Wavelength scaling of electron collision time in plasma for strong field laser-matter interactions in solids

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Although the dielectric constant of plasma depends on electron collision time as well as wavelength and plasma density, experimental studies on the electron collision time and its effects on laser-matter interactions are lacking. Here, we report an anomalous regime of laser-matter interactions generated by wavelength dependence (1.2–2.3 μm) of the electron collision time in plasma for laser filamentation in solids. Our experiments using time-resolved interferometry reveal that electron collision times are small (<1 femtosecond) and decrease as the driver wavelength increases, which creates a previously-unobserved regime of light defocusing in plasma: longer wavelengths have less plasma defocusing. This anomalous plasma defocusing is counterbalanced by light diffraction which is greater at longer wavelengths, resulting in almost constant plasma densities with wavelength. Our wavelength-scaled study suggests that both the plasma density and electron collision time should be systematically investigated for a better understanding of strong field laser-matter interactions in solids.

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It is well known that the optical properties of materials, such as light reflection, refraction, and absorption are described by dielectric constants. When a high-intensity laser interacts with a material, bound electrons are ionized, creating plasma. The complex dielectric constant for plasma¹ is given by $\epsilon(\omega) = 1 - \omega_p^2 / (\omega(\omega + i/\tau_c))$, where ω is the optical angular frequency, ω_p is the plasma frequency which is proportional to the square root of the plasma density, and τ_c represents the electron collision time. Therefore, to fully determine the optical properties of plasma, it is critical to have the information on the plasma density, as well as the electron collision time. In particular, the electron collision time plays an important role in high-density plasma, where free electrons frequently collide with other electrons, ions, and atoms. Although there exist a lot of reports on plasma density measurements, there are only a handful of experiments that measured electron collision times^{2,3} and thus τ_c is still a debatable parameter. For instance, the electron collision times used in previous work for the laser-matter interaction in fused silica^{2–4} vary significantly, ranging from 0.2 to 23 fs.

One of the key light-matter interaction phenomena involving light refraction and absorption in plasma is laser filamentation, which is high-intensity laser self-guidance due to the dynamic balance between optical Kerr effect (self-focusing) and plasma defocusing/diffraction^{5–8}. Laser filamentation has been an active topic of research because of its fundamental novelty^{6–8}, as well as various important applications, such as few-cycle pulse generation^{9,10}, terahertz generation^{11,12}, remote sensing¹³, long-lived waveguides¹⁴, rain-making¹⁵, and lightning control¹⁶. Modern laser technology enables researchers to study laser filamentation in gases^{17,18} and solids^{19–23} and other strong field laser-matter interactions, such as high-order harmonic generation (HHG)²⁴ in the mid-infrared (IR) and long-wavelength IR. Since plasma defocusing and diffraction generally increase as the driver wavelength increases, the peak intensity and plasma density in a laser filament are expected to become smaller at longer wavelengths^{25,26}. Several theoretical studies in gases/air have shown similar trends^{27,28} and moreover, recent experimental work in air²⁹ using a 10- μm CO₂ laser has reported a long megafilament with low-plasma densities ($<10^{16} \text{ cm}^{-3}$). As a comparison, a theoretical study on wavelength-scaled filamentation in a solid has predicted a similar monotonic decrease in the plasma density, with increasing wavelength for the driver wavelengths in the anomalous group-velocity dispersion (GVD) regime³⁰. However, to the best of our knowledge, there is no systematic experimental investigation on wavelength dependence of the plasma dynamics in laser filamentation.

Here, we report on experimental and theoretical wavelength scaling of the plasma dynamics in femtosecond laser filamentation in solids. Time-resolved interferometry is performed to simultaneously measure plasma densities and electron collision times in filaments by varying the pump wavelength from $\lambda = 1.2\text{--}2.3 \mu\text{m}$. The plasma densities show little change for different wavelengths, which is in contrast to the expected trend of monotonic decrease with increasing wavelength. Furthermore, the measured electron collision times are smaller than 1 femtosecond (fs) and decrease as the driver wavelength increases, which is due to hotter electron generation and increased electron excursion length in filaments at longer wavelengths. Our analysis shows that the observed wavelength dependence of the electron collision time creates an anomalous regime of light defocusing under plasma: longer wavelengths have less plasma defocusing. Simulations using the measured electron collision times successfully reproduce the measured wavelength dependence of the plasma density. Our work suggests that the electron collision time in plasma should be systematically studied for a precise understanding of strong field laser-matter interactions in solids.

