Supporting Information for:

Mechanism of short-pulse-induced solute migration in comparison to continuous-light-driven thermal diffusion

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The aim of this **Supporting Information (SI)** is to calculate the pulse energy absorbed by individual ClAlPc molecules ($\Delta\varepsilon_p$) dissolved in ethanol (EtOH) at two concentrations, 4.2×10^{17} cm^{-3} and 1.2×10^{17} cm^{-3}. Given that the Beer's law equation (eqn (5)) and the associated population-change-rate equations (eqn (6)-(8)) govern the time rate of pulse energy absorption by ClAlPc molecules in a unit of volume, collectively, we numerically derive $\Delta\varepsilon_p$ by tracking the excitation and relaxation behaviors of individual ClAlPc molecules (in addition to alternately integrating eqn (5) and eqn (6)-(8) to evaluate the intensity $I(z',r,t)$ and population densities of the S_0, S_1 and S_2 states, denoted by $N_{S0}(z',r,t)$, $N_{S1}(z',r,t)$ and $N_{S2}(z',r,t)$, respectively). Here, z' denotes the penetration depth of a pulse into the sample and falls in the range [0, L=1 mm] with L being the sample thickness, r the lateral distance relative to the laser beam axis and t the time relative to the pulse peak. In the following, we first show how we evaluate $I(z',r,t)$ and N's(z',r,t) specified by the subscripts by alternately integrating eqn (5) and eqn (6)-(8). Afterward, we explain how we derive $\Delta\varepsilon_p$ by counting the numbers of one-photon $S_0\rightarrow|\nu)S_1$, two-photon $S_0\rightarrow|\nu)S_2$ and one-photon $|0)S_1\rightarrow|\nu)S_2$ excitation processes that individual ClAlPc molecules experience in the course of pulse-matter interaction.

As mentioned in the **Main Text**, The intensity of each laser pulse incident on the sample's front surface at a certain position z relative the beam waist is denoted by $I(z'=0,r,t)=I_z(r,t)$ given by eqn (1). When each pulse has a width of τ=19 ps (HWe^{-1}M), we ignore the pulse for the time beyond $\pm3\tau$ (±57 ps) when simulating the pulse-matter interaction. This is because the intensity outside the time range $[-3\tau, 3\tau]$ is vanishingly small. When the pulse-matter interaction starts at $t=-3\tau$, ClAlPc/EtOH is in full thermodynamic (thermal, mechanical and chemical) equilibrium with $N_{S1}(z',r,-3\tau)=N_{S2}(z',r,-3\tau)$ =0 and $N_{S0}(z',r,-3\tau)$=4.2×10^{17} cm^{-3} or 1.2×10^{17} cm^{-3} throughout the whole solution.

In our numerical integration of $I(z',r,t)$ and N's(z',r,t) and derivation of $\Delta\varepsilon_p$, we slice the sample into 10 layers and divide each pulse into 5000 temporal segments. Therefore, each layer of the sample has a thickness of $\Delta z'$=0.1 mm and each temporal segment of a pulse has a width of Δt=6τ/5000=22.8 fs. Labeling each layer of the sample by an integer m ranging between 1 and 10, we denote the depths for the leading and closing edges of the mth layer by $z'_{mb}=(m-1)\Delta z'$ and $z'_{mf}=z'_{(m+1)b}=m\Delta z'$, respectively. Labeling each temporal segment of a pulse by an integer n ranging between 1 and 5000, we denote the times for the leading and the closing edges of the nth temporal segment by $t_{nb}=-3\tau+(n-1)\Delta t$ and

$t_{nf}=t_{(n+1)b}=-3\tau+n\Delta t$, respectively.

