Attoclock setup with negative ions: A possibility for experimental validation

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Attoclock setup with negative ions: A possibility for experimental validation

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The presumed connection in attoclock setups between the electron tunneling time and its asymptotic momentum has triggered vigorous debates. In neutral atomic systems investigated so far, the action of the long-range Coulomb potential on the electron momentum hinders extracting the effect of the tunneling process on the offset angle of the asymptotic electron momentum. We propose and investigate an attoclock experiment using F^- or Cl^- to circumvent this difficulty. Our calculations, performed with realistic laser parameters in the tunneling regime, could be checked directly against experiment and predict essentially a "zero" offset angle with no detectable effect of polarization.

The concept of "tunneling" by an electron through a potential barrier in strong-field ionization (SFI) with few-cycle pulses has attracted much attention in recent years. Numerous theoretical and experimental studies have been, and continue to be, performed using the so-called "attoclock setup" [1–10]. The basic idea is to relate the offset angle in the photoelectron momentum distribution (PMD) to the time it might take the electron to tunnel through a potential barrier. Several assumptions are being made to do so, and it is fair to say that the concept remains controversial [3–14].

It is generally accepted that using atomic hydrogen as the target represents the cleanest attoclock experiment, since it avoids a potential effect of electron correlation on the tunneling process, and it also enables a highly accurate theoretical description. Indeed, such an experiment was recently performed [7]. There is excellent agreement between experiment and theory when the nonrelativistic time-dependent Schrödinger equation (TDSE) is solved in full dimensionality. Details can be found in Ref. [7], where it is also explained that, in practice, elliptically (albeit with an ellipticity close to 1.0) rather than circularly polarized light needs to be used, so that a major and a minor polarization axis can be properly defined to which the offset angle is then related. Other experiments were performed with many-electron atoms such as helium [1, 3], argon [3, 4], and krypton [4], and calculations for such systems, necessarily performed with some level of approximation, were also carried out. All these experiments reported, as expected, a non-zero offset angle. The detailed origin of this angle, however, remains under discussion.

The reason for the ongoing debate is the fact that even for atomic hydrogen as the target the "cleanliness" of the study is perturbed, not only by the well-known difficulty in performing experiments with this target, but even more importantly by the fact that the Coulomb interaction between the ejected electron and the residual ion (a bare proton in the case of atomic hydrogen) strongly affects the offset angle – in addition to ambiguities in how this angle is actually defined (e.g., as the maximum in the polarization plane of the light [6], considering different geometric distributions of the momentum [15], or after some integration over momentum bins with sufficient signal-to-noise ratio [7]). Theoretical "solutions" to this problem have been the attempt to somehow figure out or eliminate the effect of the Coulomb potential after tunneling, for example, by classical backpropagating [16] or by switching off the Coulomb tail by using a Yukawa potential of "sufficiently short range".

The Yukawa potential $V_a(r) = -Z\exp(-r/a)/r$ used in [6] is, of course, unrealistic for experimental studies in atomic physics with neutral targets, since its asymptotic behavior is incorrect at both ends. In studies on atomic hydrogen, the "way out" for theorists has been to first pick the range parameter $a = 1$ a.u. and then adjust the charge Z seen by the electron near the origin by requiring that the $1s$ binding energy of -0.5 a.u. be reproduced ($Z = 1.94$ for $a = 1$ a.u). However, already for $a = 2$ a.u., one starts detecting a nonzero offset angle, and as $a \to \infty$, the results approach gradually those of the Coulomb potential [17]. Hence, one can possibly "dial in" the desired result making the use of such a potential rather arbitrary. Furthermore, since the ionization probability using the Yukawa potential is almost 1000 times smaller than in hydrogen at 10^{14} W/cm^2, mostly the exponential tail of the ground state wavefunction in the Yukawa potential is ionized [6, 14], thereby questioning whether tunneling across the entire barrier ever occurred.

Therefore, it seems virtually impossible to disentangle the roles played by the tunneling process and the long-range Coulomb force in the deflection of the electron momentum. To our knowledge, the following alternative to avoid this difficulty in attoclock experiment has not yet been investigated to date; use *anions*, ideally F^- or Cl^-, in the attoclock setup. An experiment on F^- was successfully conducted already more than a decade ago [18] to test the validity of the Keldysh-Faisal-Reiss theory, as it also required the absence of a long-range Coulomb potential. This was not an attoclock experiment, however, since it employed a very long pulse (∼100 fs), did not resolve the PMD in the polarization plane (it was assumed symmetric), and used circularly instead of elliptically polarized light. In [18], F^- was produced and accelerated in

a beam following discharge of CF_4. See also [19] for a review on anion sources. The idea of using anions was also recently proposed in the study of weak-field one-photon ionization time delay [20].

