# Theory for time-resolved measurements of laser-induced electron emission from metal surfaces

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We present theory for the laser-assisted photoelectric effect (LAPE) from metal surfaces in combined extreme ultraviolet attosecond and near-infrared femtosecond pulses. We discuss how LAPE with short pulses is used to make time-resolved measurements. We propose the use of reference states located outside the metal surface to allow for measurements of the absolute travel time of electronic wave packets formed inside the metal and to the surface. Finally, we study the variation in the magnitude of laser-induced sidebands with the length of the laser pulse.

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# I. INTRODUCTION

Laser pulses have long been used as a tool for timeresolved measurements in pump-probe experiments. Currently, ever shorter laser pulses are becoming available. It is now possible to create single laser pulses with ultraviolet frequencies and attosecond duration [1,2] and pulses in the near infrared containing only a single optical cycle [2]. These very short laser pulses open the possibility of examining electronic processes on their natural time scale.

One tool for examining fast processes is ionization by a short, extreme ultraviolet (xuv) pulse into the continuum dressed by a relatively strong near-infrared field, a process known as the laser-assisted photoelectric effect (LAPE). This is used both as a tool to study properties of the pulses [3] and to study ultrafast processes within atoms [4] and molecules [5]. Recently experiments have appeared involving LAPE from metal surfaces [6–9]. Other recent works on short-pulsed laser interactions with surfaces include Refs. [10–12].

The purpose of this paper is first to present a simple theory of LAPE from metal surfaces and to discuss how this can be used for time-resolved measurement (see Fig. 1). We discuss a single-active-electron model and present two regimes of xuv pulses that both give destinct signatures of the electron emission time. This latter point is essential for timeresolved measurements. We then discuss the necessity of a reference state for measurement and propose the use of electronic states located outside the metal surface to allow for absolute measurements of electronic travel times to the surface. Previously, only differences in travel time have been measured using reference states located inside the metal [7]. We present a calculation of emission spectra using a simple model for both the surface and the reference state, and show that indeed a reference state located outside the surface will give direct access to absolute time measurements. The third purpose of the paper is to investigate what time resolution can be obtained in the limit considered recently experimentally [6,8,9], where the pulse length of the xuv source is comparable to or longer than the laser period.

The paper is organized as follows. In Sec. II, we present our theory and discuss two limiting cases for time-resolved measurements: (i) the xuv pulse is much shorter than the laser period and (ii) the xuv pulse is comparable to or longer than the laser period. In Sec. III, we discuss the need for reference states for time-resolved measurements and propose reference states located outside the surface, allowing for direct time measurement of electron motion to the surface. In Sec. IV, we present a simple model of the surface and the reference state. In Sec. V, we present the results of our calculations which show that the proposed image charge reference state would give direct access to absolute travel times from within and to the surface. In Sec. V, we also consider the case of relatively long xuv pulses and show that sidebands can be resolved in the wings of the single xuv-induced photopeak for xuv pulse durations down to a single laser cycle at 800 nm. We also show that close to maximum sideband intensity is obtained already with a ten-cycle assisting pulse; the shorter the assisting pulse the better the temporal resolution. Section VI concludes.



FIG. 1. (Color online) Illustration of the LAPE process. (a) The spatial intensity profile I(z) of the laser pulses at a given instant of time. In vacuum, the intensity is constant in space. In the metal, the intensity drops due to the final skin depth. (b) The electrons are lifted into the continuum by the short xuv pulse. The peak of the xuv pulse is delayed relative to the peak of the laser pulse. The electrons that are freed from the metal surface by the xuv pulse are therefore exposed to more or less of the laser pulse depending on this variable time delay. In the situation depicted here, the electrons can originate from either the conduction band or from a reference state located outside the metal. In the figure, the reference state is depicted as an image charge state, but could also be a suitable state in an adsorbed atom (see Secs. III and IV).