First of all, we deal with the interaction of a 19 ps pulse with the entrance layer ($m=1$) of the sample ranging between $z'_{1b}=0$ and $z'_{1f}=0.1$ mm. By substituting the incident intensity $I(z'_{1b},r,t')=I_z(r,t')$, with $t'=t_{nb}$ ($1\leq n\leq5000$), for $I(z',r,t')$ on the right hand sides of eqn (6)-(8) and then integrating these three equations over t' from t_{nb} to t_{nf}, we obtain $N_{S0}(z'_{1b},r,t)=N_{S0}(z'_{1b},r,t_{nb})+\Delta N_{S0}(z'_{1b},r,t_{nb})$, $N_{S1}(z'_{1b},r,t)=N_{S1}(z'_{1b},r,t_{nb})+\Delta N_{S1}(z'_{1b},r,t_{nb})$ and $N_{S2}(z'_{1b},r,t)=N_{S2}(z'_{1b},r,t_{nb})+\Delta N_{S2}(z'_{1b},r,t_{nb})$ with $t=t_{nf}=t_{(n+1)b}$ ($1\leq n\leq 5000$). Here

$$\Delta N_{S0}=\left(-\frac{\sigma_{S0}N_{S0}I}{\hbar\omega}-\frac{\beta N_{S0}I^2}{2\hbar\omega}\right)\times\Delta t\,,\tag{S.1}$$

$$\Delta N_{S1}=\left(\frac{\sigma_{S0}N_{S0}I}{\hbar\omega}-\frac{\sigma_{S1}N_{S1}I}{\hbar\omega}+\frac{N_{S2}}{\tau_{S2}}\right)\times\Delta t\tag{S.2}$$

and

$$\Delta N_{S2}=\left(\frac{\beta N_{S0}I^2}{2\hbar\omega}+\frac{\sigma_{S1}N_{S1}I}{\hbar\omega}-\frac{N_{S2}}{\tau_{S2}}\right)\times\Delta t\,,\tag{S.3}$$

in which the arguments (z'_{1b},r,t_{nb}) of I, N's and ΔN's are omitted. Note that after the above integrations $N_{S0}(z'_{1b},r,t_{1b})=4.2\times10^{17}$ cm^{-3} or 1.2×10^{17} cm^{-3}, $N_{S1}(z'_{1b},r,t_{1b})=0$ and $N_{S2}(z'_{1b},r,t_{1b})=0$ remain unchanged. Next, by substituting the concentrations $N_{S0}(z'',r,t)$ and $N_{S1}(z'',r,t)$, with $t=t_{nb}$ ($1\leq n\leq5000$), for $N_{S0}(z'',r,t)$ and $N_{S1}(z'',r,t)$ on the right hand side of eqn (5) and then integrating this equation over z'' from z'_{1b} to z'_{1f}, we obtain $I(z'_{1f},r,t)=I(z'_{2b},r,t)=I(z'_{1b},r,t)+\Delta I(z'_{1b},r,t)$ with $t=t_{nb}$ ($1\leq n\leq5000$). Here

$$\Delta I=\left[-\left(\sigma_{S0}N_{S0}+\sigma_{S1}N_{S1}\right)\times I-\beta N_{S0}\times I^2\right]\times\Delta z'\,,\tag{S.4}$$

in which the arguments (z'_{1b},r,t_{nb}) of I, N's and ΔI are omitted. During this integration, we consider that N's(z'',r,t_{nb}) equals N's(z'_{1b},r,t_{nb}) and does change with z''.

Secondly, we consider the 19 ps pulse advances to the second layer of the sample ranging between $z'_{2b}=0.1$ mm and $z'_{2f}=0.2$ mm. By substituting $I(z'_{2b},r,t')$, with $t'=t_{nb}$ ($1\leq n\leq5000$), for $I(z',r,t')$ on the right hand sides of eqn (6)-(8) and then integrating these three equations over t' from t_{nb} to t_{nf}, we

obtain $N_{S0}(z'_{2b},r,t)=N_{S0}(z'_{2b},r,t_{nb})+\Delta N_{S0}(z'_{2b},r,t_{nb})$, $N_{S1}(z'_{2b},r,t)=N_{S1}(z'_{2b},r,t_{nb})+\Delta N_{S1}(z'_{2b},r,t_{nb})$, and

