrical permittivity of free space; m_e denotes the mass of electron; c represents the scalar speed of light in vacuum; λ is the wavelength of the femtosecond laser pulse.

2.2. The two-temperature model

It is assumed that $LiNbO_3$ crystal can be damaged if its lattice temperature is higher than the melting point, 1533 K [16]. The ionized crystal materials start to behave like a metal with a time varying electron density in the conduction band. The deposited lasers energy is then transferred to the lattice, and the thermomechanical relaxation and ejection of matter processes occur depending on the amount of the absorbed lasers energy. In order to analyze the heat propagation, the two temperature model has been applied, where the following two equations determine the evolution of the electron and lattice temperatures, respectively, *i.e.* [17,18]

$$c_e\frac{\partial T_e}{\partial t} = \nabla\cdot(k_e\nabla T_e) + Q_L - Q_1 \tag{8}$$

$$\rho c_l\frac{\partial T}{\partial t} = k_l\nabla^2 T + Q_1 \tag{9}$$

where T_e and T denote electron temperature and lattice temperature, respectively; c_e and c_l represent the heat capacities per unit volume of electron and lattice systems, respectively; k_e and k_l are the heat conductivities for the electron and lattice systems; ρ is the density of crystal material. The heat capacities per unit volume and the heat conductivities of electron system can be expressed by

$$c_e = \frac{3k_b n_o}{2}, \qquad k_e = \frac{3k_b^2\mu_0 n_o T_e}{2e} \tag{10}$$

where k_b denotes the Boltzmann constant; n_o represents the refraction index of crystal material; μ_0 is the mobility of crystal material.

Table 1
Properties of $LiNbO_3$ crystal used in the calculations [16,21].

Crystal	E_g (eV)	m_{eff} (m_e)	n_o	c_l (J kg^{-1} K^{-1})	k_l (W m^{-1} K^{-1})	ρ (g cm^{-3})
$LiNbO_3$	3.8	0.45	2.159	648	4.5	4.63