The main advantage of studying SFI with anions is the weakness of the long-range induced dipole potential between the neutral product and the ejected electron, as it drops much faster than the Coulomb potential. As a result, the photoelectron might still be considered as virtually free in the electromagnetic field almost immediately after tunneling. Most importantly, the assumption could be checked not only by calculations, but also by performing experiments with systems that have different dipole polarizabilities (see below).

We suggest the specific choice of F^- and Cl^-, because these atomic ions have the highest known affinity, or ionization potential, I_p, namely 3.40 eV for F^- [21] and 3.61 eV for Cl^- [22]. Therefore, SFI of these ions with few-cycle mid-range infrared pulses ($\lambda \geq 1500$ nm), which have successfully been produced by various experimental groups (e.g., [4, 18, 23, 24]), would require the absorption of at least 4 to 5 photons — a necessary condition for the ionization process to be considered tunneling. On the other hand, tunneling ionization in H^- would require very long wavelengths, well beyond current laser capabilities, due to the low affinity of only 0.75 eV [25].

We illustrate our proposal using F^- as the target by employing a single-active electron (SAE) approach that can grasp the essence of the tunneling process. As in most theoretical work [1–10] on attoclock experiments, electron correlation is not accounted for in this approach, however, Majety and Scrinzi [26] managed to compute, using fully correlated calculations, the PMD in helium (under a well-justified preponderance rule approximation) and found no appreciable effect of correlations in the asymptotic electron momentum.

Unless specified otherwise, we use atomic units (a.u.) below. We consider an effective one-electron hamiltonian $\hat{H}_0 = \hat{T} + \hat{V}$, where $\hat{T}$ is the kinetic energy operator and the potential operator $V(\boldsymbol{r}, t)$, as a function of the electron position $\boldsymbol{r}$ and the time t, takes the form

$$V(\boldsymbol{r}, t) = -\frac{Z}{r} e^{-r/a} - \left[\frac{\alpha_d \boldsymbol{E}(t) \cdot \boldsymbol{r}}{r^3} + \frac{\alpha_d}{2r^4}\right] \xi_c(r). \quad (1)$$

Here $Z = 9$ is the nuclear charge and $\alpha_d = 3.76\ a_0^3$ (with $a_0 = 0.529 \times 10^{-10}$ m denoting the Bohr radius) is the dipole polarizability of F [27]. The parameter a in (1) determines the range of the mean-field Coulomb potential created by the nucleus and the other nine electrons. This potential is "seen" by the active electron while bound. While its form could be improved by combining several terms of this kind to fit to an *ab initio* potential from a sophisticated structure calculation, the simple version used here is sufficient to illustrate the principal idea. After the detachment, the short-range potential is ultimately overcome at large distances by the induced dipole potential of the F atom. The latter potential results from the polarization of the F atom, which is in part produced by the external electric field $\boldsymbol{E}(t)$ (first term in brackets) and by the field produced by the ejected electron (second term). Including the effect of the time-dependent core polarization in SFI with circular light has, to our knowledge, not yet been studied quantum mechanically. We use the function $\xi_c(r) = 1 - \exp[-(r/r_c)^6]$, with a cutoff parameter r_c, which leads to a rapid convergence towards the correct physical form of the induced dipole potential for $r > r_c$ and to a smooth decay towards zero as $r \to 0$. Hence, in contrast to a Yukawa potential for neutral systems, which is physically incorrect on both ends, our $V(\boldsymbol{r}, t)$ has the correct asymptotic behavior for $r \to 0$ and $r \to \infty$.

The potential (1) reproduces the affinity of the $2p$ orbital in F^-, without external field, if $a \leq 0.5255$ a.u. (where the maximum corresponds to the limiting case of no induced dipole potential, i.e., $r_c \to \infty$) and $r_c \geq 0.6529$ a.u. (where the minimum corresponds to the limiting case of no short-range potential, i.e., $a \to 0$). Consequently, one has the flexibility to reproduce the $2p$ orbital energy for different choices of the set of parameters $\{a, r_c\}$. As demonstrated in more detail below, the main conclusions of our study are independent of the particular choices of a and r_c used to reproduce F^- affinity. In order to produce a potential as realistic as possible, we added the criterion that the parameters should reproduce not only the $2p$ ionization energy in F^-, but also give a reasonable binding energy for the $2s$ orbital. Since, to our knowledge, the latter has neither been measured nor calculated reliably to date for F^-, we used the binding energy of the $2s$ orbital in neutral fluorine (≈ 21.04 eV [28]), which should be a reasonable approximation. This leads to $a = 0.5061$ a.u. and $r_c = 1.492$ a.u.

