

Supplementary Information: A mid-infrared Brillouin laser using ultra-high-Q on-chip resonators

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Supplementary Note 1: Details of fabrication and optimization

A. Formation of trapezoidal chalcogenide glass on-chip device

Detailed steps for fabricating the trapezoid chalcogenide glass (ChG) resonator are given here (Figure S1a). The SiO₂ trapezoidal substructure is fabricated from an 8 μm thick thermal oxide layer grown in a furnace system (Tytan Mini 1800, TYSTAR). To control the wet-etched angle of SiO₂, a vapor of hexamethyldisilazane (HMDS) was applied to the SiO₂ layer to change its surface adhesion. The photoresist was patterned onto the SiO₂ layer using a lithography system (MA6 Mask Aligner, Carl Suss) and used as an etch mask for wet etching with a buffered oxide etchant. Then, photoresists are removed using a mixture of sulfuric acid and hydrogen peroxide. Finally, XeF₂ dry etching (XeF₂ etching system, Teraleader Co. Ltd.) is applied to the silicon substrate to separate the optical modes from the high refractive index silicon and prevent optical mode leakage due to the bending structure. More details about the fabrication and its resulting quality can be found in previous papers^{1,2}.

ChG films are directionally deposited on this SiO₂ substructure using a high vacuum (HV) thermal evaporator ($<5 \times 10^{-8}$ torr) (KVE-T2000, Korea Vacuum Tech). Four different evaporation sources can be simultaneously loaded in the chamber, allowing multi-layer deposition without breaking the vacuum. High-purity ChG ingots used for making low-loss fibers were chosen as evaporation sources: As₂S₃ (AMTIR-6, Amorphous Materials Inc.), Ge₃₃As₁₂Se₅₅ (AMTIR-1, Amorphous Materials Inc.), As₂Se₃ (Ningbo University), and Ge_{11.5}As₂₄S_{64.5} (Ningbo University). For the resonator with the highest Q-factor in this study, specially purified As₂Se₃ ingot was used as an evaporation source (details in sub-sections C and D). Before being loaded into the chamber, these sources are sonicated and then dehydrated inside a medium vacuum to remove surface contaminants. The SiO₂ structure's surface is also cleaned by O₂ plasma using an end-hall permanent magnet ion source (EH 400, KRI) for 15 minutes before film deposition. ChG films were deposited at a rate of 0.5~1 Å/s for the highest film quality while being monitored by a residual gas analyzer. Notably, we passivated the films with a thin layer of Ge-As-Se ChG, instead of the more common atomic layer deposited (ALD) layer such as Al₂O₃, to prevent crystallization. Details about the passivation are given in sub-section B. The samples are rotated and revolved at a different frequency during deposition to achieve high film thickness uniformity ($< 2\%$ over a 4-inch wafer), which is shown in Figure S1b.

After deposition, post-treatments³, thermal - and light annealing, are applied to stabilize the films and further decrease their optical loss. See sub-section C for details.

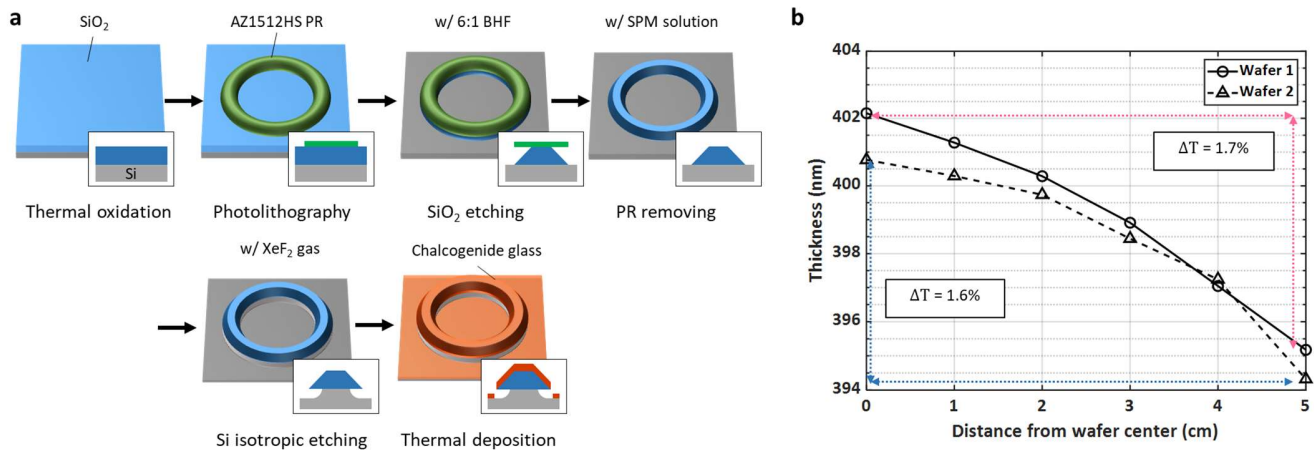


Fig.S1 | Fabrication process and thickness uniformity of thermally deposited ChG film. **a**, Schematic of the whole fabrication process. **b**, Thickness uniformity of two As₂S₃ films deposited on 4-inch wafers at a different run. The thickness distribution is below 2% from the center to the edge of the wafer. The thickness was measured by a reflectometer (K-MAC, Spectrathick).

B. Device passivation by Ge-As-Se thin layer.

Arsenic-based ChG films, especially As_2S_3 and As_2Se_3 , are known to quickly form oxide crystals (As_2O_3) under ambient conditions, which can lead to excess scatter loss. To prevent this phenomenon, passivation films such as atomic layer deposited (ALD) Al_2O_3 or fluoropolymers are conventionally added. However, substantial amounts of precursor-originating impurities commonly get incorporated into ALD films when they are deposited at temperatures below the glass transition temperature of ChG⁴, as it is generally lower than that required for the proper ALD chemical process. These films containing impurities can result in high optical losses in mid-IR resonators, even when applied with nanometer thicknesses. While fluoropolymer films have been used to address this, they have limitations at wavelengths beyond $4\text{ }\mu\text{m}$ due to their intrinsic C-F absorption bands⁵.