Results

Plasma density measurement. Time-resolved experiments are performed using variable-wavelength driver (pump) pulses from an optical parametric amplifier (OPA) that is pumped by a linearly polarized Ti:sapphire laser system [1-kHz repetition rate, 800-nm central wavelength, 45-fs full-width at half-maximum (FWHM) pulse duration]. The pump wavelength (λ) is changed between 1.2 and 2.3 μm with a 0.1- μm interval. However, we do not use 1.6- μm wavelength in our experiments because of its instability. Filaments are generated in a 2.5-cm long fused silica sample. Since zero GVD occurs near 1.27 μm for fused silica, wavelengths longer than 1.3 μm belong to the anomalous-GVD regime. The input peak powers are smaller than six times the critical power for self-focusing P_{cr} to avoid multifilaments and generate stable single filaments. We focus the pump beams using a 15-cm focal length CaF₂ lens and the geometrical foci are located 2-mm before the input face of the sample to ensure that plasma is generated through self-focusing with minimal help from CaF₂ lens. First, we characterize the input beams at all the wavelengths by measuring the focused mode sizes, pulse durations (60–130 fs), spectra, and chirp. For measurements of plasma densities in filamentation, we use femtosecond time-resolved single-shot interferometry, in which a weak collimated 800-nm beam acts as a probe and orthogonally traverses the pump-generated filamentation region. Single-shot measurements of maximum plasma densities near the onset of stable filaments are performed by decreasing the pulse repetition rate from 1 kHz to 50 Hz using optical choppers (see the “Methods” and Supplementary Note 1). We use the standard Fourier transformation technique³¹ for phase extraction and Abel inversion, assuming cylindrical symmetry for retrieving the refractive index change (Δn) via plasma from the measured phase. First, based on the assumption $\omega_{\text{pr}}\tau_c \gg 1$, where ω_{pr} is the 800-nm probe angular frequency, which is the standard assumption for most cases (e.g., gases), we retrieve the plasma density using $\Delta n = -\rho/2\rho_c$, where ρ is the plasma density, $\rho_c = \epsilon_0 m_e^* \omega_{\text{pr}}^2 / e^2$ is the critical plasma density for the probe, ϵ_0 is the vacuum permittivity, e is the electron charge, and m_e^* is the reduced electron mass. We will discuss the validity of the assumption of $\omega_{\text{pr}}\tau_c \gg 1$ and its effect on the measured plasma densities later. According to Fig. 1a, the measured plasma densities show little change for different wavelengths³². Details on data averaging and comparison of two different averaging methods are provided in Supplementary Note 2. This result is in contrast to the expected trend based on filaments in air/gases: monotonic decrease in the plasma density because of greater diffraction and plasma defocusing at longer wavelengths.

Numerical simulations. To compare with experiments and understand the underlying dynamics, we perform numerical simulations by solving the nonlinear envelope equation^{33,34} in fused silica, using the experimental parameters. The electric field propagation equation is coupled with the plasma generation equation considering optical field ionization (OFI), collisional ionization³⁵, and plasma recombination, which is given by

$$\frac{\partial \rho}{\partial t} = W(I)(\rho_0 - \rho) + \frac{\sigma_{\text{IB}} I}{U_i} - \frac{\rho}{\tau_r} \quad (1)$$

Here, ρ is the plasma density, ρ_0 is the neutral atomic density, and $W(I)$ is the OFI rate. For OFI, we use either multiphoton ionization (MPI) or full Keldysh ionization (FKI)³⁶, which describes both MPI and tunneling ionization depending on driver wavelength and intensity. The second term in Eq. (1) represents collisional ionization. Here, σ_{IB} is the inverse

