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# Supplement Information

# Nd:MgO:LiTaO$_3$ crystal for self-doubling laser applications: growth, structure, thermal and laser properties

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## Supplement 1optimization procedure for Nd:MgO:LT crystal growth

Before optimization, an iridium crucible was placed in the center of an alumina crucible with zirconium dioxide sand filling the gap. The iridium crucible, alumina crucible and coil are homocentric. On top of the iridium crucible, a mullite cap encloses an alumina tube for better insulation. Some values of physical properties applied in the numerical simulation are shown in Table S1. The crystal rotation rate and pulling rate were 6 r/min and 2 mm/h, respectively. The simulated results of the thermal and flow fields with a crystal length of 25 mm are shown in the left hand of Fig. S1. A clear interface shape was shown in the right hand of Fig. S1.

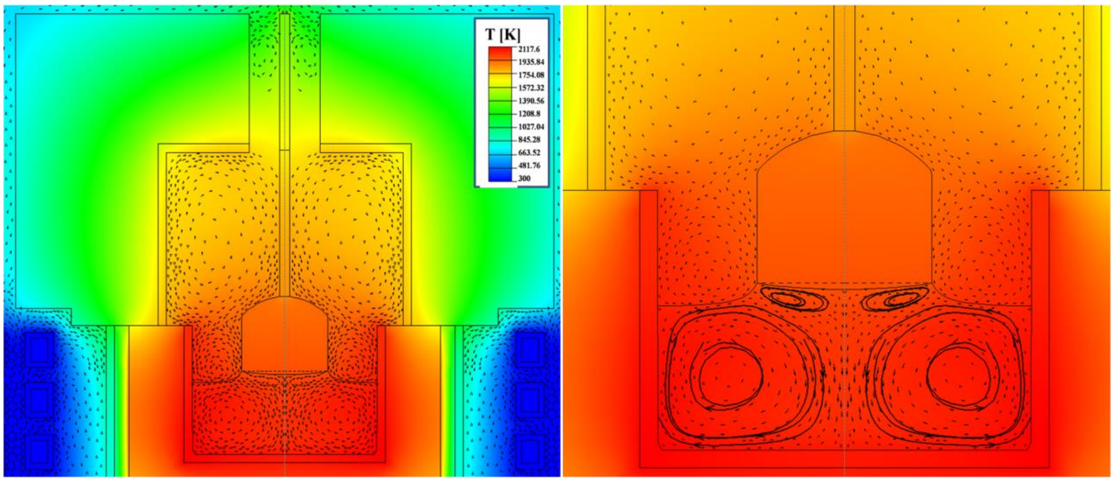


Fig. S1 The optimum thermal and flow fields (left) and the interface (right)

Table S1 Main parameters used in numerical simulation.

| Property | Value |
|---|---|
| Density of the crystal(g/cm$^3$) | 7.49 |

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| | |
|---|---|
| Emissivity of the crystal | 0.5 |
| Specific heat of the crystal(J/gk) | 0.424 |
| Thermal conductivity of the crystal(W/mk) | 5.155 |
| Density of the melt(g/cm$^3$) | 5.64 |
| Emissivity of the melt | 0.3 |
| Specific heat of the melt(J/gk) | 1100 |
| Thermal conductivity of the melt(W/mk) | 3 |
| Thermal conductivity of the Ircrucible(W/mk) | 147 |
| Emissivity of the Ir crucible | 0.8 |
| Electricity conductivity of the Ircrucible(S/m) | $2\times10^7$ |
| Specific heat of the Ircrucible(J/gk) | 130 |
| Thermal conductivity of ZrO2(W/mk) | 2.14 |
| Emissivity of ZrO2 | 0.6 |
| Thermal conductivity of alumina(W/mk) | 2.05 |
| Emissivity of alumina | 0.7 |

Optimization 1: (1) Change the inside insulation from an alumina tube to a $ZrO_2$ tube. The purpose is to increase the stability of the thermal field and to decrease the temperature gradient. (2) Decrease the pulling rate to 0.5-1 mm/h. (3) increase the rotation rate to 10 r/min in order to make the forced convection strong. Fig. S2 shows the simulated results of the thermal and flow fields with a crystal length of 25 mm and the interface shape between solid and liquid.

