
Supplementary Information

Mechanisms of Femtosecond Laser-Induced Structural Reconfiguration in Fluoride Glasses

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Note 1: Electron density calculation

This Note details the calculation process for plasma electron density.

To characterize the time evolution of femtosecond laser-induced plasma within the sample, we performed calculations using the Drude model^{1,2}. Two images (background and signal) were collected before and after femtosecond irradiation, from which the transmission intensities at 520 nm probe wavelength were extracted. Denoting the transmittances of the background and signal images as I_0 and I_1 respectively, the transient transmittance T at different time delays is expressed as:

$$T = \frac{I_1}{I_0}. \quad (1-1)$$

The laser intensity I_c at the center of the pump-probe beam is given by:

$$\frac{dI_c}{dx} = -\alpha I_c, \quad (1-2)$$

where α is the absorption coefficient, obtained via integration:

$$\alpha = -(\ln T) / x. \quad (1-3)$$

Here, x denotes the length of the electron plasma along the propagation direction of the probe beam, determined by the plasma width at each time delay (assuming plasma symmetry). The absorption coefficient is directly related to the plasma frequency ω_p :

$$\alpha = \frac{\tau}{nc} \frac{\omega_p^2}{1 + \omega^2 \tau^2}, \quad (1-4)$$

where n is the material refractive index at the probe wavelength, c is the vacuum speed of light, ω is the laser frequency, and τ is the electron collision time. The plasma frequency ω_p depends on the free electron density, as described by:

$$\omega_p = \sqrt{\frac{n_e e^2}{m_e \epsilon_0}}, \quad (1-5)$$

with e as the electron charge, m_e as the electron mass, and ϵ_0 as the vacuum permittivity. The final electron density is thus:

$$n_e = \frac{\alpha n c m_e \varepsilon_0 (1 + \omega^2 \tau^2)}{e^2 \tau}. \quad (1-6)$$

In the calculation, the refractive index of INF glass at the 520 nm probe wavelength was $n = 1.542$, which was measured in this work. The laser frequency was 5.77×10^{14} Hz, corresponding to the 520 nm probe wavelength used in this work. The electron collision time was taken as $\tau = 1.7$ fs³.

Note 2: Phase delay and stress direction calculation

This Note describes the calculation procedures for optical retardance and birefringence distribution.

Fluoride glasses can be considered isotropic materials at room temperature. When a laser is focused inside the sample, transient stress is induced, disrupting the isotropic properties of the glass. This leads to a refractive index difference between the two principal stress directions, resulting in a birefringence effect. We derived the photoelastic equation using Stokes vectors and Mueller matrices^{4,5}. The Stokes vector of light detected by the polarization camera after passing through the optical path is expressed as:

$$I_i = P_i M S_1 \quad (i = 1, 2, 3, 4). \quad (2-1)$$

The angle between the horizontal polarizer and the x-axis is β , given by:

$$P_i = \frac{1}{2} \begin{bmatrix} 1 & \cos 2(\beta_i) & \sin 2(\beta_i) & 0 \\ \cos 2(\beta_i) & \cos^2 2(\beta_i) & \sin 2(\beta_i) \cos 2(\beta_i) & 0 \\ \sin 2(\beta_i) & \sin 2(\beta_i) \cos 2(\beta_i) & \sin^2 2(\beta_i) & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \quad (2-2)$$

where $\beta_i = 0^\circ, 45^\circ, 90^\circ, 135^\circ$. M is the Mueller matrix of the material induced by transient stress, expressed as:

$$M = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos^2 2\theta + \sin^2 2\theta \cos \varphi & (1 - \cos \varphi) \sin 2\theta \cos 2\theta & -\sin 2\theta \sin \varphi \\ 0 & (1 - \cos \varphi) \sin 2\theta \cos 2\theta & \sin^2 2\theta + \cos^2 2\theta \cos \varphi & \cos 2\theta \sin \varphi \\ 0 & \sin 2\theta \sin \varphi & -\cos 2\theta \sin \varphi & \cos \varphi \end{bmatrix}. \quad (2-3)$$

Here, θ is the angle between the glass material and the x-axis, and φ is the retardation. The Stokes vector of an ideal circularly polarized light field is:

$$S_1 = Q P_0 S_0 = I [1 \quad 0 \quad 0 \quad 0]^T, \quad (2-4)$$

Q denotes the Mueller matrix of the quarter-wave plate, which can be neglected when matched to the light source wavelength. From the above equations, the stress expression in a circularly polarized field is derived as:

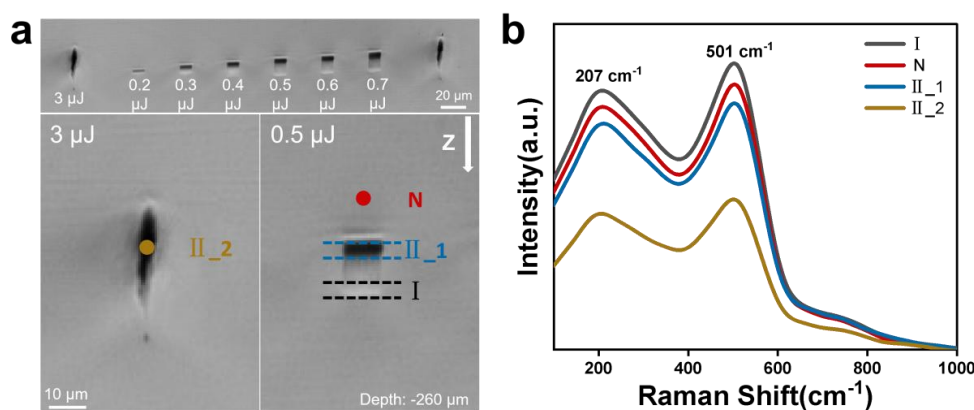
$$\varphi = \arcsin \sqrt{\frac{(I_3 - I_1) + (I_2 - I_4)}{(I_1 + I_3)(I_2 + I_4)}}. \quad (2-5)$$

$$\theta = \frac{1}{2} \arctan \left(\frac{I_3 - I_1}{I_2 - I_4} \right). \quad (2-6)$$

Note 3: Raman characterization of waveguide region

This Note presents Raman characterization results of femtosecond laser-modified regions in INF glass.

Channel waveguides were fabricated in INF glass (260 μm below the surface) using femtosecond laser repeated scanning, with single-pulse energies ranging from 0.2 to 0.7 μJ ; single-scan reference lines (3 μJ) were inscribed on both sides. Supplementary Fig. 1a shows a cross-sectional micrograph of the waveguide, where Regions I, N, II_1, and II_2 denote the positive refractive index modified region, the unmodified region, the negative refractive index modified region induced at lower pulse energy, and the more strongly modified negative-index region induced at higher pulse energy, respectively. Along the laser incidence direction, the channel waveguide exhibits a ‘sandwich-like’ contrast pattern with a ‘bright-dark-bright’ sequence, attributed to glass network polarizability changes induced by low-repetition-rate pulses⁶.



Supplementary Figure 1 | Raman characterization of femtosecond laser direct-written waveguide regions (INF glass). **a** Micrograph of a femtosecond laser direct-written waveguide region. The laser repetition rate and scanning speed are 1 kHz and 500 $\mu\text{m}/\text{s}$, respectively, with a waveguide width of 10 μm (scan interval: 1 μm). **b** Comparison of Raman spectra in waveguide regions. Regions I, II, and N denote the positive refractive index modified region, negative refractive index modified region, and unmodified region, respectively. Source data are provided as a Source Data file.

