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Temporal lenses for attosecond and femtosecond electron pulses

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Here, we describe the “temporal lens” concept that can be used for the focus and magnification of ultrashort electron packets in the time domain. The temporal lenses are created by appropriately synthesizing optical pulses that interact with electrons through the ponderomotive force. With such an arrangement, a temporal lens equation with a form identical to that of conventional light optics is derived. The analog of ray diagrams, but for electrons, are constructed to help the visualization of the process of compressing electron packets. It is shown that such temporal lenses not only compensate for electron pulse broadening due to velocity dispersion but also allow compression of the packets to durations much shorter than their initial widths. With these capabilities, ultrafast electron diffraction and microscopy can be extended to new domains, and just as importantly, electron pulses can be delivered directly on an ultrafast techniques target specimen.

With electrons, progress has recently been made in imaging structural dynamics with ultrashort time resolution in both microscopy and diffraction (ref. 1 and references therein). Earlier, nuclear motions in chemical reactions were shown to be resolvable on the femtosecond (fs) time scale using pulses of laser light (ref. 2 and references therein), and the recent achievement of attosecond (as) light pulses (for recent reviews, see refs. 3-6) has opened up this temporal regime for possible mapping of electron dynamics. Electron pulses of femtosecond and attosecond duration, if achievable, are powerful tools in imaging. The “electron recombination” techniques used to generate such attosecond electron pulses require the probing electron to be created from the parent ions (to date no attosecond electron pulses have been delivered on an arbitrary target) and for general applications it is essential that the electron pulse be delivered directly to the specimen.

In ultrafast electron microscopy (UEM) (7), the electron packet duration is determined by the initiating laser pulse, the dispersion of the electron packet due to an initial energy spread and electron-electron interactions (see, e.g., ref. 8). Because packets with a single electron can be used to image (1, 7), and the initiating laser pulse can in principle be made very short (<10 fs), the limiting factor for the electron pulse duration is the initial energy spread. In photoelectron sources this spread is primarily due to the excess energy above the work function of the cathode (8), and is inherent to both traditional photocathode sources (9) and optically induced field emission sources (10-13). Energy-time uncertainty will also cause a measurable broadening of the electron energy spread, when the initiating laser pulse is decreased below ~10 fs. For ultrafast imaging techniques to be advanced into the attosecond temporal regime, methods for dispersion compensation and new techniques to further compress electron pulses to the attosecond regime need to be developed.

A recent article by Baum and Zewail (14) has proposed a new technique for compressing free electron packets, from durations of hundreds of femtoseconds to tens of attoseconds, using spatially dependent ponderomotive potentials. The numerical results showed that a train of attosecond pulses can be created and used in ultrafast electron imaging. Because they are generated independent of the target they can be delivered to a specimen for studies of transient structures and electronic excitations on the attosecond time scale. In reference (14), the proposed compression concept was examined using numerical, electron trajectory calculations. The deflection of electrons [as in the Kapitza–Dirac effect (15)] by the ponderomotive potential of intense lasers (16) and the diffraction (17) of electrons in standing waves of laser light have been observed, and so is the possibility (described through computer modeling) of spatial/temporal focusing with combined time-dependent electric and static magnetic fields (18).

This article develops the “temporal lens” description that analytically expresses how ponderomotive compression can be used to both compensate for the dispersion and magnify (in this case compress) the temporal duration of electron packets. We obtain simple lens equations that have analogies in optics and the results of “electron ray optics” of temporal lenses reported here are entirely consistent with the findings of ref. 14, but now allow for analytical expressions and for the design of different schemes using geometrical optics. Here, we consider 2 types of temporal lenses: thin and thick.

For the realization of the temporal thin lens, a laser beam with a Laguerre–Gaussian transverse mode, radial index ρ = 0 and azimuthal index l = 0 [or, in common nomenclature, a “donut” mode (19–21)], is used. In the center of the donut mode, electrons will experience a spatially varying ponderomotive potential (intensity) that is approximately parabolic. This potential corresponds to a linear spatial force that, for chirped electron pulses, can lead to compression from hundreds of femtoseconds to <10 fs. The second type, that of a thick lens, which is the concept outlined in ref. 14, is based on the use of 2 counterpropagating laser beams to produce a spatially dependent standing wave that copropagates with the electrons. A train of ponderomotive potential wells are produced at the nodes of the standing wave, leading to compression but now with much “tighter focus” (thick lens). Because the electron copropagates with the laser fields the velocity mismatch is no longer a problem (14). Here, analytical expressions are derived showing that this lens has the potential to reach foci with attosecond duration, in agreement with the results of ref. 14. Finally, we discuss methods for creating tunable standing waves for attosecond pulse compression, and techniques for measuring the temporal durations of the compressed pulses. Space-charge dispersed packets of electrons that have a linear spatial velocity chirp (22, 23) may also be compressed with the temporal lenses described here.