## **II. THEORY**

The laser-assisted photoelectric effect is an ionization process in which two different fields are present: A low-intensity xuv field with a sufficiently high photon energy for direct ionization and a relatively high-intensity laser field which has insufficient photon energy for direct ionization, but instead dresses the continuum in the sense that the freed electrons exchange energy with the laser field. In our study of the radiation-induced dynamics from metal surfaces, we take as the effective Hamiltonian [atomic units ( $|e| = \hbar = m_e = 1$ ) are used throughout unless otherwise stated]

$$H = \sum_{i} h_{i},\tag{1}$$

with

$$h_i = \frac{p_i^2}{2} + V(\vec{r}_i) + V_L(\vec{r}_i, t) + V_X(\vec{r}_i, t), \qquad (2)$$

where  $V(\vec{r})$  is the effective one-electron potential,  $V_L(\vec{r},t) = \vec{r} \cdot \vec{E}_L(t)$  the interaction with the laser field, and  $V_X(\vec{r},t) = \vec{r} \cdot \vec{E}_X(t)$  the interaction with the xuv field. Both electromagnetic interactions are in the length gauge and dipole approximation. Below we drop the subscript *i* in our description of the one-electron Hamiltonian.

If the electrons are in thermal equilibrium with temperature T, the occupation of the electron states at energy E is determined by the Fermi-Dirac distribution function

$$f^{\rm FD}(E,T) = \frac{1}{1 + \exp[(E - \mu)/(k_B T)]},$$
(3)

where  $\mu$  is the chemical potential and  $k_B$  is Boltzmann's constant. In the limit  $T \rightarrow 0$ , the distribution approaches a step function, which is unity for energies below the chemical potential,  $E < \mu$ , and zero above. For metals, the chemical potential at temperatures below a few thousand kelvins is approximated by the Fermi energy.

The differential probability for electron emission from all the initial states into a final continuum state with momentum  $\vec{k}_f$  is given by

$$\frac{dP}{d\vec{k}_f} = \rho_e \sum_{\text{initial states}} f^{\text{FD}}(E_i, T) |T_{fi}|^2, \qquad (4)$$

where  $\rho_e = 2$  gives the density of states including the spin multiplicity of the levels for momentum-normalized continuum states. The index *i* runs through all initial states and the weight of the states is given by the Fermi-Dirac distribution, Eq. (3). The transition matrix element for a transition from state  $\Psi_i$  to state  $\Psi_f$ ,  $T_{fi}$  is modeled by

$$T_{fi} = -i \int_{-\infty}^{\infty} dt \langle \Psi_f(\vec{r}, t) | V_X(\vec{r}, t) + V_L(\vec{r}, t) | \Psi_i(\vec{r}, t) \rangle, \quad (5)$$

where  $V_L(\vec{r},t)$  gives the contribution from above-threshold ionization (ATI) and  $V_X(\vec{r},t)$  gives the direct ionization by the attosecond pulse. In the high-energy region reached by absorption of a single xuv photon, the ATI contribution is highly suppressed due to the fact that many photons are needed to reach that region and the *T*-matrix element is accurately described by

$$T_{fi} = -i \int_{-\infty}^{\infty} dt \langle \Psi_f(\vec{r}, t) | V_X(\vec{r}, t) | \Psi_i(\vec{r}, t) \rangle.$$
 (6)

In Eq. (6) the initial wave function is given by

$$\Psi_i(\vec{r},t) = \psi_i(\vec{r})e^{iE_it},\tag{7}$$

with  $E_i$  the energy of the initial state. The time dependence of the final state wave function is approximated by

$$\Psi_f(\vec{r},t) = \psi_f(\vec{r})e^{i\Phi(\vec{r},t)},\tag{8}$$

where  $\Phi(\vec{r},t)$  is the phase of a free electron in the presence of the laser field, the Volkov phase, including the energy dependence of the final state

$$\Phi(\vec{r},t) = -\int^{t} \frac{[\vec{k}_{f} + \vec{A}_{L}(t')]^{2}}{2} dt' + \vec{r} \cdot \vec{A}_{L}(t), \qquad (9)$$

and where  $\vec{A}_L$  is the integral of the laser  $\vec{E}_L$  field, given by

$$\vec{A}_{L}(t) = -\int_{-\infty}^{t} dt' \vec{E}_{L}(t').$$
 (10)

#### A. xuv pulses much shorter than the laser period

A very short xuv pulse allows a direct mapping of the laser pulse onto the energy spectrum of the emitted electrons, since the final momentum of the electron depends on the value of the laser field through the momentum shift  $-\int_{t_0}^{\infty} \vec{E}(t) dt$ , where  $t_0$  denotes the instant of ionization [3]. With a short laser pulse, this mapping gives a signature of the instant when the electron enters the laser field. This has been used to measure the difference in travel time to the surface for electrons emitted from the conduction band and electrons emitted from the more localized 4f bands of tung-sten [7].