The intensity range to be considered in an actual experiment should be large enough to be performed under the conditions of the tunneling regime, i.e., with $\gamma \lesssim 1$, where $\gamma = \omega\sqrt{2I_p}/E_{\max}$ is the Keldysh parameter [29] for the angular frequency ω, ionization potential I_p, and maximum electric field strength $E_{\max}$. In fact, Ni *et al.* [16] recently demonstrated that the major part of the ionization process comes from tunneling already for $\gamma \lesssim 1.5$. In the case of F^-, using 1500 nm elliptical light with ellipticity $\varepsilon = 0.84$, $\gamma = 1.17$ for an intensity as low as 10^{13} W/cm^2, and $\gamma = 0.74$ at 2.5×10^{13} W/cm^2, respectively. Hence, one can well be within the tunneling regime with realistic laser parameters.

Another essential condition in the study of tunneling is for the field not to become strong enough for over-the-barrier ionization (OBI) to occur. The above model potential for F^- gives an intensity for OBI always larger than 3×10^{13} W/cm^2, and specifically larger than 4.5×10^{13} W/cm^2 for our potential. Therefore, the tunneling conditions in F^- are well fulfilled for a wide range of intensities that can be produced experimentally.

In this respect, this system appears even better suited for the attoclock setup than atomic hydrogen, where the tunneling condition cannot be completely fulfilled since $\gamma \approx 1$ when the OBI intensity is reached. The experimental study of SFI in F^- [18] used a 1500 nm pulse with an intensity of $I = 2.6 \times 10^{13}$ W/cm^2.

We solve the TDSE

$$-\mathrm{i}\frac{\partial\psi}{\partial t} = [\hat{H}_0 + \hat{H}_{int}]\psi, \tag{2}$$

where ψ is F^- wavefunction and the field-atom interaction hamiltonian $\hat{H}_{int} = \boldsymbol{p}\cdot\boldsymbol{A}(t)$ is expressed in the velocity gauge. Here $\boldsymbol{p}$ is the electron momentum and

$$\boldsymbol{A}(t) = -\frac{E_0}{\omega}\, f(t)\, \frac{\varepsilon\cos(\omega t)\boldsymbol{e}_x + \sin(\omega t)\boldsymbol{e}_y}{\sqrt{1+\epsilon^2}} \tag{3}$$

is the potential vector for an N-cycle pulse with ellipticity ϵ, frequency $\omega = 0.0304$ a.u., period $T = 2\pi/\omega$, and envelope $f(t)$. The initial state is propagated from $t_{initial} = -NT/2$ to $t_{final} = NT/2$ using an accurate numerical method described in [7, 30, 31]. The pulse takes its maximum amplitude $E_0/\sqrt{1+\varepsilon^2}$ along the positive $\boldsymbol{e}_y$ axis at $t_0 = 0$, when the envelope reaches its maximum, $f(t_0) = 1$, and the potential vector is oriented along the x-axis, i.e., $\boldsymbol{A}(t_0) = A_x(t_0)\boldsymbol{e}_x$. In order to represent an initially unpolarized target, we perform calculations starting from the three initial projections of the magnetic moment of the $2p$ electron. Finally, the PMD $d^3\mathcal{P}(\boldsymbol{p})/d^3\boldsymbol{p}$ is computed in the xy-polarization plane of the light by projecting the wavefunction at the end of the pulse onto the e−F scattering state $|\boldsymbol{p}\rangle$.

Figure 1 shows the result of our calculation with the "nearly one-cycle" circularly polarized pulse ($\varepsilon = 1$) used by Torlina *et al.* [6] at an intensity $I_0 = 10^{13}$ W/cm^2. Note that this is actually a 2-cycle pulse with a very steep (half-cycle) ramp-on/ramp-off $\sin^4$ envelope function. Even though the pulse is unrealistic for current experimental setups for a number of reasons (too short, too steep, and circularly polarized, which prevents the definition of an experimental reference axis), we use it for comparison with [6]. Clearly, the PMD is centered along the positive p_x direction. This result corroborates the picture of an electron that tunnels at t_0 and further interacts with the field only, thereby scattering with a momentum $\boldsymbol{p} = -A_x(t_0)\boldsymbol{e}_x$ asymptotically.