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Results and Discussion

Preliminaries: Temporal Lens Concepts. All electron sources, both cw and pulsed, have an initial energy spread. For pulsed electron sources this is particularly relevant as electron packets created in a short time disperse as they propagate. The initial energy spread leads to an initial spread in velocities. These different velocities cause the initial packet to spread temporally, with the faster electrons traveling a further distance and the slower electrons traveling a shorter distance in a given amount of time. The dispersion leads to a correlation between position (along the propagation direction) and electron velocity (see Fig. 1). The linear spatial velocity “chirp” can be corrected for with a spatially dependent linear impulsive force (or a parabolic potential). Thus, if a pulsed, spatially dependent parabolic potential can be made to coincide appropriately with the dispersed electron packet, the slow trailing electrons can be sped up and the faster leading electrons can be slowed down. The trailing electrons, now traveling faster, can catch the leading electrons and the electron pulse will thus be compressed.

Consider a packet of electrons, propagating at a speed $v_0$ along the x axis, with a spread in positions of $\Delta x_0 = v_0 \Delta t_0$ at time $t = t_0$; see Fig. 1. At $t = 0$, a potential of the form $U(x) = \frac{1}{2}Kx^2$ interacts with the electron packet for a duration $\tau$ in the lab frame. The waist, or spatial extent of the potential (temporal lens) is chosen to be $w$, whereas the duration $\tau$ is chosen such that it is short compared with $w/v_0$. When this condition is met the impulse approximation holds, and the change in velocity is $\Delta v = -\pi m (dU(x)/dx) dx = -\pi Kx/m$, for $x < w$, where $m$ is the electron mass. After the potential is turned off, $t > \tau$, the trailing electrons will pass through the same position, $x_\text{f} = (v_0 + \Delta v)t_\text{f}$, at the focal time $t_\text{f} = -x/\Delta v = m/(K\tau)$. To include an initial velocity spread around $v_0$ (due to an initial $\Delta x_0$), consider electrons that all emanate from a source located at a fixed position on the x axis. An electron traveling exactly at $v_0$ will take a time $t_0$ to reach the center of the potential well at $x = 0$. Electrons leaving the source with other velocities $v_0 + \Delta v$ will reach a location $x = v_0 t_0$ at $t = 0$ (Fig. 2). The image is formed at a location where electrons traveling with a velocity $v_0$ and a velocity $v_0 + \Delta v$ intersect, which is, when $v_0 t_0 = x + (v_0 + \Delta v + v_k)t$. The image time $t_\text{i}$ is then $t_\text{i} = -x/\Delta v + v_k$.

For the object time, $t_\text{o} = x/v_0$, image time $t_\text{i} = -x/(\Delta v + v_k)$, and the focal time $t_\text{f} = -x/(\Delta v + v_k)$ and the temporal lens equation holds,

$$\frac{1}{t_\text{o}} + \frac{1}{t_\text{i}} = \frac{1}{t_\text{f}}$$

Ray tracing for optical lenses is often used to visualize how different ray paths form an image, and is also useful for visualizing how temporal lenses work, see Fig. 2. As derived in later sections the magnification $M$ is defined as the ratio of the electron pulse duration ($\Delta t$) at the image position to the electron pulse duration ($\Delta t_0$), and is directly proportional to the ratio of the object and image times ($t_\text{i}/t_\text{o}$) and distances ($-x_i/x_o$).

Femtosecond Thin Lens. In polar coordinates a Laguerre–Gaussian (LG) mode has a transverse intensity profile given by, $I(r,\phi) = I_0 \exp\left(\frac{1}{2} r^2 \exp(-2r^2 w^2)\right)/2\pi w^2$, where $w$ is the waist of the beam and $I_0$ the maximum intensity (19–21). This “donut” mode has an intensity maximum located at $r = \sqrt{2}w/2$ with a value of $I_0 = 2E_P\sqrt{\ln 2}/\pi(\omega^2)$ where $E_P$ is the energy of the laser pulse and $\omega$ is the full-width-at-half-maximum of the pulse duration, assuming a Gaussian temporal profile given by $\exp(-4ln2(t/\tau)^2)$. The ponderomotive energy $U_p(x)$ is proportional to intensity (24).

