Slow and Fast Light in Plasma Using Optical Wave Mixing

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Phys. Rev. Lett. **126**, 205001 — Published 19 May 2021

DOI: 10.1103/PhysRevLett.126.205001

Slow and fast light in plasma using optical wave mixing

C. Goyon,[1, *] M. R. Edwards,[1] T. Chapman,[1] L. Divol,[1] N. Lemos,[1] G. J. Williams,[1] D. A. Mariscal,[1] D. Turnbull,[2] A. M. Hansen,[2] and P. Michel[1]

[1] *Lawrence Livermore National Laboratory, Livermore, CA, USA*

[2] *University of Rochester Laboratory for Laser Energetics, Rochester, NY, USA*

(Dated: April 12, 2021)

Slow and fast light, or large changes in the group velocity of light, have been observed in a range of optical media, but the fine optical control necessary to induce an observable effect has not been achieved in a plasma. Here, we describe how the ion-acoustic response in a fully-ionized plasma can produce large and measurable changes in the group velocity of light. We show the first experimental demonstration of slow and fast light in a plasma, measuring group velocities between $0.12c$ and $-0.34c$.

Extreme manipulation of the group velocity of light in optical media produces "fast" and "slow" light, where pulses propagate superluminally or slow to an almost complete stop [1, 2]. Both phenomena have been found in a variety of media, including atomic gases [3, 4], photorefractive crystals [5], and optical fibers [6, 7]. The nonlinear optical properties of plasma are important for high-energy laser experiments in high-energy density physics and inertial confinement fusion, where, for example, plasma-mediated energy transfer predicted by linear theory is used to tune implosion symmetry [8, 9]. Additionally, plasma-based replacements for a range of standard optical components allowed the manipulation of light at extreme fluences[10]. Plasma optics include plasma mirrors, now a standard method for improving temporal contrast[11, 12]; plasma gratings, which redirect light or combine multiple laser beams[13–15]; parametric plasma amplifiers for reaching extreme laser intensities [16–18]; and plasma-based manipulation of polarization at high flux[19–22]. Efforts have also been made to manipulate the phase [23] and group [24] velocity of light in plasma, including focusing-driven focal-spot control [25–27], but experimental demonstrations of plasma-driven group-velocity modification have been limited to small variations ($0.005c$), achieved in photon deceleration studies[28, 29]. The fine control over laser and plasma properties required for significant modification of the group velocity via optical wave mixing has kept measurement of fast and slow light in plasma elusive.

In this letter, we report the first experimental demonstration of slow and fast light in plasma with order of magnitude changes in the group velocity. We used wavelength detuning between a pump and a probe laser to control the ion-acoustic plasma response and tailor the refractive index experienced by the probe beam. The linear theory of optical wave mixing in plasma predicts measurable changes in group velocity near the ion-acoustic resonances. The group velocity is sensitive to deviations of the plasma distribution function from an idealized Maxwellian, and our experimental confirmation of the presence of slow light is a more stringent test of the linear theory than would be possible by measuring only gain. Our results characterize the accuracy of linear theory while showing that the measurement of slow light could be used to characterize distribution functions inside plasmas and study kinetic effects.

