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Origin and relation of three kinds of scatter centers in KDP and DKDP crystals

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The origin of three kinds of scatter centers in KDP and DKDP crystals and their relations were investigated. The results have shown that solid inclusions will form liquid and gas inclusions in KDP (DKDP) crystals. The effect of scatter centers on Laser Damage Threshold of the crystals is not same because their sizes are different.

1 Introduction

Potassium dihydrogen phosphate (KDP) and deuterated potassium dihydrogen phosphate (DKDP) are two nonlinear optical materials commonly used for harmonic generation and electro-optic switching in ICF [1]. However, it has been observed that the usable beam fluence in both KDP and DKDP is limited by the occurrence of bulk damage. Scatter centers has been thought to be one reason of lowering laser damage threshold (LDT), no matter whether the crystals were grown by the traditional slow temperature-lowering method or the fast growth method [2-4]. Therefore, to understand the formation of the scatter centers and their behavior under high power laser illumination is very important.

Generally, there are three kinds of inclusions, i.e. solid inclusions, liquid inclusions and gas inclusions, these will form scatter centers in KDP (DKDP) crystals, but the origin and the relation between them is still not very clear.

In order to understand the origin of scatter centers in KDP and DKDP crystals and their behaviors under high power laser radiation, we present TEM results and thermal annealing effects on scatter centers in KDP and DKDP crystals. A formation mechanism is proposed and verified. The relation between scatter centers and laser damage threshold of the crystal is shown.

2 Experimental Procedure

2.1 Samples

KDP and DKDP crystals were grown by traditional temperature-lowering method, deuterium fractions of the solution can be measured by IR spectrometric method, the deuterium fraction in crystals was calculated to be around 95% [5-6].

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2.2 Measurement condition

TEM: The samples were cut into slices parallel to [001] direction of the crystal, and then fabricated into a slice about 100 nm by ion beam to let the probing beam penetrate the samples. TEM investigations were carried out with a HITACHI H800. The working voltage was 75kV.

Thermal annealing: Following the initial testing, the samples were annealed in a furnace at 140°C for approximately 96 hours. No phase transition happened in DKDP samples, and KDP crystals were still kept transparent.

3 Results and discussion

We have found by ultra-microscopy that the scatter centers in some samples became larger while with lower density after thermal annealing (Fig.1). This result is contradicted to the results from LLNL and the other of our results [7-9]. In order to understand these confusing results, TEM was applied to detect the character of the scatter centers before/after thermal annealing.

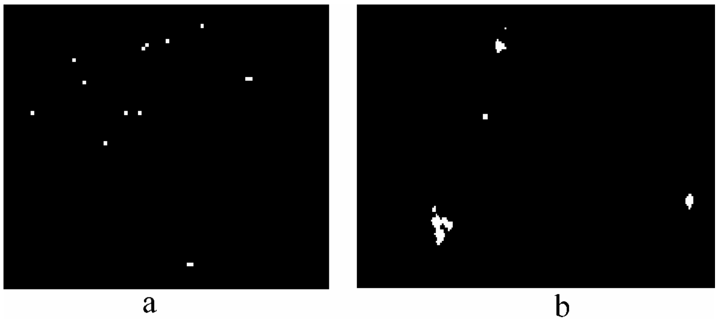

Fig. 1 Scatter centers before (a) and after (b) thermal annealing (×25).

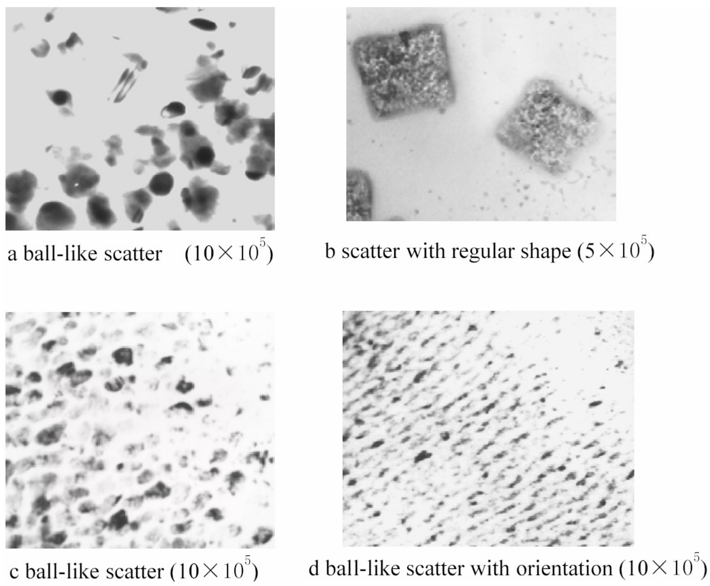

Fig. 2 Scatter in KDP (a, b) and DKDP (c, d) crystals.

Figure 2 shows the scatter centers observed by TEM. Most of the scatter centers appeare as ball-like shape, which can be understood as a kind of liquid inclusion, mainly composed of mother solution. For it can keep the lowest surface-energy. But its size is only from few nms to few tens of nms, which is much smaller than that of liquid inclusion reported in the literatures [10-12]. The other kind of scatter centers has similar shape with KDP (DKDP) crystals, and even the growth sector can be seen clearly. Considering its size, it can be ascribed to inclusion in references [10-12], but it still can give some information about the formation of scatter centers. When the temperature is higher than the growth temperature, dissolution happens, and then the scatter centers form. At the same time, some impurities and mother liquid will be included into this crystal, which is the reason why the scatters are not transparent in the photos. So the liquid inclusions are not composed of solution totally, and there are some solid impurities in it.