Raman spectroscopy was performed on these four regions using a 633 nm laser, with results shown in Supplementary Fig. 1b. The Raman spectra exhibit two distinct peaks:

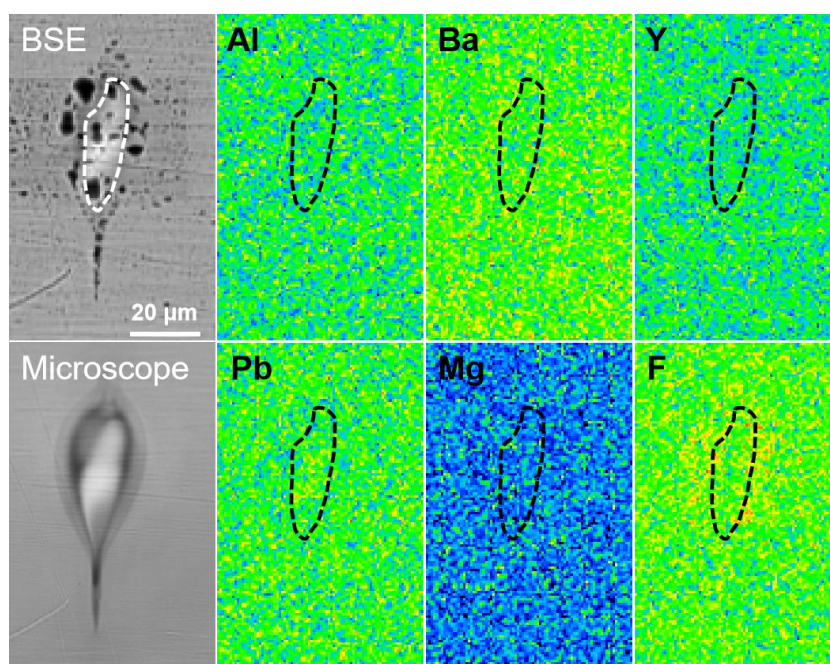
asymmetric Zn-F and Ba-F stretching vibrations at 207 cm^{-1} , and 501 cm^{-1} for octahedral $[\text{InF}_6]$ stretching vibrations⁷⁻⁹. Relative to the unmodified region (N), the Raman intensity in Region I increases by about 5–7% at both characteristic bands, whereas Region II_1 decreases by about 6%, and Region II_2 shows a much stronger reduction of about 38%. These systematic intensity differences indicate a clear structural response of the glass network to femtosecond laser irradiation^{10,11}.

Further examination of the peak-position behavior shows that the apparent peak maximum near 501 cm^{-1} exhibits a weak but discernible trend with the refractive-index variation: compared with Region N, Region I shows a slight shift toward higher wavenumbers (approximately $1\text{--}2\text{ cm}^{-1}$), Region II_1 shows no obvious shift, and Region II_2 shifts toward lower wavenumbers (approximately $2\text{--}3\text{ cm}^{-1}$). This tendency is qualitatively consistent with the refractive-index distribution. By contrast, the band near 207 cm^{-1} is broader and does not show an equally clear systematic shift among the four regions. Therefore, in the present INF glass system, the Raman response is reflected more clearly in the intensity variation, while the 501 cm^{-1} band provides additional peak-shift information related to the difference between positive- and negative-index modified regions. In particular, the pronounced intensity reduction together with the slight red shift of the 501 cm^{-1} band in Region II_2 suggests a stronger rarefaction or structural loosening of the local glass network under higher pulse energy.

Note 4: Electron probe analysis

This Note characterizes elemental redistribution in ABYPM glass under high repetition rates.

In the fluoroaluminate glass (ABYPM), single line scanning was performed using the same femtosecond laser parameters to observe the characteristics of element redistribution (femtosecond laser incident along the Z direction), as shown in Supplementary Fig. 2. From the cross-sectional microscope images of the modified area, it can be seen that the modified profiles of ABYPM and INF glass are very similar, both exhibiting a 'droplet shaped' profile. The main body of the contour has a brighter contrast compared to the modified area, which corresponds to an increase in refractive index.



Supplementary Figure 2 | Elemental distribution in the cross-section of femtosecond laser-modified regions (ABYPM glass). Backscattered electron (BSE) image, micrograph, and corresponding elemental distribution maps (from electron probe microanalysis). This region was fabricated via single-line scanning with a femtosecond laser (repetition rate: 50 kHz, scanning speed: 500 $\mu\text{m/s}$, single-pulse energy: 3 μJ).

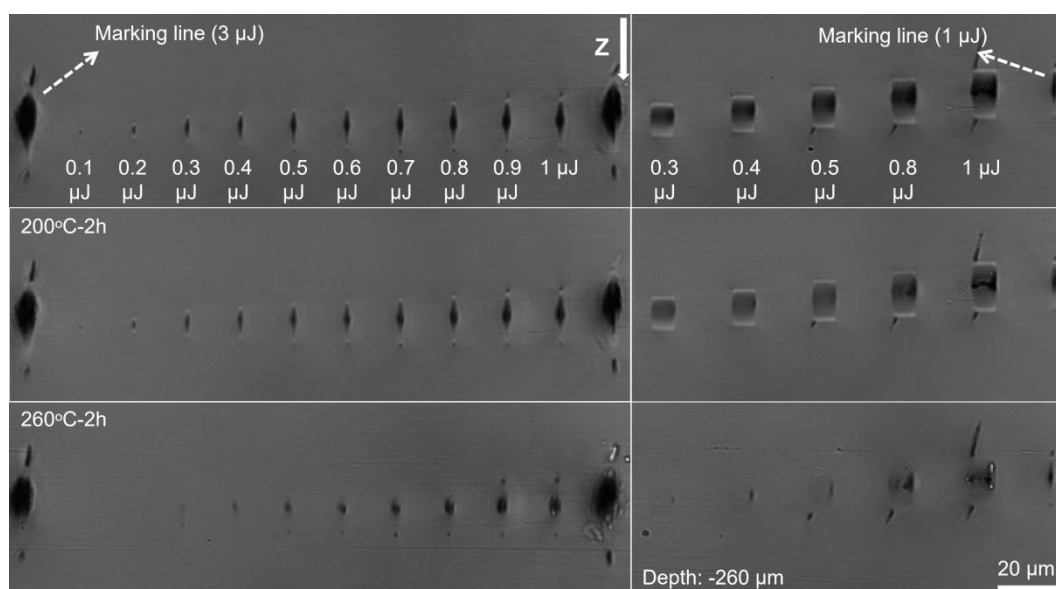
The backscatter (BSE) image also shows the same contrast pattern, with the black area due to the expansion of the gold film on the glass surface under high-energy

electron beam bombardment (which does not affect the distribution test of glass composition elements). The wavelength dispersive spectrometer (WDS) mapping results intuitively display the element redistribution under high repetition rate femtosecond laser pulses. Pb element is enriched in the high refractive index modified zone, while Ba and F elements migrate to the negative refractive index modified zone outside the modified zone. It exhibits good consistency with the element redistribution of INF glass.

Note 5: Thermal erasure comparisons

This Note provides details of thermal erasure phenomena in regions modified by femtosecond laser pulses with different single-pulse energies.

Under identical femtosecond laser pulse parameters, we compare thermal erasure phenomena across a broader range of single-pulse energies (Supplementary Fig. 3). Given that excessively high heat-treatment temperatures can induce crystallization in fluoride glasses, the maximum heat-treatment temperature was set near the glass transition temperature ($T_g \approx 257^\circ\text{C}$).



Supplementary Figure 3 | Thermal erasure in femtosecond-modified regions. Comparative micrographs of single-line scanned and multi-scan waveguide regions at different heat-treatment temperatures (200°C, 260°C). Laser parameters: 1 kHz repetition rate, 500 $\mu\text{m/s}$ scanning speed. Multi-scan waveguide width: 8 μm (scan interval: 0.5 μm).

Cross-sectional images of the modified zones reveal that contrast between modified and unmodified regions gradually diminishes with increasing temperature. The modified region fabricated with single-pulse energy $< 0.3 \mu\text{J}$ is completely erased at 260°C, while bright-contrast modified regions at higher energies are also erased. As single-pulse energy increases, the erasure degree of dark-contrast zones decreases progressively (without complete erasure) and more severe damage—including cracks

and nanocavities—appears (e.g., in regions modified with 1 μJ single-pulse energy). Thus, excessive single-pulse energy should be avoided for fabricating low-loss optical waveguides.