$$U_p(x) = \frac{1}{2} e^2 \lambda^2 \exp(1) I_0 \ln 2 \pi \sqrt{\ln 2 \pi} x^2 = \frac{1}{2} Kx^2,$$

where $m$ is the electron mass, $e$ is the electron charge and $\lambda$ the central wavelength of the laser radiation and replacing $r$ with $x$. Near the center of the donut mode focus ($r < w$) the intensity distribution is approximately parabolic, and hence the ponderomotive energy near the donut center is also parabolic. In analogy with a mechanical harmonic oscillator, the quantity in the square brackets of Eq. 2 can be referred to as the “stiffness” $K$; it has units of $J/m^2$, and at 800 nm has the numerical value of $K = 3.1 \times 10^{36} E_p/(w^2)$. For this parabolic approximation to be applicable, the spatial extent of the dispersed electron pulse, at
\[ t = 0, \Delta x(0) = v_0 \Delta t_o + \Delta v_i \Delta t_i \text{ must be much smaller than the laser waist, where the object velocity spread is } \Delta v_i = \Delta E / \sqrt{2mE} \] (8).

The effect of this parabolic potential on an ensemble of electrons emitted from a source will now be analyzed (for detailed derivation, see SI Appendix). 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 \( x_0 = -v_0 t_o \). Assuming a single donut-shaped laser pulse is applied at \( t = 0 \), and centered at \( x = 0 \), the ensemble is then influenced by the potential \( U(x) = (1/2)Kx^2 \). The \( k^{th} \) electron in the ensemble has an initial velocity \( v_0 + v_K \) and emission time \( t_o + t_K \). Using a Galilean transformation to a frame moving with velocity \( v_0 \), the propagation coordinate \( x \) (lab frame) is replaced with the moving frame coordinate \( \tilde{x} = x - v_0 t \). At \( t = 0 \) the potential exists for the ultrashort laser pulse duration \( \tau \), giving the electron an impulse (or ‘kick’) dependent on its instantaneous position in the parabolic potential. In both frames, the position of the electron at \( t = 0 \) is \( x_k(0) = \tilde{x}_k(0) = -v_0 t_k + v_k t_o - v_0 t_o \), where \( x_k(t) \) and \( \tilde{x}_k(t) \) are in the lab and moving frames, respectively. Using the impulse approximation the electron trajectory immediately after the potential is turned off becomes,

\[ \tilde{x}_k(t) = v_0 t + \tilde{x}_k(0)(1 - t/t_o) \] (3)

where \( t_o = m/(K \tau) \) is the focal time. The electron trajectories, before and after \( t = 0 \), can be plotted in both frames to give the equivalent of a ray diagram, Fig. 3. Electrons emitted at the same time, i.e., \( t_k = 0 \), but with different velocities, will meet at the image position, \( \tilde{x}_k = 0 \) in the moving frame at the image time \( t \). The image time is found by setting \( \tilde{x}_k(t_k) = 0 \), from Eq. 3, with \( t_k = 0, \tilde{x}_k(t_k) = v_0 t_k + v_k t_o(1 - t_k/t_o) = 0 \), which is equivalent to the lens equation, Eq. 1: \( t_o^{-1} + t_k^{-1} = t_i^{-1} \).

An expression for the magnification can be obtained when electrons that are emitted at different times \( t_k \) and different velocities \( v_k \) are considered (see SI Appendix for detailed derivation). If the magnification is defined as \( M = -t_i/t_o \), then the temporal duration at the image time becomes,

\[ \Delta t_i = M \Delta t_o \] (4)

where \( \Delta t_i \) and \( \Delta t_o \) are the duration of the electron packet at the object and image time, respectively. Durations achievable with a thin temporal lens follow from Eq. 4.

An experimentally realistic temporal lens would use a 50-fs, 800-nm laser pulse with 350 \( \mu \)J energy, focused to a waist of \( w = 25 \mu m \). These values result in a stiffness of \( K = 5.5 \times 10^{-9} \) N/m and a focal time of \( t_i = 0.3 \) ns; \( t_o = m/(K \tau) \). If the lens is applied 10 cm from the source, electrons emitted at \( v_0 = c/10 \) (3 keV) would have an object time of \( t_o = x_0/v_0 = 0.1/(c/10) = 3.0 \) ns. Using the temporal lens equation, Eq. 1, \( t_i \) is obtained to be 0.33 ns. Hence, a magnification of \( M = -t_i/t_o = 0.1 \). Consequently, a thin temporal lens can compress an electron packet with an initial temporal duration of \( \Delta t_o \approx 100 \) fs, after it has dispersed, to an image duration of \( \Delta t_i \approx 10 \) fs. Although the example presented here is for 3 keV electrons, the thin lens approximation holds for higher energy electrons as long as \( \tau \) is chosen to be short compared with \( w/v_0 \). Experimentally, the thin temporal lens can be used in ultrafast diffraction experiments (25), which operate at kHz repetition rates with lasers that typically possess power that exceeds the value needed for the ponderomotive compression.