We computed the offset angle $\theta_o^{(c)}$ in the circular case for different intensities. Among the various possible definitions of the offset angle, we chose $\theta_o^{(c)}$ as the angle leading to the maximum of $d^3\mathcal{P}(\boldsymbol{p})/d^3\boldsymbol{p}$ in the cartesian representation of the (p_x, p_y)-plane. The results, presented in Tab. I, reveal essentially a zero offset angle with very small deviations. The uncertainty is due to the finite momentum grid step used in our calculation. Applying the same procedure as [6] by looking instead at the maximum of $p^2 d^3\mathcal{P}(\boldsymbol{p})/d^3\boldsymbol{p}$ gives very small differences in the offset angle at a particular intensity but

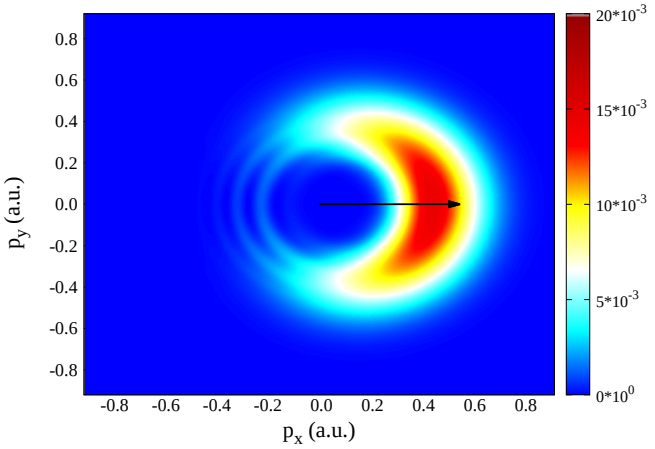

FIG. 1: PMD for a 1500 nm circularly polarized 2-cycle pulse with a $\sin^4$ envelope and a peak intensity $I_0 = 10^{13}$ W/cm^2. The arrow defines the positive x-axis.

leads to the same conclusion. Repeating the TDSE simulation with the Yukawa potential employed in [6] at a few intensities above 10^{14}W/cm^2 also resulted in offset angles with deviations from zero of the same order as in the F^- case. Our results are consistent with [6] where $\theta_o^{(c)}$ had an uncertainty of about $\pm 0.5°$. This finding also shows that tunneling ionization of an initial p-wave orbital with a centrifugal barrier does not seem to affect the offset angle. Finally, the fact that the offset angle becomes negative and decreases with intensity, is most likely due to the depletion of the F^- ground state (0.8% at $I_0 = 2\times 10^{13}$ W/cm^2), as shown in Refs. [6, 10], as well as frustrated tunneling [6, 32, 33].

We now move on to a more realistic pulse, similar to that used in the experiment of Satya *et al.* [7] on atomic hydrogen. Figure 2 shows our prediction for this case. From the beginning to the end, this is an elliptically polarized ($\varepsilon = 0.84$) 6-cycle pulse in the xy-plane with a $\sin^2$ envelope function and the y-direction as the major axis. The wavelength is kept at 1500 nm, and the peak intensity is 10^{13} W/cm^2. The offset angle θ_o^e of predominant ejection is clearly along the direction, in which the y-component of the linear momentum is close to zero. Due to the multi-cycle nature of the pulse, several fringes appear, and ejection is strong along both the positive and negative p_x axis. Nevertheless, θ_o^e should be evaluated for $p_x > 0$ as these electrons are associated with

I_0	0.5	1.0	1.5	2.0	2.5
θ_o^c	-0.4 ± 0.2	0.3 ± 0.2	-0.5 ± 0.3	-0.9 ± 0.1	-1.2 ± 0.1
θ_o^e	-0.5 ± 0.3	0.0 ± 0.2	0.6 ± 0.2	0.0 ± 0.2	0.5 ± 0.3

TABLE I: Offset angle (in degrees) for the circular $\theta_o^{(c)}$ and elliptical $\theta_o^{(e)}$ cases at different intensities given in units of 10^{13}W/cm^2 (see text for details).

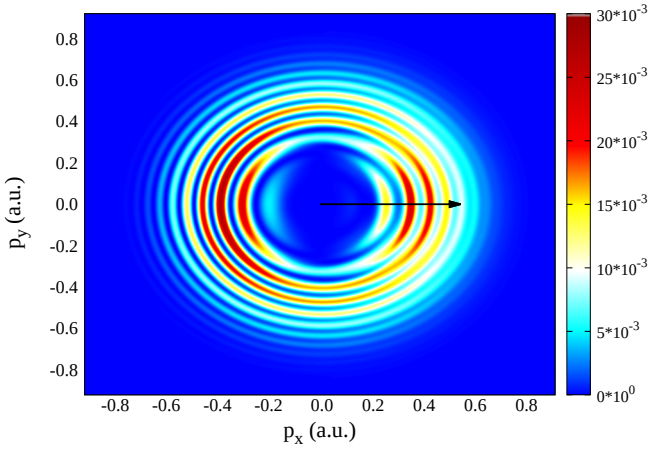

FIG. 2: PMD for a 1500 nm elliptically polarized ($\varepsilon = 0.84$) 6-cycle pulse with a $\sin^2$ envelope and a peak intensity $I_0 = 10^{13}$ W/cm^2. The arrow defines the positive x-axis.