Note 6: Quantitative carrier-thermal-elastic model for femtosecond laser-induced stress waves in fluoride glasses

The goal of this Note is to quantitatively link carrier dynamics, energy dissipation, lattice heating, and stress-wave generation in femtosecond-laser-modified fluoride glass. In the main text, time-resolved pump-probe shadowgraphy and birefringence imaging experimentally reveal the transient plasma evolution and subsequent stress-wave propagation. Here, we establish a physics-based carrier-thermal-elastic model to examine whether the experimentally relevant plasma parameters can reproduce the observed stress-wave amplitude and propagation velocity.

The model combines nonlinear carrier generation, transient Drude optical response, electron-lattice energy transfer, and thermoelastic stress-wave propagation. This framework follows established treatments of femtosecond laser interaction with transparent dielectrics, in which free-electron generation, transient optical absorption, two-temperature relaxation, and stress/strain dynamics are coupled to describe ultrafast energy deposition and subsequent mechanical response^{1-3,12-17}.

The representative simulation is performed for INF glass under a single-pulse energy of 0.8 μJ , corresponding to the saturated plasma condition discussed in the main text. To further examine the parameter sensitivity and predictive capability of the model, a lower pulse energy of 0.4 μJ is also investigated by combining time-resolved plasma/stress-wave experiments with the corresponding carrier-thermal-elastic simulations.

6.1 Physical background and motivation

When a femtosecond laser pulse is tightly focused into a transparent fluoride glass, a sequence of electronic excitation and structural evolution processes unfolds from femtosecond to nanosecond timescales.

- Carrier generation (fs–ps) occurs via multiphoton and avalanche ionization, creating a dense plasma of free electrons and holes.
- Energy transfer (ps) from hot carriers to the lattice proceeds through Drude absorption and electron-phonon coupling, leading to rapid local heating.

-
- Stress-wave formation (ps–ns) follows as the localized energy deposition induces thermal expansion and launches a high-amplitude longitudinal acoustic wave.

This sequence couples femtosecond-scale electron dynamics with picosecond-to-nanosecond mechanical response. The localized plasma acts as a transient pressure source—often described as a “plasma piston”—which generates GPa-level stress waves observable via birefringence and pump–probe imaging.

6.2 Rate equation for carrier generation

The temporal evolution of free-electron density $n_e(t)$ follows the rate equation:

$$\frac{dn_e}{dt} = W_{\text{MPI}}(I)(N_0 - n_e) + \sigma_{\text{av}} I(t) n_e - \frac{n_e}{\tau_r}, \quad (6-1)$$

where

- N_0 – atomic density of the glass,
- $W_{\text{MPI}}(I) = \sigma_K I^K$ – multiphoton ionization rate,
- σ_{av} – avalanche ionization coefficient,
- τ_r – electron recombination time,
- $I(t)$ – instantaneous laser intensity.

The laser pulse is modelled as a Gaussian envelope:

$$I(r, t) = I_0 (1 - R) \exp\left(-\frac{2r^2}{w_0^2}\right) \exp\left[-4 \ln 2 \left(\frac{t^2}{\tau_p^2}\right)\right], \quad (6-2)$$

where τ_p is the pulse duration (400 fs), R is the reflectivity of the material, w_0 is the beam waist radius. For femtosecond timescales ($t < 2$ ps), recombination is negligible, giving:

$$\frac{dn_e}{dt} \approx W_{\text{MPI}}(I)(N_0 - n_e) + \sigma_{\text{av}} I(t) n_e \quad (6-3)$$

This equation is numerically integrated to obtain $n_e(t)$ under 0.8 μJ irradiation, as shown in Supplementary Fig. 4a.

6.3 Transient optical response and free-carrier absorption

The high intensity of the femtosecond pulse induces a time- and space-dependent dielectric response. The generated free-electron plasma modifies the optical response of the glass, which is described using the Drude model^{1-3,13}. The complex dielectric function is expressed as:

$$\varepsilon(\omega, t) = n_0^2 - \frac{n_e(t)e^2}{\varepsilon_0 m_e (\omega^2 + i\omega / \tau_c)}, \quad (6-4)$$

where n_0 is the refractive index of the unperturbed material, m_e is the electron mass, ε_0 is the vacuum permittivity, ω is the laser angular frequency, e is the elementary charge, and τ_c is the electron collision time.

The complex refractive index is described as:

$$n^* = n + i\kappa = \sqrt{\varepsilon(\omega, t)}, \quad (6-5)$$

where n and κ are the real refractive index and extinction coefficient, respectively.

The reflectivity R of the material can be calculated using the Fresnel expression:

$$R = \left| \frac{n^* - 1}{n^* + 1} \right|^2. \quad (6-6)$$

The absorption coefficient of laser energy is then given by:

$$\alpha = \frac{4\pi\kappa}{\lambda}, \quad (6-7)$$

where λ is the laser wavelength. The local volumetric power absorbed per unit time is expressed as:

$$S(r, t) = \alpha I(r, t), \quad (6-8)$$

where S is subsequently used as the laser source term in the two-temperature model. Integrating the absorbed power over the pulse gives the deposited energy density:

$$Q(r) = \int S(r, t) dt. \quad (6-9)$$

This treatment retains the original free-carrier absorption and energy-deposition logic while incorporating the transient optical response calculated from the Drude model.

6.4 Two-temperature model and lattice heating

The femtosecond timescale of the pulse-material interaction necessitates the decoupling of the temperature of the electron and lattice system. On this timescale, the electron system absorbs the incident pulse energy first and then transfers the absorbed energy to the lattice through electron-phonon coupling. To model this, modified forms of the heat equation can be coupled to simulate the temperatures of the two systems^{2,14,15}, given as:

$$C_e \frac{\partial T_e}{\partial t} = \nabla \cdot (K_e \nabla T_e) - G(T_e - T_l) + S(r, t), \quad (6-10)$$

$$C_l \frac{\partial T_l}{\partial t} = \nabla \cdot (K_l \nabla T_l) + G(T_e - T_l), \quad (6-11)$$

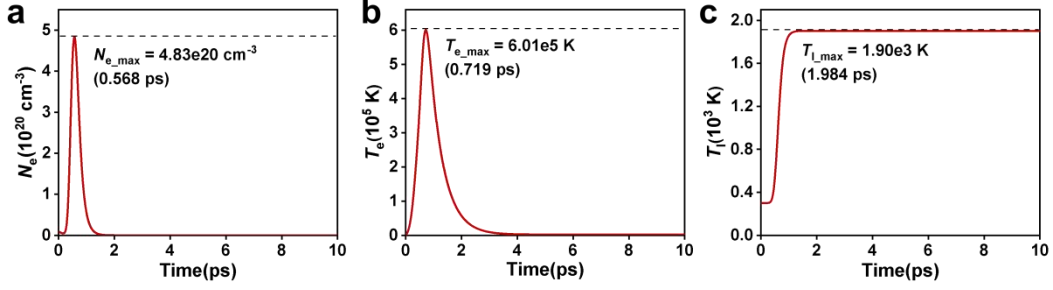
where C_e and C_l are the electron and lattice heat capacities, K_e and K_l are the electron and lattice thermal conductivities, T_e and T_l are the electron and lattice temperatures, respectively, and G is the electron-lattice coupling factor.

The electron-lattice coupling factor determines the rate of energy transfer from the electron system to the lattice and can be expressed as:

$$G = \frac{C_e}{\tau_{el}}, \quad (6-12)$$

where τ_{el} is the electron-lattice coupling time.

