Sidebands in Electron Energy upon Multi-Photon Ionization Depend on the Relative Helicity in $(\omega, 2\omega)$ Fields

S. Eckart1, D. Trabert1, K. Fehre1, A. Geyer1, J. Rist1, K. Lin1, F. Trinter1,2, L. Ph. H. Schmidt1, M. S. Schöffler1, T. Jahnke1, M. Kunitski1, and R. Dörner1

1 Institut für Kernphysik, Goethe-Universität, Max-von-Laue-Str. 1, 60438 Frankfurt, Germany
2 Molecular Physics, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

(Dated: May 11, 2020)

We experimentally and theoretically compare multi-photon ionization by counter-rotating two-color (CRTC) laser fields and co-rotating two-color (CoRTC) laser fields. The CRTC and the CoRTC fields are both a superposition of circularly polarized laser pulses at a central wavelength of 390 nm and 780 nm (intensity ratio $I_{390}/I_{780} \approx 250$). Unexpectedly, we find that the measured electron energy spectra strongly depend on the relative helicity in circularly polarized two-color laser fields. For the CRTC field, the measured electron energy spectrum is dominated by peaks that are spaced by 3.18 eV (corresponds to light at 390 nm). For the CoRTC field, we observe additional energy peaks (sidebands). Comparison of the experimental results with our semi-classical, trajectory-based model shows excellent agreement. We conclude that the sideband visibility is modulated by a sub-cycle interference, which sensitively depends on the relative helicity in circularly polarized two-color fields.

When a single atom or molecule is irradiated with a highly intense light field, it can be ionized by non-resonant absorption of more photons than necessary to overcome the binding energy [1]. This phenomenon of above threshold ionization (ATI) [2–6] leads to peaks in the electron energy spectrum that are spaced by the photon energy. When photons at a second wavelength are added to the light field, the question arises what determines the relative amount of photons that is absorbed from each of the two single colors. A trivial control parameter is the relative intensity of the two colors. Here we show that the relative helicity of the two single colors is an additional, very effective control parameter.

For light at a central wavelength of 390 nm, the ATI comb in the electron energy spectrum has a spacing of $E_{390}^{ph} = 3.18$ eV. If the light field comprises photons of a second energy $E_{780}^{ph} = 1.59$ eV in addition, the electrons can have discrete energies of:

$$E_{elec} = N_{390} \cdot E_{390}^{ph} + N_{780} \cdot E_{780}^{ph} - U_p - I_p$$

Equation 1 indicates that in addition to the ionization potential $I_p$, also the ponderomotive potential $U_p$ has to be taken into account [4,7]. To demonstrate the helicity dependence, we choose a two-color field that is the superposition of an intense circularly polarized light field at a central wavelength of 390 nm ($I_{390} = 1.2 \cdot 10^{14}$ W/cm$^2$, peak electric field of 0.041 a.u.) and a much weaker circularly polarized light field at a central wavelength of 780 nm ($I_{780} = 4.7 \cdot 10^{13}$ W/cm$^2$, peak electric field of 0.0026 a.u.). Thus, we expect $N_{780} = 0$ or $N_{780} = \pm 1$ in most cases. According to Eq. 1 one expects the electron energy spectrum to show a comb of discrete energies that are spaced by $E_{780}^{ph}$. We refer to the peaks in the electron energy spectrum which result from the photons of the dominating color (at a central wavelength of 390 nm) as ATI peaks and those in between, which require the absorption or emission of at least one photon of $E_{780}^{ph}$, as sidebands.

Figure 1 shows the measured electron energy spectra upon the single ionization of argon comparing the counter-rotating two-color field (CRTC, opposite helicity of the two colors) and the co-rotating two-color field (CoRTC, same helicity of the two colors). The intensity of the two single-color pulses is identical in both cases. It is evident that the CRTC field produces ATI peaks which are spaced by $E_{390}^{ph}$ and that the sidebands are hardly visible. For the CoRTC field, the sidebands are as intense as the ATI peaks resulting in an energy spectrum that is a comb with a spacing of $E_{780}^{ph}$.

Previous related work has shown that the pathways in multi-photon ionization are different comparing counter-rotating two-color and orthogonally polarized two-color fields [8]. Further, it was discovered that the ionization probability in tunnel ionization is different for CRTC...
fields and CoRTC fields. Both findings were explained by referring to intermediate states and the related selection rules for light-matter interaction. Moreover, CRTC fields and CoRTC fields have been used to, e.g., retrieve properties of the wave function of the electron, obtain attosecond time information, investigate non-adiabatic offsets in momentum space at the tunnel exit, observe sub-cycle interference, and to control non-sequential double ionization.