Fig. 1 Plasma density scaling with wavelength in filamentation.

a Measured plasma density scaling with wavelength. Simulated plasma density scaling using **b** multiphoton ionization (MPI) and collisional ionization, and **c** full Keldysh ionization (FKI) and collisional ionization. Simulations are performed with the measured parameters (measured para.,) [red diamond and black square] and also with the same (input beam) parameters (same para.) ($P = 5P_{cr}$, $\tau_p = 70$ fs, $w_0 = 60$ μm and no chirp) [blue triangle and magenta downward triangle]. The full list of measured parameters is in the table displayed in **d**. In the simulations, constant electron collision times $\tau_c = 1.7$ and 3 fs are used for all the wavelengths. **d** Table of experimental parameters; λ : wavelength, τ_p : FWHM pulse duration, w_0 : $1/e^2$ beam radius, and P/P_{cr} : normalized input peak power with respect to critical power. The error bars in **a** correspond to the standard deviation of 35 laser shots.

Bremsstrahlung coefficient, which is given by the real part of $\sigma_c(\omega) = \omega_0^2 \tau_c (1 + i\omega\tau_c) / [n(\omega)c\rho_c(1 + \omega^2\tau_c^2)]$, where ω_0 is the driver laser angular frequency, $n(\omega)$ is the refractive index, and ρ_c is the critical plasma density for the driver laser. U_i is the fused silica bandgap energy (9 eV) and I is the laser intensity. The plasma recombination time τ_r in fused silica is 150 fs (ref. 37). More details about the nonlinear index measurement via z -scan³⁸ and the simulation are provided in Supplementary Notes 3 and 4, respectively.

We first use the MPI rate for OFI and constant τ_c 's for all the wavelengths in the simulation. In detail, we perform two sets of simulations with two constant τ_c 's: $\tau_c = 1.7$ fs (ref. 2) and 3 fs (ref. 33). The calculated plasma density with MPI (Fig. 1b) shows a monotonic decrease with increasing wavelength for both τ_c 's, which thus cannot reproduce the experiment. Next, we use the FKI rate for OFI, which is known to be more accurate than the MPI rate. However, even with FKI, the calculated plasma density still monotonically decreases with increasing wavelength (Fig. 1c). As shown in Fig. 1d, our experimental parameters such as pulse durations and beam sizes are different at different wavelengths.

Fig. 2 Measurement of the electron collision times in filament-produced plasma.

Examples of two-dimensional profiles of phase and electric field absorption (normalized probe electric field amplitude) during filamentation for **a, b** 1.2 μm and for **c, d** 2.3 μm measured by single-shot femtosecond time-resolved interferometry. **e** Measured electron collision time versus wavelength. The error bars in **e** correspond to the standard deviation of 35 laser shots.

Thus, we also perform simulations using the same parameters for all the wavelengths (input power of $5P_{cr}$, pulse duration of 70 fs, beam radius of 60 μm and no chirp) to investigate the effect. However, the simulations with the same parameters also show a similar monotonic decrease (dashed lines in Fig. 1b, c). We hypothesize that the discrepancy between the experiment and the simulation comes from uncertainties in the electron collision time because $\sigma_c(\omega)$, which critically affects both collisional ionization and light defocusing/absorption, is a function of τ_c . As mentioned, the electron collision times used in previous work using fused silica vary significantly⁴, ranging between 0.2 and 23 fs for 800-nm lasers.