The limit of a very short xuv pulse is defined by

$$\tau_X \ll \frac{2\pi}{\omega_L},\tag{11}$$

where  $\tau_X$  is the duration of the xuv pulse and  $\omega_L$  is the angular frequency of the laser. Since the LAPE interaction term  $V_X(\vec{r},t)$  in Eq. (6) is only nonzero for a short time in this limit, the laser field in the phase of the final state wave function of Eq. (8) can be considered constant over  $\tau_X$ . As we shall see below, this allows an analytical evaluation of the time integral in Eq. (6).

The attosecond pulse is modeled by the vector potential

$$\vec{A}_X(t) = \vec{\epsilon} A_{0X} \exp\left(-\frac{(t-t_0)^2}{2\tau_X^2}\right) \sin(\omega_X t + \phi)$$
$$\sim \vec{\epsilon} A_{0X} \exp\left(-\frac{(t-t_0)^2}{2\tau_X^2}\right) e^{-i(\omega_X t + \phi)}, \qquad (12)$$

where  $\vec{\epsilon}$  is the linear polarization,  $A_{0X}$  is the amplitude,  $t_0$  denotes the peak of the attosecond pulse and the instant of

ionization into the laser-dressed continuum,  $\omega_X$  is the angular frequency, and  $\phi$  is the carrier-envelope phase difference, which we set equal to zero. The rotating wave approximation has been used to eliminate the non-energy-conserving term. The latter is a valid approach for the presently considered weak xuv fields. In the limit where  $A_L$  is considered constant over the xuv pulse, the time integration can then be carried out to give

$$|T_{fi}|^{2} = 2\pi A_{0X}^{2} \Delta E(t_{0})^{2} e^{-\tau_{X}^{2} [\omega_{X} - \Delta E(t_{0})]^{2}} \\ \times |\langle \psi_{f}(\vec{r})| \vec{\epsilon} \cdot \vec{r} e^{-i\vec{A}_{L}(t_{0}) \cdot \vec{r}} |\psi_{i}(\vec{r})\rangle|^{2},$$
(13)

with  $\Delta E(t_0) = \{ [\vec{k}_f + \vec{A}_L(t_0)]^2 / 2 - E_i \}$ . We have tested this approximation against the full numerical integration of Eq. (6) for the simple jellium model presented in Sec. IV. For xuv pulses with a duration  $\leq 150$  as, the approximation is very accurate for the presently considered 800 nm laser pulses. Note that, once this approximation has been made, only a single spatial integration is required and in the jellium model, even this integration can be done analytically [see Eq. (22)]. Monitoring the energy of the streaked electron, the spectrum allows for a determination of the time  $t_0$  that an electron was released into the laser field. This can be used for time measurements, using a variable delay between the xuv and the laser pulse in a pump-probe experiment.

# B. xuv pulses longer than the laser period

When the xuv pulse duration is longer than the period of the laser field, a series of sidebands appear on both sides of the central xuv-induced photopeak. The magnitude of these peaks depends on the laser intensity at the instant the electron emerges into the laser-dressed continuum. Hence the sideband magnitude gives a signature of the emission time and this effect has been used to measure the Auger lifetimes in atoms [13,14] and very recently in adsorbates [9]. Sidebands have been observed in LAPE from metal surfaces as well [6,8], and this allows for similar time-resolved measurements. For example, in Ref. [9], the lifetime of the  $4d^{-1}$  core level of Xe on a Pt(111) surface was measured using a technique based on these sidebands. In this experiment sidebands were measured as a function of time delay for direct and Auger electrons from Xe. The difference in sideband maximum as a function of time delay is a measure of the lifetime of the core level.

