SI Appendix

Femtosecond Thin Lens Derivation. In polar coordinates a Laguerre-Gaussian $\left(LG_0^1\right)$ mode has a transverse intensity profile given by,

$$I(r,\varphi) = I_0 \exp(1)\frac{2r^2}{w^2}e^{-2(r/w)^2}, \quad \text{[S.1]}$$

where w is the waist of the focus and I_0 the maximum intensity (1). This 'donut' mode has an intensity maximum located at $r = \sqrt{2}w/2$ with a value of,

$$I_0 = 2\frac{E_\mathrm{p}}{w^2\tau}\sqrt{\frac{\ln 2}{\pi^3}}, \quad \text{[S.2]}$$

where E_p is the energy of the laser pulse and τ is the full-width-at-half-maximum of the pulse duration, assuming a Gaussian temporal profile given by $\exp\left(-4\ln 2(t/\tau)^2\right)$. The ponderomotive energy $U_\mathrm{P}(r)$ is proportional to intensity,

$$U_\mathrm{P}(r) = \frac{e^2\lambda^2}{8\pi^2 m\varepsilon_0 c^3} I(r,\varphi), \quad \text{[S.3]}$$

where m is the electron mass, e is the electron charge and λ the central wavelength of the laser radiation. Near the center of the donut mode focus (or $r \ll w$) the intensity distribution is approximately parabolic, and hence the ponderomotive energy near the donut center is also parabolic. The potential can be approximated by,

$$U_\mathrm{P}(x) = \frac{1}{2}\left[\frac{e^2\lambda^2 \exp(1) E_\mathrm{p}}{\pi^3 m\varepsilon_0 c^3 w^4 \tau}\sqrt{\frac{\ln 2}{\pi}}\right]x^2 \equiv \tfrac{1}{2}Kx^2, \quad \text{[S.4]}$$

in which x is used instead of r. For this parabolic approximation to be applicable, the spatial extent of the dispersed electron pulse, at $t = 0$,

$$\Delta x(0) = v_0 \Delta t_o + \Delta v_o t_o\,, \qquad \text{[S.5]}$$

must be much smaller than the laser waist, where the object velocity spread is $\Delta v_o = \Delta E \big/ \sqrt{2mE}$ (2). In analogy with a mechanical harmonic oscillator, the quantity in the square brackets of Eq. [S.4] can be referred to as the *stiffness* K; it has units of $\mathrm{J/m^2} = \mathrm{N/m}$, and at 800 nm has the numerical value of,

$$K \approx 3.1\times 10^{-36}\,\frac{E_\mathrm{p}}{w^4 \tau}\,. \qquad \text{[S.6]}$$

The effect of this parabolic potential on an ensemble of electrons emitted from a source will now be analyzed. The velocity distribution of the ensemble is centered around v_0, with an emission time distribution centered on $-t_o$, where all electrons are emitted from the same location $x_o = -v_0 t_o$. Assuming a single donut-shaped laser pulse is applied at $t = 0$, and centered at $x = 0$, the electron ensemble is then influenced by the potential $U(x) = \frac{1}{2} K x^2$. The k^{th} electron in the ensemble has an initial velocity $V_k = v_0 + v_k$ and emission time $T_k = -t_o + t_k$. Using a Galilean transformation to a frame moving with velocity v_0, the coordinate x is replaced with the moving frame coordinate $\tilde{x} = x - v_0 t$. Before the electron is influenced by the pulsed potential, the trajectory in the lab frame is,

$$x_k(t) = -v_0 t_o + V_k (t - T_k)\,, \qquad \text{[S.7a]}$$

and in the moving frame, the trajectory is,

$$\tilde{x}_k(t) = -v_0 t_k + v_k t + v_k t_o - v_k t_k\ . \qquad \text{[S.7b]}$$

At $t = 0$ the potential exists for the ultrashort laser pulse duration τ, giving the electron an impulse (or 'kick') dependent on its instantaneous position in the parabolic potential.