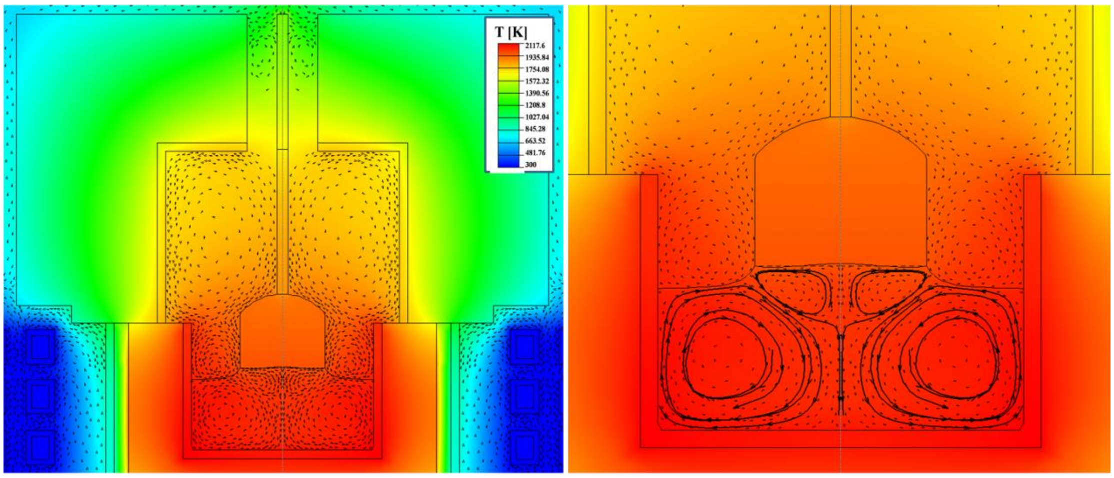
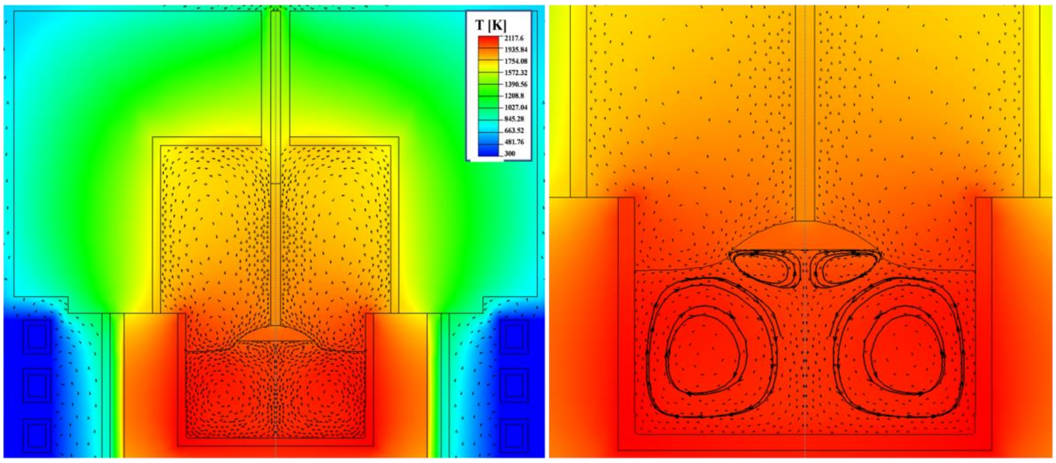
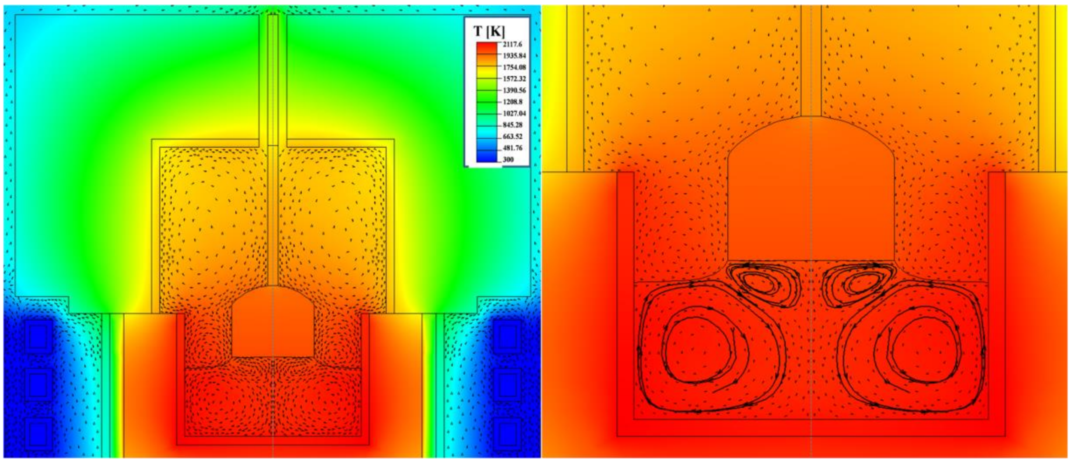