Electron collision time measurement. To confirm our hypothesis, we directly measure electron collision times in laser filaments as a function of driver wavelength. Our analysis shows that time-resolved interferometry enables simultaneous measurements of the plasma density and electron collision time in a single shot via phase shift and probe absorption, for which the expression is given by (see Supplementary Note 5 for further details)

$$\omega_{pr}\tau_c = \frac{\Delta\phi}{\ln\left(\frac{E_0}{E_p}\right)} \quad (2)$$

Here, $\Delta\phi$ is the measured phase change and $\ln(E_0/E_p)$ represents the probe absorption due to plasma, where E_p and E_0 are the probe electric fields with and without plasma, respectively. Examples of single-shot two-dimensional phase and amplitude are shown in Fig. 2a, b and Fig. 2c, d for $\lambda = 1.2$ and 2.3 μm, respectively. The positive phases in Fig. 2a and Fig. 2c are due to cross-phase modulation generated by pump pulses. The measured electron collision time versus wavelength is shown in Fig. 2e. The

Fig. 3 Simulation results with the measured electron collision times.

a Measured plasma density analysis with (black circle) and without (red square) the assumption of $\omega_{pr}\tau_c \gg 1$, where ω_{pr} is the probe angular frequency and τ_c is the electron collision time. **b** Simulated plasma densities versus wavelength with constant $\tau_c = 0.4 \text{ fs}$ (red dashed line), 0.7 fs (blue dotted line), and with measured collision times (black solid line). Simulation results using the measured collision times agree well with the observed wavelength scaling of plasma density. The error bars correspond in **a** to the standard deviation of 35 laser shots.

measured electron collision times are very small ($<1 \text{ fs}$) and not constant, showing a decrease from $\tau_c \sim 0.7$ to $\tau_c \sim 0.4 \text{ fs}$ with increasing wavelength. Examples of extracted two-dimensional electron collision times are provided in Supplementary Note 6. Our observation is reproducible and one more data set, which shows similar wavelength scaling, is provided in Supplementary Note 7.

Discussion

Since the measured electron collision times are small, the assumption of $\omega_{pr}\tau_c \gg 1$ is not quite valid, and thus the plasma density measurement via interferometry should be reexamined. We reextract the plasma densities using the index change

$$\Delta n = -\rho/2\rho_c^*, \text{ where } \rho_c^* = \varepsilon_0 m_e^* \omega_{pr}^2 \left(1 + (\omega_{pr}\tau_c)^{-2} \right) / e^2 \text{ is the}$$

modified critical plasma density considering the electron collision time. As shown in Fig. 3a (black solid line), although consideration of τ_c increases the extracted plasma densities, the trend remains almost the same (almost constant plasma densities for different driver wavelengths). Next, we perform numerical simulations using the measured collision times with the FKI and collisional ionization models (Eq. 1). As shown in Fig. 3b (black solid line), the simulations are in very good agreement with the experiments. In contrast, the simulations using a couple of measured electron collision times ($\tau_c = 0.7$ and 0.4 fs), but assuming constant τ_c for all λ 's still show a decrease with increasing wavelength [$\tau_c = 0.4$ (red dashed line) and 0.7 fs (blue dotted line) in Fig. 3b].

Fig. 4 Calculations of collisional ionization cross-section and plasma defocusing as a function of electron collision time.

a The inverse Bremsstrahlung coefficient $\sigma_{IB} = \text{Real}[\sigma_c(\omega)]$, which is related to collisional ionization, versus electron collision time for a few wavelengths. **b, c** Dependence of plasma defocusing ($\text{Imag}[\sigma_c(\omega)]$) on electron collision time (τ_c) at $\lambda = 1.2 \mu\text{m}$ (black solid line), $\lambda = 1.7 \mu\text{m}$ (red dashed line), $\lambda = 2 \mu\text{m}$ (blue dotted line), and $\lambda = 2.3 \mu\text{m}$ (magenta dashed-dotted line). Plasma defocusing increases with increasing wavelength for $\tau_c > 1 \text{ fs}$, whereas it decreases with increasing wavelength for $\tau_c < 1 \text{ fs}$ as shown in **c**, which is a zoomed-in figure for the shaded area in **b**.