the main tunneling event at the maximum of the field t_0 when $\boldsymbol{A}(t_0)$ is along the x-axis. Note that the ionization probability for $p_x < 0$ is larger because it results from ionization at two local extrema of the electric field. The total ionization probability is equal to 1.4% at an intensity 2×10^{13}W/cm^2, thus indicating that a relatively strong signal should be obtained experimentally and that complete tunneling across the barrier occurred.

We computed θ_o^e as before for different intensities. The results are also shown in Tab. I, where all angles are virtually zero. If one interprets this picture as the hand of a clock measuring the tunneling time (with the horizontal axis corresponding to zero), this result would, indeed, support the notion of instantaneous tunneling. On the other hand, we do not observe the angle becoming negative in the elliptical case despite depletion of the ground state. This is likely the result of the complex dynamical behavior of the short-distance part of the wavepacket in a multi-cycle field and will require further investigation. However, this finding corroborates the results on hydrogen [7] with a 6 fs elliptical pulse for which no inflection in the variation of the offset angle with intensity was found.

We repeated these calculations for a few intensities using different parameters of the potentials, while still reproducing the $2p$ binding energy of F^-. Again, we obtained a practically zero offset angle. This shows that the principal results, at the intensities studied, depend neither on the specific details of the mid- to long-range potential in strong-field photodetachment, nor on the shape of the initial highest occupied valence orbital for an unpolarized target. On the other hand, for an oriented p-orbital, we found that the offset angle can be different than zero, as was recently also shown in [34].

The Cl^- anion also appears to be a very interesting candidate. It has an even slightly higher electron affinity than F^-, and the Cl atom has a much higher dipole polarizability ($\alpha_d = 14.7\, a_0^3$ [27]) than F. To check whether this may have a significant effect on our final conclusion, we performed additional calculations using the form of the potential (1) and setting "reasonable parameters" to reproduce the Cl^- $3p$ binding energy as well as that of the $3s$ orbital, this time using neutral chlorine as a guide for the latter. Once again, we found virtually zero offset angles. Hence, ionizing the $3p$ orbital with an additional radial node, and for a higher polarization potential, does not modify the conclusion.

The effect of a time-dependent dipole polarizability in SFI has been recently studied [35] for linearly polarized light with a semi-classical two-step model. The authors predict a narrowing of the PMDs and imprints in interference structures. At the intensities considered, and for elliptical light, we indeed noted small effects on the shape of the PMD due to the polarizability. However, we found no noticeable impact on the offset angle. Hence, we conclude that the induced dipole potential in these anions does not affect, at least not to an extent that could likely be resolved by current experimental setups, the direction of the asymptotic momentum of the tunneling electron. This conclusion is based on the mid- to long-range physics, which is very accurately treated in the present SAE approach. It is fortunate that in the intensity considered here this polarization term has no significant effect. A dipole aligned along a rotating field covering several time periods will likely leave a fingerprint at larger intensities.

To summarize: We suggested and investigated an attoclock setup with negative ions that could realistically be employed in an experimental study *without* being affected by the long-range Coulomb interaction between the ejected electron and the residual ion. We find that F^- and Cl^- would both be appropriate, allowing a range of intensities for which tunneling ionization below the barrier should be the dominant ionization/detachment process. We predict that the leading mid- to long-range induced dipole potential, determined solely by the physical properties of the target and including polarization induced by the electron and the external field, has no significant effect on the asymptotic photoelectron momentum offset angle for unpolarized F^- and Cl^- at below-the-barrier intensities. This finding is independent on electron correlation effects. Any offset angle in this system would be the result of short-range physics. Such attoclock experiments could explore what they originally aimed at, i.e., the ultrafast dynamics in the core region of the target, without effect of one-electron long-range physics.

Our calculations produce an offset angle of essentially zero in the momentum plane. Whether or not this angle should be interpreted as zero tunneling time depends on the reader's attitude towards the tunneling concept altogether. Independent of this, we suggest that experimentalists consider such attoclock setups in future studies.

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