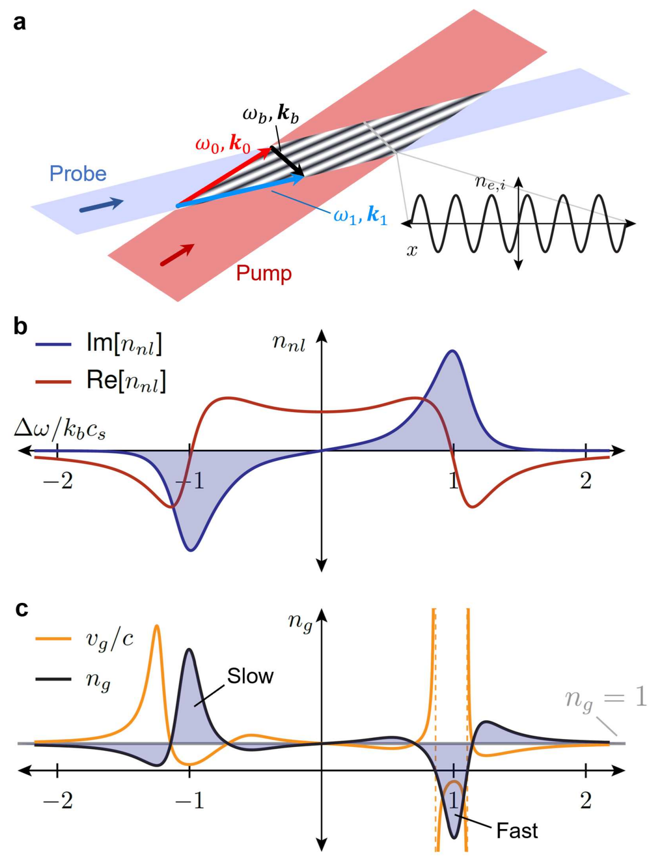

FIG. 1. **Group velocity modification in plasma. a,** Schematic of the wave-mixing arrangement, with inset graph showing ion-wave response. **b,** The real and imaginary components of the nonlinear refractive index as a function of the frequency separation between pump and probe: $\Delta\omega = \omega_1 - \omega_0$. **c,** The corresponding group index and group velocity.

* email: goyon1@llnl.gov

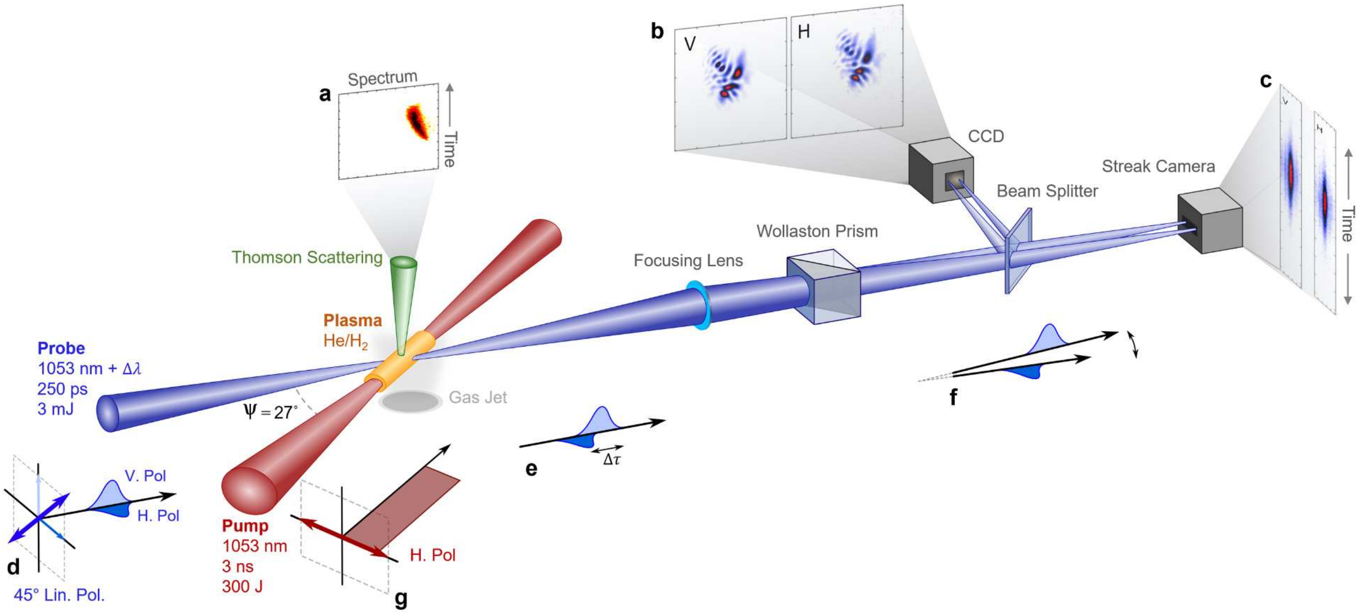

FIG. 2. **Experiment schematic.** The probe and pump beams interact in a plasma formed by the pump. **a,** Thomson scattering of pump beam used to measure density and temperature. **b,** CCD measures probe focal spot and spatial distribution of gain. **c,** Streak camera gives relative timing between probe polarization components and gain. **d,** Incident probe is linearly polarized with H and V components. **e,** Plasma introduces delay in probe H component. **f,** Wollaston prism separates the polarization components. **g,** Pump beam is polarized in the horizontal plane.