The two-color fields are generated using a 200 μm BBO to frequency double 780 nm laser pulses (KMLabs Dragon, 40 fs FWHM, 8 kHz) using the same optical setup as in Refs. [13, 18, 20]. We estimate the uncertainty of the absolute intensity for 780 nm and 390 nm to be 50% and 20%, respectively. The three-dimensional electron momentum distributions from single ionization of argon presented in this work have been measured using cold-target recoil-ion momentum spectroscopy (COLTRIMS). The length of the electron and ion arm was 378 mm and 67.8 mm, respectively. Homogeneous electric and magnetic fields of 11.4 V cm\(^{-1}\) and 8.6 G, respectively, guided electrons and ions towards time- and position-sensitive microchannel plate detectors with hexagonal delay-line anodes. During the measurement, we switch the helicity of the laser pulse at 780 nm every 240 seconds to minimize systematic errors. The total ionization rate does not depend significantly on the relative helicity.

To explain the almost complete suppression of sidebands in the CRTC field, for the remainder of this paper we switch from the time-independent perspective of photon absorption to the time-dependent perspective of wave packet creation by tunnel ionization. In a time-dependent formulation, the discrete ATI peaks in the electron energy spectrum result from the periodicity of the electron release times.

Figures 2(a) and 2(b) depict the combined laser electric fields and the negative vector potentials that are used in the experiment. The Lissajous curves of the CRTC field and the CoRTC field are very similar. The weak laser pulse at a central wavelength of 780 nm only induces a small distortion and gives rise to a three-fold (one-fold) symmetry in the CRTC (CoRTC) field. Figure 2(c) shows the measured electron momentum distribution in the plane of polarization for the CRTC field. This electron momentum distribution shows almost no three-fold symmetric features and is completely dominated by the ATI peaks from the laser pulse at 390 nm. Upon inversion of the helicity of the laser pulse at a central wavelength of 780 nm, the measured electron momentum distribution changes drastically, as can be seen in Fig. 2(d). A strong one-fold symmetric pattern of alternating half-rings is observed. In particular, regions in between the rings in Fig. 2(c) are not empty anymore, giving rise to the sidebands in the electron energy spectrum (see Fig. 1). The alternating half-ring pattern by itself is a well-documented phenomenon. The fact that the existence of sidebands depends on the relative helicity of the two single colors has not yet been described to the best of our knowledge. What is the microscopic origin of this huge difference in the energy spectra comparing CoRTC and CRTC fields?

In the following, we show that the intensity of the ATI peaks and the sidebands are modulated by a sub-cycle interference. For CoRTC fields, this sub-cycle interference has recently been termed holographic angular streaking of electrons (HASE) and modeled using a trajectory-based, semi-classical model. The semi-classical model can be summarized as follows: We neglect the Coulomb potential after tunneling and assume that the electrons are released with an initial momentum (which is perpendicular to the tunnel direction and
zero along the light propagation direction). The final electron momentum is the vectorial sum of the initial momentum upon tunneling and the vector potential at the electron’s release time. Since for every angle in the plane of polarization there are two different vector potentials, there are exactly two possible initial release times \((t_1 \text{ and } t_2)\) within one cycle of the two-color laser field that lead to the same final electron momentum \(\vec{p}_f\). One cycle of the two-color laser field has a periodicity of \(T_{780}\) (duration of one optical cycle of light at a wavelength of 780 nm). For each final electron momentum in the plane of polarization \((p_x,p_y\)-plane\), an optimization algorithm searches for the initial release times \(t_1 \text{ and } t_2\). Having found the two initial release times \(t_1 \text{ and } t_2\) within one cycle of the two-color laser field, one can consider a second, subsequent, cycle of the two-color laser field. This allows for the modeling of sub- and inter-cycle interference on the same footing \([12]\). Therefore, the release times \(t_3 = t_1 + T_{780}\) and \(t_4 = t_2 + T_{780}\) are the equivalent release times in the second light cycle that lead to the same final momentum \(\vec{p}_f\). Thus, within two cycles of the laser field, exactly four release times \(t_n\) can be identified for each final momentum \(\vec{p}_f\) (with the trajectory number \(n \in \{1, 2, 3, 4\}\)). Knowing these release times and neglecting Coulomb interaction after tunneling, the semi-classical phase for these trajectories is given by \([12]\):

\[
\phi_n(\vec{p}_f, t_f) = \frac{1}{\hbar} \left( I_p t_n - \int_{t_n}^{t_f} \frac{p_y^2(t) + p_x^2(t)}{2m_e} dt \right)
\]