$N_{S2}(z'_{2b},r,t)=N_{S2}(z'_{2b},r,t_{nb})+\Delta N_{S2}(z'_{2b},r,t_{nb})$, with $t=t_{nf}=t_{(n+1)b}$ (1≤n≤5000). Here $\Delta N_{S0}(z'_{2b},r,t_{nb})$,

$\Delta N_{S1}(z'_{2b},r,t_{nb})$ and $\Delta N_{S2}(z'_{2b},r,t_{nb})$ can be derived from eqn (S.1)-(S.3) with (z'_{2b},r,t_{nb}) being the

arguments of I, N's and $\Delta N'$s. Note that after the above integrations $N_{S0}(z'_{2b},r,t_{1b})=4.2\times10^{17}$ cm^{-3} or

1.2×10^{17} cm^{-3}, $N_{S1}(z'_{2b},r,t_{1b})=0$ and $N_{S2}(z'_{2b},r,t_{1b})=0$ remain unchanged. Next, by substituting the

concentrations $N_{S0}(z'',r,t)$ and $N_{S1}(z'',r,t)$, with $t=t_{nb}$ (1≤n≤5000), for $N_{S0}(z'',r,t)$ and $N_{S1}(z'',r,t)$ on the

right hand side of eqn (5) and then integrating this equation over z'' from z'_{2b} to z'_{2f}, we obtain

$I(z'_{2f},r,t)=I(z'_{3b},r,t)=I(z'_{2b},r,t)+\Delta I(z'_{2b},r,t)$ with $t=t_{nb}$ (1≤n≤5000). Here $\Delta I(z'_{2b},r,t_{nb})$ can be derived from

eqn (S.4) with (z'_{2b},r,t_{nb}) being the arguments of I, N's and ΔI. During this integration, we consider

that N's(z'',r,t_{nb}) equals N's(z'_{2b},r,t_{nb}) and does change with z''.

Finally, by repeating the same procedures, we advance the 19 ps pulse, in increments of 0.1 mm, to

the last layer of the sample ranging between $z'_{10b}=0.9$ mm and $z'_{10f}=1.0$ mm. In each layer of the sample,

we derive $I(z',r,t)$ as well as $N_{S0}(z',r,t)$, $N_{S1}(z',r,t)$ and $N_{S2}(z',r,t)$ with $z'=z'_{1b}$ and z'_{10f} (1≤m≤10) as well

as $t=t_{1b}$ and t_{nf} (1≤n≤5000). In summary, all N's(z',r,t) at $t=t_{nf}$ can be related to those at $t=t_{nb}$ (1≤n≤5000)

$$N_{S1}\left(t_{nf}\right)=N_{S1}\left(t_{nb}\right)+\left[\frac{\sigma_{S0}N_{S0}\left(t_{nb}\right)I\left(t_{nb}\right)}{\hbar\omega}-\frac{\sigma_{S1}N_{S1}\left(t_{nb}\right)I\left(t_{nb}\right)}{\hbar\omega}+\frac{N_{S2}\left(t_{nb}\right)}{\tau_{S2}}\right]\times\Delta t ,\tag{S.5}$$

$$N_{S2}\left(t_{nf}\right)=N_{S2}\left(t_{nb}\right)+\left[\frac{\beta N_{S0}\left(t_{nb}\right)I^2\left(t_{nb}\right)}{2\hbar\omega}+\frac{\sigma_{S1}\left(t_{nb}\right)I\left(t_{nb}\right)}{\hbar\omega}-\frac{N_{S2}\left(t_{nb}\right)}{\tau_{S2}}\right]\times\Delta t \tag{S.6}$$

and

$$N_{S0}\left(t_{nf}\right)=N_{S0}\left(-3\tau\right)-N_{S1}\left(t_{nf}\right)-N_{S2}\left(t_{nf}\right).\tag{S.7}$$

For simplicity, we have dropped the arguments z' and r of $N_{S0}(z',r,t)$, $N_{S1}(z',r,t)$ and $N_{S2}(z',r,t)$ in eqn

(S.5)-(S.7).