The theory of slow and fast light is well-established for gases, crystals and fibers[2]. To derive an equivalent theory for plasma, we note that for small probe amplitude the pump (frequency ω_0, wavenumber k_0) is unaffected by the probe (ω_1, k_1) and that the refractive index (n) experienced by the probe is the sum of linear (n_0) and nonlinear (n_{nl}) contributions:

$$n = n_0 + n_{nl}, \tag{1}$$

where $n_0 = \sqrt{1 - \omega_{pe}^2/\omega_1^2}$ for electron plasma frequency ω_{pe}. The nonlinear component results from the steady-state plasma response to ponderomotive forcing by the pump-probe interference beat wave ($\omega_b = \omega_0 - \omega_1$, $\boldsymbol{k}_b = \boldsymbol{k}_0 - \boldsymbol{k}_1$), as illustrated in Fig. 1a. The non-linear contribution can be expressed in terms of electron and ion susceptibilities (χ_e and χ_i) [19, 30]:

$$n_{nl} = n_0 \left[\frac{\chi_e(1+\chi_i)}{1+\chi_e+\chi_i}\right]^*_{\omega_b,k_b} \frac{k_b^2}{8k_1^2} J_0 \cos^2\psi, \tag{2}$$

where $J_0 = 0.731 \times 10^{-18} I_{0,[\mathrm{W/cm^2}]} \lambda^2_{0,[\mu\mathrm{m}]}$ for vacuum pump wavelength $\lambda_0 = 2\pi/k_0$ and ψ is the angle between $\boldsymbol{k}_0$ and $\boldsymbol{k}_1$. The susceptibilities are given by:

$$\chi_\alpha = \frac{4\pi q_\alpha^2}{k_b^2 m_\alpha} \int \boldsymbol{k}_b \cdot \frac{\partial f_\alpha/\partial \boldsymbol{v}}{\omega_b - \boldsymbol{k}_b\cdot\boldsymbol{v}} d^3v, \tag{3}$$

where q_α, m_α and $f_\alpha(v)$ are the charge, mass, and velocity distribution function of the electrons ($\alpha = e$) or ions ($\alpha = i$). If the velocity distributions are Maxwellian, then the susceptibilities can be expressed in terms of the plasma dispersion function Z,[31] as $\chi_\alpha = -\frac{1}{2}(k_b\lambda_{D\alpha})^{-2} Z'[\omega_b/(\sqrt{2}k_b v_{T_\alpha})]$, where $\lambda_{D\alpha}$ is the Debye length, $Z' = dZ/dv$, and $v_{T\alpha} = \sqrt{T_\alpha/m_\alpha}$ is the thermal velocity with T_α the temperature.

The real and imaginary components of n_{nl}—connected via the Kramers-Kronig relations [32]—are plotted in Fig. 1b. The imaginary part of n_{nl} corresponds to an exponential growth or decay of the probe amplitude along its propagation direction due to energy exchange with the pump. This is referred to as the gain G,

$$G = -2L\frac{\omega_1}{c}\frac{\mathrm{Im}(n_{nl})}{n_0}, \tag{4}$$

where L corresponds to the plasma length over which the pump and probe beams are overlapping. The real part of n_{nl} affects the probe's phase velocity. At the optical resonances, where $\omega_b, \boldsymbol{k}_b$ satisfy the ion-acoustic wave dispersion relation, the imaginary part of n_{nl} (and therefore energy transfer to or from the pump) reaches extrema, and the real part rapidly varies with ω_1 for fixed ω_0. This dependence leads to a strong variation of the group index, defined as $n_g = c/v_g$ (where $v_g = \partial\omega/\partial k$ is the group velocity) and expressed as:

$$n_g = \mathrm{Re}\left[n + \omega_1 \frac{\partial n}{\partial \omega_1}\right]_{\omega_1,k_1}. \tag{5}$$

For ω_b of order $\pm k_b c_s$, where $c_s = \sqrt{Z_i T_e/m_i}$ is the ion sound speed for which Z_i is the ion charge state, the dominant contribution to n_g is the large variation of n_{nl} with ω_1 i.e. $n_g \approx 1 + \mathrm{Re}[\omega_1 \partial n_{nl}/\partial\omega_1]$, which peaks near the optical resonances as shown in Fig. 1c. For an interaction length L through the pump-plasma system, the time delay experienced by the probe compared to its propagation in the absence of interaction (i.e. with $n_{nl} = 0$) is:

$$\Delta\tau = \frac{L}{c}(n_g - 1) \tag{6}$$

where $n_g \gg 1$ or $n_g \ll 1$ will produce slow or fast light, respectively. Direct measurement of $\Delta\tau$ is our primary diagnostic for determining the plasma group index.

