Theoretical and Experimental Study on Femtosecond Laser-induced Damage in MgF_2 Crystals

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We study single-shot damage morphology of MgF_2 irradiated under 800 nm laser using scanning electronic microscope (SEM). The dependence of damage threshold on laser pulse width is measured by the linear relationship between the area of damage spot and the logarithm of laser energy. The pulse duration ranges from 55 fs to 750 fs. We develop the multiple-equation model (*Phys. Rev. Lett.* Vol.92, pp.187401, 2004), in which two-photon absorption in the conduction band is considered. The experimental results agree well with our model.

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1. Introduction

These years, there has been considerable interest in femtosecond laser-induced damage in transparent dielectric.[1-8] However, the reports on damage for MgF_2 are very few. In this paper, we extend our interest to the damage in MgF_2, a potential material as window, lens and fiber guide. Considering the important role of the high kinetic electrons, Rethfeld[9] developed a multiple rate equation (MRE) model. It can keep track of the transient energy distribution of electrons on ultrashort time scales by solving single-photon absorption multiple rate equation. Although this model can explain some experimental results, it has not considered the two-photon absorption in conduction band (CB). The effect of two-photon absorption in CB has been studied in recent years, which was considered to be very important in the damage of transparent material induced by ultrashort pulse.[10,11]

In this paper, we study single-shot damage morphology of MgF_2 irradiated under 800 nm laser using scanning electronic microscope (SEM). The dependence relation of the damage threshold and laser pulse width is measured by the linear relationship between the area of damage and the logarithm of the laser energy for pulse duration ranging from 55 fs to 750 fs. We calculate the intraband absorption of second order in the conduction band, develop an advanced multiple equation (AME) model, and investigate damage mechanism of Magnesium Fluoride irradiated by femtosecond laser pulse.

2. Experimental setup and results

The experiments are conducted with a chirp pulse amplified Ti: Sapphire laser system. Its standard output is 50 fs, 0.6 mJ with a wavelength centered at 800 nm. By using a half wave plate and a polarizer, we can adjust the pulse energy continuously. Pulse duration is adjusted by compressing grating. The pulse is perpendicularly focused on the front surface of sample to a radius of ~25 μm. The MgF_2 crystal is polished on both sides and is set on a three-dimensional translation stage. Each site on the sample was irradiated by only one laser pulse. The area of spot was measured with the higher-resolution optical microscopy (NA~0.65).

Figure 1 shows the typical electron microscope picture irradiated by femtosecond laser pulse. We can see the clear edge with no indication of melting, which is different from the morphology of irradiated by long pulse (10 ps~ns). When the material is irradiated by long pulse, electrons transfer laser energy to ions by electron-phonon scattering. Material is heated, molten and ablation occurs. Thermodynamic effect plays more important role in this damage process. However, as for femtosecond pulse, pulse duration is less than the timescale of electron-phonon scattering, material damage is no more dominantly associated with thermodynamic effect, which is a 'cool ablation' process.

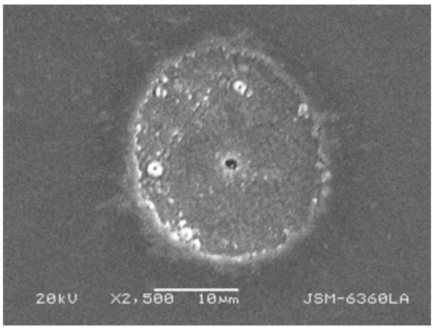

Fig. 1 Damage morphology of MgF_2 irradiated under 800 nm laser, F=10.17 J/cm^2 and τ=55 fs.

We define the minimum laser fluence causing permanent detectable damage as the damage threshold of material. There are many ways by which we can measure the material damage threshold.[1,2,4,6,12,13] In this paper, we measure the damage areas, and find the linear relation between the areas of spot and the logarithm of laser energies.[12] From figure 2 we can calculate the damage threshold (F_{th}) and a $1/e^2$ radius ω_0 by intercept and slope, respectively.