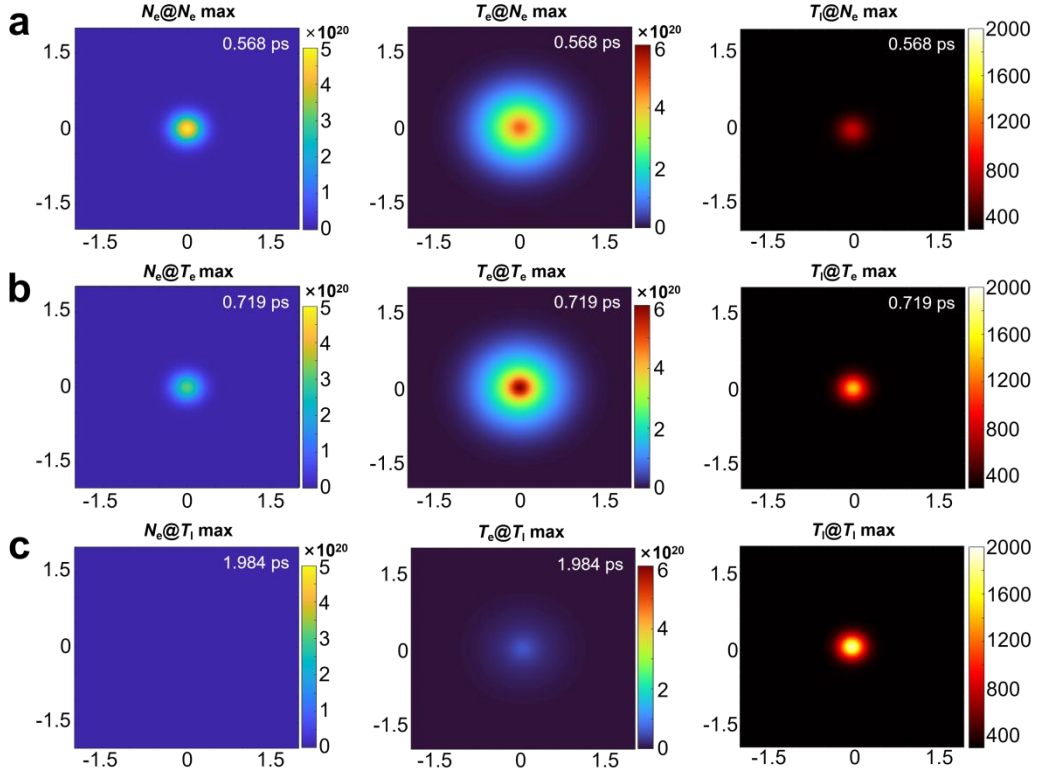
Supplementary Fig. 4 shows the simulated temporal evolution of the free-electron density, electron temperature, and lattice temperature under 0.8 μJ femtosecond laser irradiation. The electron density reaches a maximum of $4.834 \times 10^{20} \text{ cm}^{-3}$ at 0.568 ps. The electron temperature reaches a maximum of $6.01 \times 10^5 \text{ K}$ at 0.719 ps, while the lattice temperature reaches $1.90 \times 10^3 \text{ K}$ at 1.984 ps. The delayed peak of the lattice temperature reflects the energy-transfer process from the electron system to the lattice.



Supplementary Figure 4 | Temporal evolution of carrier and thermal parameters in INF glass. **a** Simulated temporal evolution of free-electron density $n_e(t)$. **b** Simulated temporal evolution of electron temperature $T_e(t)$. **c** Simulated temporal evolution of lattice temperature $T_l(t)$. The single-pulse energy used in the simulation is 0.8 μJ . Source data are provided as a Source Data file.

6.5 Spatial evolution of carrier and thermal fields

To further examine the spatial behavior of energy deposition and energy transfer, we calculated the two-dimensional distributions of n_e , T_e , and T_l in the transverse plane.



Supplementary Figure 5 | Simulated spatial distributions of carrier and thermal fields in INF glass under 0.8 μJ . **a** Two-dimensional distributions of free-electron density (n_e), electron

temperature (T_e), and lattice temperature (T_l) at the time of maximum n_e (0.568 ps). **b** Two-dimensional distributions of n_e , T_e , and T_l at the time of maximum T_e (0.719 ps). **c** Two-dimensional distributions of n_e , T_e , and T_l at the time of maximum T_l (1.984 ps).

Supplementary Fig. 5 shows the simulated spatial distributions at representative delay times. The free-electron density is initially strongly localized near the laser focal region. The electron temperature distribution becomes broader as the electron system absorbs and redistributes the laser energy. The lattice temperature increases later and remains spatially confined around the energy-deposition region. This sequence directly visualizes the transfer of laser energy from the ionized free-electron system to the lattice.

6.6 Simulation of fs laser-induced strain dynamics

Based on thermoelastic dynamics, the lattice-temperature field calculated in the previous section is adopted as the transient photo-excited heat source. In the present calculation, the evolutionary process in the x/y-plane is considered. The temporal evolution of displacement within the material is calculated using the equation of motion¹⁶:

$$\rho \frac{\partial^2 \mathbf{u}(x, y, t)}{\partial t^2} = \nabla \cdot [\mathbf{P}(x, y, t) + \mathbf{P}^{(\text{thermal})}(x, y)], \quad (6-13)$$

where t is the time after photoexcitation, ρ is the density of the material, $\mathbf{u}(x, y, t)$ is the displacement vector at t , $\mathbf{P}(x, y, t)$ is the stress tensor due to displacement, and $\mathbf{P}^{<\text{thermal}>}(x, y)$ is the thermal stress tensor caused by the temperature change. According to Hook's law, the components of $\mathbf{P}(x, y, t)$ can be expressed as:

$$P_{xx} = (\lambda + 2\mu)S_{xx} + 2\mu S_{yy}, \quad (6-14)$$

$$P_{yy} = \lambda S_{xx} + (\lambda + 2\mu)S_{yy}, \quad (6-15)$$

$$P_{xy} = 2\mu S_{xy}, \quad (6-16)$$

where P_{ij} and S_{ij} are the components of the stress and strain tensors, respectively, and λ and μ are Lamé constants:

$$\lambda = \frac{E\nu}{(1+\nu)(1-2\nu)}, \quad (6-17)$$

$$\mu = \frac{E}{2(1+\nu)}. \quad (6-18)$$

The strain tensor is calculated by

$$S_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right). \quad (6-19)$$

The thermal stress tensor is expressed as

$$P^{(\text{thermal})} = -\frac{E\beta}{1-2\nu} \Delta T, \quad (6-20)$$

where β is the thermal expansion coefficient of the material, E is the Young's modulus, and ν denotes the Poisson's ratio. ΔT is the temperature increment. Although glass is intrinsically isotropic, the energy deposition under intense laser irradiation can be modulated by the polarization direction, leading to an effective anisotropic spatial distribution. To capture this effect in the calculated stress pattern, a polarization-dependent term $\cos^2(\varphi-\theta)$ is introduced in the spatial heat-source distribution, where θ denotes the azimuthal angle and φ denotes the polarization angle of the incident laser¹⁷.

The displacement distribution of the crystal after the generation of photo-induced transient thermal stress can be obtained by numerically solving the above equations.

