Indium arsenide phosphide (InAsP) nanowires (NWs), a member of the III–V semiconductor family, have been used in various photonic and optoelectronic applications thanks to their unique electrical and optical properties such as high carrier mobility and adjustable band gap. In this work, we synthesize InAsP NWs and further explore their nonlinear optical properties. The ultrafast carrier dynamics and nonlinear optical response are thoroughly studied based on the nondegenerate pump probe and Z-scan experimental measurements. Two different characteristic carrier lifetimes (∼2 and ∼15 ps) from InAsP NWs are observed during the excited-carrier relaxation process. Based on the physical model analysis, the relaxation process can be ascribed to the carrier cooling process via carrier-phonon scattering and Auger recombination. In addition, based on the measured excited-carrier lifetime and Pauli-blocking principle, we discover that InAsP NWs show strong saturable absorption properties at the wavelengths of 532 and 1064 nm. Last, we demonstrate for the first time a femtosecond (∼426 fs) solid-state laser based on an InAsP NWs saturable absorber at 1.04 μm. We believe that our work provides a better understanding of the InAsP NWs optical properties and will further advance their photonic applications in the near-infrared range. © 2021 Chinese Laser Press

1. INTRODUCTION

In the past two decades, nanowires (NWs) have been employed as versatile building blocks [1–3] used in various optoelectrical devices [4–7] because of their tailored optical and electronic properties [8,9]. Semiconductor NWs exhibit many unique physical characteristics such as significant surface and size effects [10], building blocks for nanoelectronics [7], and Majorana fermions [11]. For example, the electrical and optical properties of semiconductor NWs can be tuned by controlling their sizes, shapes, and compositions [12,13]. For instance, alloyed III–V semiconductor ternary GaAs/InGaAs nanowires show continuous tuning of the bandgap (0.91–1.52 eV) by adjusting the ratio of In and Ga elements [12]. In addition, the bandgap of semiconductor NWs decreases with increasing size [13]. Thus, semiconductor NWs are useful and multifunctional nanomaterials working as various building blocks to advance the research of photonic and optoelectronic devices, such as solar cells [1,14], lasers [2,15], and photodetectors [3,16].

Owing to the large electron g-factor and small electron effective mass [17–19], III–V semiconductor NWs have been applied in nanoscale optoelectronic devices [17], infrared detectors [18], and spin electronics [19]. Known as the representative of the III–V semiconductor family, InAsP NWs which were first synthesized by Pettersson in 2006 [3] show great potential for infrared photodetectors due to the high carrier mobility. Since then, great efforts have been devoted to pursuing InAsP NW-based optoelectronic devices, such as high-speed electronics [20,21] and near-infrared light emitters and detectors [22–24]. In theory, the bandgap of InAsP NWs can be tailored from 0.35 to 1.35 eV by adjusting the alloy composition, covering the important telecommunication wavelength band from 1.3 to 1.55 μm [25]. Besides, the growth of InAsP NWs has larger tolerance for lattice mismatch than thin-film epitaxial growth, which allows adjusting the optical nonlinear sensitivity through external bending or twisting strain [26,27] and has more flexibility in substrate selection and mechanical properties. Thus, there is a demand to study the intrinsic optical properties (e.g., carrier dynamics, nonlinear optical absorption) of InAsP NWs in the near-infrared wavelength range, which remains unexplored.

In this work, we fabricate high-quality InAsP NWs by directly growing them on a quartz substrate using the Au
nanoparticle-assisted vapor-liquid-solid (VLS) method. Then, the carrier dynamics of InAsP NWs is investigated by non-degenerate pump-probe measurements, which show that the excited carrier in InAsP NWs exhibits one fast (∼2 ps) and one slow (∼15 ps) relaxation processes. Furthermore, we study the nonlinear absorption properties of InAsP NWs by tuning the light intensity and wavelength using the Z-scan technique. We find that InAsP NWs have a large nonlinear absorption coefficient ($\beta_{\text{eff}} \sim 10^2 \text{cm/MW}$) and positive nonlinear refractive index ($n_2 \sim 10^{-13} \text{m}^2/\text{W}$), which is much higher than the reported values of other nanomaterials including graphene, transition metal dichalcogenides (TMDs), and black phosphorus (BP). The studies on the ultrafast carrier relaxation dynamics and nonlinear optical absorption properties show the potential of InAsP NWs in the application field of optical switching. By using InAsP NWs as a saturable absorber (SA), a passively mode-locked laser at 1.04 μm is demonstrated with the shortest pulse width of 426 fs. Our results prove that InAsP NWs could be very promising candidates as excellent nonlinear optical modulators used in photonic and electronic devices.