When deriving $N_{S0}(z',r,t)$, $N_{S1}(z',r,t)$ and $N_{S2}(z',r,t)$ by integrating eqn (S.5)-(S.7), we additionally

track the excitation and relaxation behaviors of individual ClAlPc molecules by counting the number

of one-photon $S_0\rightarrow|v\rangle S_1$, two-photon $S_0\rightarrow|v\rangle S_2$ and one-photon $|0\rangle S_1\rightarrow|v\rangle S_2$ excitation processes that

individual ClAlPc molecules on $|0\rangle S_1$ and $|0\rangle S_2$ have experienced up to any time $t\leq3\tau$. Accordingly,

we decompose $N_{S1}(z',r,t)$ and $N_{S2}(z',r,t)$ into their components $N_{ES1}(i;z',r,t)$ and $N_{ES2}(i;z',r,t)$ which designate the concentrations of ClAlPc molecules on $|0)S_1$ and $|0)S_2$ that have absorbed i photons ($i \geq 1$) up to $t \leq 3\tau$. $N_{ES1}(i;z',r,t) + N_{ES2}(i;z',r,t)$, denoted by $N_E(i;z',r,t)$ henceforth, designates the concentrations of ClAlPc molecules on S_0, $|0)S_1$ or $|0)S_2$ (all the ClAlPc molecules indeed) that have individually absorbed i photons with energy of $i\hbar\omega$ ($i \geq 1$) up to $t \leq 3\tau$. Because ClAlPc molecules relaxing to S_0 from $|0)S_1$, with a fluorescent lifetime τ_f (12.1 ns) greatly longer than τ (19 ps), are neglected during the pulse-matter interaction, $N_{S0}(z',r,t)$ does not actually contribute to $N_E(i;z',r,t)$ for $i \geq 1$. For simplicity, we drop the arguments z', r or t, of $I(z',r,t)$, $N_{S0}(z',r,t)$, $N_{S1}(z',r,t)$, $N_{S2}(z',r,t)$, $N_{ES1}(i;z',r,t)$, $N_{ES2}(i;z',r,t)$ and $N_E(i;z',r,t)$, fully or partially, in the following unless it is necessary to express them explicitly.

Given $N_{ES1}(i;t_{1b}) = N_{S1}(t_{1b}) = 0$, $N_{ES2}(i;t_{1b}) = N_{S2}(t_{1b}) = 0$ and $N_{S0}(t_{1b}) = 4.2 \times 10^{17}$ cm^{-3} or 1.2×10^{17} cm^{-3}, $|0)S_1$ and $|0)S_2$ are respectively populated, in the time range $[t_{1b}, t_{1f}]$, by one-photon $S_0 \rightarrow |v)S_1$ and two-photon $S_0 \rightarrow |v)S_2$ excitations of the solute molecules pertaining to $N_{S0}(t_{1b})$. Therefore, after the first temporal segment of a pulse, falling in the time range $[t_{1b}, t_{1f}]$, traverses the sample, both $N_{ES1}(i;t_{1f})$ and $N_{ES2}(i;t_{1f})$ have a single nonzero component

$$N_{ES1}\left(1;t_{1f}\right) = N_{S1}\left(t_{1f}\right) \tag{S.8}$$

and

$$N_{ES2}\left(2;t_{1f}\right) = N_{S2}\left(t_{1f}\right). \tag{S.9}$$

Both $N_{S1}(t_{1f})$ and $N_{S2}(t_{1f})$ on the right hand side of eqn (S.8) and (S.9) are derived from eqn (S.5) and (S.6) with n set to be 1.