Therefore, all stress components can be derived:

$$\sigma_{xx} = \frac{E}{(1+\nu)(1-2\nu)} \left[(1-\nu) S_{xx} + \nu S_{yy} \right], \quad (6-21)$$

$$\sigma_{yy} = \frac{E}{(1+\nu)(1-2\nu)} \left[(1-\nu) S_{yy} + \nu S_{xx} \right], \quad (6-22)$$

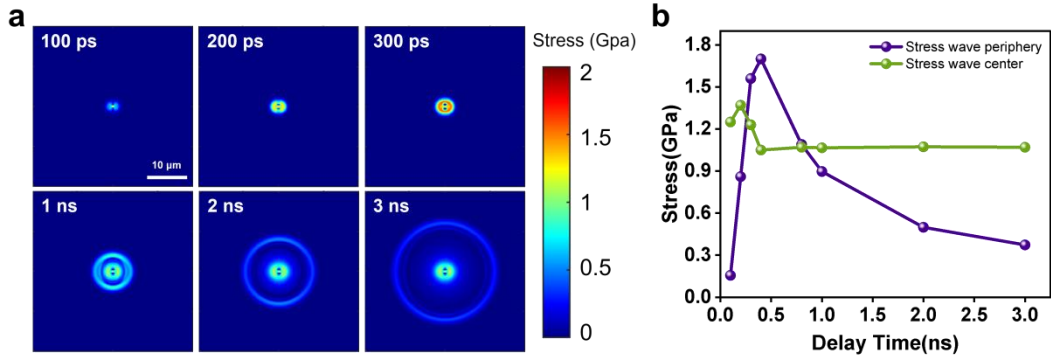
$$\sigma_{xy} = 2\mu S_{xy}. \quad (6-23)$$

The principal stresses and principal stress directions are given by:

$$\sigma_{1,2} = \frac{\sigma_{xx} + \sigma_{yy}}{2} \pm \sqrt{\left(\frac{\sigma_{xx} - \sigma_{yy}}{2}\right)^2 + \sigma_{xy}^2}, \quad (6-24)$$

$$\tan(2\theta) = \frac{2S_{xy}}{S_{xx} - S_{yy}}. \quad (6-25)$$

Ultimately, these two equations describe stress magnitude and direction measured in transient stress birefringence experiments. Supplementary Fig. 6 shows the simulated stress-wave evolution and the temporal stress variation in the central and peripheral regions.



Supplementary Figure 6 | Simulated stress-wave evolution in INF glass under 0.8 μJ. a Simulated spatial distributions of the stress wave at different time delays. **b** Simulated temporal evolution of stress in the central and peripheral regions. Source data are provided as a Source Data file.

6.7 Analytical summary

Combining the above relationships, the chain of processes is shown as follows:

$$I(t) \Rightarrow n_e(t) \Rightarrow \Delta T_e(t) \Rightarrow \Delta T_l(t) \Rightarrow \mathbf{P}^{(\text{thermal})}(t) \Rightarrow \mathbf{P}(x, y, t) \quad (6-26)$$

This compact scaling quantitatively connects plasma energy dissipation to stress-wave amplitude.

6.8 Numerical implementation and comparison with experiment

The numerical calculation was performed for INF glass under a single-pulse energy of 0.8 μJ. The laser wavelength and pulse duration were set to 1040 nm and 400 fs, respectively, consistent with the experimental conditions. The carrier-generation

equation, transient Drude optical response, two-temperature model, and thermoelastic strain-dynamics equations were solved sequentially.

The simulation gives a peak free-electron density of $4.834 \times 10^{20} \text{ cm}^{-3}$, which agrees well with the experimentally measured saturated electron density of INF glass. The calculated peak lattice temperature is $1.90 \times 10^3 \text{ K}$. Taking room temperature as approximately 300 K, the corresponding lattice temperature rise is about $1.6 \times 10^3 \text{ K}$, close to the experimentally inferred temperature rise of approximately $1.5 \times 10^3 \text{ K}$ from stress analysis.

The calculated stress wave expands outward from the laser-excited region. By tracking the wavefront position as a function of time, the stress-wave propagation velocity is estimated to be approximately $4.5 \text{ } \mu\text{m/ns}$, which is close to the experimentally measured velocity of approximately $4.3 \text{ } \mu\text{m/ns}$. The calculated central stress reaches approximately 1.4 GPa at around 200 ps, while the peripheral stress reaches approximately 1.7 GPa at around 300–400 ps. These values are quantitatively comparable to the GPa-level stresses obtained from birefringence measurements.

The comparison between simulation and experiment is summarized below:

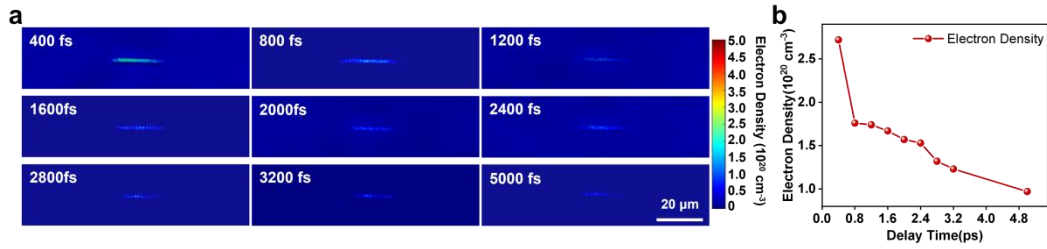
Quantity	Experiment	Simulation
Peak electron density	$4.73 \times 10^{20} \text{ cm}^{-3}$	$4.834 \times 10^{20} \text{ cm}^{-3}$
Peak lattice temperature rise	$\sim 1.5 \times 10^3 \text{ K}$	$\sim 1.6 \times 10^3 \text{ K}$
Central stress amplitude	$\sim 1.47 \text{ GPa}$	$\sim 1.4 \text{ GPa}$
Peripheral stress amplitude	$\sim 1.23 \text{ GPa}$	$\sim 1.7 \text{ GPa}$
Stress-wave velocity	$\sim 4.3 \text{ } \mu\text{m/ns}$	$\sim 4.5 \text{ } \mu\text{m/ns}$

The simulated peak electron density and lattice temperature rise are in good agreement with the experimentally derived values. More importantly, the model reproduces the GPa-level stress amplitude and the stress-wave propagation velocity observed in the transient birefringence measurements. The slightly higher simulated peripheral stress may arise from the simplified two-dimensional treatment, idealized heat-source geometry, and the absence of dissipative stress relaxation in the present model. Nevertheless, the agreement in magnitude and propagation velocity supports

the quantitative consistency between plasma energy deposition and stress-wave generation.

6.9 Parameter-sensitivity validation under lower pulse energy

To further examine the parameter sensitivity of the carrier-thermal-elastic model, we performed additional experiments and simulations under a lower single-pulse energy of 0.4 μJ . Compared with the representative 0.8 μJ condition discussed above, the 0.4 μJ condition provides a useful test case for evaluating whether the model can capture the reduced plasma generation, lattice heating, and stress-wave response when the deposited laser energy is decreased.

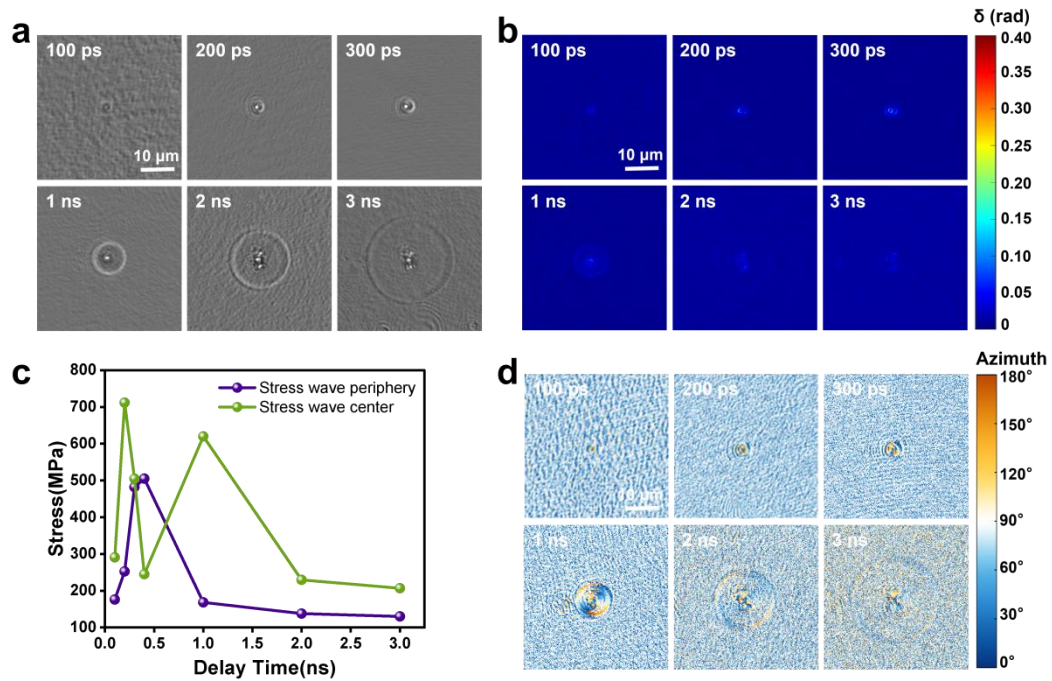


Supplementary Figure 7 | Experimental time-resolved plasma response in INF glass under 0.4 μJ . **a** Spatiotemporal electron-density distributions of femtosecond-laser-induced plasma. **b** Temporal evolution of the transient peak electron density. Source data are provided as a Source Data file.