To resolve these issues, all of our presented devices were passivated using a crystallization-resistant⁶, low-loss⁷ ChG material, Ge-As-Se, as mentioned in the main text. To test its passivation ability, a 10 nm-thick Ge-As-Se layer was deposited on an As_2S_3 film with a rate of $0.5\text{ }\text{\AA}/\text{s}$ using a $\text{Ge}_{33}\text{As}_{12}\text{Se}_{55}$ (AMTIR-1) evaporation source. As shown in Figure S2, the passivated As_2S_3 film stays unoxidized for at least 12 hours under ambient conditions. No signs of crystallization were observed even after being placed in the same environment for several months.

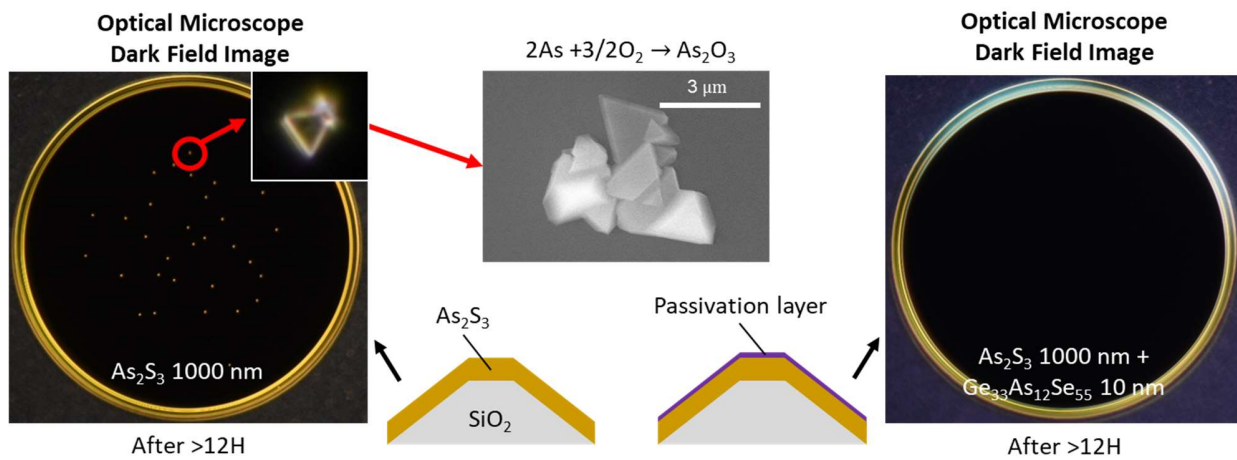


Fig.S2 | Passivation performance of Ge-As-Se layer applied on As_2S_3 films. Images of $1\text{ }\mu\text{m}$ thick As_2S_3 film deposited onto trapezoid resonators and surrounding silicon substrates, taken using darkfield optical microscopy. The left device is not passivated while the right has an additional 10 nm Ge-As-Se passivation layer on top. After being exposed to the atmosphere for 12 hours, the left sample developed numerous As_2O_3 crystals (SEM image shown in the middle) while the passivated sample remained unchanged.

C. Improvements in optical losses in the mid-IR

Thermally deposited ChG films contain much more inhomogeneities than melt-quenched bulk ingots, resulting in higher optical loss⁸. Thermal and light annealing treatments are known to relax these local structural defects, such as homopolar bonding (As-As, S-S, and Se-Se) and crystalline states, by providing the energy for bond realignment^{3,9-11}, and to minimize the change of the index in the resonators working at high optical powers. All resonators used in this paper were thermally annealed at a temperature of 130 °C under a medium vacuum ($< 10^{-3}$ torr) for 12 hours. The temperature was set to be below the glass transition temperature (T_g) of all ChG materials forming the multi-layer resonator (T_{g, As_2S_3} : 182 °C, T_{g, As_2Se_3} : 178 °C, and $T_{g, Ge_{33}As_{12}Se_{55}}$: 353 °C). A green LED light ($\lambda = 540$ nm) was then employed for light annealing after the device was cooled to room temperature. Figure S3a shows an overall enhancement of mid-IR Q-factors by this annealing process, which coincides with the previously reported loss reduction measured in the near-IR regime⁸.

The material loss of ChG films starts to matter when the resonator Q-factor goes beyond 10^7 . Research on low-loss ChG fibers shows that their material loss at mid-IR wavelength strongly depends on the level of included chemical impurities¹². Overall enhancement of mid-IR Q-factors was also observed on our on-chip resonators by using a higher purity source material (Figure S3b). The sources used for depositing the As_2Se_3 core layer of two resonators shown in Figure S3b have different purities: one was purified only once by the method given in sub-section D, while the other was purified twice by the same method. The purity of the cladding material (As_2S_3) and all geometric parameters were the same for both resonators. The resonator with a doubly purified core shows a higher Q-factor over the whole wavelength range, confirming that the purity of the ChG source does affect the loss of thermally deposited films. All resonators presented in the main text having an As_2Se_3 core layer used the doubly purified ingot as their evaporation source.