In the limit of long monochromatic assisting laser pulses, certain features of the spectrum of emitted electrons are known analytically [15]. The main features are a central peak, centered around  $E_f = \omega_X + E_i$ , with  $E_i$  the initial state energy, and a series of sidebands separated by the laser photon energy. The ratio of the first sideband to the central peak without the assisting laser field is given by the square of the generalized Bessel function of integer order 1,

$$J_1^2 \left( \frac{\vec{p} \cdot \vec{E}_0}{\omega_L^2}, \frac{U_p}{2\omega_L} \right), \tag{14}$$

where  $\vec{p}$  is the final momentum,  $\vec{E}_0$  the electric field amplitude of the laser field, and  $U_p = E_0^2 / 4\omega_L^2$  the ponderomotive

potential. In the weak laser field limit, the ponderomotive potential can be neglected and the generalized Bessel function reduces to the ordinary Bessel function, since  $J_1(x,0) = J_1(x)$ . When the electric field and the kinetic energy of the free electron are sufficiently small, the Bessel function may be Taylor expanded to first order and the ratio of the first sideband to the single laser-field-free photopeak is

$$J_{1}^{2} \left( \sqrt{\frac{2E_{0}^{2}E_{f}}{\omega_{L}^{4}}} \right) \sim \frac{E_{0}^{2}E_{f}}{2\omega_{L}^{4}},$$
(15)

where we have assumed that the final momentum and the polarization of the driving laser field are parallel; see also the discussion in Refs. [6,8]. In the limit of very short xuv pulses these sidebands vanish: As the pulse gets temporally shorter, it gets spectrally wider and, in the limit of short pulses, the xuv pulse is spectrally wider than the laser frequency. When the sidebands do appear, however, the fact that their magnitude depends on the intensity of the laser field allows for a pump-probe experiment very similar to the streaking experiment. Varying the delay between the laser and the xuv pulse maps out the intensity profile of the laser and this allows for time-resolved measurements [9].

In contrast to the streaking described above, we are here working in a low-intensity regime, perhaps more suitable for studying surface dynamics as space-charge effects and contributions from above-threshold ionization are less significant.

# III. REFERENCE STATES FOR ABSOLUTE TIME MEASUREMENTS

To make any kind of time-resolved measurement, we need a signal to start and a signal to stop our clock. In the monitoring of the dynamics of electrons these signals will usually be in the form of electrons emitted from different steps in the process under investigation. For example, in the case of laser-assisted Auger decay, the signal to start the clock will be the direct emission of the inner-shell electron and the signal to stop the clock will be the Auger electron [9,13,14]. In other measurements it might not be as obvious how to choose these signals.

In a recent experiment, electron emission from the conduction band of tungsten was used as the start signal and emission from the lower-lying band of 4f electrons was used for stopping the clock [7]. This gives a measurement of the difference in travel time to the surface of electron wave packets formed in the conduction band and in the 4f band. It does not, however, give us the absolute travel time for either of the two. In order to measure the absolute travel time, we propose the use of electronic reference states located *outside* the metal. Electrons emitted from these states will be created directly in the infrared field and present a natural time zero for travel time measurements.

We propose two different kinds of reference states for these absolute measurements. First, classical electrodynamics tells us that a charge outside a conducting surface sees an attractive potential. Quantum mechanically, this gives rise to a series of bound states located outside the surface [16]. For metals with a band gap around the continuum edge, these states can have fairly long lifetimes of several tens of femtoseconds for the ground state and much longer for higherlying states [17,18]. This should be sufficient time to make it possible to prepare a system in a state with a high population in these states.

Another and perhaps more easily realizable system is to adsorb a few atoms onto the metal surface. The atomic species should be chosen so that electrons emitted from a certain level in the adsorbed atoms do not coincide in energy with any of the electrons from the metal. Then the electrons emitted from this atomic state serve as a time-zero signal (they are already at the surface) and if the amount of adsorbed atoms is sufficiently small then it should not alter the travel time for electrons emitted from the surface.