2. RESULTS AND DISCUSSION

A. Preparation and Characterization of InAsP NWs

InAsP NWs samples are grown on quartz substrates inside a horizontal flow atmospheric pressure metal-organic vapor phase epitaxy (MOVPE) system. The detailed process can be found in Appendix A.1. Scanning electron microscopy (SEM) is used to examine the surface morphology of the prepared NWs. Figure 1(a) depicts the as-grown InAsP NWs that have an average diameter of ∼50 nm with smooth surfaces (shown in the inset). Atomic force microscopy (AFM) image together with the height profile provides further evidence for the uniform formation of InAsP NWs. As shown in Figs. 1(b) and 1(c), the diameter of InAsP NWs is determined to be ∼50 nm (average value), which is consistent with Au particle diameter. What is more, we did the energy-dispersive X-ray spectroscopy (EDX) along the growth direction to study the crystal structure of InAsP NWs, as shown in Fig. 1(d), which illustrates the high quality of our InAsP NWs sample solely consisting of In, As, and P without additional elements and the ratio of In, As, and P elements is about 2:2:1. Figure 1(e) shows the Raman spectrum of the InAsP NWs excited by a 532 nm laser with the power of 2 mW. The peaks observed at 217.6 and 247.8 cm$^{-1}$ are InAs-like modes which correspond to a transverse optical (TO) and a longitudinal optical (LO) phonon modes. The peaks around 298.1 cm$^{-1}$ are InP-like modes corresponding to a transverse optical phonon mode (TO$_2$), which is consistent with the previous research [28]. Photoluminescence (PL) measurement [Fig. 1(f)], performed at room temperature, reveals only one PL peak at 1280 nm, corresponding to the band-to-band transition in bulk form.

B. Ultrafast Carrier Dynamics of InAsP NWs

Pump-probe spectroscopy with femtosecond resolution is employed at room temperature to study the relaxation process of the excited carriers in InAsP NWs. A schematic of the experimental setup is shown in Fig. 2(a) (details in Appendix A.2). Pump light with wavelength of 400 nm is used to stimulate electrons from the ground state to the excited state, while the probing is done at the wavelength of 675 nm. It is worth noting that pump-probe spectroscopy is a nondestructive method, so no damage is induced to the samples. Figure 2(b) shows the photoinduced bleaching signal (positive $\Delta T/T$) under different pump energy intensities (from 47.7 to 197.4 μJ/cm$^2$). The increase in the transmission right after excitation indicates the saturable absorption of the InAsP NWs due to
The values of the short carrier lifetimes (magnitude larger than the duration of the pump light (in ps order) can be ascribed to the carrier cooling process via slow carrier-phonon scattering, which is consistent with the phenomenon observed in graphite [30]. At the same time, the slow carrier lifetimes is consistent with carrier capture by defects via Auger scatterings [31]. Because the defect-assisted Auger recombination is a three-particle effect in which carrier–carrier interactions become increasingly significant at high carrier densities; the lifetime is inversely proportional to the carrier concentration, and as the carrier concentration increases, the defect concentration is constant, so the proportion of this process is gradually decreasing.

The photoinduced carrier dynamics relaxed from the excited states to the valence band is commonly described by a three-term rate equation [32–34]:

$$-\frac{dn}{dt} = \alpha n + \beta n^2 + \gamma n^3,$$

where \(t\) is the relaxation time, \(n\) is the excited carrier density, and \(\alpha\), \(\beta\), and \(\gamma\) are the first, second, and third terms, respectively, which represent first-order Shockley-Read-Hall (SRH) recombination, free-carrier recombination, and Auger recombination. Figure 2(c) plots the maximum bleach signal measured as a function of the pump energy density, which is modeled by the phenomenological expression [35,36]

$$\Delta T/T_{max} = \frac{A n_0}{B n_0 + C},$$

where \(A\), \(B\), and \(C\) are fitting parameters. The initial charge carrier density \(n_0\) can be calculated by \(n_0 = j \alpha_0\), where \(\alpha_0\) is the InAsP NWs absorption coefficient, and \(j\) is the pump fluence (photons per cm²). Therefore, we can correlate differential transmission \(\Delta T/T\) to the density of photogenerated charge carriers using the above expression.