We carry out further analysis on the effect of the measured electron collision times on collisional ionization and plasma defocusing. Figure 4a shows calculations of the inverse Bremsstrahlung coefficient σ_{IB} versus τ_c for a few wavelengths, which shows a decrease in σ_{IB} as τ_c decreases for $\tau_c < 1 \text{ fs}$. Since collisional ionization is proportional to σ_{IB} (Eq. 1), this indicates that collisional ionization should be greater at larger electron collision times for $\tau_c < 1 \text{ fs}$. However, as shown in Fig. 3b, the simulation with $\tau_c = 0.4 \text{ fs}$ shows higher plasma densities than that with $\tau_c = 0.7 \text{ fs}$. This suggests that plasma defocusing should be more important than collisional ionization. According to the calculated imaginary part of $\sigma_c(\omega)$ related to plasma defocusing (Fig. 4b, c), an anomalous regime of light defocusing under plasma is created for $\tau_c < 1 \text{ fs}$: longer wavelengths have less plasma defocusing. This is in contrast to the well-known normal regime, where plasma defocusing becomes greater with increasing wavelength, which is the case for gases/air (trend in Fig. 4b for $\tau_c > 1 \text{ fs}$). In this,

Fig. 5 Calculations of clamping intensities inside filaments, ponderomotive energies, excursion lengths, and electron collision times. Calculated **a** clamping intensities with measured τ_c 's and constant $\tau_c = 1.7$ fs, **b** ponderomotive energies (U_p), and **c** excursion lengths (x_c) are shown for filamentation at different wavelengths. **d** The calculated electron collision time as a function of electron temperature. Clamping intensities from simulations in **a** (black solid line) are used to calculate the ponderomotive energies in **b** and excursion lengths in **c**.

observed anomalous regime, plasma densities and thus filament intensities should be large even at long wavelengths. Our calculations confirm that the clamping intensities in filaments at long wavelengths with the measured τ_c 's are larger than those with a constant $\tau_c = 1.7$ fs (see Fig. 5a and additional simulation data in Supplementary Note 8). These relatively high-intensity filaments at long wavelengths should enhance HHG and may facilitate attosecond pulse generation in solids^{39–41} due to enhanced ponderomotive energy (cycle-averaged kinetic energy of a free electron). However, an important question arises here: why do the plasma density and filament intensity not increase with increasing wavelength in the anomalous plasma defocusing regime? We think it is because the anomalous plasma defocusing is counterbalanced by light diffraction, which is greater at longer wavelengths.

Finally, we attribute the observed decrease in electron collision time with increasing wavelength to two reasons: first, it is because of larger ponderomotive energy $U_p \propto \lambda^2 I$ for longer-wavelength filaments (shown in Fig. 5b), which results in more heating of free electrons and frequent electron-neutral collisions. Second, it is because of larger free electron excursion length ($x_c \propto \lambda^2 I^{0.5}$) at longer wavelengths (shown in Fig. 5c), which results in higher probabilities of electron collision with neighbor atoms. The simulation with the measured τ_c 's shows that the filament intensity does not decrease much even at long wavelengths (only ~35% decrease as shown in the black solid line of Fig. 5a). Note that both U_p and x_c depend on λ^2 and thus, the wavelength is more important than the intensity for their scaling. For further details related to electron heating, the electron-neutral collision frequency (ν_{en})^{42,43} is given by $\nu_{en}[\text{s}^{-1}] = 2 \times 10^{-7} (\rho_0 - \rho) T_e^{1/2}$, where ρ_0 is the neutral atomic density in cm^{-3} and T_e is the free electron temperature in eV. Since the electron temperature should increase at longer wavelengths due to larger ponderomotive energy, the electron collisional frequency (time) should increase (decrease) with increasing wavelength (i.e., $\nu_{en} = 1/\tau_c \propto T_e^{1/2}$). Note that ν_{en} should dominate over electron-electron and electron-ion collisions due to weak ionization (<0.1%) in fused silica under the filamentation regime. We calculate τ_c considering various types of electron collisions as a function of T_e (Supplementary Fig. S14a). Figure 5d shows that the calculation

successfully reproduces the measured decrease of τ_c with increasing wavelength based on the simple, but reasonable assumption of $U_p \propto T_e$ (more precisely $U_p = 3T_e/2$). More details on calculations of electron collision time are provided in Supplementary Note 9, wherein we also discuss the effect of pulse durations on electron collision times.