Here, \(m_e = 1 \text{ a.u.}\) is the electron’s mass, \(\hbar = 1 \text{ a.u.}\) is the reduced Planck constant, and \(I_p = 15.76 \text{ eV}\) is the ionization potential of argon. In Eq. \(2\) the first summand models the phase evolution of the electron in its bound state and the second term models the phase evolution after tunneling by the integral of the electron’s kinetic energy with respect to time. Note that the choice of \(t_f\) only affects the absolute phase of all four trajectories but not the relative phase of the trajectories. We always choose \(t_f = t_4\) as in Ref. \([12]\). Assuming that all four trajectories have the same probability to exist (see Ref. \([12]\) for details), the semi-classically modeled wave function at a given final electron momentum \(\vec{p}_f\) is:

\[
\Psi(\vec{p}_f) = \sum_{n=1}^{4} \exp(i \phi_n(\vec{p}_f, t_f))
\]

Within this model, \(P_{\text{complete}}(\vec{p}_f) = |\Psi(\vec{p}_f)|^2\) describes intensity modulations in momentum space that are due to sub-cycle and inter-cycle interference. Trajectories are only calculated for final electron momenta \(\vec{p}_f\) with an absolute value between 0.2 a.u. and 1 a.u.

The results from the semi-classical model are shown in Fig. \(3\). Figures \(3(a)\) and \(3(b)\) show the absolute value of the negative vector potential \(|-\vec{A}(t)|\) for two optical cycles of the CRTC and the CoRTC field, respectively.

FIG. 3. (a) [(b)] shows the absolute value of the negative vector potential \(|-\vec{A}(t)|\) for two optical cycles of the CRTC [CoRTC] field. The four colored regions indicate the temporal windows of the corresponding electron release times \(t_n\) and the corresponding trajectory number \(n\). The duration of one cycle of the combined electric field is indicated. (c) [(d)] shows the semi-classically modeled intensity in final electron momentum space \(P_{\text{complete}}(\vec{p}_f) = |\sum_{n=1}^{4} \exp(i \phi_n(\vec{p}_f))|^2\) for the CRTC [CoRTC] field (see text related to Eq. \(3\)). Horizontal dashed gray lines guide the eye in (c) and (d).

The possible ranges of the release times \(t_1, t_2, t_3, \text{ and } t_4\) are colored in blue, green, yellow, and red and are labeled with the corresponding trajectory number \(n\). It should be noted that for every point in final momentum space, the values of \(t_n\) can differ but the allowed ranges, that are indicated with colors, do not change as a function of \(\vec{p}_f\) \([12, 24]\). Figure \(3(c)\) \([3(d)]\) shows the semi-classically modeled intensity \(|\Psi(\vec{p}_f)|^2\) in final electron momentum space for the CRTC [CoRTC] field. Strikingly, the semi-classical result for the CRTC field shows almost no intensity at the energies that correspond to the sidebands. The alternating half-ring structure is only observed for the CoRTC field. For the CRTC field, only a weak modulation in intensity as a function of the angle in the plane of polarization is visible. This is all in excellent agreement with the experimental findings presented in Fig. \(2\).

What is the reason for the obvious difference comparing the CRTC and the CoRTC field? Since our semi-classical results only model sub-cycle and inter-cycle interference, we inspect the relative phases of the interfering semi-classical trajectories. To this end, the intensity \(P_{\text{sub}} = |\exp(i \phi_1) + \exp(i \phi_2)|^2\) is visualized in Fig. 3(a)
FIG. 4. (a) [(b)] shows the intensity due to sub-cycle interference \( P_{\text{sub}} = |\exp(i\phi_1) + \exp(i\phi_2)|^2 \) for the CRTC [CoRTC] field. The possible release times of the trajectories for \( \phi_1 \) and \( \phi_2 \) are labeled with \( n = 1 \) and \( n = 2 \) in Fig. 3. In (c) and (d) the intensity due to inter-cycle interference \( P_{\text{inter}} = |\exp(i\phi_1) + \exp(i\phi_3)|^2 \) is shown (release times are labeled with \( n = 1 \) and \( n = 3 \) in Fig. 3). (e) shows the interference patterns from (a), (c), and Fig. 3(c) as a function of the electron energy for the CRTC field. (f) shows the same as (e) for the CoRTC field. Vertical gray dashed lines indicate the peaks of the inter-cycle interference. The dashed gray lines in (a)-(d) are the same in all four panels and guide the eye.