![Fig. 2](image)

(a) Experimental setup of the nondegenerate pump-probe measurement. (b) Differential transmission of InAsP NWs at different pump pulse energies with a 675 nm probe laser. (c) Relationship between maximum differential transmission and initial photoinduced carrier density. (d) Linear fit to \((n_0/n)^2\) as a function of pump-probe delay time and initial photoinduced carrier density.

The Pauli-blocking effect. To further analyze the carrier dynamics, we employ a biexponential decay model [29]:

$$\frac{\Delta T}{T_0}(t) = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2},$$

where \(\tau_1\) and \(\tau_2\) (\(\tau_1 < \tau_2\)) represent the carrier lifetimes, and \(A_1\) and \(A_2\) are the corresponding relative amplitudes. Good fit to experimental data shown in Fig. 2(b) is obtained with Eq. (1), and the fitted parameters are listed in Table 1. Two clearly different characteristic carrier lifetimes \(\tau_1\) and \(\tau_2\) are identified. It is worth noting that their time scale is about two orders of magnitude larger than the duration of the pump light (\(\sim 150\) fs). The values of the short carrier lifetimes \(\tau_1\) are \(\sim 2\) ps, independent of the pump intensity. Such ultrafast relaxation time (in ps order) can be ascribed to the carrier cooling process via carrier-phonon scattering, which is consistent with the phenomenon observed in graphite [30]. At the same time, the slow time scale (\(\sim 20\) ps) decreases with the increase of the pump intensity, and the pump fluence dependence of the measured.

### Table 1. Fitted Parameters on the Carrier Relaxation Time of InAsP NWs from Eq. (1)

<table>
<thead>
<tr>
<th>Pump Energy ((\mu J/cm^2))</th>
<th>(\tau_1) (ps)</th>
<th>(\tau_2) (ps)</th>
<th>(A_1)</th>
<th>(A_2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>47.7</td>
<td>1.89 ± 0.93</td>
<td>19.27 ± 0.89</td>
<td>0.192</td>
<td>0.803</td>
</tr>
<tr>
<td>79.6</td>
<td>2.53 ± 1.65</td>
<td>11.17 ± 0.67</td>
<td>0.145</td>
<td>0.855</td>
</tr>
<tr>
<td>127.3</td>
<td>2.26 ± 0.65</td>
<td>10.79 ± 0.49</td>
<td>0.228</td>
<td>0.772</td>
</tr>
<tr>
<td>175.1</td>
<td>1.77 ± 0.59</td>
<td>10.12 ± 0.55</td>
<td>0.250</td>
<td>0.750</td>
</tr>
<tr>
<td>197.4</td>
<td>2.65 ± 1.20</td>
<td>7.95 ± 0.78</td>
<td>0.252</td>
<td>0.748</td>
</tr>
</tbody>
</table>
After converting the measured $\Delta T / T$ signal into photogenerated carrier density, the third-order rate equation can be obtained by eliminating the first- and second-order terms in Eq. (2) and assuming the numbers of photogenerated electrons and holes are equal, which can be expressed as follows:

$$\frac{n_0^2}{n_t^2} - 1 = k n_0^2 t,$$

where $k$ is the third-order rate constant for the recombination process. By linear fitting of Eq. (4), we obtain the initial photoinduced carrier density ($n_0$) of $13 \times 10^{18}$ cm$^{-3}$, as shown in Fig. 2(d). Then the Auger recombination rate of $9.5 \pm 0.2) \times 10^{-27}$ cm$^6$ s$^{-1}$ is obtained.