#### **IV. MODEL**

#### A. Jellium model

To model a metallic surface, we use the very simple jellium model

$$V(\vec{r}) = -V_0 \Theta(-z), \tag{16}$$

where  $\Theta(z)$  is the Heaviside step function and where  $V_0$  is the sum of the Fermi energy and the work function for the metal. The energy eigenstates are analytically known for this simple potential, which has been extensively used in the description of ion-surface collisions (see, e.g., Refs. [19–24] and references therein) and to study the interaction of short pulses with metal surfaces [25]. For simplicity we restrict the study to grazing incidence with the linear polarization of the laser and the xuv pulse perpendicular to the surface. In this case, the dimensionality of the problem reduces to one.

For the spatial part of the initial and final wave functions of Eqs. (7) and (8), we use the eigenstates corresponding to the potential of Eq. (16) with the appropriate boundary conditions [26],

$$\psi_i^k(z) = \frac{\Theta(-z)}{\sqrt{2\pi}} \left( e^{ikz} + \frac{k - i\gamma}{k + i\gamma} e^{-ikz} \right) + \frac{\Theta(z)}{\sqrt{2\pi}} \frac{2k}{k + i\gamma} e^{-\gamma z},$$
(17)

$$\psi_f^k(z) = \frac{\Theta(-z)}{\sqrt{2\pi}} \sqrt{\frac{k}{q}} \frac{2q}{k+q} e^{ikz} + \frac{\Theta(z)}{\sqrt{2\pi}} \sqrt{\frac{k}{q}} \left( e^{iqz} - \frac{k-q}{k+q} e^{-iqz} \right),$$
(18)

where k is the momentum within the metal,  $q = \sqrt{k^2 - 2V_0}$  is the momentum outside the metal, and  $\gamma = \sqrt{2V_0 - k^2}$ . In this case, the sum over initial states becomes an integral and the differential emission probability given by Eq. (4) is now given by

$$\frac{dP}{dE_f d\Omega} = 2q_f \int dk_i f^{\rm FD}(E_{k_i}, T) |T_{fi}|^2, \qquad (19)$$

where the integration is over all initial bound states,  $k_i$ , and where we have used  $dE = k^2 dk d\Omega$ .

For the interaction inside the metal, we use the classical skin depth  $\delta$  to include the weakening of the electromagnetic field [27],

$$E_X(z,t) = E_X(t)\exp(z/\delta), \quad z < 0, \tag{20}$$

where  $\delta = \sqrt{2/\mu \sigma \omega_X}$  with  $\mu$  the permeability and  $\sigma$  the conductivity of the metal. For this case, the spatial part of Eq. (13) should be replaced with

$$M_{fi}(t_0) = \left| \langle \psi_f(z) | z [\Theta(z) + \Theta(-z) \exp(z/\delta)] e^{-iA_L(t_0)z} | \psi_i(z) \rangle \right|^2,$$
(21)

and explicit calculation with the wave functions in Eqs. (17) and (18) gives the analytical result

$$\begin{split} M_{fi}(t_0) &= \frac{1}{2\pi} \sqrt{\frac{k_f}{q_f}} \bigg[ \frac{2k_i}{k_i + i\gamma} \bigg( \{\gamma + i[A_L(t_0) + q_f]\}^{-2} \\ &- \frac{k_f - q_f}{k_f + q_f} \{\gamma + i[A_L(t_0) - q_f]\}^{-2} \bigg) \\ &- \frac{2q_f}{k_f + q_f} \bigg( \{\delta^{-1} - i[A_L(t_0) + k_f - k_i]\}^{-2} \\ &+ \frac{k_i - i\gamma}{k_i + i\gamma} \{\delta^{-1} - i[A_L(t_0) + k_f + k_i]\}^{-2} \bigg) \bigg]. \end{split}$$
(22)