**Attosecond Thick Lens.** In the previous section it was analytically shown that free electron packets can be compressed from hundreds to tens of femtoseconds using a temporal thin lens, which would correspond to a magnification of \( \approx 0.1 \). However, the analytic solutions used rely on an impulse approximation that may not generally hold for the temporal lens described in ref. 14. For example, ref. 14 numerically showed that a \( \approx 300 \)-fs duration electron packet can be compressed to a train of \( \approx 15 \) as pulses, which corresponds to a magnification of \( M = 5 \times 10^{-5} \); the associated image time of \( t_i = 2 \) ps is comparable with the laser pulse duration of \( \tau = 0.3 \) ps. Because \( M \ll 1 \) and \( t_i \) and \( \tau \) are the same order, it is unclear whether the impulse approximation would accurately describe the results.

Another issue that needs to be addressed is how an initial
source velocity spread and dispersion affects the ability for the temporal lens in ref. 14 to compress electron packets. The simulation done in ref. 14 predicted \( \approx 15 \) as electron pulses, where the duration was determined solely by the sinusoidal deviation from the optimal parabolic potential, and did not allow the electron packet to disperse before compression. In the thin lens case, an initial velocity spread results in a nonzero pulse duration even with a parabolic potential. Developing a model incorporating an electron packet that is allowed to disperse before encountering a parabolic potential will answer whether or not an initial velocity spread is detrimental to attosecond pulse compression.

The attosecond compression scheme presented in ref. 14 relies on the presence of a standing wave that copropagates with an ultrashort electron pulse. The copropagating standing wave is created by using 2 different optical frequencies, constructed by having a higher frequency (\( \omega_1 \)) optical pulse traveling in the same direction as the electron packet and a lower frequency (\( \omega_2 \)) traveling in the opposite direction. When the optical frequencies \( \omega_1, \omega_2 \) 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 (14) (see Fig. 4). If the electron has a velocity \( v_0 = c/3 \), and \( \omega_1 = 2\omega_2 \) then the copropagating standing wave has a ponderomotive potential of the form (24),

\[
U_p(x) = \frac{1}{2} \left( \frac{\epsilon^2 \tilde{E}_0^2}{8 \pi mc^2} \right) \cos^2(\tilde{k}x),
\]

where \( \tilde{E}_0 \) is the peak electric field, \( \tilde{k} \) the Doppler shifted wavelength (14). The envelopes of the laser pulses are ignored in this derivation, but they can be engineered so that the standing wave contrast is optimized (26).

To find an analytic solution in the thick lens geometry, each individual potential in the standing wave is approximated by a parabolic potential that matches the curvature of the sinusoidal potential, \( U_p(x) = (1/2)(\epsilon^2 \tilde{E}_0^2/(2mc^2)) |x|^2 \equiv (1/2)Kx^2 \). Using the exact solution to the harmonic oscillator the focal time is,

\[
t_f = \cot(\omega_p \tau)/\omega_p + \tau,
\]

where \( \omega_p = \sqrt{Km} \) and \( \tau \) is the duration that the lens is on. For \( \tau \rightarrow 0 \), \( t_f \rightarrow m/(K\tau) \), which is identical to the thin lens definition. The image time, \( t_i \), has a form,

\[
t_i = (1/\omega_p^2 + t_0 \tau - t_0 \tau + \tau^2)/(t_0 - t_f + \tau),
\]

and after the 2 assumptions, \( \tau \rightarrow 0 \) and \( t_0 \gg 1/(t_0 \omega_0^2) \) becomes equivalent to Eq. 1, the lens equation: \( t_0^2 + t_i^2 = t_f^2 \).

The standard deviation of the compressed electron pulse at arbitrary time \( t_0 \) is,

\[
\Delta t_s = \sqrt{\frac{4t_0^4 + 4t_0^2 \Delta t_{\theta}^2 + t_0^2 \Delta \mu^2 - 2t_0 \mu \Delta \mu}{48t_0^2 \mu_0^2}},
\]

which is valid for an individual well (detailed derivation in the SI Appendix). The time when the minimum pulse duration occurs is \( t_0 = \lambda^2/(4c^2 \Delta \mu^2) \approx t_f \) 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. Because 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,

\[
\Delta t = \sqrt{\frac{t_f^4 \lambda^4 \Delta \mu^2}{12\mu_0^2(\lambda^2 + 4c^2 \Delta \mu^2) \mu^2}} \approx t_f \Delta \mu_0/\mu_0^2 2/3,
\]

which is equivalent to Eq. 6 in ref. 14. It should be noted that neither the temporal focal length nor the temporal duration are directly dependent on the Doppler shifted wavelength \( \lambda \), as long as the condition \( t_0 < \nu_0 \Delta \mu_0/\mu_0 \) is met.