### C. Nonlinear Optical Response of InAsP NWs

The open-aperture (OA) and closed-aperture (CA) Z-scan techniques are effective ways to investigate the nonlinear optical (NLO) responses of the InAsP NWs through the measurement of their nonlinear absorption and refraction. The details are given in Appendix A.3. The intensity-dependent normalized transmittance as a function of $Z$ position at 532 and 1064 nm is shown in Figs. 3(a) and 3(c). For a certain incident laser energy, the transmittance curve is symmetrical at the focus point which has a Gaussian-like shape, arising from the saturation absorption of the InAsP NWs. To obtain the saturable absorption phenomena, various lenses with different focus lengths are used for the different excitation lasers. Typically, the normalized transmittance for InAsP NWs can be written as [37]

$$T(Z) = \sum_{n=0}^{\infty} (-\beta \text{eff} I_0 L_\text{eff})^n / \left(1 + \frac{Z^2}{Z_0^2}\right)^n (n + 1)^{3/2},$$

$$= 1 - \beta \text{eff} I_0 L_\text{eff} / 2 \left(1 + \frac{Z^2}{Z_0^2}\right),$$

where $Z_0$ and $I_0$ are the Rayleigh length and the peak on-axis intensity at the focus ($Z = 0$), respectively. $L_\text{eff}$ and $\beta \text{eff}$ are the effective thickness and effective nonlinear absorption coefficient of the InAsP NWs, respectively. $\beta \text{eff}$ can be obtained by fitting the OA Z-scan measurement data with different incident pulse energies. The results show that $\beta \text{eff} = -207 \pm 2.3$ and $-144 \pm 0.7$ cm/MW at 532 and 1064 nm, separately. It is worth noting that the value of $\beta \text{eff}$ remains constant for different incident energies, which indicates that the nonlinear absorption process of InAsP NWs is dominated by the single-photon saturable absorption. The values of $\beta \text{eff}$ are much larger than those of the most 2D materials such as graphene [38], transition metal dichalcogenides (TMDs) [38,39], black phosphorus (BP) [40], and MXenes [41], as shown in Table 2. In addition, it is also comparable to our previous results on InAs NWs [42], indicating a strong saturable absorption capability. The imaginary part of the third-order nonlinear optical susceptibility ($\text{Im} \chi^{(3)}$) can be calculated with the following formula [41]:

$$\text{Im} \chi^{(3)} = \frac{2 e_0 c^2 n_1^3}{3 \omega} \beta \text{eff},$$

where $e_0$ is the vacuum permeability, $c$ is the vacuum light speed, $\omega$ is the angular frequency, and $n_1$ is the linear refractive index.

---

**Fig. 3.** Characterization of the NLO properties of the InAsP NWs. OA Z-scan measurements of the InAsP NWs at (a) 532 nm and (c) 1064 nm. CA Z-scan measurements of the InAsP NWs at (b) 532 nm and (d) 1064 nm.
MXenes (

3

tion intensities

σ

ratively. To further characterize the saturable absorption properties of InAsP NWs, the optical transmittance as a function of the incident laser intensity is fitted based on a two-level system (see Appendix C). The values of \( I_s \) and \( \Delta R \) for different wavelengths are summarized in Table 3 by fitting the experimental data. The highest modulation depths are determined to be 33.7% ± 0.2% and 19.2% ± 0.2%, while the average saturation intensities \( I_s \) are 0.8 ± 0.02 and 0.25 ± 0.008 GW/cm² at 532 and 1064 nm, respectively. The saturation intensity is inversely proportional to the wavelength that is in agreement with the two-level energy system model (\( I_s = h\nu/\sigma_r \)), where \( \sigma \) is the absorption cross section, \( h\nu \) is the photon energy, and \( \tau \) is the decay time. The observed saturation intensities of the InAsP NWs saturable absorber are lower than that based on graphene (170 ± 51 GW/cm²) [38], TMDs (114 ± 63 GW/cm²) [38,39], BP (459 GW/cm²) [40], and MXenes (69.3 GW/cm²) [41], which is very promising for mode-locking process.

