

深紫外铁电氟化物晶体BaMgF₄由于具有极短的紫外截止波长（125nm），有望作为非线性光学倍频晶体，实现深紫外全固态激光器的制造。目前对其非线性性质的研究还不够充分。我们课题组通过实验测量了BaMgF₄三个晶轴方向上的三阶非线性系数，发现其具有较大的各项异性。其中最大值为 $2.35 \times 10^{-18} \text{m}^2/\text{W}$ ，这在已知的铁电晶体中几乎是最大的。这说明具BaMgF₄有明显的三阶非线性现象。于是，我们研究了它的自位相调制效应，并与传统的铁电晶体LiNbO₃进行了比较。另外，我们还进行了初步的铁电畴极化实验，发现BaMgF₄的单畴呈现六边形。

The BaMgF₄ single crystal is grown by Bridgman method. The third-order nonlinear refractive indices along three crystallographic axes are determined by Z-scan technique with femtosecond laser for the first time. The largest one which has a value of $2.35 \times 10^{-18} \text{m}^2/\text{W}$ is along the c-axis and the corresponding third-order nonlinear susceptibility is $1.24 \times 10^{-12} \text{esu}$. This value is compared with LiNbO₃ through self-phase modulation effect. Furthermore, the poling experiment is performed to investigate its ferroelectric property.

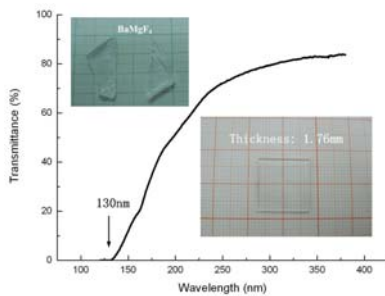


Figure 1 | UV/UVU transmittance spectrum of the BaMgF₄ single crystal with a thickness of 1.76 mm shown in the inset.

图1 | BaMgF₄晶体紫外透过率谱

The VUV absorption edge of BaMgF₄ is about 130 nm. There is a relatively high transmittance in the VUV region so that many nonlinear processes in BaMgF₄ can be observed in this region. According to the absorption edge, its band gap can be estimated at 9.5 eV.

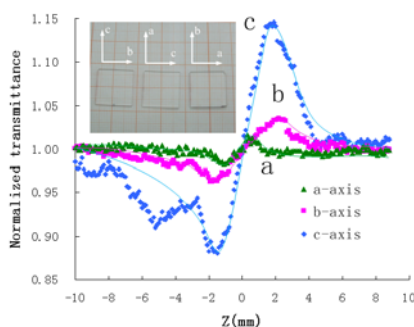


Figure 2 | Z-scan traces and fittings of the BaMgF₄ crystal along a-, b- and c-axis, respectively. The irradiance at the focus is $4.3 \text{GW}/\text{cm}^2$.

图2 | 沿着BaMgF₄晶体三个轴向上的Z-scan曲线

The Z-scan measurement shows that the third-order nonlinear refractive indices of BaMgF₄ have strong anisotropy. The largest one is along c-axis, possessing a value of $2.35 \times 10^{-18} \text{m}^2/\text{W}$.

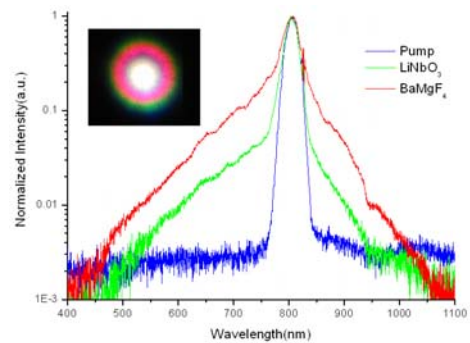


Figure 3 | Supercontinuum spectra of BaMgF₄ and LiNbO₃. The propagation distance in the crystal is 1 mm and the irradiance is $1 \times 10^{18} \text{W}/\text{m}^2$

图3 | BaMgF₄晶体和LiNbO₃晶体超连续谱

In the same environment, the spectrum of BaMgF₄ is obviously wider than that of LiNbO₃, and it conversely demonstrates that BaMgF₄ has larger third-order nonlinear refraction because of the self-phase modulation effect.

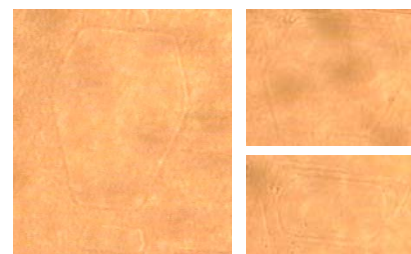


Figure 4 | Micrograph of single domains on the BaMgF₄ wafer with thickness of 0.5 mm

图4 | BaMgF₄单畴显微照片

After poling experiment, the profile of single domains on the +Z surface of the BaMgF₄ wafer can be observed. Most of the shapes are hexagon. The orientation of the sides may depend on the crystallographic planes of BaMgF₄ crystal.