An example illustrates what temporal foci are obtainable. A source emits electrons with an energy distribution of 1 eV and a temporal distribution of 100 fs. Electrons traveling at \( v_0 = c/3 \) and having an energy \( E = 31 \) keV gives a velocity distribution of \( \Delta v_0 = 1670 \) m/s. If the distance between the source and the temporal lens is 10 cm, \( t_0 = 1.0 \) ns is less than \( \nu_0 \Delta v_0/\mu_0 = 6.0 \) ns, satisfying the condition \( t_0 < \nu_0 \Delta \mu_0/\mu_0 \) and Eq. 9 is then valid. If the 2 colors used for the laser beams are 520 nm and 1040 nm, the Doppler-shifted wavelength is \( \lambda = 740 \) nm. For a laser intensity of \( 3 \times 10^{22} \) Wcm\(^{-2} \) (available with repetition rates up to megahertz), the oscillation frequency in the potential well is \( \nu_0 = 2 \times 10^{13} \) rad/s, which gives a focal time of \( t_f \approx 1 \) ps. With these parameters, Eq. 9 gives a temporal duration at the focus \( \Delta t \approx 5 \) as. To support this \( \approx 5 \) as electron pulse, time-energy uncertainty demands an energy spread of \( \approx 50 \) eV. The ponderomotive compression impacts an energy spread to the electron pulse, which can be estimated from \( \Delta E \approx m\nu_0(2\nu_0) \), giving \( \approx 50 \) eV similar to the uncertainty limit. This \( \Delta E \) is very small relative to the accelerating voltage in microscopy (200 keV) and only contributes to a decrease of the temporal coherence. In optical spectroscopy such pulses can still be used as attosecond probes despite the relatively large \( \Delta E \) when the chirp is well characterized (27). Combining the anharmonicity broadening of 15 as (as discussed in ref. 14), we conclude that ultimately temporal pulse durations in the attosecond regime can be reached.

**Tunable Thick Lens.** In the temporal thick lens case, the use of \( \omega_1 \) and \( \omega_2 \) to create a copropagating standing wave requires \( v_0 = c/3 \). However, the velocity of the electrons, \( v_0 \), can be tuned by changing the angle of the 2 laser pulses. A copropagating standing wave can still be obtained by forcing the Doppler-shifted frequencies of both tilted laser pulses to be equal. A laser pulse that propagates at an angle \( \theta \) with respect to the electron propagation direction has a Doppler-shifted frequency \( \omega = \gamma\omega_0(1 \pm (\nu/c)\cos \theta) \), where \( \gamma \) is the angular frequency in the lab frame, \( \beta = \nu/c \) is the electron velocity, and \( \nu = 1/\sqrt{1 - \nu^2/c^2} \) (28). When the 2 laser pulses are directed as shown in Fig. 4, a copropagating standing wave occurs for an electron with a velocity \( v_0 = (c/k_1 - k_2)/(\nu_0 \cos \theta_1 + k_2 \cos \theta_2) \), where the laser pulse traveling with the electron packet has a wave vector of magnitude \( k_1 \) and makes an angle of \( \theta_1 \) with the electron propagation axis; the second laser pulse traveling against has a wave vector magnitude of \( k_2 \) and angle \( \theta_2 \) in the lab frame. An
electron moving at $v_0$ will see a standing wave with an angular frequency,

$$\dot{\omega} = \frac{2(\cos \theta_1 + \cos \theta_2)}{2\cos \theta_1 + \cos \theta_2} v_0 (1 - \beta),$$  \hspace{1cm} [10]$$

where $2\lambda = k_1 = 2k_2$ for experimental convenience, $\omega = kc$, and the wavelength is $\lambda = 2\pi/\tilde{\lambda} = 2\pi/\lambda$.

The standing wave created with arbitrary angles $\theta_1$ and $\theta_2$ will be tilted with respect to the electron propagation direction, which will temporally smear the electron pulse. This tilting of the standing wave can be corrected for by constraining the angles $\theta_1$ and $\theta_2$ to be: $\theta_2 = \arcsin(2\sin \theta_1)$. For $\theta_1 = 15^\circ$ (forcing $\theta_2 \approx 31^\circ$), electrons with velocity $v_0 = 0.36c$ ($E \approx 33$ keV) see a standing wave. A 1 eV electron energy distribution at the source gives a velocity distribution of $\Delta v_0 = 1630$ m/s, at 33 keV. Using the same laser intensity as in the thick lens case, and the new $v_0$ and $\Delta v_0$, the condition $t_0 < v_0\Delta t_0/\Delta v_0$ is still satisfied, allowing Eq. 10 to be used, resulting in a duration at the focus of $\Delta t_0 \approx 4.6$ as. Using the tunable thick lens makes the experimental realization more practical, allowing for easy optical access and electron energy tuning, while at the same time keeping $\Delta t_0$ approximately the same. For additional tunability, an optical parametric amplifier can be used so that the laser pulse frequencies are not restricted to $\omega$ and $2\omega$.