The CA Z-scan technique is used to characterize the refractive index \( (n_2) \) of the sample. The normalized transmittances of the closed aperture Z-scan results under different incident energies at the wavelength of 532 and 1064 nm are shown in Figs. 3(b) and 3(d). The normalized transmittance can be fitted using the following formula [43]:

\[
T = 1 + \frac{4kL_\text{eff} n_2 I_0 Z}{Z_0 (Z^2/Z_0^2 + 9) (Z^2/Z_0^2 + 1)},
\]

where \( k = 2\pi/\lambda \) and \( n_2 \) is the nonlinear refractive index. The valley–peak shapes of the normalized transmittance CA/OA Z-scan curves, which result from the Kerr effect induced self-focusing effect in InAsP NWs, suggest the positive refractive index. The obtained nonlinear refractive index is 2.39 ± 0.03 and 2.73 ± 0.02 × 10⁻¹³ m²/W at 532 and 1064 nm. The real part of the third-order nonlinear optical susceptibility (\( \chi^{(3)} \)) can be expressed as [40]

\[
\chi^{(3)} = \frac{4\varepsilon_0 c m_2}{3} n_2.
\]

The \( \chi^{(3)} \) is \( (7.17 ± 0.01) \times 10⁻⁷ \) esu (at 532 nm) and \( (8.2 ± 0.01) \times 10⁻⁷ \) esu (at 1064 nm). As listed in Table 2, \( n_2 \) of InAsP NWs is slightly higher than that of typical 2D materials, such as graphene [35], TMDs [35,36], and MXene [38].

### D. Ultrafast Photonic Applications

Because InAsP NWs exhibit strong third-order nonlinear optical response and ultrafast saturation recovery time, it is meaningful to further explore their ability on the generation of ultrashort pulses. Here, a mode-locked solid-state laser based on InAsP NWs saturable absorber is assembled. Based on the mode-locking theory, when the mode-locking pulse energy is larger than the minimum intracavity pulse energy, the stable continuous-wave (CW) laser can be demonstrated. Therefore, considering the SA parameters, the following formula should be satisfied [41]:

\[
F_{\text{sat,} A} \Delta R < \frac{(PT)^2}{F_{\text{sat,} A} A_{\text{eff,} A} A_{\text{eff,} L}} = \frac{(PT)^2}{hc} \times \frac{\pi \omega_{\text{eff,} A}^2}{\pi \omega_{\text{eff,} L}^2} \times \frac{\sigma_{\text{em,} L} \lambda}{4\varepsilon_0 c m_2},
\]

where \( P \) is the intracavity pulsed laser power, \( T \) is the round-trip time, and \( F_{\text{sat,} A} \) and \( F_{\text{sat,} L} \) are the saturation fluence of SA and laser gain medium, respectively. \( A_{\text{eff,} L} \) and \( A_{\text{eff,} A} \) are the effective laser mode on the laser gain medium and SA, respectively, \( b \) is the Planck constant, \( c \) is the light velocity, \( \sigma_{\text{em,} L} \) is the emission cross section of the laser crystal, \( \omega_{\text{eff,} L} \) and \( \omega_{\text{eff,} A} \) are the effective laser modes radii on the laser gain medium and SA, respectively, and \( m \) is a cavity constant; \( m = 1 \) for a ring cavity, and \( m = 2 \) for a linear cavity. Based on ABCD propagation matrix, a 3.5 m long Z-type resonator is designed. The setup details are given in Appendix A.4. Here, the \( \omega_{\text{eff,} L} \) and \( \omega_{\text{eff,} A} \) are calculated as 37 and 54 μm, respectively. The left-hand side of Eq. (9), \( F_{\text{sat,} A} \Delta R \), is calculated to be 2.5 μJ/cm², which is smaller than the right-hand side (11.2 μJ/cm²). Therefore, based on the as-designed and the as-prepared InAsP NWs SA, stable CW mode-locked lasers can be obtained.

When the absorbed pump power exceeds 4.26 W, stable CW mode-locked (CWML) operation is established. Figure 4(a) depicts the relationship between the absorbed pump power and average output power. The obtained maximum average output power is 333 mW under the absorbed pump power of 7.17 W. Once the absorbed pump power exceeds 7.17 W, the CWML operation is broken. The output power instabilities (RMS) are measured to be less than 2% over 1 h. By sech² pulse shape fitting listed in Fig. 4(b), the pulse duration is measured to be 426 fs. The inset shows that the spectrum is centered at 1043 nm with a full-width-at-half-maximum (FWHM) of 3.1 nm, corresponding to the time-bandwidth product of 0.364, which is higher than the Fourier transform-limited value (0.315). It indicates that the output pulses are slightly chirped. Also, the radio-frequency spectra are recorded with different spans. As shown in Fig. 4(c), the fundamental peak is located at 42.93 MHz with