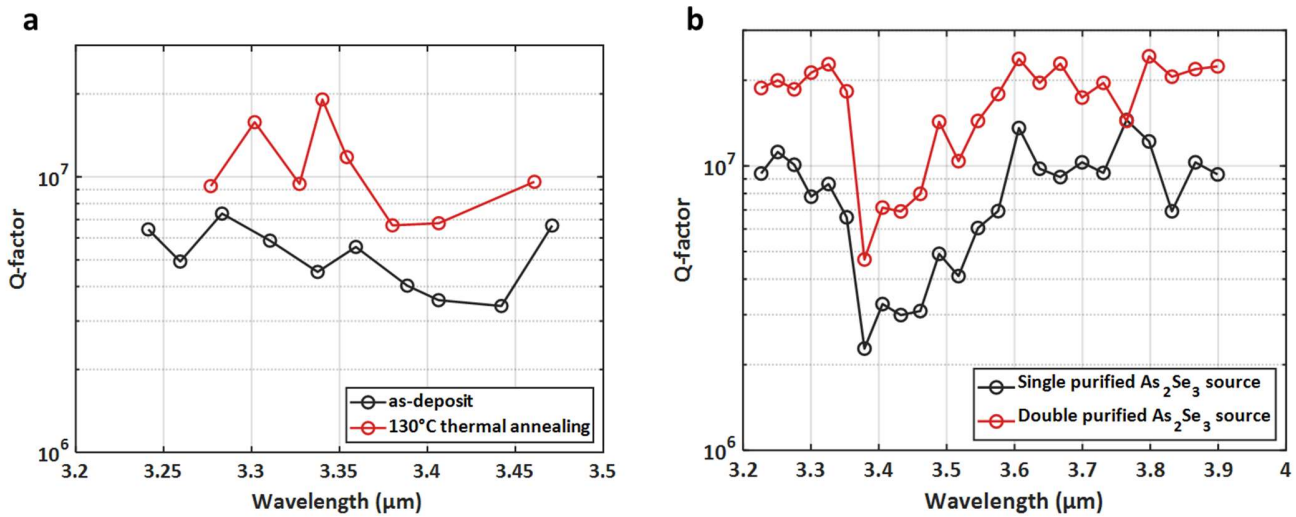


Fig.S3 | Q-factor enhancement by film post-treatment and evaporation source purification. **a**, Mid-IR Q-factor enhancement after thermal annealing. The annealing was done at 130 °C for 12 hours inside a medium vacuum chamber. **b**, Q-factor of two resonators having As_2Se_3 core layers with different chemical purity. The core layer of the resonator corresponding to the black curve was deposited using a singly purified As_2Se_3 source. The other resonator (red curve) used a doubly purified As_2Se_3 ingot as an evaporation source. The overall Q-factor is enhanced around two times by using a higher purity source.

D. Purification of As_2Se_3 deposition source material.

The high-purity As_2Se_3 glass ingots used as deposition sources were distilled using three steps (Figure S4).

First, to remove Se-H impurities, a dynamic distillation method was applied with a hydrogen removal agent (Figure S4a). The temperature of the 1st quartz tube is set to 350 °C, while the other tubes remain at room temperature. The Se raw material rapidly evaporates and condenses in the 2nd tube due to the pressure difference. Then the 1st and 2nd tubes are set to the same temperature and pressure to ensure that Se and As fully mix into the As_2Se_3 compound, which is then distilled and condensed in the 3rd tube. Then, the 1st, 2nd, and 3rd tubes are set to the same temperature to ensure that As_2Se_3 compounds fully mix with the TeCl_4 hydrogen removal agent. TeCl_4 reacts with the Se-H impurity generating highly volatile HCl gas, which is pumped out. After the dynamic distillation is completed, the As_2Se_3 compound is melted at a temperature of 650 °C in a rocking furnace for 12 hours to fully homogenize the compound. Subsequently, it is cooled down to 450 °C and then air quenched. Finally, it is annealed at a temperature starting from 170 °C, which slowly drops to room temperature for 15 hours to reduce the stress inside the glass.

Second, a static distillation technique was used to remove As-O and OH impurities from the glass (Figure S4b). The ready-made As_2Se_3 glass and Mg are put together on the left side of the H-type quartz tube. The tube is then put in a furnace that has a precisely controlled dual-temperature region. When the reaction between As_2Se_3 and Mg is done, As_2Se_3 evaporates into the right part of the tube due to the pressure difference caused by the different temperature settings of the two tube sides. The O- and OH-related impurities stay on the left side of the tube as MgO .

When oxygen is extracted from the OH-impurities in step 2, the remaining hydrogens are reintroduced to the glass in the form of H-related impurities. These impurities are eliminated as a last step by applying the same dynamic distillation process once more (Figure S4c).

For the highest quality resonators in this work, extra-pure As_2Se_3 ingots that were purified by applying this entire process twice were used as the evaporation source. More details about the proceeds and their results are described in another paper¹³.

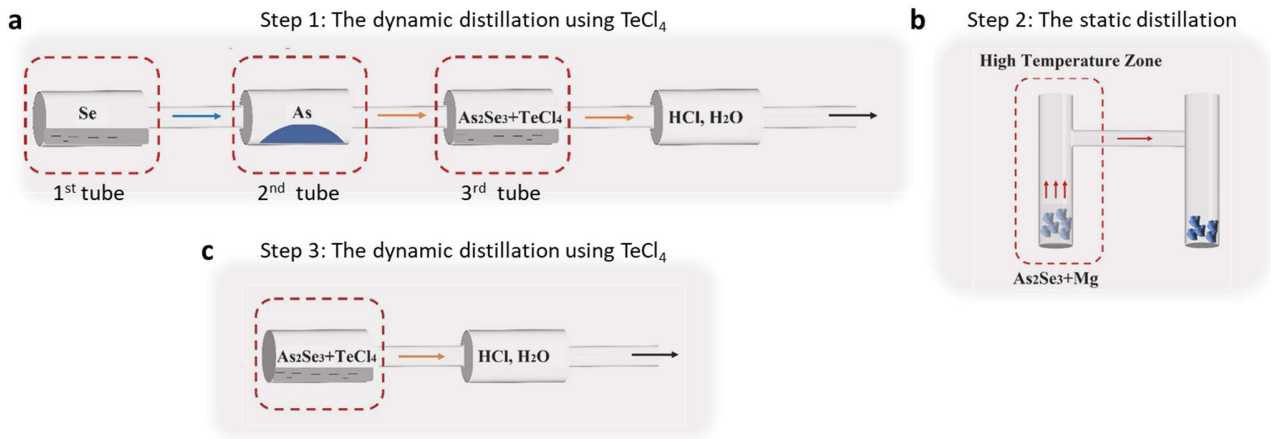


Fig.S4 | The preparation process of high-purity As_2Se_3 glass. The high-purity As_2Se_3 glass ingot is fabricated and distilled in three steps. **a**, Step 1 : Removal of Se-H impurities by dynamic distillation using TeCl_4 as a hydrogen removal agent. **b**, Step 2: Elimination of oxygen from As-O and -OH impurities using static distillation. **c**, Step 3: Removal of hydrogen impurities using the same dynamic distillation as step 1, which are reintroduced from the oxygen extracted -OH impurities.

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Supplement Note 2: Mid-IR Q-factor of resonators without an upper cladding.