To examine this effect experimentally, two Nd:glass beamlines at the Jupiter Laser Facility (Lawrence Livermore National Laboratory) with a tunable wavelength separation ($\Delta\lambda$) of up to ± 4 Å were crossed in a gas jet at a 27° angle. The pump beam provided 300 J at 1053 nm in a rectangular 3 ns pulse spread by a phase plate to a 600 μm speckled focal spot ($\sim 4 \times 10^{13}$ W.cm^{-2}) and the probe beam gave 3 mJ in a 250 ps (FWHM) Gaussian pulse focused to an irregular 200 μm-diameter spot at 1053 nm ($\sim 4 \times 10^{10}$ W.cm^{-2}). The timing of the probe with respect to the pump was controlled to within 120 ps before each shot using a streak camera and measured on-shot with photodiodes upstream from the target chamber.

A 3-mm inner-diameter supersonic gas jet nozzle released an H_2/He mixture (50% H, 50% He by atomic fraction) to form a gas column. The mixture ratio was chosen to control the level of ion wave damping inside the plasma, which sets the width of the resonance peaks in $\mathrm{Im}[n_{nl}]$ (Fig. 1b). Increasing the fraction of hydrogen leads to stronger damping, broadened resonance peaks, and lower gains. It also shortens the transient response of the plasma [33] as a larger fraction of the lighter and faster hydrogen ions are resonant with the ion acoustic waves, enhancing Landau damping. We used the Thomson scattered signal from the pump beam [34, 35] to estimate the electron plasma density and temperature. An achromatic lens imaged a scattering volume ($200 \times 200 \times 600\ \mu\mathrm{m}^3$) through a spectrometer onto a streak camera. The pump entirely ionized the gas, reaching a temperature $T_e = 240 \pm 50$ eV and density $1.1 \pm 0.2 \times 10^{19}$ cm^{-3} by the time the probe arrived, 1.6 ns after the leading edge of the pump.

The pump beam was horizontally polarized (H) in the plane of the interaction (Fig. 2g). The probe beam was linearly polarized near 45°, allowing decomposition into horizontal and vertical (V) components (Fig. 2d). The pump and the horizontal probe component drove a beat-wave plasma response, modifying the propagation of the probe beam H-polarization but leaving the V-polarization unaffected. The collected probe passed through a Wollaston prism, which split the polarization components into two distinct beams. These beams were split in two again by a non-polarizing beamsplitter, with each component of the probe imaged onto both a charge-coupled device (CCD) and a streak camera. The relative delay between the probe polarization components imparted by the plasma was measured using a single streak camera, allowing picosecond precision. The exponential gain resulting from the imaginary component of n_{nl} was measured by both the streak camera and the CCD.

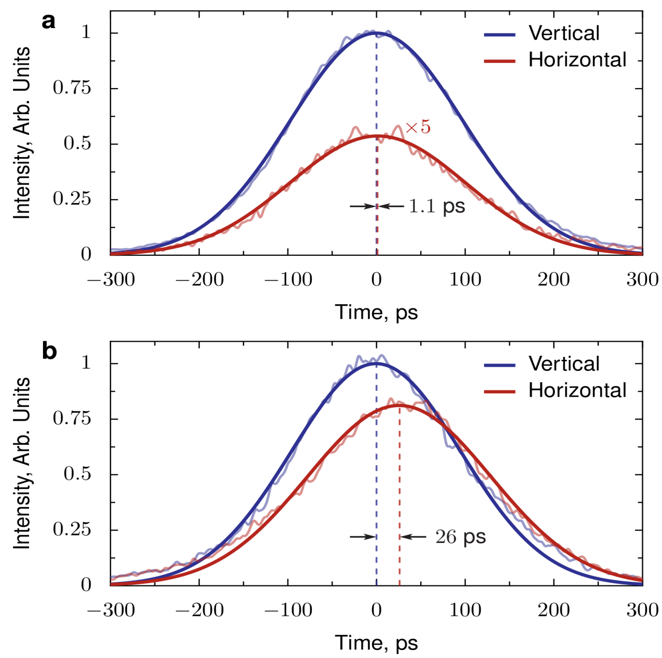

FIG. 3. **Time-resolved probe envelope. a,** Streak camera traces of the incident probe horizontal and vertical polarization components, measured without plasma or pump. **b**, Corresponding traces for both components after transmission through a pump-plasma system with probe detuning near the gain resonance leading to slowing of the horizontal component. A Gaussian curve is fit to all traces.