### Table 2. Comparison of \( \beta_{\text{eff}} \) and \( n_2 \) Values between InAsP NWs and Other Nanomaterials

<table>
<thead>
<tr>
<th>Materials</th>
<th>Laser Parameters</th>
<th>( \beta_{\text{eff}} ) (cm/GW)</th>
<th>( n_2 ) (m²/W)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Graphene</td>
<td>1030 nm, 1 kHz, 340 fs</td>
<td>(-19.27 ± 0.89 \times 10^{-2})</td>
<td>(-13.7 \times 10^{-16})</td>
<td>[38]</td>
</tr>
<tr>
<td>MoS₂</td>
<td>532 nm, 10 kHz, 100 ps</td>
<td>(-26.2 ± 8.8)</td>
<td>(-2.5 ± 1.2 \times 10^{-16})</td>
<td>[38]</td>
</tr>
<tr>
<td>WS₂</td>
<td>1064 nm, 20 Hz, 25 ps</td>
<td>(-5.1 ± 0.26)</td>
<td>((5.83 ± 0.18) \times 10^{-15})</td>
<td>[39]</td>
</tr>
<tr>
<td>BP</td>
<td>800 nm, 10 kHz, 100 fs</td>
<td>(-3.8 \times 10^{-2})</td>
<td>—</td>
<td>[40]</td>
</tr>
<tr>
<td>MXene</td>
<td>800 nm, 1 kHz, 95 fs</td>
<td>(-0.07)</td>
<td>(-3.47 \times 10^{-20})</td>
<td>[41]</td>
</tr>
<tr>
<td>InAs NWs</td>
<td>1064 nm, 50 kHz, 100 ns</td>
<td>—</td>
<td>—</td>
<td>[42]</td>
</tr>
<tr>
<td>InAsP NWs</td>
<td>532 nm, 200 kHz, 10 ps</td>
<td>(-2.07 ± 0.02 \times 10^{3})</td>
<td>((2.39 ± 0.03) \times 10^{-13})</td>
<td>This work</td>
</tr>
<tr>
<td>InAsP NWs</td>
<td>1064 nm, 200 kHz, 10 ps</td>
<td>(-1.44 ± 0.01 \times 10^{2})</td>
<td>((2.73 ± 0.02) \times 10^{-13})</td>
<td>This work</td>
</tr>
</tbody>
</table>
a signal-to-noise ratio of 60 dB. No spurious frequency modulations are observed in a wide span of 1 GHz with a resolution bandwidth (RBW) of 75 kHz, which proves clean CWML operation based on InAsP NWs. What is more, the CWML pulse trains show good amplitude stability with a time span of 2 ms [Fig. 4(d)]. The mode-locked laser based on InAsP NWs shows a relatively short pulse width and relatively high output power, which indicates the InAsP NWs are promising SA for the ultrafast laser generation.

3. CONCLUSIONS

In summary, we fabricate InAsP NWs by using an Au nanoparticle-assisted VLS growth method. The NLO properties and ultrafast carrier dynamics of InAsP NWs have been studied by Z-scan and nondegenerate pump-probe measurements for the first time. The excited carriers of InAsP NWs exhibit two characteristic carrier lifetimes (fast ∼2 ps and slow ∼15 ps), which can be attributed to a carrier cooling process via carrier-phonon scattering and an Auger recombination process. Besides, Z-scan measurements demonstrate that InAsP NWs have excellent NLO properties with a large nonlinear absorption coefficient of ∼10^2 cm/MW and nonlinear refractive index of ∼10^{-13} m^2/W. Furthermore, we successfully demonstrate the femtosecond mode-locked solid-state laser based on InAsP NWs SA. Our work indicates that InAsP NWs are excellent candidates for applications in photonic and electronic devices.