After thermal annealing, the same samples were measured again with TEM and compared with the results obtained before thermal annealing (Table 1).

Table 1 Thermal annealing on scatter centers in KDP and DKDP crystals.

Before thermal annealing			After thermal annealing		
Samples	Size (nm)	Density (/cm^2)	Samples	Size (nm)	Density (/cm^2)
Fig.2-a	30-100	10^7-10^8	Fig.3a, 3b	<10 or >50	$\leq 10^5$
Fig.2-b	>1000	-	Fig.3c	>1000	-
Fig.2-c	10-50	$\approx 10^8$	Fig.3d	> 50	$\leq 10^3$
Fig.2-d	10-100	10^8-10^9	Fig.3e*	-	-

*: There are still some gas/solid scatter centers in this sample, but most parts of the sample became clear. This table gives the results of the main character of scatter centers after thermal annealing. The size of the scatter centers can be measured and calculated.

At least three differences can be seen. 1) The density of the scatter centers decreased evidently. 2) The scatter centers became larger or smaller. 3) Most part of the sample became clear with no scatter centers (Fig.3). Even these blank place are no same, the difference can be seen clearly from Fig.3-d and 3-e. Fig.3-e shows the macrosteps with certain orientation, which can explain the orientation of scatter in DKDP crystal (Fig.2-d). Besides, there is still some black material left as shown in Fig.3-c. Its component is not very clear, but as comparing with Fig.2-b, it is thought to be the impurities, which left after the KDP material in mother liquid begins to crystallize into the crystal.

These three characters truly reflected the effect of thermal annealing and help us understand the origin of the three kinds of scatter centers and their relations.

The appearance of the larger scatter centers can explain ultra-microscopy results. But there is still some solid scatters with smaller size can be seen.

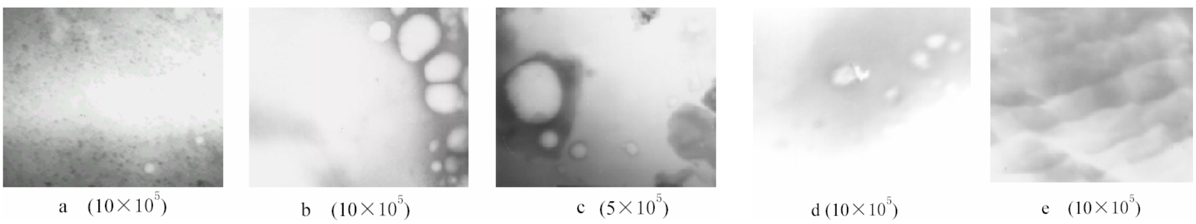

Fig. 3 TEM results after thermal annealing of KDP (a, b, c) and DKDP (d, e) a,b,d,e,with (10×10^5), c with(5×10^5).

From Fig.2, it can be seen that there is always some mother solution in the scatter centers, so two possibilities happened during the crystal growth process. 1) The size of the scatter centers became smaller, which indicated that the materials crystallized around, which happened when the content of mother solution was relative small. 2 When the content of mother solution in the scatter centers was high, the solution in scatter centers began to dissolve the KDP material in crystal around it when the temperature was raised during thermal process,. So, the larger inclusions formed. After this process, because the temperature fell down quickly, not only the solution in scatter centers crystallized to the crystal around it, but also the inside solution began to crystallize, then the bubble formed. During this process, those formed bubbles aggregated together to form large bubble (Fig. 3-b, c). This process will also happen in crystal growth when the temperature disturbing happens.

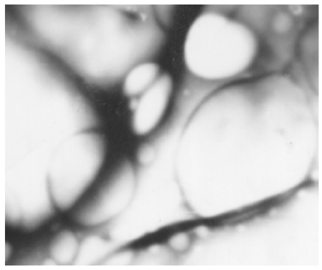

Fig. 4 TEM result of laser damage site of KDP crystals(5×10^5).

From above analysis, it can be seen that solid scatter is the key reason of causing scatter centers in KDP crystals. When the impurities absorbed onto the crystal surface, the growth steps were inhibited, then the inclusion formed and mother liquid was also included into the scatter. So, the solid inclusion also contained some liquid. Along with the temperature fell down, the crystallization happened also in scatter centers, then the "gas scatter" formed. Therefore, the difference among these three inclusions is that the ratios among solid, liquid and gas are not the same.

Considering of the size of the difference of the three scatter centers, the effects of them on the laser damage threshold (LDT) are also not the same. For example, in sample of Fig. 2-b, after thermal annealing, the LDT falls from 14J/cm^2 to11J/cm^2. And there are lots of gas inclusions at the damage site (Fig.4). There are two possibilities of this phenomenon. 1) Inclusion induces damage; 2) Under the laser beam with high power, the micro-inclusion aggregated together, similar to the process of thermal annealing, forming larger inclusions, which absorbed more probing beam, causing damage. From this point of view, those scatter centers with larger size, such as liquid and gas scatter centers do more harm to the LDT of the crystals.

4 Conclusion

The process of thermal annealing changes the existence of scatter centers in KDP and DKDP crystals, which revealed the origin of three kinds of scatter centers. The KDP material in mother liquid can be crystallized into the crystal during the thermal annealing process, and then the bubbles are left and aggravated, causing absorption of laser beam and the collapse of the crystal at last.

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