In both frames, the position at $t=0$ is $x_k\left(0\right)=\tilde{x}_k\left(0\right)\equiv x_k^*=-v_0t_k+v_kt_o-v_kt_k$ and the acceleration is $a=-(\partial U_P(x)/\partial x)/m=-(K/m)x_k^*$. The change in velocity is then,

$$\Delta v=a\tau=-\left(K\tau/m\right)x_k^*=-x_k^*\Big/t_f\ , \qquad \text{[S.8]}$$

where $t_f=m\big/\left(K\tau\right)$ is the focal time. Immediately after the potential is turned off the electron trajectory becomes,

$$x_k(t)=x_k^*+\left(V_k-x_k^*\big/t_f\right)t=v_kt+v_0t+\left(-v_0t_k+v_kt_o-v_kt_k\right)\left(1-t\big/t_f\right), \qquad \text{[S.9a]}$$

$$\tilde{x}_k(t)=x_k^*+\left(v_k-x_k^*\big/t_f\right)t=v_kt+\left(-v_0t_k+v_kt_o-v_kt_k\right)\left(1-t\big/t_f\right), \qquad \text{[S.9b]}$$

in the lab and moving frames, respectively. The electron trajectories, before and after $t=0$, can be plotted in both frames to give the equivalent of a ray diagram. Electrons emitted at the same time, i.e. $t_k=0$, but with different velocities, will meet at the *image position*, $x_i=v_0t_i$ in the lab frame at the *image time* t_i, whereas in the moving frame, the *image position* is located at $\tilde{x}_k=0$ with the same *image time*. The image time is found by setting $\tilde{x}_k\left(t_i\right)=0$, from Eq. [S.9b], with $t_k=0$,

$$\tilde{x}_k(t_i)=v_kt_i+v_kt_o\left(1-t_i/t_f\right)=0 \quad , \qquad \text{[S.10]}$$

which is equivalent to the lens equation: $t_o^{-1}+t_i^{-1}=t_f^{-1}$.

Electrons that are emitted at different times, i.e., $t_k\neq 0$, will now be considered. To find the duration of an electron packet, needed first is the time, t_a, it takes the k^{th} electron to reach an arbitrary position x_a after the lens. By equating $x_k\left(t\right)$ to v_0t_a, or,

$$v_0t_a=v_kt+v_0t+\left(-v_0t_k+v_kt_o-v_kt_k\right)\left(1-t/t_f\right), \qquad \text{[S.11]}$$

and then solving for time gives,

$$t(t_k, v_k) = \frac{v_0 t_a + v_0 t_k - v_k t_o + v_k t_k}{v_0 + v_k + \frac{v_0 t_k}{t_f} - \frac{v_k t_o}{t_f} + \frac{v_k t_k}{t_f}}. \qquad [S.12]$$

The arrival times of electrons with different t_k and v_k, for an arbitrary position x_a in the lab frame, determines the temporal duration of the electron packet. The temporal duration is the time difference of the k^{th} electron and the ensemble average ($v_k = t_k = 0$), $t_k^a(t_k, v_k) = t(t_k, v_k) - t(0,0)$, and has the form,

$$t_k^a(t_k, v_k) = t_a t_f \frac{v_k t_o\left(\frac{1}{t_f} - \frac{1}{t_a} - \frac{1}{t_o}\right) + t_k(v_0 + v_k)\left(\frac{1}{t_a} - \frac{1}{t_f}\right)}{t_f(v_0 + v_k) + t_k(v_0 + v_k) - v_k t_o}. \qquad [S.13]$$

If $t_k/t_o \ll 1$ and $v_k/v_0 \ll 1$, Eq. [S.13] becomes,

$$t_k^a(t_k, v_k) = t_a\left[\frac{v_k t_o}{v_0 + v_k}\left(\frac{1}{t_f} - \frac{1}{t_a} - \frac{1}{t_o}\right) + t_k\left(\frac{1}{t_a} - \frac{1}{t_f}\right)\right]. \qquad [S.14]$$

The relationship between t_k and t_k^a at the image time t_i (or $a = i$ in Eq. [S.14]) gives a relationship for the temporal magnification of the electron packet. Using Eq. [S.14], the relationship between t_k^i and t_k is simply $t_k^i = -t_i t_k / t_o$. If the magnification is defined as $M = -t_i/t_o$ then the temporal duration at the image time becomes,

$$\Delta t_i = M \Delta t_o, \qquad [S.15]$$

where Δt_o and Δt_i are the duration of the electron packet at the object and image time, respectively. Durations achievable with a thin temporal lens follow from Eq. [S.15].