After the second temporal segment, falling in the time range $[t_{2b}, t_{2f}]$, of this pulse traverses the sample, the solute molecules on $|0)S_1$ may have absorbed one or two photons according to eqn (S.5)-(S.7) in combination with eqn (S.8) and (S.9). Hence $N_{ES1}(i;t_{2f})$ has the following two nonzero components

$$N_{ES1}\left(1;t_{2f}\right) = N_{ES1}\left(1;t_{2b}\right) + \left[\frac{\sigma_{S0}N_{S0}\left(t_{2b}\right)I\left(t_{2b}\right)}{\hbar\omega} - \frac{\sigma_{S1}N_{ES1}\left(1;t_{2b}\right)I\left(t_{2b}\right)}{\hbar\omega}\right] \times \Delta t \tag{S.10}$$

and

$$N_{ES1}\left(2;t_{2f}\right) = \frac{N_{ES2}\left(2;t_{2b}\right)}{\tau_{S2}} \times \Delta t. \tag{S.11}$$

When the first term on the right hand side of eqn (S.10), i.e., $N_{ES1}(1;t_{2b}=t_{1f})$ derived from eqn (S.8),

is the initial condition, the second and third terms denote the change of $N_{ES1}(i=1)$ induced in this temporal segment by one-photon $S_0 \rightarrow |v\rangle S_1$ and $|0\rangle S_1 \rightarrow |v\rangle S_2$ excitation of the solute molecules pertaining to $N_{S0}(t_{2b})$ and $N_{ES1}(1;t_{2b})$. Eqn (S.11) shows the change of $N_{ES1}(i=2)$ induced in this temporal segment by $|v\rangle S_2 \rightsquigarrow |0\rangle S_2 \rightsquigarrow |v\rangle S_1 \rightsquigarrow |0\rangle S_1$ relaxation of the solute molecules pertaining to $N_{ES2}(2;t_{2b})$. Here, $\tau_{S2}=900$ fs (the $|v\rangle S_2 \rightsquigarrow |0\rangle S_2 \rightsquigarrow |v\rangle S_1 \rightsquigarrow |0\rangle S_1$ relaxation lifetime) is much longer than the temporal segment width of $\Delta t=22.8$ fs. The initial condition $N_{ES1}(2;t_{2b})=0$ is omitted herein. Analogously, $N_{ES2}(i;t_{2f})$ has only one nonzero component

$$N_{ES2}\left(2;t_{2f}\right) = N_{ES2}\left(2;t_{2b}\right) \\ + \left[\frac{\beta N_{S0}\left(t_{2b}\right)I^2\left(t_{2b}\right)}{2\hbar\omega} + \frac{\sigma_{S1}N_{ES1}\left(1;t_{2b}\right)I\left(t_{2b}\right)}{\hbar\omega} - \frac{N_{ES2}\left(2;t_{2b}\right)}{\tau_{S2}}\right]\times\Delta t . \tag{S.12}$$

When the first term on the right hand side of eqn (S.12), $N_{ES2}(2;t_{2b}=t_{1f})$ derived from eqn (S.9), is the initial condition, the second through the fourth terms respectively denote the changes of $N_{ES2}(i=2)$ induced in this temporal segment by two-photon $S_0 \rightarrow |v\rangle S_2$ excitation of the solute molecules pertaining to $N_{S0}(t_{2b})$, one-photon $|0\rangle S_1 \rightarrow |v\rangle S_2$ excitation of the solute molecules pertaining to $N_{ES1}(1;t_{2b})$ and $|v\rangle S_2 \rightsquigarrow |0\rangle S_2 \rightsquigarrow |v\rangle S_1 \rightsquigarrow |0\rangle S_1$ relaxation of the solute molecules pertaining to $N_{ES2}(2;t_{2b})$. $N_{S0}(t_{2b})$ in the second terms of both eqn (S.10) and (S.12) is derived from eqn (S.5)-(S.7) with n set to be 1.