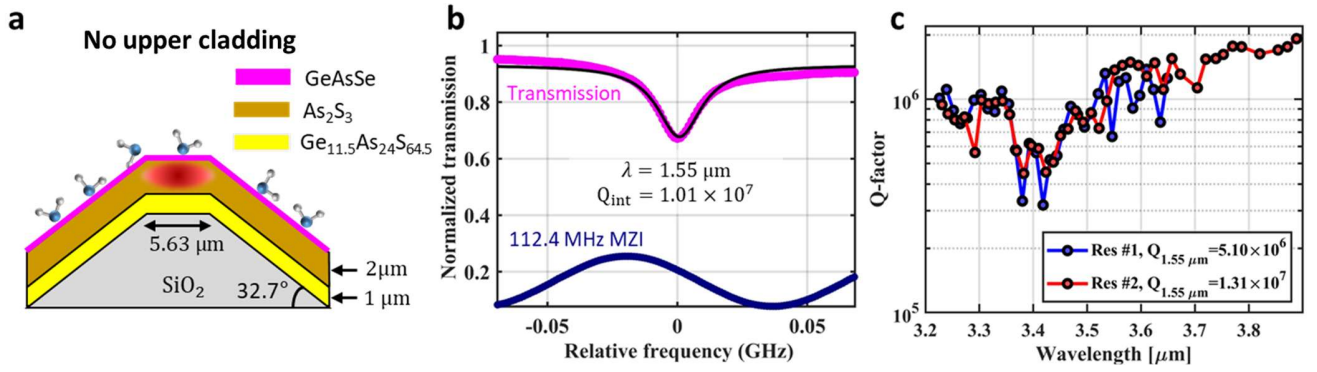


Fig.S5 | Near-IR and mid-IR Q-factor of resonators without an upper cladding. **a**, Cross-section drawing of the resonator with no upper cladding, discussed in Figure 3a of the main text. The core material was chosen as As_2S_3 , unlike the resonators with upper cladding, which have As_2Se_3 as their core. The bottom cladding material was also modified to $\text{Ge}_{11.5}\text{As}_{24}\text{S}_{64.5}$, which has a lower index than As_2S_3 . **b**, Near-IR Q-factor of the resonator measured at $1.55 \mu\text{m}$ wavelength. The magenta line corresponds to the waveguide transmission while the navy line indicates the 112.4 MHz Mach-Zehnder interferometer (MZI) signal. **c**, Mid-IR Q-factor spectra of two different resonators without an upper cladding. Both resonators are nearly identical in geometry but have different scatter-limited near-IR Q-factors due to surface defects.

The mid-IR Q-factor of resonators with no upper cladding (bilayer resonators) presented in the main text Figure 3a is discussed in more details. To confirm that their low mid-IR Q-factor ($Q \sim 10^6$) is not a consequence of other general loss factors mentioned in the main text, especially surface scattering, their Q-factors were measured at $1.55 \mu\text{m}$ near-IR wavelength where the scattering loss dominates the total optical loss.

To exactly obtain the scatter-limited loss (Q-factor) of bilayer resonators, their material absorption loss should be negligible. For this purpose, As_2S_3 was chosen as the core material of bilayer resonators (Figure S5a) instead of As_2Se_3 , which has a two-photon absorption loss near $1.55 \mu\text{m}$ wavelength. The measured $Q = 1.01 \times 10^7$ at $1.55 \mu\text{m}$ wavelength shown in Figure S5b, which is an order higher than the mid-IR values, confirms that the scatter loss is not the reason for the low mid-IR Q.

For more certain proof, mid-IR Q-factors of two bilayer resonators with different near-IR Q-factors (scatter loss) were compared (Figure S5c). Resonator sample 1 (Res #1) has nearly the same geometric parameters as sample 2 (Res #2) but its near-IR Q-factor is two times lower due to surface defects coming from fabrication errors. However, their Q-factor spectra over the mid-IR wavelengths are nearly identical, confirming that their mid-IR Q-factor is not limited by surface scatter loss. Finally, we note that the coincidence in the mid-IR Q-factor of two resonators is also not a consequence of SiO_2 absorption coming from the substrate. Optical simulations confirm that the $1 \mu\text{m}$ thick $\text{Ge}_{11.5}\text{As}_{24}\text{S}_{64.5}$ bottom cladding, as drawn in Figure S5a, is enough to decrease silica absorption loss below 0.02 dB/cm ($Q > 8 \times 10^6$) over the $3 - 4 \mu\text{m}$ wavelength.

Supplement Note 3: Linear proportionality between the mid-IR loss and surface electric field intensity

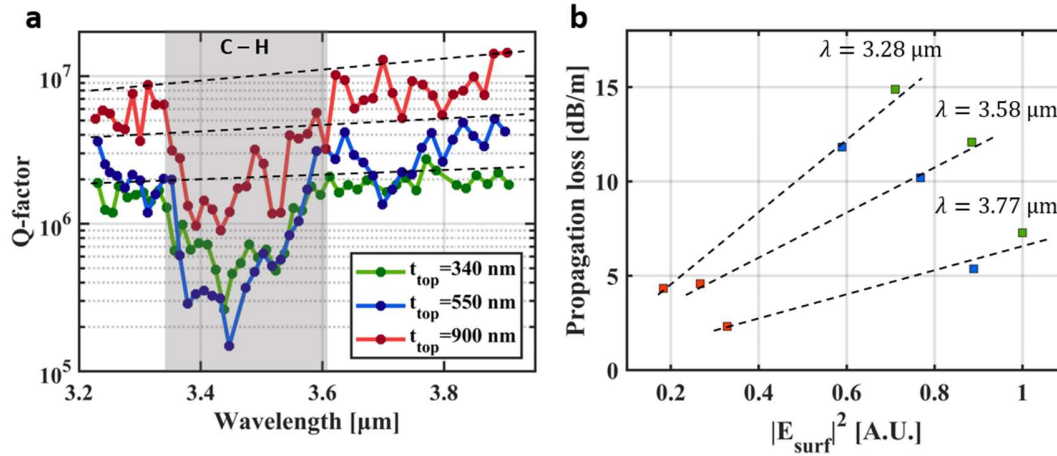


Fig.S6 | Mid-IR Q-factor spectrum versus surface electric field intensity. **a**, Q-factor spectrum of three resonators with different upper cladding thicknesses (t_{top}). An increase in overall Q-factor is observed as the upper cladding gets thicker. The grey dashed lines are drawn as guides to the eye, indicating the Q-factor limit imposed by surface-adsorbed water absorption. The grey-scaled region indicates the hydrocarbon absorption band. The material composition and layer thickness of the three resonators slightly vary which is given in Table S2. **b**, Propagation loss of three resonators plotted versus the normalized electric field intensities at their outermost surfaces. The loss values were converted from Q-factors shown in Figure a, with the same color sets used to indicate the upper cladding thickness. The linear proportionality between the field intensity and propagation loss is displayed for three different wavelengths (3.28, 3.58, and 3.77 μm). The grey dashed lines are drawn as guides to the eye.