Fig. S2 The optimum thermal and flow fields (left) and the interface (right)

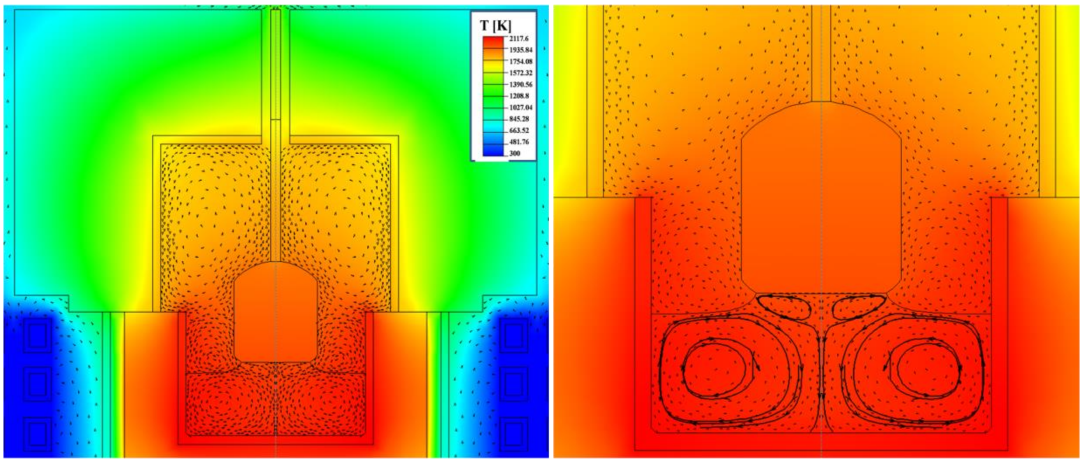
Optimization 2: (1) Increase the thickness of the mullite cap to decrease the temperature gradient. (2) Adopt different pulling rates 0.5-2 mm/h at different stages. Generally, the pulling rate is 2 mm/h at the seeding stage and decreases slowly through the shoulder-extending stage to reach 0.5 mm/h at the iso-diameter pulling stage. In this way, not only is the crystal growth period shortened, but the crystal quality is also optimized. (3) Decrease the rotation rate to 8 r/min in order to make the forced convection match the natural convection. (4) Increase a tailing stage, i.e. automatically pull the crystal up away from the melt slowly with a pulling rate of 8 mm/h (The crystal was pulled up away from melt manually at a quick rate), as the crystal pictures shown in Fig. 2. The simulated results of the thermal and flow fields at different stages with different pulling rates are shown Fig. S3, as well as the clear interface shape.