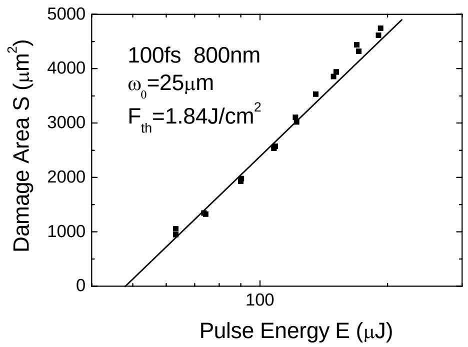

Fig. 2 The linear relationship between the irradiated area and laser energy, τ=100fs.

By this means, we measure the dependence of MgF_2 damage threshold on laser pulse width irradiated by 800 nm wavelength laser. The triangles in figure 3 show the experimental results.

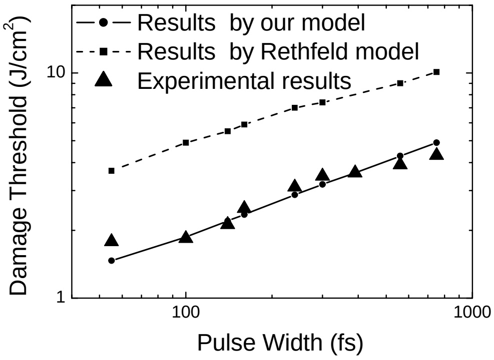

Fig. 3 The dependence of damage threshold on pulse width.

3. Theoretical model in damage

The avalanche model is a popular model on the damage mechanism in dielectric. In this model, impact ionization is dealt to depend on the total free-electron density, which is somewhat simple. Rethfeld[9] thought only electrons with high kinetic energy are important to impact ionization and the transient distribution of electrons should be considered. But she only considered the single-photon absorption in CBE. Belsky et al[10] reported the work about multiple-photon absorption in CB during the interaction of laser pulse and material. In our previous works[3,11], we investigated multiple-photon absorption in CB in theory and experiment. These results reveal that during the interaction of femtosecond laser pulse and material, two-photon absorption in the CB is very important with increasing laser intensity.

Ignoring electrons relaxation processes,[9] we introduce two-photon absorption in the conduction band in high pulse intensity and develop an advanced multiple equations model. Single-photon and two-photon absorption rates are calculated with second-order and third-order perturbation theory.[3,11] Figure 4 shows the conduction electron cascade processes. When the transparent dielectric is irradiated by high intensity laser pulse, the electrons in valence band (VB) top transit to CB bottom at rate $\dot{n}_{pi}$ with approximate zero kinetic energy ($\varepsilon_0 \approx 0$). This process can be dealt by Keldysh photoionization theory. We define discrete kinetic energy levels $\varepsilon_j := \varepsilon_{j-1} + \hbar\omega_L$ with the electrons density n_j, where ω_L is the laser frequency. The low kinetic energy electrons absorb single photon or two-photon energy from laser pulse with rates $W_{1pt}(\varepsilon_j)$ and $W_{2pt}(\varepsilon_j)$. When electrons kinetic energy $\varepsilon_k = \varepsilon_0 + k\hbar\omega_L$ exceeds the critical energy for impact ionization ε_{crit}, these electrons generate impact ionization with a probability α. k is determined by $k = \left\lceil \frac{\varepsilon_{crit}}{\hbar\omega_L} \right\rceil$. This process can be described with multiple difference equations.

$$
\begin{aligned}
\dot{n}_0 &= \dot{n}_{pi} + 2\alpha n_k - W_{1pt}(\varepsilon_0)n_0 - W_{2pt}(\varepsilon_0)n_0 \\
\dot{n}_1 &= W_{1pt}(\varepsilon_0)n_0 - W_{1pt}(\varepsilon_1)n_1 - W_{2pt}(\varepsilon_1)n_1 \\
\dot{n}_2 &= W_{1pt}(\varepsilon_1)n_1 + W_{2pt}(\varepsilon_0)n_0 - W_{2pt}(\varepsilon_2)n_2 \\
&\quad - W_{1pt}(\varepsilon_2)n_2 \\
\dot{n}_3 &= W_{1pt}(\varepsilon_2)n_2 + W_{2pt}(\varepsilon_1)n_1 - W_{2pt}(\varepsilon_3)n_3 \\
&\quad - W_{1pt}(\varepsilon_3)n_3 \qquad (1) \\
&\vdots \\
\dot{n}_{k-1} &= W_{1pt}(\varepsilon_{k-2})n_{k-2} + W_{2pt}(\varepsilon_{k-3})n_{k-3} \\
&\quad - W_{2pt}(\varepsilon_{k-1})n_{k-1} - W_{1pt}(\varepsilon_{k-1})n_{k-1} \\
\dot{n}_k &= W_{1pt}(\varepsilon_{k-1})n_{k-1} + W_{2pt}(\varepsilon_{k-2})n_{k-2} \\
&\quad + W_{2pt}(\varepsilon_{k-1})n_{k-1} - \alpha n_k
\end{aligned}
$$