The relative delay ($\Delta\tau$) between the two components was found from the difference in maxima of Gaussian fits to each component, as shown in Fig. 3, and could be measured with an uncertainty of 3.4 ps. For each shot, one or more low-energy probe-only measurements were taken without plasma to capture the intrinsic delay of the measurement line and the relative strength of the probe polarization components; delay and gain were then found by comparing the full-energy with-plasma data to the probe-only measurements. The 250-ps Gaussian envelope of the probe laser was not strongly distorted under either high gain or high extinction conditions. The experimental gains reported in this paper are defined as the natural logarithm of the change in pulse energy; the streak camera and CCD values agreed with each other.

The gain (Fig. 4a) and delay (Fig. 4b) curves were mapped by varying the wavelength detuning between the pump and probe ($\Delta\lambda = \lambda_1 - \lambda_0$); under these plasma and beam conditions both the slow and fast light interactions could be captured within the $\Delta\lambda = \pm 4$ Å facility capability. Both slow and fast light were successfully measured. As anticipated, the delay varies rapidly with $\Delta\lambda$ near resonance (most positive gain) and anti-resonance

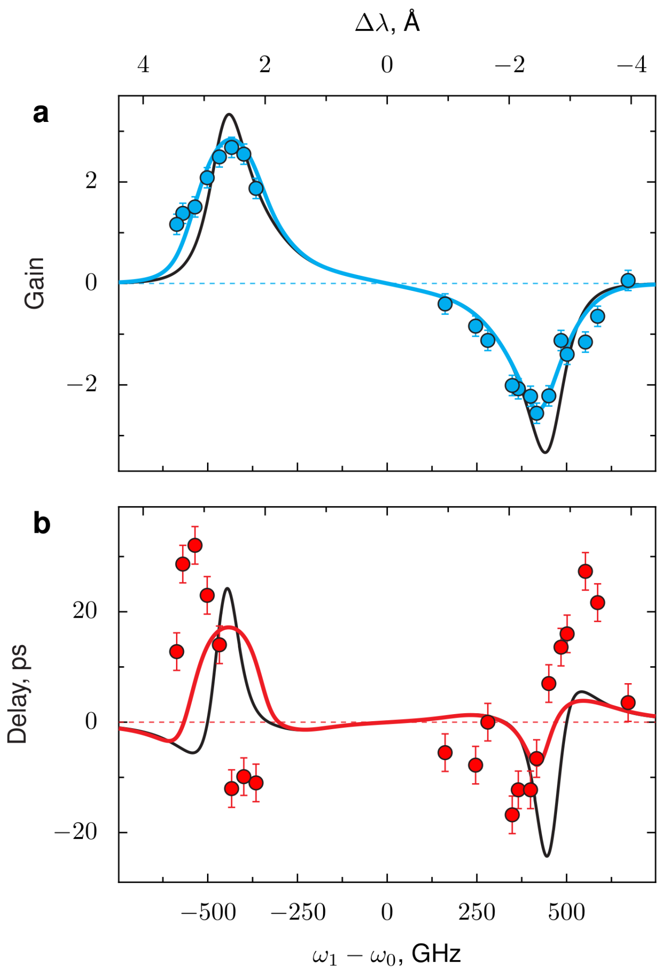

FIG. 4. **Experimental measurement of slow and fast light compared to theory.** **a,** The measured gain (blue circles) compared to linear theory calculations with (blue) and without (black) compensation for the finite f-number of the pump ($f/6.7$) and probe ($f/10$) for both the slow light ($\Delta\lambda <$ 0) and fast light ($\Delta\lambda < 0$) sides. **b**, The measured pulse delay compared to linear theory calculations with (red) and without (black) compensation for the finite f-number. For both plots, the theory curves are calculated for $n_e = 1.25 \times 10^{19}$ cm^{-3}, $T_e = 256$ eV, $T_i = 45$ eV, and $I_0 = 3.25 \times 10^{13}$ W/cm^2.