Supplementary Fig. 7 shows the experimentally measured time-resolved electron-density evolution in INF glass under 0.4 μJ irradiation. The plasma signal is weaker than that under the saturated 0.8 μJ condition. The peak electron density reaches approximately $2.72 \times 10^{20} \text{ cm}^{-3}$ at around 400 fs and then gradually decreases with increasing pump-probe delay. This value is clearly lower than the saturated electron density measured under 0.8 μJ irradiation, indicating that carrier generation is strongly dependent on the pulse energy.

The corresponding time-resolved stress-wave measurements under 0.4 μJ irradiation are shown in Supplementary Fig. 8. The dynamic optical transmission images show a radially propagating stress wave similar to that observed under 0.8 μJ irradiation. The

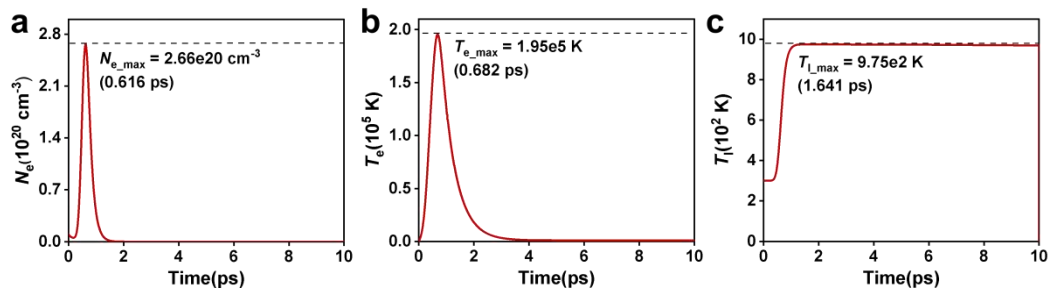
propagation velocity is approximately $4.3 \mu\text{m}/\text{ns}$, indicating that the stress wave velocity is mainly governed by the acoustic properties of the glass rather than by the pulse energy. However, the stress amplitude is significantly reduced. Based on the optical retardance calculation, the maximum stress values in the central and peripheral regions are approximately 711 MPa and 505 MPa, respectively, occurring at around 200 ps and 400 ps. These values correspond to lattice-temperature rises of approximately 714 K and 507 K, respectively. Compared with the $0.8 \mu\text{J}$ condition, the stress amplitude decreases more rapidly after approximately 1 ns, which is consistent with weaker energy deposition and faster attenuation of the thermoelastic stress wave during radial propagation.



Supplementary Figure 8 | Experimental time-resolved stress-wave response in INF glass under $0.4 \mu\text{J}$. **a** Dynamic optical transmission images at different time delays. **b** Optical retardance images at different time delays. **c** Temporal evolution of stress in the central and peripheral regions. **d** Slow-axis azimuth distributions of the transient birefringence. Source data are provided as a Source Data file.

The carrier-thermal simulation under $0.4 \mu\text{J}$ irradiation is shown in Supplementary Fig. 9. In this calculation, the same material parameters and modeling framework are used,

while the pulse energy is reduced from 0.8 μJ to 0.4 μJ . The simulated electron density reaches a maximum of $2.66 \times 10^{20} \text{ cm}^{-3}$ at 0.616 ps, which agrees well with the experimentally measured value of $2.72 \times 10^{20} \text{ cm}^{-3}$. The electron temperature reaches $1.95 \times 10^5 \text{ K}$ at 0.682 ps, while the lattice temperature reaches $9.75 \times 10^2 \text{ K}$ at 1.641 ps. Taking room temperature as approximately 300 K, the calculated lattice-temperature rise is approximately 675 K, close to the experimentally inferred temperature rise of approximately 714 K. This agreement indicates that the model captures the reduced carrier generation and lattice heating under lower pulse energy.



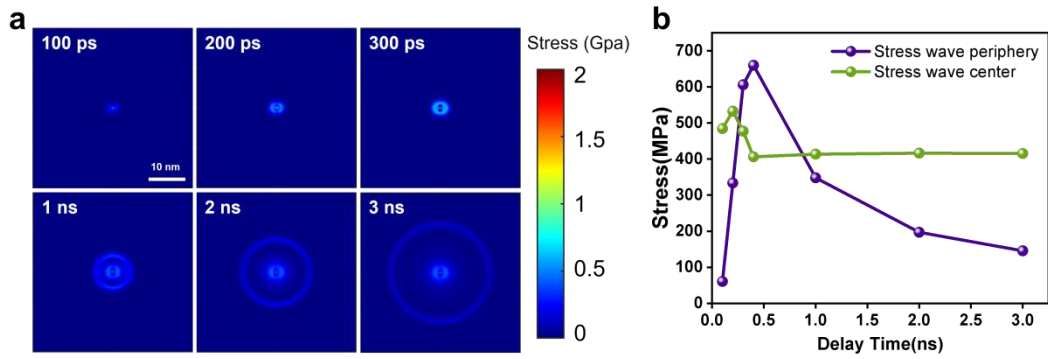
Supplementary Figure 9 | Simulated carrier and thermal dynamics in INF glass under 0.4 μJ .

a Simulated temporal evolution of free-electron density $n_e(t)$. **b** Simulated temporal evolution of electron temperature $T_e(t)$. **c** Simulated temporal evolution of lattice temperature $T_l(t)$. Source data are provided as a Source Data file.

Supplementary Fig. 10 shows the simulated stress-wave evolution under 0.4 μJ irradiation. The simulated stress wave propagates outward from the laser-excited region with a velocity of approximately 4.35 $\mu\text{m/ns}$, in good agreement with the experimental value of approximately 4.3 $\mu\text{m/ns}$. The simulated central stress reaches approximately 0.53 GPa at around 200 ps, while the peripheral stress reaches approximately 0.66 GPa at around 400 ps. Although the simulated center/periphery stress ratio differs slightly from the experimental values, the model captures the main trend that lowering the pulse energy reduces the stress amplitude to the sub-GPa level while maintaining a similar stress-wave velocity.

The slightly higher simulated peripheral stress can be understood from the fact that the calculated quantity is the principal stress difference associated with the

propagating stress wave. During radial propagation, the stress gradient is concentrated near the wavefront, so the local principal stress difference in the peripheral wavefront region can exceed that in the more isotropic central heated region. In addition, the simplified two-dimensional treatment, idealized heat-source geometry, and the absence of dissipative stress relaxation may enhance the simulated wavefront stress. Nevertheless, the calculated values are in the same order of magnitude as the experimental results and reproduce the key energy-dependent trend.



Supplementary Figure 10 | Simulated stress-wave evolution in INF glass under 0.4 μJ . **a** Simulated spatial distributions of the stress wave at different time delays. **b** Simulated temporal evolution of stress in the central and peripheral regions. Source data are provided as a Source Data file.