After the third segment, falling in the time range [t_{3b}, t_{3f}], of this pulse traverses the sample, the solute molecules on $|0\rangle S_1$ may have absorbed one or two photons and those on $|0\rangle S_2$ may have absorbed two or three photons according to eqn (S.5)-(S.7) in combination with eqn (S.10)-(S.12). Hence $N_{ES1}(i;t_{3f})$ and $N_{ES2}(i;t_{3f})$ respectively contain two nonzero components

$$N_{ES1}\left(1;t_{3f}\right) = N_{ES1}\left(1;t_{3b}\right) + \left[\frac{\sigma_{S0}N_{S0}\left(t_{3b}\right)I\left(t_{3b}\right)}{\hbar\omega} - \frac{\sigma_{S1}N_{ES1}\left(1;t_{3b}\right)I\left(t_{3b}\right)}{\hbar\omega}\right]\times\Delta t, \tag{S.13}$$

$$N_{ES1}\left(2;t_{3f}\right) = N_{ES1}\left(2;t_{3b}\right) - \left[\frac{\sigma_{S1}N_{ES1}\left(2;t_{3b}\right)I\left(t_{3b}\right)}{\hbar\omega} - \frac{N_{ES2}\left(2;t_{3b}\right)}{\tau_{S2}}\right]\times\Delta t, \tag{S.14}$$

$$N_{ES2}\left(2;t_{3f}\right) = N_{ES2}\left(2;t_{3b}\right) \\ + \left[\frac{\beta N_{S0}\left(t_{3b}\right)I^2\left(t_{3b}\right)}{2\hbar\omega} + \frac{\sigma_{S1}N_{ES1}\left(1;t_{3b}\right)I\left(t_{3b}\right)}{\hbar\omega} - \frac{N_{ES2}\left(2;t_{3b}\right)}{\tau_{S2}}\right]\times\Delta t \tag{S.15}$$

and

$$N_{\text{ES2}}\left(3;t_{3f}\right)=\frac{\sigma_{\text{S1}}N_{\text{ES1}}\left(2;t_{3b}\right)I\left(t_{3b}\right)}{\hbar\omega}\times\Delta t \,. \tag{S.16}$$

When the first terms on the right hand side of eqn (S.13)-(S.15) obtained from eqn (S.10)-(S.12) with t_{2f} replaced by t_{3b}, are the initial conditions, the subsequent terms denote the changes induced in this temporal segment by one-photon $S_0\rightarrow|v)S_1$ and $|0)S_1\rightarrow|v)S_2$ excitation of the solute molecules pertaining to $N_{\text{S0}}(t_{3b})$ and $N_{\text{ES1}}(1;t_{3b})$ respectively (related to eqn (S.13)), one-photon $|0)S_1\rightarrow|v)S_2$ excitation and $|v)S_2\rightsquigarrow|0)S_2\rightsquigarrow|v)S_1\rightsquigarrow|0)S_1$ relaxation of the solute molecules pertaining to $N_{\text{ES1}}(2;t_{3b})$ and $N_{\text{ES2}}(2;t_{3b})$, respectively (related to eqn (S.14)) as well as two-photon $S_0\rightarrow|v)S_2$ excitation, one-photon $|0)S_1\rightarrow|v)S_2$ excitation and $|v)S_2\rightsquigarrow|0)S_2\rightsquigarrow|v)S_1\rightsquigarrow|0)S_1$ relaxation of the solute molecules pertaining to $N_{\text{S0}}(t_{3b})$, $N_{\text{ES1}}(1;t_{3b})$ and $N_{\text{ES2}}(2;t_{3b})$, respectively (related to eqn (S.15)). The initial condition of eqn (S.16) $N_{\text{ES2}}(3;t_{3b})=0$ is omitted herein. The only term on the right hand side denotes the change of $N_{\text{ES2}}(i=3)$ induced in this temporal segment by one-photon $|0)S_1\rightarrow|v)S_2$ excitation of the solute molecules pertaining to $N_{\text{ES1}}(2;t_{3b})$. $N_{\text{S0}}(t_{3b})$ in the second terms of both eqn (S.13) and (S.15) is derived from eqn (S.5)-(S.7) with n set to be 2.

