1. MILLER INDEX AND CRYSTAL PLANE

The notation of the Miller index defines the crystal planes and directions. A single crystal is composed of infinite unit cells, which correspond to the simplest possible units in the crystal. These unit cells are categorized into seven fundamental patterns, and the ordered structure characterizes various optical and material properties of the crystal, such as refractive index, absorption, cleavage, plastic deformation, and crystal anisotropy [1, 2].

When the three lattice vectors \(a\), \(b\), and \(c\) are used to denote a unit cell, the following notation \([h k l]\) expresses the direction:

\[
ha + kb + lc,
\]

where \(h\), \(k\), and \(l\) are relatively prime integer numbers. For instance, \([100]\) denotes the direction along the \(a\)-axis. A negative number indicates the opposite direction and is written as \([100]\).

The crystal plane is also denoted with such indices as well as the direction so that \([h k l]\) gives a plane that intercepts the three points \(a/h\), \(b/k\), and \(c/l\). With a cubic lattice structure, the \([h k l]\) direction is normal to the \((h k l)\) plane.

Specifically, \(\text{MgF}_2\) is known as a uniaxial crystal, and it is slightly birefringent depending on the polarization and the direction of the incident light. Therefore, \(\text{MgF}_2\) material is often cut with the optical axis perpendicular to the plane of the window to avoid birefringence in commercial products. Such window materials are referred to as \(c\)-cut or \(z\)-cut, in which the optical axis is normal to the \((001)\) plane.

2. SINGLE CRYSTAL DIAMOND TOOL

Figure S1(a) shows schematics of the single crystal diamond tool used in this work. The shape of the tool is fixed to the turning machine, as shown in Fig. S1(a). We used two types of single crystal diamond tools, and their properties were as follows: Tool #1 had a 0.2 mm nose radius, a \(-20^\circ\) rake angle, and a \(10^\circ\) clearance angle [Fig. S1(b)]; Tool #2 had a 0.01 mm nose radius, a \(0^\circ\) rake angle, and a \(10^\circ\) clearance angle [Fig. S1(c)]. Tool #1 had a negative rake angle and a large nose radius, which made it suitable for efficiently manufacturing millimeter size workpieces such as crystalline microresonators. Therefore, we used Tool #1 for the orthogonal cutting experiment and the rough turning stage in the ultra-precision turning. On the other hand, Tool #2 used in the finish turning process had a sharper edge with a rake angle of \(0^\circ\), and this made it possible to achieve stable cutting because the small rake angle reduced the thrust force on the material [3].

3. SUPPLEMENT ON ULTRA-PRECISION TURNING

A. Detailed cutting condition for cylindrical turning

When choosing suitable parameters and a diamond tool for use in manufacturing, we must take account of the relationship between the edge radius of the tool, the undeformed chip thickness, and the critical depth of cut, namely the size effect [4]. If the undeformed chip thickness is much smaller than the edge radius of the tool, the cutting process does not occur (the plowing effect occurs instead). The critical thickness at which material removal begins is called the minimum chip thickness, and chip formation
occurs when the undeformed chip thickness reaches this critical value [4].

Furthermore, the undeformed chip thickness must not exceed the critical depth of cut, which indicates the boundary of the ductile-brittle transition to minimize excess surface roughness. Considering the discussion above, the depth of cut should exceed the tool edge radius and be smaller than the critical depth of cut. The tool edge radii of both Tool #1 and Tool #2 are less than approximately 10 nm. Hence, we can see that a depth of cut of 50 nm is a feasible value for our ultra-precision turning.

Table S1-S3 show the cutting condition for each step in ultra-precision turning. The effective cutting speed is given by the rotation speed multiplied by \(2\pi R\), where \(R\) is the radius of the cylinder workpiece, and feed per revolution (mm/rev) is given by the feed rate (mm/min) divided by the rotation speed (min\(^{-1}\)). In this work, we employed rotation speeds of 500 min\(^{-1}\) and 1000 min\(^{-1}\) for 3 mm and 0.5 mm diameter workpieces, respectively. Nevertheless, it should be noted that the rotation speed is less important than the other parameters (i.e., feed rate, depth of cut, and the choice of the diamond tool) as regards machined surface quality [5].

### Table S1. Cutting condition for rough turning

|             | Rotation speed | Feed rate | Depth of cut | Diamond tool |
|-------------|----------------|-----------|--------------|--------------|
| Rotation speed | 1000           | 20        | 2.0          | Tool #1      |

### Table S2. Cutting condition for pre-finish turning

|             | Rotation speed | Feed rate | Depth of cut | Removed thickness | Diamond tool |
|-------------|----------------|-----------|--------------|--------------------|--------------|
| Rotation speed | 500 or 1000    | 0.5       | 100          | 8.0                | Tool #1      |

### Table S3. Cutting condition for finish turning and microresonator fabrication

|             | Rotation speed | Feed rate | Depth of cut | Removed thickness | Diamond tool |
|-------------|----------------|-----------|--------------|--------------------|--------------|
| Rotation speed | 500 or 1000    | 0.1       | 50           | 2.0                | Tool #2      |