The proportionality between the measured Q-factor and the outermost surface electric field (E-field) intensity is presented in more detail. Mid-IR Q-factors of three resonators having different upper cladding thicknesses are measured as shown in Figure S6a. An increase in overall Q-factor is observed as the upper cladding becomes thicker owing to the decrease in surface E-field intensity. The outermost surface material of the three resonators is the same, meaning the concentration of surface contaminants (especially water) can be considered nearly identical under the same environment. The Q-factors residing in the hydrocarbon absorption band (grey-scaled region) deviate from this tendency since their surface concentration varies greatly depending on minor fabrication details.

The propagation loss converted from the measured Q-factor is plotted as a function of E-field intensity at the outermost surface (Fig. S6b). The E-field intensity was calculated by a finite element method solver (COMSOL) using measured geometric parameters. The direct proportionality between the loss and E-field intensity is observable for three sample wavelengths outside the hydrocarbon band, confirming that the mid-IR loss is indeed being dominated by surface absorption. On a single device, the loss is larger at shorter wavelengths since the increase in water absorption coefficient (maximum near 2.9 μm wavelength) more than compensates for the decrease in surface field intensity.

The material composition and layer thicknesses of the three resonators used here have slight differences but have a minor effect on the loss tendency. The exact parameters are given in Table S2 below.

Table S2 | Material type and layer thicknesses of the three resonators are presented in Fig. S6.

Resonator #	Bottom cladding	Core	Upper cladding
#1: $t_{top} = 340 \text{ nm}$	$\text{Ge}_{11.5}\text{As}_{24}\text{S}_{64.5}$, $t = 1 \mu\text{m}$	As_2Se_3 , $t = 2 \mu\text{m}$	GeAsSe , $t = 340 \text{ nm}$
#2: $t_{top} = 550 \text{ nm}$	As_2S_3 , $t = 1 \mu\text{m}$	As_2Se_3 , $t = 1.5 \mu\text{m}$	GeAsSe , $t = 550 \text{ nm}$
#3: $t_{top} = 900 \text{ nm}$	As_2S_3 , $t = 1 \mu\text{m}$	As_2Se_3 , $t = 1.5 \mu\text{m}$	GeAsSe , $t = 900 \text{ nm}$

Supplement Note 4: Estimation of loss intensity coming from surface adsorbed water

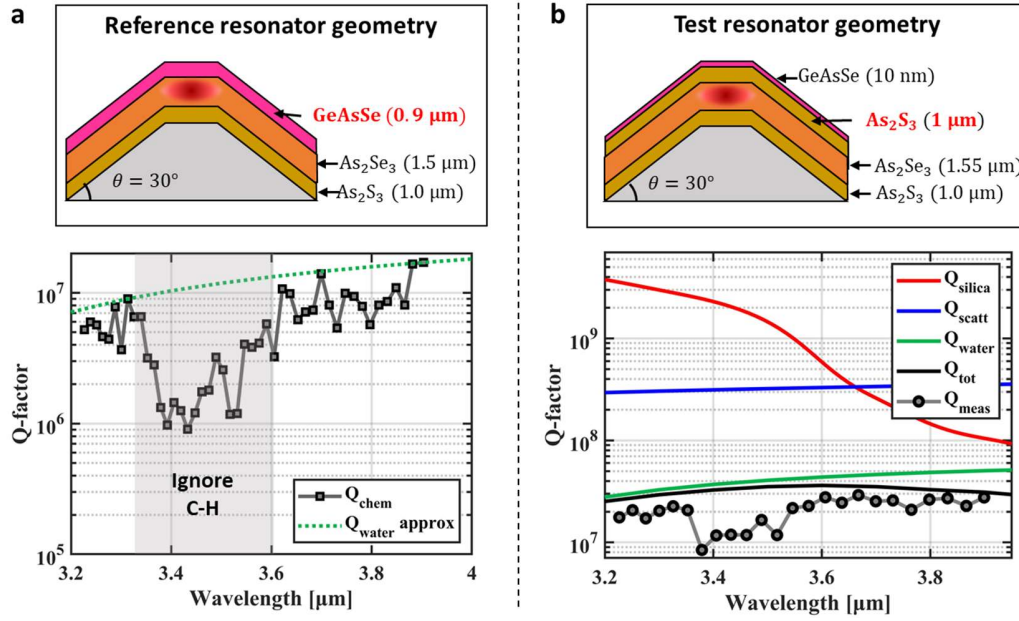


Fig.S7 | Estimating water absorption limited Q-factor (Q_{water}). **a**, Extraction of Q_{water} spectrum from a reference resonator. **b**, Predicting Q_{water} of a new test resonator having different geometric parameters with the reference.

A more detailed explanation for numerically calculating the surface water absorption loss ($\alpha_{water} \propto 1/Q_{water}$) on resonators is presented. Using this method, mid-IR Q-factors of any arbitrary-shaped resonators having Ge-As-Se as their outermost layer can be quantitatively predicted over the 3 – 4 μm wavelength.

Generally, estimation of absorption loss requires the knowledge of the target chemical concentration, which is tricky for surface adsorbed-chemicals. Here instead, we directly scale the water absorption loss of a reference resonator with the surface E-field intensity to obtain the loss of new resonators, which is possible by assuming the concentration of surface water is identical for all resonators with the same surface material.