APPENDIX A: EXPERIMENTAL METHODS

1. Growth of InAsP NWs

InAsP NWs were grown on quartz substrates inside a horizontal flow atmospheric pressure metal-organic vapor phase epitaxy (MOVPE) system using an Au nanoparticle-assisted vapor-liquid-solid (VLS) growth method. Trimethylindium (TMIn), tertiarybutylarsene (TBAs) and tertiarybutylphosphine (TBP) were used as precursors. First, the substrates were cleaned in an ultrasonic bath with isopropanol (IPA) and acetone, rinsed in deionized water, and then treated with a poly-L-lysine (PLL) solution for 120 s. Next, the surface was treated with 40 nm diameter colloidal gold (Au) nanoparticles solution (BBI International, UK) for 120 s. Prior to the growth, the substrates were annealed in situ at 600°C for 10 min under hydrogen flow to desorb surface contaminants. The NW growth temperature was fixed at 410°C for 5 min with the TMIn, TBAs, and TBP flows of 2.8, 14.4, and 1276 μmol/min, respectively. The nominal V/III ratio during the growth was ∼460. After the growth, the TBP source kept on until the reactor cooled down to 250°C to protect the NW surface from desorption. Hydrogen was used as a carrier gas, and the total reactor gas flow rate was ∼5 L/min. The growth temperatures reported in this work are thermocouple readings of the
lamp-heated graphite susceptor, which are slightly higher than the real substrate surface temperature. The structural analysis of the NWs was performed using scanning electron microscopy (SEM) (Zeiss Supra 40).

2. Nondegenerate Micro Pump-Probe Measurements

The setup was established using Ti:sapphire oscillators (800 nm, 80 MHz, 150 fs), separated to two components. One beam was used to drive the optical parametric oscillator to generate the pulses from 1000 to 1500 nm. After frequency doubling through a BBO crystal, the wavelength of the beam was changed to 500–750 nm. The wavelength of 650 nm was used as the probe light. The other beam was fixed at 400 nm and was used as the pump light. Both the pump and probe light were focused onto the sample by a 20× objective lens, and the pump fluence was 20 times higher than the probe fluence. The CCD camera (Thorlabs, DCC1545M) was used to image the sample and to check that the centers of pump and probe light coincide. The pump-induced change of the probe was detected by an adjustable gain balanced photoreceiver (Newport, 2317NF). The chopper frequency was fixed to 1 kHz for the spectral range of 1000–1100 nm via a PMMA-mediated method. Yb³⁺-ion-doped (5%, atom fraction) Yb:KGW crystal (35 × 3 mm × 2 mm) was used as the gain medium. It was wrapped in indium foil and cooled by running water at a temperature of 18°C to accelerate thermal conduction. The pump source was a 27 W fiber-coupled laser diode (numerical aperture of 0.22, core diameter of 105 μm) emitting at 976 nm, which was focused into the laser gain medium via a 1.8:1 coupling optics system with a diameter of 58 μm and well matched with the diameter of the TEM₀₁ cavity mode (74 μm) calculated by ABCD propagation matrix theory. A two-plane Gires-Tournois interferometer (GTI) mirror was used to compensate the intracavity group delay dispersion (GDD), which provided a total GDD of ~ −800 fs² per round. We used a plane OC with a transmittance of 1% for the spectral range of 1000–1100 nm.