(a) shoulder-extend stage with pulling rate of 1 mm/h




(b) iso-diameter pulling stage with pulling rate of 0.5 mm/h




(c) tailing stages with pulling rate of 8 mm/h

Fig. S3 The optimum thermal and flow fields (left) and the interface (right) at different stages with different pulling rates

## Supplement 2variation of Curie temperature at head and tail of crystal with different neodymium and magnesium concentrations

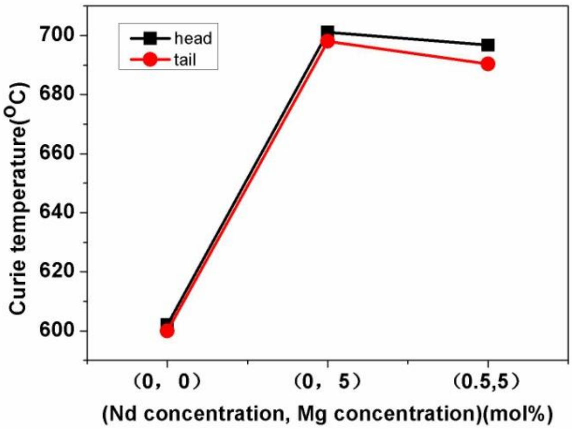



Fig. S4 Variation of Curie temperature with the different neodymium and magnesium concentrations. The black and red lines correspond respectively to the wafer from the head and the tail of LT crystal body.

From Fig. S4, doping with magnesium (within 5 mol%) increases the Curie temperature of the LT crystal, but neodymium (within 0.5 mol%) decreases it. For the MgO:LT crystal, the Curie temperature at the head of the crystal body is larger than that at the tail. For the Nd:MgO:LT crystal, the Curie temperature at the head decreases less than that at the tail. As is well known, the segregation coefficient of magnesium in the LT crystal is greater than 1. So the magnesium concentration at the head of the crystal body is a little larger than that at the tail, which leads to a higher Curie temperature at the head. Likewise, the segregation coefficient of neodymium is less than 1, which makes the Curie temperature at the head decrease less than that at the tail. It can be concluded that the Curie temperature measurement verifies the prior findings on the segregation coefficient of magnesium and neodymium in the LT crystal.

## Supplement 3 Crystallinity characterized by high resolution X-ray rocking curve

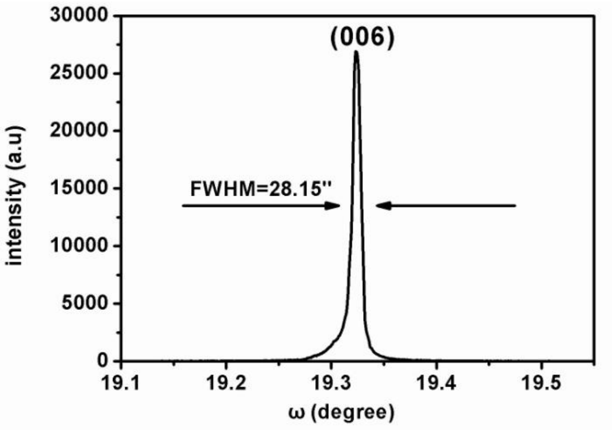



Fig. S5 X-ray rocking curve of the poled Nd:Mg:LT crystal.

The peak is highly symmetrical without any splitting, and its full width at half-maximum (FWHM) is only 28.15", indicating a complete lattice structure for the crystal and few defects. The high crystal quality provides a basis for better physical and optical properties that lead to high laser output.