First, to extract the water absorption loss of a reference resonator, the total chemical limited Q-factor (Q_{chem}) is obtained from the measured Q-factors (Q_{meas}) by subtracting the calculated silica absorption and scatter limited Q-factor (Q_{silica} , Q_{scatt}).

$$\frac{1}{Q_{chem}} = \frac{1}{Q_{meas}} - \frac{1}{Q_{silica}} - \frac{1}{Q_{scatt}} \quad (1)$$

Ignoring the effect of hydrocarbon absorption, which is too random to predict, the upper bound of Q_{chem} spectrum is linearly approximated which we define as the water absorption limited Q-factor (Q_{water}), as shown in Figure S7a.

This value now can be used to predict the Q_{water} of a new resonator having a different geometry. This is done by scaling the reference Q_{water} with the E-field intensity of the resonators' outermost surface, calculated using a finite element method simulation (COMSOL).

$$Q_{water,new} = Q_{water,ref} \cdot \frac{|E_{surf,ref}|^2}{|E_{surf,new}|^2} \quad (2)$$

The green line in Figure S7b shows the predicted Q_{water} for the drawn test resonator geometry. Adding this value with the calculated Q_{silica} and Q_{scatt} , the total mid-IR Q-factor (Q_{tot}) can be predicted, which agrees well with the measured Q-factor as shown in the figure.

Supplement Note 5: Expected mid-IR Q-factor spectrum as a function of the upper cladding thickness.

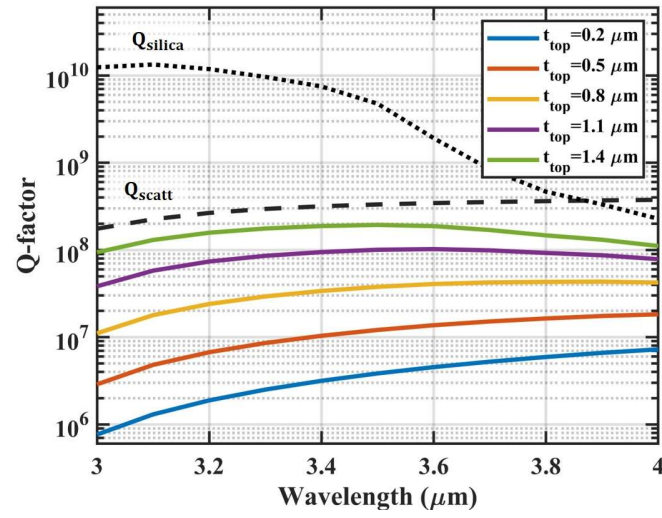


Fig.S8 | Estimated total Q-factor versus the upper cladding thickness. The colored solid lines represent calculated total Q-factors (Q_{tot}) for resonators with different upper cladding thicknesses (t_{top}). Other geometric parameters of these resonators are all identical. The black dashed lines indicate the Q_{silica} and Q_{scatt} values calculated for the $t_{top} = 1.4$ μm resonator.

The dependence of the resonator total Q-factor ($Q_{tot} = 1/(1/Q_{silica} + 1/Q_{scatt} + 1/Q_{water})$) was calculated for varying upper cladding thickness (t_{top}), as shown in Figure S8. Other geometric parameters were fixed identically to the record Q-factor resonator drawn in Figure 3b of the main text. The calculated Q_{tot} monotonically increases with t_{top} due to the rise in Q_{water} until it becomes clamped by Q_{satt} and Q_{silica} around $t_{top} = 1.4$ μm.

However, t_{top} of the actual record Q resonator was compromised as 0.8 μm considering the critical coupling condition. In our top-to-top flip-chip coupling scheme, a rise in t_{top} not only increases the Q but also decreases the resonator-to-waveguide coupling strength. Resonators with too large t_{top} will fail to achieve critical coupling since the clamped Q-factor is unable to compensate for the decreasing coupling coefficient. Thus, we selected $t_{top} = 0.8$ μm as a safe choice, where the clamping in Q-factor is negligible.

Supplement Note 6: Brillouin phonon gain bandwidth calculation

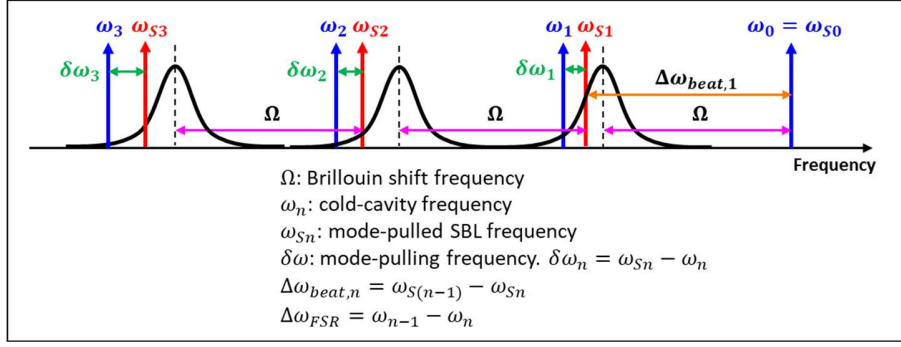


Fig.S9 | Frequency pulling in cascade Stokes waves. Schematic drawing and notations for the frequency (mode) pulling effect in cascade Stokes wave generation. The blue arrows indicate the cold cavity resonance frequencies (ω_n) while the red arrows show the frequency pulled Stokes wave frequencies (ω_{Sn}). The black curve indicates the Brillouin phonon gain bandwidth.

Derivation of the frequency mismatch per pump wavelength detuning (Hz/nm) parameter used to calculate the Brillouin phonon gain bandwidth is shown here. In the following section, ω_n indicates the n th cold cavity resonance frequency and ω_{Sn} corresponds to the frequency of the n th Stokes wave. The pump frequency is set as $\omega_0 = \omega_{S0}$. For other variables, check the notations summarized in Figure. S9.

In the main script, the clamped power of the 2nd Stokes wave was measured as a function of the pump wavelength λ_p . This power has a quadratic relation with the frequency mismatch between the Brillouin gain maximum and the actual 3rd Stokes frequency, $(\omega_{S2} - \Omega) - \omega_{S3} = \Delta\omega_{beat,3} - \Omega$.

