

## Supplemental material

### Crystal Orientation

A Laue x-ray diffraction pattern was recorded to determine the orientation of the crystalline sample in this work. Figure S1 shows the reflected diffraction pattern and the simulated pattern based on the lattice parameters of single crystal  $\text{LiYF}_4$ . Excellent agreement was found between the measured and simulated patterns, confirming both the composition and orientation of the crystal. The lack of distortion in Laue spots confirmed the single crystal nature of the sample and absence of internal stress. The orientation of crystal axes with respect to faces of the cut and polished sample are illustrated in Figure S2.

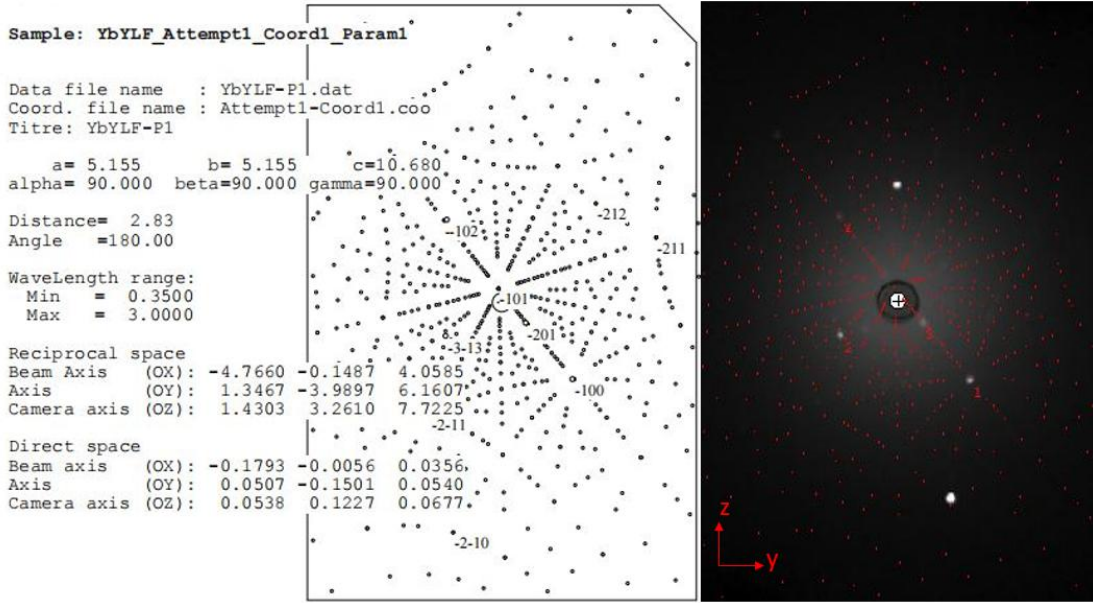


Figure S1: Simulated and measured Laue diffraction pattern. The excellent agreement confirms the crystal type and orientation.

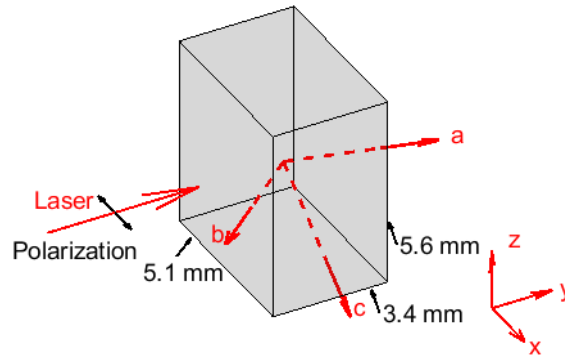


Figure S2: The crystal orientation and crystal cut. The crystal axes are indicated by the rotation matrix.

The sample was polished to maintain 3 pairs of parallel surfaces and the lab frame coordinates  $x$ ,  $y$ ,  $z$  correspond to normals to various surfaces as indicated in Fig. S2. Axes  $x$ ,  $y$ ,  $z$  were parallel to the 5.1 mm, 3.4 mm, 5.6 mm sample edges respectively. The pump laser propagated along  $y$ , with a polarization parallel to  $x$  for laser cooling measurements. The unit vectors of axes  $x$ ,  $y$ ,  $z$  are defined to be

$$\hat{x} = \begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix}, \hat{y} = \begin{bmatrix} 0 \\ 1 \\ 0 \end{bmatrix}, \hat{z} = \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix}. \quad (\text{S1})$$

The crystal axes are related to the lab frame by the rotation equation

$$\begin{bmatrix} a \\ b \\ c \end{bmatrix} = R \begin{bmatrix} x \\ y \\ z \end{bmatrix} \quad (\text{S2})$$

where

$$R = R_z(\gamma)R_y(\beta)R_x(\alpha), \quad (\text{S3})$$

where

$$R_x(\alpha) = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos(\alpha) & -\sin(\alpha) \\ 0 & \sin(\alpha) & \cos(\alpha) \end{bmatrix}, \quad (\text{S4})$$

$$R_y(\beta) = \begin{bmatrix} \cos(\beta) & 0 & \sin(\beta) \\ 0 & 1 & 0 \\ -\sin(\beta) & 0 & \cos(\beta) \end{bmatrix}, \quad (\text{S5})$$

$$R_z(\gamma) = \begin{bmatrix} \cos(\gamma) & -\sin(\gamma) & 0 \\ \sin(\gamma) & \cos(\gamma) & 0 \\ 0 & 0 & 1 \end{bmatrix} \quad (\text{S6})$$

represent rotations of  $\alpha$ ,  $\beta$ ,  $\gamma$  about axes  $x$ ,  $y$ ,  $z$ . The Euler angles  $\alpha$ ,  $\beta$ ,  $\gamma$  were  $181.8^\circ$ ,  $337.7^\circ$ ,  $38.6^\circ$ , respectively and the calculated rotation matrix was

$$R = \begin{bmatrix} 0.7231 & 0.6326 & 0.2774 \\ 0.5767 & -0.7739 & 0.2614 \\ 0.3802 & -0.0289 & -0.9245 \end{bmatrix}. \quad (\text{S7})$$

The unit vectors of crystal axes  $a$ ,  $b$ ,  $c$  were therefore

$$\hat{\mathbf{a}} = R \cdot \hat{\mathbf{x}} = \begin{bmatrix} 0.7231 \\ 0.5767 \\ 0.3802 \end{bmatrix}, \quad (\text{S8})$$

$$\hat{\mathbf{b}} = R \cdot \hat{\mathbf{y}} = \begin{bmatrix} 0.6326 \\ -0.7739 \\ -0.0289 \end{bmatrix}, \quad (\text{S9})$$

$$\hat{\mathbf{c}} = R \cdot \hat{\mathbf{z}} = \begin{bmatrix} 0.2774 \\ 0.2614 \\ -0.9245 \end{bmatrix}. \quad (\text{S10})$$