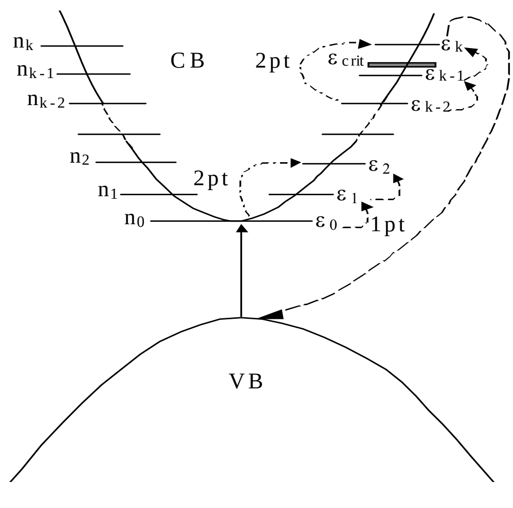

Fig. 4 Electrons cascade processes irradiated by high intensity laser pulse.

Figure 5 shows the evolutions of electron density for 100 fs pulse at 1.865 J/cm^2 by MRE model and AME model, respectively. We can see that electron density reached 10^{18} cm^{-3} if only considering the single-electron absorption. It can reach 10^{21} cm^{-3} when two-photon absorption is considered. From some experimental results we know only the fluence above 10 kJ/cm^3 can cause material damage,[15,16] i.e., free-electron density reaches 10^{21} cm^{-3}, which agrees well with the free-electron density of plasma resonate aborption.[17]

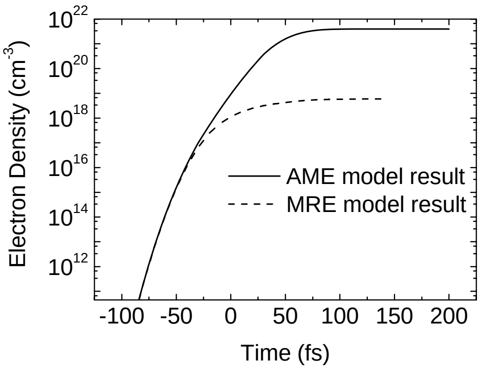

Fig. 5 The evolutions of electron density for 100 fs pulse at 1.865 J/cm^2 by MRE model and AME model, respectively.

In figure 3, solid line shows the dependence relation between laser pulse width and material damage threshold by AME model; dash line shows the calculated results by MRE model. We can see the AME model agrees experiment very well because of considering the two-photon absorption in conduction band. If ignoring the two-photo absorption, the theoretical damage threshold is as twice as experimental threshold. Therefore, we can conclude that two-photo absorption plays important role in the damage of dielectric irradiated by femtosecond laser pulse.

4. Conclusion

We study single-shot damage morphology of MgF_2 irradiated under 800 nm laser wavelength using scanning electronic microscope (SEM). By the linear relationship between the area of damage spot and the logarithm of the laser energy, we measure the dependence of damage threshold on pulse width ranging from 55 fs to 750 fs.

We consider the transient distribution in CBE and introduce two-photon absorption in CBE based on avalanche model. An advanced multiple-equation model is developed. The results show that when we introduce the two-photon absorption in CBE, the calculated threshold agrees the experimental measurement very well; but if not considering the two-photon absorption in CBE, the theoretical result is as twice as the experimental threshold. So two-photon absorption plays indispensable role in the damage of dielectric irradiated by femtosecond laser pulse.

Acknowledgements

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