When n exceeds 3 and is even, we analogize the above processes to obtain

$$N_{\text{ES1}}\left[(n/2+1);t_{nf}\right]=\frac{N_{\text{ES2}}\left[(n/2+1);t_{nb}\right]}{\tau_{\text{S2}}}\times\Delta t \,, \tag{S.17}$$

$$N_{\text{ES1}}\left(i;t_{nf}\right)=N_{\text{ES1}}\left(i;t_{nb}\right)-\left[\frac{\sigma_{\text{S1}}N_{\text{ES1}}\left(i;t_{nb}\right)I\left(t_{nb}\right)}{\hbar\omega}-\frac{N_{\text{ES2}}\left(i;t_{nb}\right)}{\tau_{\text{S2}}}\right]\times\Delta t \tag{S.18}$$

and

$$N_{\text{ES2}}\left(i+1;t_{nf}\right)=N_{\text{ES2}}\left(i+1;t_{nb}\right)+\left[\frac{\sigma_{\text{S1}}N_{\text{ES1}}\left(i;t_{nb}\right)I\left(t_{nb}\right)}{\hbar\omega}-\frac{N_{\text{ES2}}\left(i+1;t_{nb}\right)}{\tau_{\text{S2}}}\right]\times\Delta t \tag{S.19}$$

with $2\leq i\leq n/2$. On the other hand, when n exceeds 3 and is odd, we simply omit eqn (S.17) and retain eqn (S.18) and (S.19) for $2\leq i\leq(n+1)/2$. No matter n is even or odd, for the case of $i=1$, we have

$$N_{\text{ES1}}\left(1;t_{nf}\right)=N_{\text{ES1}}\left(1;t_{nb}\right)+\left[\frac{\sigma_{\text{S0}}N_{\text{S0}}\left(t_{nb}\right)I\left(t_{nb}\right)}{\hbar\omega}-\frac{\sigma_{\text{S1}}N_{\text{ES1}}\left(1;t_{nb}\right)I\left(t_{nb}\right)}{\hbar\omega}\right]\times\Delta t \tag{S.20}$$

and

$$\begin{aligned}N_{\text{ES2}}\left(2;t_{nf}\right)=&N_{\text{ES2}}\left(2;t_{nb}\right)\\&+\left[\frac{\beta N_{\text{S0}}\left(t_{nb}\right)I^2\left(t_{nb}\right)}{2\hbar\omega}+\frac{\sigma_{\text{S1}}N_{\text{ES1}}\left(1;t_{nb}\right)I\left(t_{nb}\right)}{\hbar\omega}-\frac{N_{\text{ES2}}\left(2;t_{nb}\right)}{\tau_{\text{S2}}}\right]\times\Delta t\end{aligned}^{\cdot} \tag{S.21}$$

At the desired z, z' and r, we sequentially integrate eqn (S.5)-(S.21) to derive $N_{\text{ES1}}(i;z',r,3\tau)$ with i

ranging between 1 and 2501 and $N_{ES2}(i;z',r,3\tau)$ with i ranging between 2 and 2501. Accordingly, we derive $N_E(i;z',r,3\tau)$ as a function of i at three incident pulse energy levels (1.1, 4.5, and 8.7 µJ), two concentrations, 4.2×10^{17} cm^{-3} and 1.2×10^{17} cm^{-3} and three penetration depths (0.1, 0.5 and 1.0 mm). The results are shown in Fig. 4 of the **Main Text**.