APPENDIX B: THE FITTING PARAMETERS OBTAINED FROM Z-SCAN CHARACTERIZATION

As shown in Table 3, it summarizes the fitting parameters obtained from Z-scan characterization of InAsP NWs at the wavelength of 532 and 1064 nm, which shows the strong saturable absorption properties of InAsP NWs.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Input Energy (μJ)</th>
<th>β_eff (cm/MW)</th>
<th>Imχ(3) (x10⁻⁷ esu)</th>
<th>I₀ (GW/cm²)</th>
<th>ΔR (x10⁻¹³ m²/W)</th>
<th>n₂ (x10⁻⁷ esu)</th>
<th>Reχ(3) (x10⁻⁷ esu)</th>
</tr>
</thead>
<tbody>
<tr>
<td>532</td>
<td>0.25</td>
<td>-207.4 ± 2.3</td>
<td>-2.63 ± 0.03</td>
<td>0.8 ± 0.02</td>
<td>0.337 ± 0.002</td>
<td>2.39 ± 0.03</td>
<td>7.17 ± 0.01</td>
</tr>
<tr>
<td>1064</td>
<td>0.16</td>
<td>-205.4 ± 1.7</td>
<td>-2.62 ± 0.04</td>
<td>0.81 ± 0.04</td>
<td>0.322 ± 0.002</td>
<td>2.46 ± 0.02</td>
<td>7.38 ± 0.01</td>
</tr>
<tr>
<td></td>
<td>0.055</td>
<td>-203.4 ± 2.8</td>
<td>-2.59 ± 0.04</td>
<td>0.78 ± 0.05</td>
<td>0.324 ± 0.002</td>
<td>2.44 ± 0.03</td>
<td>7.32 ± 0.01</td>
</tr>
<tr>
<td></td>
<td>1.01</td>
<td>-144 ± 0.7</td>
<td>-3.7 ± 0.02</td>
<td>0.25 ± 0.008</td>
<td>0.192 ± 0.002</td>
<td>2.73 ± 0.02</td>
<td>8.2 ± 0.01</td>
</tr>
<tr>
<td></td>
<td>0.55</td>
<td>-146 ± 0.9</td>
<td>-3.7 ± 0.03</td>
<td>0.24 ± 0.006</td>
<td>0.148 ± 0.002</td>
<td>2.75 ± 0.03</td>
<td>8.3 ± 0.01</td>
</tr>
<tr>
<td></td>
<td>0.33</td>
<td>-142 ± 1.1</td>
<td>-3.6 ± 0.03</td>
<td>0.24 ± 0.006</td>
<td>0.105 ± 0.002</td>
<td>2.76 ± 0.02</td>
<td>8.3 ± 0.01</td>
</tr>
</tbody>
</table>

3. Z-scan Measurement

Z-scan measurements were performed using a homemade mode-locked Yb fiber laser (center wavelength 1064 nm, repetition rate 100 kHz–1 MHz, pulse duration 10 ps), which can generate a signal with the wavelength of 532 nm by doubling the frequency through a BBO crystal. The pulses were divided to two parts: one was set as a reference light, collected by a power meter (Thorlabs S470C); the other was focused by a lens into the samples. Lenses of different focusing lengths were used for different wavelength of incident light, f = 50 mm for 532 nm and f = 75 mm for 1064 nm, and the focused beam waist was estimated to be 26 and 52 μm. The sample was fixed to a stepper motor which was controlled by a computer program.

4. CW Mode-Locking Operation

We transferred the as-grown InAsP NWs grown on quartz onto the real substrate surface temperature. The structural analysis of the sample and to check that the centers of pump and probe light coincide. The pump-induced change of the probe was detected by an adjustable gain balanced photoreceiver (Newport, 2317NF). The chopper frequency was fixed to 1 kHz as the reference frequency of the lock-in amplifier. The initial charge carrier density (n₀) can be calculated by the following formula:

\[
n₀ = \frac{\lambda E a₀}{\omega₀^2 \pi \hbar c},
\]

where \( \lambda \) is the wavelength of excitation light, \( E \) is the pump energy fluence, \( \omega₀ \) is the beam waist (~5 μm), \( h \) is the Planck constant, \( c \) is the speed of light in vacuum, and the absorption coefficient (\( a₀ \)) of InAsP NWs is ~1.02 × 10⁵ cm⁻¹.

APPENDIX C: THE NONLINEAR TRANSMITTANCE CURVE OF InAsP NWs

As shown in Fig. 5, the curve of the nonlinear transmittance with respect to the intensity of incident light was obtained by extracting the experimental data of the open-aperture (OA) Z-scan. The basic parameters of the InAsP NWs, which are summarized in Table 3, including saturation intensity and modulation depth, can be fitted using the following formula [37]:

\[
T(Z) = 1 - \frac{\Delta R}{1 + \frac{Z}{\lambda_0}} - A_{se},
\]
where $\Delta R$, $I_s$, and $A_{ns}$ are modulation depth, saturable intensity, and nonsaturable loss, respectively.

**APPENDIX D: THE EXPERIMENTAL SETUP OF THE MODE-LOCKED SOLID-STATE LASER**

In our experiment, a z-type resonator with the cavity length of 3.49 m is applied, as shown in Fig. 6.

**Fig. 6.** Experimental setup of the mode-locked solid-state bulk laser based on an InAsP NWs SA.

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