Attosecond Thick Lens Derivation. The co-propagating standing wave is created by using two different optical frequencies, constructed by having a higher frequency (ω_1)

optical pulse traveling in the same direction as the electron packet and a lower frequency (ω_2) traveling in the opposite direction. When the optical frequencies ω_1, ω_2, and the electron velocity v_0 are chosen according to $v_0 = c(\omega_1 - \omega_2)/(\omega_1 + \omega_2)$, a standing wave is produced in the rest frame of the electron (3). If the electron has a velocity $v_0 = c/3$, and $\omega_1 = 2\omega_2$ then the co-propagating standing wave has a ponderomotive potential of the form,

$$U_\mathrm{P} = \frac{1}{2}\left(\frac{e^2\tilde{\lambda}^2 E_0^2}{8\pi^2 mc^2}\right)\cos^2(\tilde{k}x), \qquad \text{[S.16a]}$$

$$\tilde{k} = \tilde{\omega}/c = \omega_1\sqrt{(c-v_0)/(c+v_0)} = \omega_2\sqrt{(c+v_0)/(c-v_0)}, \qquad \text{[S.16b]}$$

where E_0 is the peak electric field, $\tilde{\lambda}$ the Doppler shifted wavelength and $\tilde{k}$ is the Doppler shifted wavenumber (3). The envelopes of the laser pulses are ignored in this derivation, but they can be engineered so that the standing wave contrast is optimized (4).

To find an analytic solution in the thick lens geometry, each individual potential well in the standing wave is approximated by a parabolic potential that matches the curvature of the sinusoidal potential, $U_\mathrm{P}(x) = \frac{1}{2}\left[\frac{e^2}{2mc^2}E_0^2\right]x^2 \equiv \frac{1}{2}Kx^2$. The motion of the electron in the parabolic potential is given by the solution to the harmonic oscillator, $x(t) = C_1\sin(\omega_\mathrm{P}t) + C_2\cos(\omega_\mathrm{P}t)$, where $\omega_\mathrm{P} = \sqrt{K/m}$ and C_1 and C_2 are determined by the initial conditions. As in the thin lens model, the potential is turned on at $t = 0$ and stays on for a duration τ. The initial conditions are $\tilde{x}_k(0) = -v_0 t_k + v_k t_o - v_k t_k$ and $\tilde{v}_k(0) = v_k$ in the co-propagating frame, which gives $C_1 = v_k/\omega_\mathrm{P}$, and $C_2 = \tilde{x}_k(0)$. While the potential is on, $0 < t < \tau$, the equations of motion for the k^{th} electron are given by,

$$\tilde{x}_k(t, v_k, t_k) = \frac{v_k}{\omega_P}\sin(\omega_P t) + \tilde{x}_k(0)\cos(\omega_P t), \quad \text{[S.17a]}$$

$$\tilde{v}_k(t, v_k, t_k) = v_k \cos(\omega_P t) - \omega_P \tilde{x}_k(0)\sin(\omega_P t). \quad \text{[S.17b]}$$