As describe in the reference¹⁴, this frequency mismatch can be written again as a function of the pump wavelength detuning from the maximum gain position, $\Delta\lambda_p$, to match the experiment:

$$\Delta\omega_{beat,3} - \Omega = \Delta\lambda_p \cdot \left(\frac{d\Delta\omega_{beat,3}}{d\lambda_p} - \frac{d\Omega}{d\lambda_p} \right) = \Delta\lambda_p \cdot C \quad (3)$$

Using this relation, the measured FWHM of the pump detuning 13.91 nm, shown in main text Figure 5d, can be converted to the phonon gain bandwidth Γ by multiplying the frequency mismatch per pump wavelength detuning parameter $C = \left(\frac{d\Delta\omega_{beat,3}}{d\lambda_p} - \frac{d\Omega}{d\lambda_p} \right)$.

The Brillouin shift frequency Ω depends on the pump wavelength as $\Omega/2\pi = 2nV_A/\lambda_p$, where n is the refractive index, V_A is the acoustic velocity of the phonon, and λ_p is the pump wavelength. This gives its pump wavelength dependence $d\Omega/d\lambda_p = -\Omega/\lambda_p = -2\pi \cdot 1.09$ MHz/nm.

To calculate $\frac{d\Delta\omega_{beat,3}}{d\lambda_p}$, the well-known mode (frequency) pulling effect in Brillouin lasers should be taken into account. When the cold cavity free-spectral-range (FSR) $\Delta\omega_{FSR} = \omega_{n-1} - \omega_n$ does not match the Brillouin shift frequency Ω , mode pulling causes the Stokes wave to be generated at a frequency detuned from the cold cavity resonance. For single Stokes wave generation, the amount of detuning depends on the linewidth of interacting photons (γ) and phonons (Γ) as follows¹⁵.

$$\Delta\omega_{beat,1} = \omega_0 - \omega_{S1} = \frac{1}{1 + \frac{\gamma}{\Gamma}} \left(\Delta\omega_{FSR} + \frac{\gamma}{\Gamma} \Omega \right) = \frac{1}{1 + \frac{\gamma}{\Gamma}} \left(\omega_0 - \omega_1 + \frac{\gamma}{\Gamma} \Omega \right) \quad (4)$$

We expand this equation to cascade Stokes wave generation. The n th mode pulling frequency $\delta\omega_n \equiv \omega_{Sn} - \omega_n$ and linewidth ratio factor $r \equiv \gamma/\Gamma$ is defined for convenience. The commonly applied assumption $\gamma \ll \Gamma$ is not used in this derivation because these two linewidths are rather comparable in our resonator as will be shown in the end of this section. Moreover, the mode-pulling effect of the anti-Stokes process¹⁵ is neglected here for simplicity.

The mode pulling frequency of the 1st Stokes wave is given as

$$\delta\omega_1 = \omega_{S1} - \omega_1 = \Delta\omega_{FSR} - \Delta\omega_{beat,1} = \frac{r}{1 + r} (\Delta\omega_{FSR} - \Omega) \quad (5)$$

leading to the pulled Stokes frequency

$$\omega_{S1} = \omega_1 + \frac{r}{1+r}(\Delta\omega_{FSR} - \Omega) \quad (6)$$

For the 2nd Stokes wave,

$$\delta\omega_2 = \omega_{S2} - \omega_2 = \frac{r}{1+r}(\omega_{S1} - \omega_2 - \Omega) = \frac{r}{1+r}\left(1 + \frac{r}{1+r}\right)(\Delta\omega_{FSR} - \Omega) \quad (7)$$

where $\omega_{S1} - \omega_2$ is inserted in the place of $\Delta\omega_{FSR}$ of equation (5) since the frequency of 1st Stokes wave acting as a pump is already detuned from ω_1 to ω_{S1} .

By mathematical induction, it can be proven that

$$\delta\omega_n = (\Delta\omega_{FSR} - \Omega) \cdot \sum_{k=1}^n \left(\frac{r}{1+r}\right)^k \quad (8)$$

$$\Delta\omega_{beat,n} = (\omega_{n-1} + \delta\omega_{n-1}) - (\omega_n + \delta\omega_n) = \Delta\omega_{FSR} - \left(\frac{r}{1+r}\right)^n (\Delta\omega_{FSR} - \Omega) \quad (9)$$

The cold cavity FSR shift $\frac{d\Delta\omega_{FSR}}{d\lambda_p} = 2\pi \cdot 0.0163$ MHz/nm can be calculated using an optical mode simulation.

The equation (9) written for $\Delta\omega_{beat,3}$ contains $r = \gamma/\Gamma$ and forms a complete set of nonlinear equations with $\Gamma = C \cdot \Delta\lambda_{FWHM} = \left(\frac{d\Delta\omega_{beat,3}}{d\lambda_p} - \frac{d\Omega}{d\lambda_p}\right) \cdot \Delta\lambda_{FWHM}$. By iteration, we obtain a self-consistent phonon bandwidth value of $\Gamma = 2\pi \cdot 14.51$ MHz, which is comparable with the used photon linewidth $\gamma = 2\pi \cdot 9.085$ MHz converted from the resonator $Q_{load} = 10^7$.

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Supplement Note 7: Parameters used for Brillouin power dynamics calculation.

Table S3 | Cold cavity characteristics and Brillouin parameters used for calculating the Stokes wave power dynamics curve (Main text Figure 5c).
Values without references are measured or simulated numbers.