Overall, the 0.4 μJ results demonstrate that the carrier-thermal-elastic model captures the pulse-energy-dependent reduction in plasma density, lattice heating, and stress-wave amplitude. At the same time, the stress-wave velocity remains close to the material-governed acoustic propagation velocity. This lower-energy validation indicates that the model is not limited to the representative 0.8 μJ condition, but provides a parameter-sensitive framework for predicting the transient plasma-to-stress response under different pulse-energy conditions.

6.10 Discussion and physical implications

The simulation results confirm that femtosecond laser-induced plasma formation and subsequent relaxation act as the principal drivers of thermoelastic stress generation in fluoride glass. The coupling between electronic excitation, energy dissipation, lattice

heating, and mechanical response bridges femtosecond and nanosecond regimes, forming a quantitative picture of femtosecond laser–matter interaction.

Within the sub-picosecond timescale, multiphoton and avalanche ionization produce dense plasma with $n_e \approx 10^{20} \text{ cm}^{-3}$. The transient plasma modifies the dielectric response through the Drude term, leading to strong free-carrier absorption. Subsequently, electron–phonon coupling transfers the absorbed energy to the lattice within several picoseconds, generating a localized high-temperature region. The rapid thermal expansion of this heated volume produces a thermoelastic pressure source, which launches a stress wave into the surrounding glass matrix.

The calculated pressure and stress profiles show that the absorbed energy density is sufficient to generate GPa-level transient stress. The simulated stress-wave velocity is also close to the measured propagation velocity. These results indicate that the experimentally measured plasma parameters can quantitatively account for the observed stress-wave behavior.

The additional lower-energy validation at 0.4 μJ further demonstrates the parameter sensitivity of this framework. When the pulse energy is reduced from 0.8 μJ to 0.4 μJ , both experiment and simulation show decreased peak electron density, weaker lattice heating, and reduced stress-wave amplitude, while the stress-wave velocity remains nearly unchanged. This behavior is consistent with the physical expectation that the stress amplitude is mainly governed by deposited energy density, whereas the propagation velocity is determined primarily by the elastic and acoustic properties of the glass.

Overall, this carrier-thermal-elastic model provides a quantitative and parameter-sensitive link between femtosecond plasma evolution, transient lattice heating, and stress-wave generation in fluoride glass. It therefore supports the physical consistency of the plasma-to-stress pathway observed experimentally and provides a useful framework for understanding and predicting femtosecond laser-induced mechanical response in transparent dielectric materials.

Note 7: Gradient-driven ion redistribution model

This Note presents a gradient-driven ion redistribution model for analyzing the elemental migration behavior observed in femtosecond-laser-modified INF glass. In the main text, WDS elemental mapping shows that Pb and In are enriched in the central positive-index region, whereas F and Ba are redistributed toward the peripheral negative-index region. To clarify the physical origin of this ion-selective redistribution, we established a drift-diffusion model that incorporates concentration-gradient relaxation, thermal-gradient-driven diffusion, pressure-gradient-driven drift, and ion-specific mobility.

The purpose of this model is to examine whether the experimentally measured thermal and pressure fields can naturally produce the observed redistribution trend. In particular, two cases are compared: a thermal-gradient-only condition with the pressure field set to zero, and a pressure-assisted condition using the experimentally constrained GPa-level pressure field.

7.1 Transport equation

The radial flux of each ionic species is expressed as the sum of concentration-gradient diffusion, thermal-gradient-driven diffusion, and pressure-gradient-driven drift. This form follows the general drift-diffusion description based on non-equilibrium thermodynamics and stress-coupled diffusion in solids^{18,19}:

$$J_i = -D_i \frac{\partial c_i}{\partial r} - D_i S_{T,i} c_i \frac{\partial T}{\partial r} + D_i \frac{\Omega_i^{\text{eff}}}{k_B T} c_i \frac{\partial P}{\partial r}, \quad (7-1)$$

where J_i is the radial flux of species i , c_i is the normalized concentration, D_i is the temperature-dependent diffusion coefficient, $S_{T,i}$ is the thermal diffusion coefficient, T is the local temperature, P is the local pressure, k_B is the Boltzmann constant, and Ω_i^{eff} is an effective pressure-coupling coefficient. The first term represents concentration-gradient relaxation, the second term represents thermal-gradient-driven diffusion, and the third term represents pressure-gradient-driven drift.

The effective pressure-coupling coefficient Ω_i^{eff} is used to describe the tendency of an ionic species to be stabilized in the compressed region. It should be understood as an

effective transport parameter rather than a strict thermodynamic partial molar volume. A larger Ω_i^{eff} indicates stronger pressure-gradient stabilization.

The temperature-dependent mobility is described using an Arrhenius-type relation²⁰:

$$D_i(T) = D_{0,i} \exp\left(-\frac{E_{a,i}}{k_B T}\right), \quad (7-2)$$

where $D_{0,i}$ is the diffusion prefactor and $E_{a,i}$ is the activation energy. The concentration evolution is calculated using the cylindrical continuity equation:

$$\frac{\partial c_i}{\partial t} = -\frac{1}{r} \frac{\partial(rJ_i)}{\partial r}, \quad (7-3)$$

where r is the radial coordinate measured from the laser-modified center. This geometry is used because the observed elemental redistribution and stress-wave response show an approximately radial character around the modified zone.

7.2 Temperature and pressure fields

The transient temperature field is approximated as a localized Gaussian source with exponential temporal relaxation:

$$T(r, t) = T_0 + \Delta T_0 \exp\left(-\frac{r^2}{2\sigma_T^2}\right) \exp\left(-\frac{t}{\tau_T}\right), \quad (7-4)$$

where T_0 is the ambient temperature, ΔT_0 is the peak temperature rise, σ_T is the effective thermal radius, and τ_T is the thermal relaxation time.

For the pressure-assisted case, the pressure field is also described using a Gaussian profile:

$$P(r, t) = P_0 \exp\left(-\frac{r^2}{2\sigma_P^2}\right) \exp\left(-\frac{t}{\tau_P}\right), \quad (7-5)$$

where P_0 is the peak pressure, σ_P is the characteristic pressure radius, and τ_P is the pressure relaxation time. In the thermal-gradient-only case, $P_0 = 0$. In the pressure-assisted case, P_0 is set to 1.5 GPa, close to the experimentally measured stress amplitude and the calculated GPa-level pressure field.

7.3 Simulation cases

Two cases were calculated to separate the roles of thermal and pressure-gradient effects.

In the thermal-gradient-only case, the pressure field is set to zero. This case is used to determine whether the transient thermal gradient alone can drive the experimentally observed redistribution.

In the pressure-assisted case, the experimentally constrained pressure field is included together with the same temperature field. This case is used to examine whether the coupled thermal and pressure-gradient field can reproduce the central enrichment of Pb/In and the peripheral redistribution of F/Ba observed in the WDS maps.

The initial concentration of each species is set to $c_i = 1$, corresponding to a uniform elemental distribution before laser irradiation. The calculated concentration variation is expressed as $\Delta c/c_0$.