After the potential is turned off, $t > \tau$, the equations of motion for electrons in the co-propagating and lab frames are respectively,

$$\tilde{x}_k(t, v_k, t_k) = -\tilde{x}_k(\tau) + \tilde{v}_k(\tau)(t - \tau), \quad \text{[S.18a]}$$

$$x_k(t, v_k, t_k) = -\tilde{x}_k(\tau) + (\tilde{v}_k(\tau) + v_0)(t - \tau), \quad \text{[S.18b]}$$

with,

$$\tilde{x}_k(\tau) = (v_k / \omega_P)\sin(\omega_P \tau) + \tilde{x}_k(0)\cos(\omega_P \tau), \quad \text{[S.18c]}$$

$$\tilde{v}_k(\tau) = v_k \cos(\omega_P \tau) - \omega_P \tilde{x}_k(0)\sin(\omega_P \tau). \quad \text{[S.18d]}$$

For $v_k = 0$, the *focal time* t_f can be found in the co-propagating frame by solving Eq. [S.18a] with $\tilde{x}_k(t_f, v_k = 0, t_k) = 0$. The focal time is then,

$$t_f = \frac{1}{\omega_P}\cot(\omega_P \tau) + \tau . \quad \text{[S.19]}$$

For $\tau \to 0$, $t_f \to m/(K\tau)$, which is identical to the thin lens definition. As in the thin lens case the *image time,* t_i, is defined in the co-propagating frame as the time it takes electrons emitted at the same time $t_k = 0$, with an arbitrary v_k, to arrive at the origin. Using Eq. [S.18a] with $\tilde{x}_k(t_i, v_k, 0) = 0$ gives,

$$t_i = \frac{1/\omega_P^{\,2} + t_o t_f - t_f \tau + \tau^2}{t_o - t_f + \tau}. \quad \text{[S.20]}$$

Additionally, the equation for the image time can be rewritten in the form,

$$\frac{1}{t_i}+\frac{t_f-\tau}{t_o t_f+1/\omega_P{}^2-t_f\tau+\tau^2}=\frac{t_o}{t_o t_f+1/\omega_P{}^2-t_f\tau+\tau^2}, \quad \text{[S.21]}$$

which after the two assumptions, $\tau \to 0$ and $t_o \gg 1/\left(t_f \omega_P{}^2\right)$ becomes equivalent to the lens equation: $t_o^{-1}+t_i^{-1}=t_f^{-1}$.

To find the temporal duration of the electron packet in the co-propagating frame (similar to that in the thin lens section) at an arbitrary position x_a after the lens, Eq. [S.19a] is set equal to $v_0 t_a$,

$$v_0 t_a=-\tilde{x}_k(\tau)+\left(\tilde{v}_k(\tau)\right)\left(t-\tau\right), \quad \text{[S.22]}$$

and solving for time gives,

$$t\left(t_k,v_k\right)=\frac{v_0 t_a+\tilde{x}_k(\tau)+\left(\tilde{v}_k(\tau)\right)\tau}{\tilde{v}_k(\tau)}. \quad \text{[S.23]}$$

The arrival times of electrons, with t_k and v_k at the arbitrary position x_a, will determine the temporal duration of the electron packet. The time t_k^a is found by taking the difference for time between the k^{th} electron and the center of the electron ensemble, $t_k^a\left(t_k,v_k\right)=t\left(t_k,v_k\right)-t\left(0,0\right)$, and has the form,

$$t_k^a\left(t_k,v_k\right)=t_a t_f\cos\left(\omega_P\tau\right)\frac{v_k t_0\left(\frac{1}{t_f}-\frac{1}{t_a}\left(1-\frac{\omega_P\tau\cot\left(\omega_P\tau\right)-1}{t_o t_f\omega_P{}^2}\right)-\frac{1}{t_o}\left(1-\frac{\tau}{t_f}\right)\right)+t_k\left(v_0+v_k\right)\left(\frac{1}{t_a}-\frac{1}{t_f}\right)}{v_0\left(t_f-\tau\right)+\left(t_k\left(v_0+v_k\right)+v_k\left(t_f-t_o-\tau\right)\right)\cos\left(\omega_P\tau\right)}. \quad \text{[S.24]}$$