**Detection Proposals.** The ability to create electron pulses with duration from ~10 fs to ~10 as raises a challenge regarding the measuring of their duration and shape. Two different schemes are presented here for measuring pulses compressed by thick and thin temporal lenses. For measuring the thin lens compressed electron packet, the focused packet could be intersected by a laser pulse with a Gaussian spatial focus (Fig. 5). An optical delay line would control the time delay between the measuring laser pulse and the compressed electron packet. As the time delay, $\Delta t$, is varied, so is the average energy of the electrons, as shown in Fig. 5. If the delay time is zero, then the average electron energy will be unaffected, because there is no force. If the delay line is changed so that the Gaussian pulse arrives early (late), then the average energy will decrease (increase). The change in the average energy depends on the duration of the electron pulse, and the intensity of the probing laser pulse. If the electron pulse is longer than the duration of the measuring laser pulse, then the change in the average energy will be reduced. The steepness of the average energy as a function of delay time, $E(\Delta t)$, is a direct measure of the electron pulse duration, and by using femtosecond-pulsed electron energy loss spectra (29) this scheme can be realized.

For the thick lens a similar method was proposed in ref. 14 and described in more detail here. At the focal position and time of the compressed temporal electron packet, a second copropagating potential is introduced. The positions of the individual wells in the second copropagating standing wave can be moved by phase shifting one of the two laser beams that create the probing potential (Fig. 5). By varying the phase shift, the potential slope (and hence the force) that the electrons encounter at the focus is changed. If no phase shift is given to the probing standing wave, no average energy shift results. When a phase shift is introduced, the electrons will be accelerated (or decelerated) by the slope of an individual well in the standing wave, and as long as the phase stability between the electrons and the probing standing wave is appropriate, attosecond resolution can be achieved. As the electron pulse duration becomes less than the period of the standing wave, the average electron energy change increases. The electron temporal duration of the compressed electron packet can be determined directly by the steepness of the $E(\phi)$ curve.

**Conclusion and Outlook.** The attosecond electron imaging regime is possible only when the electron pulses are compressed to the attosecond duration. Current efforts in employing attosecond technologies (primarily electron recombination) require the target specimen to be part of the electron generation, causing the pump and the probe to be coupled. Here, we have described 2 temporal lens designs and obtained analytical expressions consistent with the proposal of Baum and Zewail (14). With a thin temporal lens, the capability to image an electron packet temporally is illustrated with compressibility (or magnification) of the pulse duration from 100 fs to 10 fs. A thick temporal lens can focus an initial dispersed electron packet (hundreds of femtoseconds) into a train of attosecond pulses; this case is discussed in ref. 14 using electron trajectory simulations.

The ultrashort pulses produced by the temporal lenses described here, or by using other compression schemes (18, 30, 31), will have a wide range of applications in UEM (1, 7) and femtosecond-pulsed electron energy loss spectroscopy (29). Although single electron packets develop a linear velocity chirp due to dispersion, packets with linear chirp can be imaged with the temporal lenses described in this article. For an initial packet with an ellipsoidal distribution of electrons, electron-electron interactions (space charge) result in a linear spatial chirp in the propagation direction (23). This linear chirp due to space charge can be used to increase the temporal resolution of ultrafast...
electron diffraction experiments through energy filtering (22) or pulse compression with radio-frequency fields (32). In principle, the concepts presented here can also be used to compress space-charge dominated pulses of electrons extending the range of applicability for temporal lenses.

Finally, temporal lenses have the potential to increase by orders of magnitude the quantum degeneracy of an electron packet (33). The first use of electron quantum degeneracy in free space—the demonstration of the Hanbury Brown and Twiss effect for electrons (antibunching)—reached a degeneracy value of $10^{-4}$ (34, 35) and in other sources could be $10^{-6}$ or less. This was done with a continuous electron beam from a field emission tip (34). By using pulsed sources and temporal lenses the degeneracy factor can become significantly closer to the quantum limit of one because of the increased current density during the pulse duration. In this limit, the potential for “electron quantum optics” becomes real.