for the CRTC field as a function of the final electron momentum in the plane of polarization. High intensities indicate constructive interference and therefore a phase difference \( \phi_2 - \phi_1 \) that is close to multiples of \( 2\pi \). This interference pattern is due to a sub-cycle interference since the difference in the release time of the two contributing trajectories is \( t_2 - t_1 \), which is shorter than one light cycle (see Fig. 3). Figure 4(b) shows the same for the CoRTC field. Comparison of Figs. 4(a) and 4(b) reveals that the phase difference \( \phi_2 - \phi_1 \) depends more strongly on the angle in the plane of polarization for the CoRTC field. This is underlined by comparing the interference pattern near the gray dashed circles that guide the eye in Figs. 4(a) and 4(b). However, in order to understand the interference patterns \( P_{\text{complete}} \) from Fig. 3, one has to consider not only two trajectories \( n = 1 \) and \( n = 2 \) but all four trajectories. To this end, the relative phase \( \phi_3 - \phi_1 \) is investigated. Figures 4(c) and 4(d) visualize \( P_{\text{inter}} = |\exp(i\phi_1) + \exp(i\phi_3)|^2 \). Because the time difference \( t_3 - t_1 \) is longer than one light cycle, the interference is an inter-cycle interference. Figures 4(c) and 4(d) show the same distribution and this distribution is independent of the angle in the plane of polarization. The two inner rings with high intensity in Figs. 4(c) and 4(d) are highlighted with gray dashed circles. If high intensity is observed for \( P_{\text{sub}} (\phi_2 - \phi_1) \) is close to multiples of \( 2\pi \) and for \( P_{\text{inter}} (\phi_3 - \phi_1) \) is close to multiples of \( 2\pi \) then \( \phi_3 - \phi_2 \) is also close to multiples of \( 2\pi \). Further, the relative phase \( \phi_4 - \phi_3 \) is the same as \( \phi_2 - \phi_1 \) because of the periodicity of the light field (see Fig. 3). This allows to conclude, that a high intensity for \( P_{\text{sub}}(\tilde{p}_{\text{y}}f) \) and \( P_{\text{inter}}(\tilde{p}_{\text{y}}f) \) implies that all four trajectories are interfering constructively for this \( \tilde{p}_{\text{y}}f \). This leads to high intensity in final electron momentum space \( P_{\text{complete}}(\tilde{p}_{\text{y}}f) \). Comparison of Fig. 3(d), Fig. 4(b), and Fig. 4(d) reveals that it is in fact the interplay of sub- and inter-cycle interference that determines the final electron momentum distribution \( P_{\text{complete}} \) and gives rise to the alternating half-ring pattern for the CoRTC field. Also the weak modulations for the CRTC field that are seen in Fig. 3(c) can be explained in full analogy.

These insights allow for a microscopic explanation of the experimental findings presented in Fig. 2. The lowest frequency component of the laser electric field in the time domain (defined by one full cycle of the two-color field \( T_{780} \)) defines the lowest frequency component in the electron energy spectrum (which is \( E_{780} \)). The corresponding time difference is \( t_3 - t_1 = T_{780} \) (see above). Hence, the inter-cycle interference \( P_{\text{inter}} \), that is seen in Figs. 4(c) and 4(d), reflects a comb of allowed final electron energies that is independent of the angle in the polarization plane. (Interestingly, this was already evident from Eq. 1.) The sideband visibility depends on the relative helicity of the two colors because the sub-cycle interference depends on the angle in the polarization plane and is very different comparing the CRTC and the CoRTC field. Figures 4(c) and 4(f) illustrate this finding in energy space showing the energy spectra of the momentum distributions \( P_{\text{sub}}, P_{\text{inter}}, \) and \( P_{\text{complete}} \). It is evident that the sub-cycle interference is very different comparing the CRTC and the CoRTC field: For the CRTC field, the sub-cycle interference strongly suppresses sidebands compared to ATI peaks. For the CoRTC field, the sub-cycle interference affects ATI peaks and sidebands similarly.

In conclusion, our experimental results show an unexpected dependence of sideband intensity on the relative helicity in circularly polarized two-color light fields. We have used a trajectory-based, semi-classical model that reproduces the suppression of sideband intensity as a function of the relative helicity. We conclude that the
modulation of the sideband intensity is a consequence of the differences of the sub-cycle interference comparing CRTC and CoRTC fields. Our findings enable a better understanding of above-threshold ionization and are an important insight regarding the coherent control of electrons. The overall ionization probability is governed by the $2\omega$ field which is about 100 times more intense than the $\omega$ field. The emission of an entire class of electrons (sideband electrons) can be switched on and off on femtosecond timescales by employing an $\omega$ field with time-dependent polarization $^{[25]}$. This is similar to a transistor in electronics and would represent an electron emitter that is powered by a strong laser field at $2\omega$ (“transistor’s collector”) and can be controlled by the helicity (which is equivalent to a relative phase for a light wave or to angular momentum for a photon) of a relatively weak laser pulse at a different frequency $\omega$ (“transistor’s base”).

ACKNOWLEDGMENTS

This work was funded by the German Research Foundation (DFG) through priority program SPP 1840 QUTIF.