In conclusion, we experimentally and theoretically investigate wavelength scaling of the plasma density and electron collision time in laser filamentation in fused silica with driver wavelengths ranging between 1.2 and 2.3 μm . The measured plasma densities show little change for different wavelengths, which is in contrast to the expected trend of monotonic decrease with increasing wavelength. As a comparison, the measured electron collision times are smaller than 1 fs and decrease with increasing wavelength. This is attributed mainly to hotter electron generation in longer-wavelength filaments via ponderomotive heating. Most importantly, the observed electron collision times create an anomalous regime of light defocusing under plasma: longer wavelengths have less plasma defocusing. Our simulation using the measured electron collision times successfully reproduces the measured wavelength scaling of plasma density. Furthermore, the simulations predict that the filament peak intensities in the mid-IR can be comparable to those in the near-IR, suggesting enhanced HHG and attosecond pulse generation in solids, using mid-IR filaments. Although our wavelength-scaled study is performed in laser filamentation, the analysis presented here suggests that the electron collision time is an important parameter in all fields of laser-plasma interactions, since it can critically affect the dielectric constant of plasma and collisional ionization. Therefore, both plasma density and electron collision time should be systematically investigated to precisely understand strong field laser-matter interactions in solids.

Methods

OPA beam characterization. We measure the focused mode size of each wavelength by imaging it onto a pyroelectric camera, using a 10-cm focal length imaging lens. For pulse duration measurements, a second-harmonic generation autocorrelator is used with a BBO crystal and a Si photodiode for $\lambda = 1.2\text{--}1.9$ μm , and an AgGaS₂ crystal and an InGaAs photodiode for $\lambda = 2\text{--}2.3$ μm . The magnitude of chirp is estimated by $C^2 = [t_p^2(C)/t_p^2(0) - 1]/t_p^4(C)$, where $t_p(C)$ is the measured

pulse duration via autocorrelation, and $t_p(0)$ is the transform limited-pulse duration, which is estimated from the measured spectrum at each wavelength. The sign of the chirp is determined by propagating each pulse through fused silica samples of various thicknesses. The chirp is positive for $\lambda = 1.2\text{--}1.7\ \mu\text{m}$, and negative for $\lambda = 1.8\text{--}2.3\ \mu\text{m}$. The experimental parameters for all the wavelengths are provided in Fig. 1d.

Time-resolved single-shot interferometry. The experimental setup (shown in Supplementary Fig. S1a) is composed of a standard pump-probe scheme and spatial interferometry. An OPA pump beam ($1.2\text{--}2.3\ \mu\text{m}$) generates a filament in a 2.5-cm long fused silica sample and a low-energy collimated 800-nm probe beam orthogonally traverses the pump-generated filamentation region. Then, the probe is magnified and collimated using a $10\times$ objective lens and a 50-cm focal length lens ($4\text{-}f$ scheme) and is sent to an interferometer, in which a charge-coupled device camera records an interferogram. To achieve single-shot interferometry measurements, we decrease the pulse repetition rate from 1 kHz to 50 Hz, using synchronized optical choppers in both pump and probe arms. We control the time delay between pump and probe using a motorized translational stage in the probe arm, but we set at near-zero delay, for which we observe the maximum plasma densities near the onset of stable filaments. In addition, filament formation is monitored by a combination of a $4\times$ objective lens and a digital camera located above the fused silica sample. To check the accuracy of our interferometry analysis for extracting plasma densities and electron collision times, we perform simulations using the conditions similar to those in our experiments (see Supplementary Note 10).

Data availability

All relevant data are available from the authors upon request.

Code availability

The computer codes used for data analysis and numerical simulations are available from the corresponding author upon reasonable request.

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Author contributions

G.C.N. and B.S. conceived and designed the study. G.C.N. and D.D. performed the experiments, and analyzed the data under the supervision of B.S. G.C.N. and B.S. performed the numerical modeling and simulations. All the authors wrote the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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