For $\omega_P\tau \ll 1$, and ignoring $v_k t_k$ terms, Eq. [S.24] can be simplified, resulting in the following relation:

$$t_k^a\left(t_k,v_k\right)=\frac{t_a}{v_0}\left(t_k v_0\left(\frac{1}{t_f}-\frac{1}{t_a}\right)+v_k t_o\left(\frac{1}{t_f}-\frac{1}{t_a}-\frac{1}{t_o}\right)\right). \quad \text{[S.25]}$$

To this point, the derivation of the thick lens case has been very similar to that of the thin lens. However, for the latter, the spatial extent of the temporal lens is chosen such that it is much larger than the dispersed electron packet. When this condition is met all electrons are captured by the parabolic potential. In contrast, for the thick lens case the dispersed electron pulse is much larger than any individual potential well. Only a portion of the electron packet enters each well, which limits the combinations of initial velocities and emission times (t_k, v_k) that are focused by a particular well. The combinations of t_k and v_k, that enter an individual well also change as the electron packet disperses. In the co-propagating frame, the position of the k^{th} electron is located at,

$$L_k = v_k t_o - t_k v_0 , \qquad \text{[S.26]}$$

for a time, $t = 0$, or when the lens is first turned on. For electrons to be focused by the center well the condition, $-\tilde{\lambda}/4 < v_k t_o - t_k v_0 < \tilde{\lambda}/4$ must be satisfied. From Eq. [S.26] we see that as the electron packet is allowed to propagate and disperse (or the value of t_o increases) the allowed values for t_k and v_k to reach a position L_k change; this results in two different regimes. The first regime is when $t_o < v_0 \Delta t_o / \Delta v_o$ and the second is when $t_o > v_0 \Delta t_o / \Delta v_o$, where Δt_o and Δv_o are the full widths of the independent temporal and velocity distributions at t_o. Only the first regime, $t_o < v_0 \Delta t_o / \Delta v_o$ is considered here due to its experimental relevance. For this regime electrons that have values of t_k that satisfy the condition $-\left(\tilde{\lambda}/4 + t_0 \Delta v_0 /2\right)/v_0 < t_k < \left(\tilde{\lambda}/4 + t_0 \Delta v_0 /2\right)/v_0$ are focused by the central well.

To find the temporal duration of the electron pulse at an arbitrary position after the thick lens is turned off, Eq. [S.26] is solved for t_k and substituted into Eq. [S.25]. The standard deviation of the compressed electron pulse at arbitrary time t_a is then,

$$\Delta t_a = \sqrt{\left\langle t_k^a\left(t_k, v_k\right)^2\right\rangle - \left\langle t_k^a\left(t_k, v_k\right)\right\rangle^2} = \sqrt{\frac{t_f^2\left(\tilde{\lambda}^2 + 4t_a^2\Delta v_o^2\right) + t_a^2\tilde{\lambda}^2 - 2t_f t_a \tilde{\lambda}^2}{48 t_f^2 v_0^2}}\,, \qquad \text{[S.27]}$$

where the limits for L_k are used along with an appropriately normalized square probability distribution for v_k. The time when the minimum pulse duration occurs is,

$$t_a = t_f \frac{\tilde{\lambda}^2}{\tilde{\lambda}^2 + 4t_f^2\Delta v_o^2} \simeq t_f\,, \qquad \text{[S.28]}$$

and for experimentally realistic parameters is equal to t_f. This implies that the thick lens does not image the initial temporal pulse; it temporally *focuses* the electrons that enter each individual well. Since there is no *image* in the thick lens regime, the minimum temporal duration is not determined by the magnification *M* as in the thin lens section, but is a given by,

$$\Delta t_f = \sqrt{\frac{t_f^2\tilde{\lambda}^2\Delta v_o^2}{12 v_0^2\left(\tilde{\lambda}^2 + 4t_f^2\Delta v_o^2\right)}} \simeq \frac{t_f \Delta v_o}{v_0 2\sqrt{3}}\,. \qquad \text{[S.29]}$$

It should be noted that neither the temporal focal length nor the temporal duration are directly dependent on the Doppler shifted wavelength $\tilde{\lambda}$, as long as the condition $t_o < v_0 \Delta t_o / \Delta v_o$ is met.

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