Pump frequency (ω_{pump})	$2\pi \times 90.92$ THz (3297 nm)
Optical cavity decay rate (γ)	$2\pi \times 7.20$ MHz
External decay rate (γ_{ext})	$2\pi \times 2.54$ MHz
Resonator length (L)	$2\pi \cdot 4.65 = 29.2$ mm
Group index (n_g)	2.8278
Brillouin gain coefficient at ω_{pump} (G_B)	$58.8092 \text{ m}^{-1}\text{W}^{-1}$ ($G_{B,max} = 59.9002 \text{ m}^{-1}\text{W}^{-1}$)
Brillouin amplification rate per photon ($\mu = \left(\frac{\hbar\omega c^2 G_B}{2n_g^2 L}\right)^{16}$)	0.6815 Hz
Intrinsic loss (α)	1.20 dB/m
Coupling loss correction factor (β)	1.43
Theoretical S1 threshold ^{16,17} , $P_{th}^1 = \frac{\omega^2 n_g^2 L Q_{ext}}{4 G_B Q_T^3 c^2} = \frac{\hbar \omega_{pump} \gamma^3}{64 \gamma_{ext} \mu} \cdot (1 + 1)^3$	64.2 μW

Table S4 | Mechanical and optical parameters of materials used for the Brillouin simulation.

	Density (kg/m ³)	Poisson ratio (ν)	Young's modulus E (GPa)	Refractive index ($\lambda = 3.3 \mu\text{m}$)
As ₂ S ₃	3200 ¹⁸	0.305 ¹⁸	16.2 ¹⁸	2.3921 ²²
As ₂ Se ₃	4660 ¹⁹	0.244 ¹⁹	18.5 ¹⁹	2.774 ²³
Ge-As-Se (Ge ₃₃ As ₁₂ Se ₅₅)	4430 ²⁰	0.215 ²⁰	21.2 ²⁰	2.5168 ²⁰
SiO ₂	2220 ²¹	0.17 ²¹	73.6 ²¹	1.4153 ²¹

Table S5 | Elastic (stiffness, c_{ij}) and photo-elastic (elasto-optic, p_{ij}) tensor components of materials used for the Brillouin simulation. The c_{ij} values were derived from the Poisson ratio and the Young's modulus given in Table S4. The p_{ij} values were copied from reference 25-27 as noted in the table.

	c_{11} (GPa)	c_{12} (GPa)	c_{44} (GPa)	p_{11}	p_{12}	p_{44}
As ₂ S ₃ ²⁴	19.75	8.7	5.525	0.308	0.299	0.0045
As ₂ Se ₃ ²⁵	21.96	8.04	6.96	0.314	0.266	0.024
Ge-As-Se (Ge ₃₃ As ₁₂ Se ₅₅) ²⁶	24.2	6.826	8.69	0.21	0.211	0
SiO ₂ ²⁵	78.6	16.1	31.2	0.12	0.27	-0.075

Supplement Note 8: Comparison of optical loss with diverse mid-IR on-chip platforms

Fig. S10 compares the record optical loss presented in this work with that of other on-chip platforms based on various materials: ChG, Si, Ge, Si₃N₄, SiGe, and III-V semiconductors (same as Figure 4 in the main script). Additionally, the loss of state-of-the-art As₂Se₃ optical fiber, extruded from the same doubly purified ingots explained in Supplement 1D, is drawn as a grey line. Our resonator's loss values are converted from the measured Q-factors (Q) using $\alpha = \frac{2\pi n_g}{\lambda Q}$ (α =optical loss(m⁻¹), $n_g \approx 2.82$ (group refractive index), λ =wavelength).

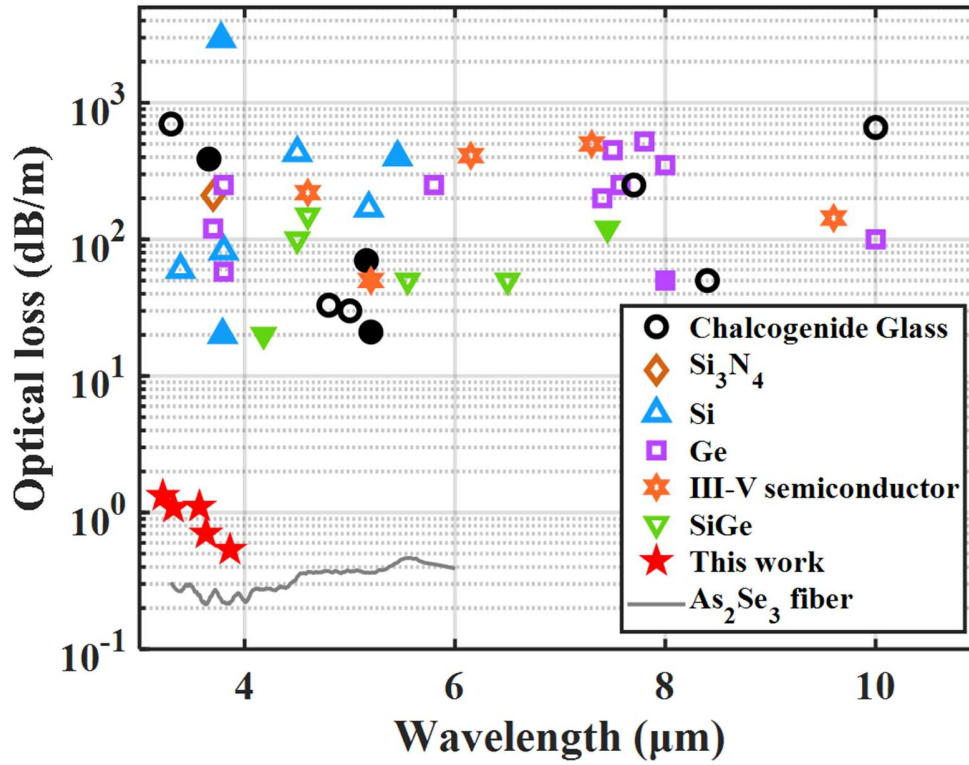


Fig. S10 | Comparison of optical losses with different material platforms on a chip. The propagation losses of diverse mid-IR on-chip resonators and waveguides are compared with this work. Different markers indicate different material platforms: chalcogenide glasses^{5,27-34} (black, circle), silicon nitride³⁵ (brown, diamond), silicon³⁶⁻⁴² (blue, triangle), germanium⁴³⁻⁵² (purple, square), III-V semiconductors⁵³⁻⁵⁷ (orange, hexagram), SiGe⁵⁸⁻⁶³ (green, reversed triangle), and this work (red, pentagram). The solid symbols represent resonators, and the open symbols represent waveguides. Loss of the state-of-the-art As₂Se₃ optical fiber¹³ is also drawn as a grey line for comparison.

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