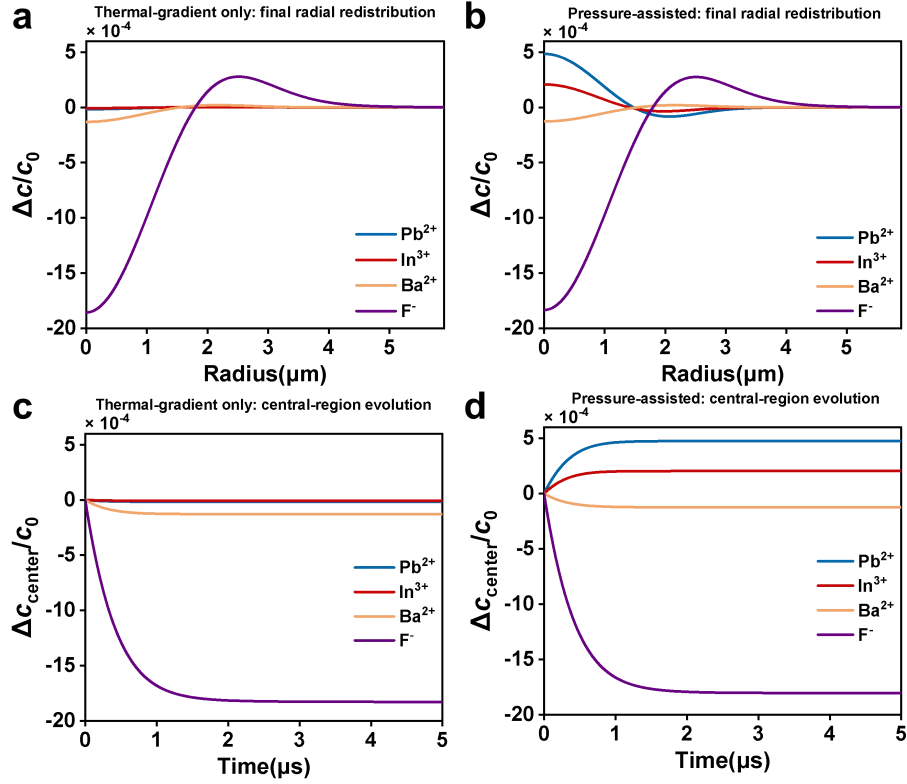
7.4 Simulation results

The thermal-gradient-only result is shown in Supplementary Fig. 11a and c. In this case, F⁻ exhibits the most obvious outward redistribution, while Ba²⁺ also shows weaker outward migration. In contrast, Pb²⁺ and In³⁺ do not show central enrichment. This result indicates that thermal-gradient-driven redistribution alone can account for the outward migration of more mobile species, but it cannot explain the Pb/In accumulation observed in the positive-index central region.

The pressure-assisted result is shown in Supplementary Fig. 11b and d. When the experimentally constrained GPa-level pressure field is included, Pb²⁺ and In³⁺ become enriched in the central region. Meanwhile, F⁻ and Ba²⁺ remain depleted in the center and are redistributed toward the peripheral region. This trend agrees with the WDS elemental maps in Fig. 6, where Pb/In enrichment corresponds to the positive-index region and F/Ba redistribution corresponds to the surrounding negative-index region.

The comparison between these two cases shows that the observed elemental segregation cannot be explained by thermal diffusion alone. Instead, it arises from coupled thermal and pressure-gradient transport. The pressure-gradient term stabilizes

highly polarizable cations such as Pb^{2+} and In^{3+} in the compressed central region, whereas thermal-gradient-driven redistribution and charge-compensated transport dominate the outward migration of F^- and Ba^{2+} .



Supplementary Figure 11 | Gradient-driven ion redistribution model in INF glass. **a** Final radial redistribution under the thermal-gradient-only condition. **b** Final radial redistribution under the pressure-assisted condition. **c** Temporal evolution of the center-region concentration change under the thermal-gradient-only condition. **d** Temporal evolution of the center-region concentration change under the pressure-assisted condition. Source data are provided as a Source Data file.

7.5 Physical implication

This model clarifies that the elemental redistribution in femtosecond-laser-modified INF glass should not be interpreted as a simple heavy-ion/light-ion separation. The different migration tendencies arise from the relative magnitudes of thermal diffusion, pressure-gradient drift, concentration/chemical-potential relaxation, and ion-specific mobility.

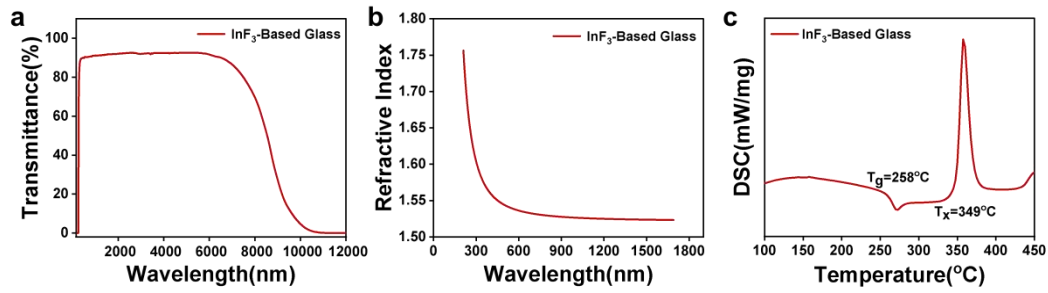
The pressure-assisted calculation shows that the experimentally measured GPa-level pressure field is sufficient to reverse the migration tendency of Pb^{2+} and In^{3+} , leading to their central enrichment. At the same time, F^- and Ba^{2+} are governed mainly by thermal-gradient-driven redistribution and charge-compensated transport, leading to peripheral enrichment. This coupled transport mechanism explains the composition-dependent polarizability redistribution and the resulting refractive-index sign inversion across the modified region.

Note 8: Fluoride glass fabrication parameters

This Note provides the fundamental-property characterization of the fluorindate (INF) glass used in this work, together with the fabrication parameters of the fluoride glasses employed in the experiments.

8.1 Fundamental-property characterization of INF glass

The basic physical properties of the INF glass used in this study are summarized in Supplementary Fig. 12, including its transmittance spectrum, refractive-index profile and differential thermal analysis (DTA) curve.



Supplementary Figure 12 | Fundamental-property characterization of INF glass. a Transmittance spectrum of fluorindate glass (200 nm–12 μm). **b** Refractive index profile of fluorindate glass (200 nm–1700 nm). **c** Differential thermal analysis curve of fluorindate glass (100–450°C). Source data are provided as a Source Data file.

Supplementary Fig. 12a shows the transmittance spectrum of INF glass: compared to chalcogenide glasses (which have longer transmission windows), INF glass achieves higher transmittance ($> 90\%$) over 0.2–7 μm , whereas chalcogenide glasses only reach ~ 50 – 70% transmittance in this range²¹. This broadband high-transmittance property is irreplaceable. The intrinsic refractive index of the glass is critical for simulating and calculating transient physical processes. As shown in Supplementary Fig. 12b, the refractive indices of INF glass are 1.542 at the probe beam wavelength (520 nm) and 1.526 at the pump beam wavelength (1040 nm). Supplementary Fig. 12c presents the differential thermal analysis curve of INF glass, with a glass transition temperature (T_g) of $\sim 250^\circ\text{C}$ and crystallization temperature (T_x) of $\sim 349^\circ\text{C}$, yielding a ΔT of 99°C —indicating excellent fiber-drawing capability.

8.2 Fluoride glass fabrication parameters

Fluoroaluminate (ABYPM) and fluorozincate glasses (AZF/AZFD) are prepared using a melt-quenching method, similar to fluoroindate (INF) glass, with the key difference being the annealing temperature. The specific preparation procedure for fluoroindate glass is provided in the main text; here, we focus on the compositions and preparation temperatures of fluoroaluminate and fluorozincate glasses.

8.2.1 Glass compositions

ABYPM: 30 AlF₃ - 12 BaF₂ - 20 YF₃ - 28 PbF₂ - 10 MgF₂

AZF: 32 ZnF₂ - 12 BaF₂ - 11 YF₃ - 13 SrF₂ - 32 AlF₃

AZFD: 32 ZnF₂ - 12 BaF₂ - 10 YF₃ - 13 SrF₂ - 32 AlF₃ - 1 DyF₃

8.2.2 Preparation temperatures

Name	Fluorination temperature/time	Firing temperature/time	Annealing temperature/time
ABYPM	400°C/0.5h	950°C/3h	360°C/5h
AZF/AZFD	400°C/0.5h	950°C/3h	350°C/5h

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