(most negative gain). The measured maximum delay of 32 ps corresponds to a group velocity of $\sim 0.12c$ along the 1.3 mm plasma length. The minimum delay of -17 ps corresponds to a group velocity of -0.34c. The negative group velocity indicates that the peak of the input pulse appears to travel through the plasma faster than the speed of light and is sometimes referred to as backward propagation. However, the information contained in the pulse still propagates slower than c (i.e. an input square pulse would be distorted by the interaction). [36]

The experimental results show clear variation of the probe seed group velocity with $\Delta\lambda$. In Fig. 4a, comparisons of the measured gain to two theory curves are made: i) the gain G of a pump and probe interaction calculated using $\psi = 27°$ (see caption for other parameters) given by Eq. 4, and ii) the gain $\bar{G} = \ln(\langle\exp(G)\rangle)$ for which the finite f-numbers of the pump and probe are accounted for using an approach similar to Ref. 37: $\langle \ldots \rangle$ denotes averaging over all ψ, where the wave vectors are sampled from a uniform near-field distribution determined by the f-numbers of the circular apertures. The theory curves assume that the distribution functions f_α of all plasma species α remain Maxwellian, free of flows, and homogeneous across the pump-probe interaction region. As in prior work[22], the gain measured in experiments is in excellent agreement with established linear theory.

Similarly, in Fig. 4b, comparisons of the measured delay to two theory curves are made: i) the delay $\Delta\tau$ given by Eq. 6, calculated using $\psi = 27°$, and ii) the gain-weighted delay $\Delta\bar{\tau} = \langle\exp(G)\Delta\tau\rangle/\langle\exp(G)\rangle$. The goal of the gain-weighting is to account for the multiple angles contained in the finite f-number of each beam. The higher (or less negative) gain will dominate the experimentally measured delay. For instance, the magnitude of maximum delay for fast light (lowest gain) is reduced in the gain-weighted case showing a better agreement with experimental results.

This work advances our understanding of the limitations and accuracy of the linear theory. Indeed, the differences between the predicted and measured locations and magnitudes of the delay peaks are likely dominated by the high sensitivity of the group index to the details of the distribution functions f_α for each species α. From Eq. 4, the gain is dependent on the imaginary part of n_{nl}. In contrast, $\Delta\tau$ depends on the real part of the derivative of n_{nl}, per Eq. 5. Since n_{nl} depends on $\partial f_\alpha/\partial v$ via the susceptibilities χ_α (see Eq. 3), $\Delta\tau$ ultimately depends on $\partial^2 f_\alpha/\partial v^2$, so it is more sensitive to deviations of f_α from a Maxwellian than G. The sensitivity of $\Delta\tau$ to $\partial^2 f_\alpha/\partial v^2$ is somewhat unusual; measurable quantities typically depend to a large degree on f_α or $\partial f_\alpha/\partial v$, the latter determining Landau damping and therefore the response of the employed Thomson scattering diagnostic. A similar dependence to the distribution function has been exploited to evidence ion-trapping frequency shift[38]. In our case, several mechanisms such as inter-species drift (including heat transport), bulk flows, and inhomogeneity may all contribute to the observed $\Delta\tau$ exceeding theoretical estimates for the magnitude and the larger than expected shift between the peaks of G and $\Delta\tau$. Indeed, the gain-weighting of the theoretical time delay performed here is only a lowest-order approximation to an interaction that is inherently three-dimensional. Although beyond the scope of what we present here, more precise future experiments and large-scale three-dimensional simulations could provide insight into these details of the plasma response.

In summary, we have manipulated the group velocity of light in a plasma using optical wave mixing between a pump, a probe, and the plasma, leading to demonstrations of both slow and fast light. By varying the wavelength separation between the pump and probe beams, we have adjusted the group velocity of a 250 ps pulse between 0.12c and -0.34c. Precise manipulation of light in plasma is challenging but nonetheless necessary for a rapidly expanding number of applications, from inertial confinement fusion and laboratory astrophysics to laser-plasma particle accelerators and plasma optics for high-power lasers. The measurement of slow and fast light in plasma is a demonstration of an exquisite degree of control over a laser-plasma system, but it also suggests that higher-precision characterization of high-energy-density light-plasma interactions will require methods outside the standard toolbox of plasma diagnostics. For example, we have shown that despite the close agreement of gain measurements with expectations, experimental group velocity measurements differ from theoretical predictions. These quantitative differences hint at kinetic effects and additional complexity in the plasma distribution function. Further experiments focused on quantifying these effects could provide an additional constraint on the properties of a plasma. Group velocity measurements might therefore become one component of a suite of new plasma diagnostics for probing and controlling the subtleties of non-Maxwellian plasma distributions, offering a future where plasma is available as an optical medium with the flexibility and precision of crystals or glass.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344, and was funded by the Laboratory Research and Development Program at LLNL under Project Tracking Codes No. 18-ERD-046 and No. 20-ERD-057.

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