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Femtosecond Thin Lens Derivation. In polar coordinates a Laguerre-Gaussian \( \text{LG}_0^1 \) mode has a transverse intensity profile given by,

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

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,

\[
I_0 = 2 \frac{E_p}{w^2 \tau} \sqrt{\frac{\ln 2}{\pi^3}},
\]

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

\[
U_p(r) = \frac{e^2 \lambda^2}{8\pi^2 me_0 c^3} I(r, \varphi),
\]

where \( m \) is the electron mass, \( e \) is the electron charge and \( \lambda \) 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,

\[
U_p(x) = \frac{1}{2} \left[ \frac{e^2 \lambda^2}{\pi^3 me_0 c^3 w^4 \tau} \sqrt{\frac{\ln 2}{\pi}} \right] x^2 \equiv \frac{1}{2} K x^2,
\]

in which \( x \) is used instead of \( r \). For this parabolic approximation to be applicable, the spatial extent of the dispersed electron pulse, at \( t = 0 \),
\[ \Delta x(0) = v_0 \Delta t_o + \Delta v_o t_o, \]  
[S.5]

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

\[ K \approx 3.1 \times 10^{-36} \frac{E_p}{w^4 \tau}. \]  
[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,

\[ x_k(t) = -v_0 t_o + V_k (t - T_k), \]  
[S.7a]

and in the moving frame, the trajectory is,

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

At \( t = 0 \) the potential exists for the ultrashort laser pulse duration \( \tau \), 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 $x_k(0) = \bar{x}_k(0) \equiv x^*_k = -v_{k,t_0} + v_{k,t_f} - v_{k,t_k}$ and the acceleration is $a = -(\partial U_p(x) / \partial x) / m = -(K/m)x^*_k$. The change in velocity is then,

$$\Delta v = a\tau = -(K\tau/m)x^*_k = -x^*_k/t_f,$$  \hspace{1cm} [S.8]

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

$$x_k(t) = x^*_k + \left( V_k - x^*_k/t_f \right) t = v_k t + v_{o,t_k} + \left( -v_{o,t_k} + v_{k,t_0} - v_{k,t_k} \right) \left( 1 - t/t_f \right),$$  \hspace{1cm} [S.9a]

$$\bar{x}_k(t) = x^*_k + \left( v_k - x^*_k/t_f \right) t = v_k t + \left( -v_{o,t_k} + v_{k,t_0} - v_{k,t_k} \right) \left( 1 - t/t_f \right),$$  \hspace{1cm} [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, $x_i = v_{i,t_i}$ in the lab frame at the image time $t_i$, whereas in the moving frame, the image position is located at $\bar{x}_k = 0$ with the same image time. The image time is found by setting $\bar{x}_k(t_i) = 0$, from Eq. [S.9b], with $t_k = 0$,

$$\bar{x}_k(t_i) = v_k t_i + v_{k,t_o} \left( 1 - t_i/t_f \right) = 0,$$  \hspace{1cm} [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, $t_o$, it takes the $k^{th}$ electron to reach an arbitrary position $x_a$ after the lens. By equating $x_k(t)$ to $v_{o,t_o}$, or,

$$v_{o,t_a} = v_k t + v_{o,t} + \left( -v_{o,t_k} + v_{k,t_0} - v_{k,t_k} \right) \left( 1 - t/t_f \right),$$  \hspace{1cm} [S.11]

and then solving for time gives,
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 ($v_k = t_k = 0$),

$$t_k^{(t_k, v_k)} = t(t_k, v_k) - t(0,0),$$

and has the form,

$$t_k^{(t_k, v_k)} = t_o t_f \frac{v_k t_o \left( \frac{1}{t_f} - \frac{1}{t_o} - \frac{1}{t_f} \right)}{t_f (v_0 + v_k) + t_k (v_0 + v_k) - v_k t_o}.$$

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

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

The relationship between $t_k$ and $t_k^{(t_k, v_k)}$ 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 $t_k^{(t_k, v_k)}$ and $t_k$ is simply $t_k^{(t_k, v_k)} = -t_i t_k / t_o$. If the magnification is defined as $M = -t_i / t_o$ then the temporal duration at the image time becomes,

$$\Delta t_i = M \Delta t_o,$$

where $\Delta t_o$ and $\Delta t_i$ 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 ($\omega_l$)
optical pulse traveling in the same direction as the electron packet and a lower frequency \( (\omega_2) \) traveling in the opposite direction. When the optical frequencies \( \omega_1, \omega_2 \), 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,

\[
U_p = \frac{1}{2 \left( \frac{e^2 \tilde{\lambda}^2 E_0^2}{8\pi^2 mc^2} \right)} \cos^2(\tilde{k}x),
\]