MgF\(_2\) has various crystal planes due to its complex crystal structure, which is a rutile structure (tetragonal structure). In addition to MgF\(_2\) crystal, CaF\(_2\) is a representative crystalline material exhibiting similar optical properties to MgF\(_2\) except for its thermal property, which induces unstable oscillatory behavior due to the opposite signs of the thermo-optic and thermal expansion coefficients [6]. Nevertheless, CaF\(_2\) has a simpler and symmetric cubic structure, namely a fluorite structure and is easier to understand in terms of crystal anisotropy [3, 5, 7]. The cleavage planes that lead to brittle fracture are also different in these crystals [i.e., (110) for MgF\(_2\), and (111) for CaF\(_2\)]. We note that strontium fluoride (SrF\(_2\)) and barium fluoride (BaF\(_2\)), which can be used for nonlinear experiments [8–10], exhibit the same crystal structure as CaF\(_2\). Therefore, we believe that the cutting condition used for CaF\(_2\) can also be applied to SrF\(_2\) and BaF\(_2\) crystal. Besides, lithium niobate (LiNbO\(_3\)) and lithium tantalate (LiTaO\(_3\)) crystals that are used for quantum optics experiments [11] have more complex structures, namely trigonal structure; and therefore further investigations of cutting parameters are needed. Table S4 shows crystal structure and lattice constant of major crystalline resonator materials.

In terms of hardness of crystal material (cf., Knoop hardness), MgF\(_2\) is 2.6 times harder than CaF\(_2\), and CaF\(_2\) is 1.8 times harder than BaF\(_2\) [12]. With ultra-precision turning, the workpiece material has a fundamental influence on diamond tool wear related to tool life as well as the cutting condition [13].

C. Cleaning with ultrasonic method

The detailed cleaning procedure is as follows. First, we used an acetone solution to reduce the amount of oil lubricant. Then, we cleaned the resonator with ethanol or isopropanol. Next, the resonator surface was treated using a dust-free dry nitrogen blower. And finally, we carefully observed the resonator surface using a stereomicroscope. We repeated this cleaning procedure several times to ensure that no particles or chips remained close to the resonator surface.

The above cleaning can be performed with an ultrasonic cleaner as well as by conventional wipe cleaning with lens tissue. Figure S3 shows the setup we used for the ultrasonic cleaning of the crystalline microresonator with solvent. The resonator...
Table S4. Crystal structure and lattice constant for crystal materials [2]

| Material | Structure      | Lattice constant (Å) |
|----------|----------------|----------------------|
| MgF₂     | Tetragonal (rutile) | \(a = 4.62, c = 3.05\) |
| CaF₂     | Cubic           | \(a = 5.46, c = a\)  |
| SrF₂     | Cubic           | \(a = 5.78, c = a\)  |
| BaF₂     | Cubic           | \(a = 6.20, c = a\)  |
| LiNbO₃   | Trigonal (R3c)  | \(a = 5.15, c = 13.9\) |
| LiTaO₃   | Trigonal (R3c)  | \(a = 5.15, c = 13.8\) |

is fixed in position with a jig and cleaned using a commercial ultrasonic cleaner with a frequency of 40 kHz (Branson M1800-J, BRANSON). The cleaning has continued for several minutes. After the cleaning, we observed small particles floating in the solvent, which convinced us that the lubricant or attached specks of dust and particles had been removed from the crystalline microresonator.

D. Triangular WGM microresonator fabrication

Figure S2(a) shows the procedure for manufacturing a triangular whispering gallery mode (WGM) microresonator. For both the spherical and triangular WGMs, the turning was performed under the finish turning condition to shape the microresonator structure as designed. The triangular WGM structure is designed with an apex angle of 120° and a diameter of 502 µm. The recorded Q value was \(1.03 \times 10^7\) at 1552.2 nm as shown in Fig. S2(b).

![Fig. S2.](image)

Fig. S2. (a) Fabrication flow of triangular WGM microresonator by ultra-precision turning. (b) Normalized transmission spectra of fabricated CaF₂ resonator, yielding a loaded Q of 10.3 million.

![Fig. S3.](image)

Fig. S3. (a) Setup for the ultrasonic cleaning of a crystalline microresonator. Adhered dust and lubricant oil on the resonator surface are removed by cleaning with solvent. (b) Comparison of solvent before and after ultrasonic cleaning.

![Fig. S4.](image)

Fig. S4. (a) and (b) TEM images of the subsurface damage of machined and polished single crystal samples, respectively. The subsurface damage is described in detail Ref. [14]. (c) and (d) SEM images of the surface condition after the machining and polishing processes, respectively.

4. OBSERVATION OF MACHINED AND POLISHED SURFACE

Here, we evaluate the surface and subsurface damage of a crystalline microresonator fabricated with ultra-precision turning and polishing with diamond slurry. We observed the subsurface damage using a transmission electron microscope (TEM), where the samples were cut into pieces of approximately 0.1 × 0.1 mm from the cylinder workpieces. For comparison, we prepared a CaF₂ microresonator fabricated by hand polishing and yielding a Q of \(1 \times 10^9\).

Figures S4(a) and S4(b) are TEM images of a machined surface and a polished surface, respectively. We observed clear subsurface damage with a depth of 11 nm in the machined sample; on the other hand, no significant damage was observed in the sample fabricated with polishing. These results suggest that the
difference in the subsurface layer could influence the Q-factor obtained with the ultra-precision turning and polishing method. To compare the surface condition, we observed the machined and polished surface conditions of fabricated resonators using a SEM. Figures S4(c) and S4(d) show the SEM images of machined and polished surfaces, respectively. On the machined surface, we can see that chips have adhered and there are periodic tracks caused by the regular motion of the ultraprecision lathe. However, we find that the polished surface is very smooth, although a few grooves are irregularly observed when the manual process is used.

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