# B. Image charge reference state

We now consider an equally simple model for a reference state (Sec. III) which makes absolute travel time measurements possible, namely, the image charge state outside the metal. From classical electrodynamics, it is known that a charge Q, outside a metal surface induces a charge in the surface, which again sets up an electric field in the region outside the metal. This gives rise to a potential

$$V_{\rm img}(\vec{r}) = -\frac{|Q|}{4z}, \quad z > 0,$$
 (23)

where z is the distance to the surface, and a corresponding series of bound states, like one-dimensional hydrogen states, with energies

$$E_n^{\rm img} = -\frac{Q^2}{32n^2},$$
 (24)

where n is the principal quantum number. Here, for simplicity, we are concerned only with the first image charge state, which has a simple exponential falloff,

$$\psi_{\rm img}(z) = \frac{1}{\sqrt{2}} \exp(-z/4), \quad z > 0.$$
 (25)

This is of course an idealized picture of the image charge states, but it shows that they are largely indifferent to the nature of the metal, which is a particular advantage of these states when they are considered them as reference states. Much more advanced models of the image charge states exist [16], but the conclusions we will draw in this paper do not depend on the specific form of the electronic states and we will therefore settle for this simple model.

# **V. RESULTS**

Earlier, experiments have been made measuring the different travel times for electrons from 4f states and valence states in tungsten. The difference was measured to be 110 attoseconds in [7]. We propose to use the image charge states to make an absolute measurement of the travel time. The electron from the image charge state is in the laser pulse immediately upon ionization, and therefore gives a reference point for an absolute measurement. The expected travel time from the valence states is about 60 as and from the 4f states about 150 as [7].

We propose to perform a LAPE streaking experiment from a metallic surface with reference states located outside the surface. If the interest is in determining the absolute travel time for relatively deeply bound core electrons, for example, 4f states as in Ref. [7], only a single type of measurement with populated image charge reference states suffices. In this case the reference states and the core states are energetically well separated and by monitoring the temporal displacement of the streaking spectra we may infer the travel time. If, on the other hand, the interest is in the travel time of the valence electrons to the surface, for each delay, two types of measurement should be performed, one from the plain metal surface and one from the same metal surface, but following a third pulse which resonantly excites some of the valence electrons to the image charge state. This third pulse should have a photon energy corresponding to the difference between the Fermi level of the metal and the binding energy of the image charge state. In this way, the reference signal for the time measurement actually comes from a different measurement.

A calculation of the LAPE spectra of a thermal metal surface modeled as described in Sec. IV A and a surface on which electrons have been excited to the image charge state described in Sec. IV B are presented in Fig. 2. In the case of a fully populated image charge state, this state is dominant in the spectrum. The valence band states are of course still occupied, but the density of states is sufficiently low that the peak from the image charge state is orders of magnitude greater. As seen by comparing the top and middle figures in Fig. 2, this results in a sharper peak, as the signal from the image charge state is not broadened by the width of the valence band. The energy of the image charge state electrons is higher, as they are more loosely bound compared to the valence electrons.

These figures are made with a delay of the valence band electrons of 2 atomic units ( $\sim$ 50 as). It is very difficult to infer this time delay from the spectra in the top and the middle of Fig. 2, and therefore, as in Ref. [7], a center-ofenergy analysis has been carried out from the two spectra, plotting in the bottom of Fig. 2 the quantity

$$\int dE \frac{dP}{dE}(E,t)E - E_{\rm NTO},$$
(26)

where  $E_{\rm NTO}$  is the center of energy when the two pulses have no temporal overlap (NTO). The center-of-energy analysis displays the order of shift that can be expected.



FIG. 2. (Color online) LAPE spectra from a thermally populated metal surface (top) and from a metal surface with a fully populated image charge state (middle). At the bottom a center-of-energy analysis of the above spectra is shown [see Eq. (26)]. The dashed (red) curve corresponds to the fully populated image charge state. The signal from the image charge state has been shifted by 50 as compared to the conduction band signal, shown as the full (blue) curve.

#### A. Sidebands and pulse length

As discussed in Sec. II A, sidebands appear in the longpulse limit that can be used for time-resolved measurements. On the other hand, in order to obtain a good temporal resolution, laser pulses with a short duration are preferred. It is therefore interesting to examine how short the pulses can be made before the sidebands disappear. In the limit of very short xuv pulses they disappear as seen directly from Eq. (13). In the limit of very short infrared pulses, the carrier frequency is no longer well determined and the sidebands are seen to vanish as well.