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, \( U_p(x) = \frac{1}{2} \left[ \frac{e^2}{2mc^2} E_0^2 \right] x^2 \equiv \frac{1}{2} K x^2 \). The motion of the electron in the parabolic potential is given by the solution to the harmonic oscillator, \( x(t) = C_1 \sin(\omega_p t) + C_2 \cos(\omega_p t) \), where \( \omega_p = \sqrt{K/m} \) 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 \( \tau \). The initial conditions are \( \tilde{x}_k(0) = -v_o t_k + v_k t_o - v_k t_k \) and \( \tilde{v}_k(0) = v_k \) in the co-propagating frame, which gives \( C_1 = v_k / \omega_p \), 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,
\[ \tilde{x}_k(t, v_k, t_k) = \frac{v_k}{\omega_p} \sin \left( \omega_p t \right) + \tilde{x}_k(0) \cos \left( \omega_p t \right), \]  
[S.17a]

\[ \tilde{v}_k(t, v_k, t_k) = v_k \cos \left( \omega_p t \right) - \omega_p \tilde{x}_k(0) \sin \left( \omega_p t \right). \]  
[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,

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

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

with,

\[ \tilde{x}_k(\tau) = \frac{(v_k / \omega_p) \sin (\omega_p \tau) + \tilde{x}_k(0) \cos (\omega_p \tau)}{\omega_p} \]  
[S.18c]

\[ \tilde{v}_k(\tau) = v_k \cos (\omega_p \tau) - \omega_p \tilde{x}_k(0) \sin (\omega_p \tau). \]  
[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,

\[ t_f = \frac{1}{\omega_p} \cot \left( \omega_p \tau \right) + \tau. \]  
[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,

\[ t_i = \frac{1}{\omega_p^2} + \frac{t_o t_f - t_f \tau + \tau^2}{t_o - t_f + \tau}. \]  
[S.20]

Additionally, the equation for the image time can be rewritten in the form,
\[
\frac{1}{t_i} + \frac{t_f - \tau}{t_{i,f} + 1/\omega_p^2 - t_f + \tau^2} = \frac{t_o}{t_o t_f + 1/\omega_p^2 - t_f + \tau^2}, \tag{S.21}
\]

which after the two assumptions, \( \tau \to 0 \) and \( t_o \gg 1/(t_f \omega_p^2) \) 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 \( v_o t_a \),

\[
v_o t_a = -\ddot{x}_k(\tau) + (\ddot{v}_k(\tau))(t - \tau), \tag{S.22}
\]

and solving for time gives,

\[
t(t_k, v_k) = \frac{v_o t_a + \ddot{x}_k(\tau) + (\ddot{v}_k(\tau))\tau}{\ddot{v}_k(\tau)}. \tag{S.23}
\]

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,

\[
t_k^a(t_k, v_k) = t(t_k, v_k) - t(0, 0), \tag{S.24}
\]

and has the form,

\[
t_k^a(t_k, v_k) = t_o t_f \cos(\omega_p \tau) \left( \frac{1}{t_f} - \frac{1}{t_o} \left( \frac{1 - \omega_p^2 \cot(\omega_p \tau) - 1}{t_o t_f \omega_p^2} \right) \right) + t_f \left( \frac{1}{t_o} - \frac{1}{t_f} \right) \frac{v_k v_0 + v_0 + v_k (t_f - t_o - \tau)}{v_0 (t_f - \tau) + v_k (t_f - t_o - \tau) \cos(\omega_p \tau)}. \tag{S.25}
\]

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

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

\[
L_k = v_k t_o - t_k v_o, \quad [S.26]
\]

for a time, \(t = 0\), or when the lens is first turned on. For electrons to be focused by the center well the condition, \(-\tilde{\lambda}/4 < v_k t_o - t_k v_o < \tilde{\lambda}/4\) 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 \(t_o < v_o \Delta t_o/\Delta v_o\) and the second is when \(t_o > v_o \Delta t_o/\Delta v_o\), where \(\Delta t_o\) and \(\Delta v_o\) are the full widths of the independent temporal and velocity distributions at \(t_o\). Only the first regime, \(t_o < v_o \Delta t_o/\Delta v_o\) is considered here due to its experimental relevance. For this regime electrons that have values of \(t_k\) that satisfy the condition \(-\tilde{\lambda}/4 + t_o \Delta v_o/2)/v_o < t_k < \tilde{\lambda}/4 + t_o \Delta v_o/2)/v_o\) 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,

$$\Delta t_a = \sqrt{\left\{ t_k^2 \left( t_k, v_k \right) \right\} - \left\{ t_k^2 \left( t_k, v_k \right) \right\}^2} = \sqrt{\frac{t_f^2 \left( \tilde{\lambda}^2 + 4t_f^2\Delta v_o^2 \right) + t_k^2\tilde{\lambda}^2 - 2t_f t_o \tilde{\lambda}^2}{48t_f^2v_o^2}}, \quad [S.27]$$

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,

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

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,

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

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 $t_o < v_o \Delta t_o/\Delta v_o$ is met.