In the present studies, we have used a near-infrared laser pulse with a central wavelength of 800 nm and an intensity of  $3.2 \times 10^{10}$  W/cm<sup>2</sup> as in Ref. [9] and an xuv pulse with a central photon energy of 90 eV. In the long-pulse limit, this set of parameters leads to a sideband magnitude of  $\approx 13\%$ 



FIG. 3. (Color online) Electron emission spectrum from a jellium surface. We have used a near-infrared laser pulse with a central wavelength 800 nm and an intensity of  $3.2 \times 10^{10}$  W/cm<sup>2</sup> as in Ref. [9] and an xuv pulse with a central photon energy of 90 eV. The first sideband can be seen as the shoulder to the contribution from the main peak. The laser pulse length is three optical cycles and the duration of the xuv pulse equals a single laser cycle.

(see Fig. 4) of the single laser-field-free xuv-induced peak. The aim of this study is to determine how these sidebands depend on the pulse length for shorter pulses. The sidebands are still almost fully developed when the duration of the xuv pulse is comparable to the period of the laser field. To illustrate this point, we show in Fig. 3 the emission spectrum from a jellium surface using an xuv pulse with a duration of a single laser cycle, and a three-cycle laser pulse.

In the figure, the first sideband is seen to form even with this very short laser pulse duration.

Since the laser pulse length is the key factor in achieving a good temporal resolution when using the sidebands as a signature of emission time, it is interesting to study how the magnitude of the sidebands depends on the duration of the laser pulse. The magnitude of the first sideband versus the duration of the laser pulse is shown in Fig. 4. It is seen that the signal is saturated already at pulse lengths around ten optical cycles. In order to achieve good temporal resolution, it is therefore preferable to use pulses no longer than this. We also see from the figure that for the present set of laser parameters it is not very accurate to apply the first-order approximation to the Bessel function in Eq. (15).

#### VI. CONCLUSION

In conclusion, we have presented a theory for timeresolved measurements using the laser-assisted photoelectric effect with combined atto- and femtosecond pulses. We have shown that laser-induced sidebands appear around the xuvinduced photopeak for xuv pulses of duration down to the order of a laser cycle at 800 nm. For such sources, the sideband magnitude reaches its maximum for laser pulses containing  $\gtrsim 10$  cycles, meaning that optimum temporal resolution in this case is obtained with ten cycles or even shorter



FIG. 4. (Color online) Magnitude of the first sideband compared to the single laser-field-free xuv-induced photopeak as a function of the laser pulse length in number of cycles for a 800 nm pulse with peak intensity  $3.2 \times 10^{10}$  W/cm<sup>2</sup>. The full increasing curve is the result of a fully time-dependent calculation within our jellium model. The two horizontal lines are the long-pulse-limit results, given in Sec. II B; the full line being the Bessel function and the dashed line the first-order Taylor expansion, both given in Eq. (15).

laser pulses if the sideband magnitude is sufficient to give a measurable signal.

We have discussed the role of a reference state for these measurements. LAPE from metal surfaces is a new field [6-9] and we have proposed the use of reference states located outside the metal to allow absolute measurements of travel times to the surface. An electron emitted from a state outside the metal gives a natural reference for measuring delays originating from the travel through the metal. Both sparse adsorbates on the surface and image charge states can be used as this reference. The absolute travel time to the surface will be valuable when using the conventional threestep model for photoelectron emission, where the emission process is separated into (i) optical excitation within the solid, (ii) transport to the surface, and (iii) escape to the vacuum [28]. Measurements of travel times will also provide valuable data for testing surface and solid state models, since they have signatures of both the mean free path in the solid and the high-energy region of the electronic band structure. While the present model provides an accurate description of the light-matter interaction and captures essential temporal issues of the processes, we have considered only the conduction band electrons and in order to describe deeper-lying levels a more detailed model of the solid is required. Such a model might also allow for a description of adsorbates on the surface and show how the electronic states in adsorbates are different from gas phase atoms and molecules.

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