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Circularly Polarized High Harmonics in Argon: Dipole and Non-Dipole Effect

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Abstract

The interaction of the intense laser pulse, which forms the basis of the strong laser field and non-linear optical physics, with atoms, molecules, and solids leads to the High Order Harmonic Generation (HHG). There are many theoretical and experimental research related to this process defined by the Semi-Classic Model which is called the Three Step Model. In this article, the dipole and non-dipole effects specified in the theoretical Lewenstein model to be used in the Argon atom interacting with the strong circular laser field (800nm) and the resulting higher order harmonic spectrum will be investigated. We compared the results obtained using the non-collinear beams with opposite circular polarizations with those obtained using a single circularly polarized beam or a linearly polarized beam. It could be said that the circular polarization can significantly affect the HHG process in an argon atom exposed to a laser field with 800 nm wavelength and 1015 W/cm2 intensity.

Keywords: Argon, high harmonic generation, dipole, non-dipole

Argonda Dairesel Polarize Yüksek Harmonikler: Dipol ve Dipol Olmayan Etki

Öz

Güçlü lazer alanının ve doğrusal olmayan optik fiziğin temelini oluşturan yoğun lazer darbesinin atomlar, moleküller ve katılarla etkileşimi, Yüksek Dereceli Harmonik Üretime (HHG) yol açar. Üç Adım Modeli olarak adlandırılan Yarı Klasik Model ile tanımlanan bu süreçle ilgili birçok teorik ve deneysel araştırma mevcuttur. Bu makalede, güçlü dairesel lazer alanı (800nm) ile etkileşen Argon atomunda kullanılacak teorik Lewenstein modelinde belirtilen dipol ve dipol olmayan etkiler ve bunun sonucunda ortaya çıkan yüksek dereceli harmonik spektrum incelenecektir. Zıt dairesel polarizasyonlara sahip doğrusal olmayan ışınlar kullanılarak elde edilen sonuçları, tek bir dairesel polarize ışın veya doğrusal polarize bir ışın kullanılarak elde edilen sonuçlarla karşılaştırdık. 800 nm dalga boyunda ve 10^{15} W/cm^2 yoğunlukta bir lazer alanına maruz bırakılan bir argon atomunda dairesel polarizasyonun HHG sürecini önemli ölçüde etkileyebileceği söylenebilir.

Anahtar Kelimeler: Argon, yüksek harmonik üretimi, dipol, dipol olmayan

INTRODUCTION

The most recent advances in ultrafast spectroscopy technology used in various applications over the last 25 years are sources of high harmonic production and high-resolution imaging of molecules, atoms, and nanostructures (Alp, 2017; Cavalieri et al., 2007; Goulielmakis et al., 2010; Itatani et al., 2004; Li et al., 2008; Marangos et al., 2008).

Higher Order Harmonic Generation (HHG), one of the subjects of nonlinear optical science is one of the fastest-developing research areas in atom-intense laser interaction. In various experimental and theoretical studies, the most basic principle of understanding the origin of high-order harmonics is defined as a semi-classical three-step model. This model has been a particularly useful description for the ‘cut-off and plateau region’ in the HHG spectrum (Krause, Schafer, and Kulander, 1992). This process is called photoionization. During the process, the electron gets the energy and momentum via the laser field. Initially, the electron gets out atom and is excited into the continuum states, where the electron is free to move and gain energy and momentum from the laser field. The second motion is considered classically and consists primarily of free charge

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oscillation which may return near the core and come back to the ground state in the laser field. Once it yields the photon energy that is defined as $E_{kin} + I_p$ where E_{kin} is kinetic energy, and I_p is ionization potential which may be released.

The harmonic photon energy, $E_C = 3.2U_p + I_p$, is given by the cut-off law where U_p is ponderomotive energy $\left(U_p\left[eV\right] = E_0^2/4\omega_0^2\right)$ (Krause et al., 1992). The cut-off region occurs in the harmonic spectrum at harmonics order which is expressed as $N_{\max} = \left(3.2U_p + I_p\right)/\hbar\omega$. In the third step, the electron is finally ejected from the atom. This photoionization process plays an important role in many areas of physics, including laser-matter interaction, atomic and molecular physics, and quantum optics. It is also used to study the interaction between atoms and light, as well as the structure and dynamics of atoms.

As a result of intense research using linear polarization, it has been observed that the electron's classical trajectories pass through periodically the nucleus, therefore permitting recombination radiatively and harmonics generation. Although great progress has been done in the use of linearly polarized beams that have been made in the high harmonic generation, it was very challenging to directly obtain HHG circularly polarized pulses. In contrast, elliptically polarized pump lasers (or circularly polarized lasers) are strongly suppressed by the possibility of electron collision and therefore do not emit harmonics (Dietrich, Burnett, Ivanov, and Corkum, 1994; Weihe et al., 1995). Recent studies have indicated the production of a high harmonic circular polarized attosecond pulse train which is modified by the deeper physical insight of HHG with the combining of collinear counter-rotating light of different degrees and macroscopic phase matching (Fan et al., 2015; Fleischer, Kfir, Diskin, Sidorenko, and Cohen, 2014; Kfir et al., 2015; Medišauskas, Wragg, Van Der Hart, and Ivanov, 2015). In addition to circular polarization, the HHG process can be influenced by the linear polarized laser field. The polarization state of a laser field influences the electron dynamics and, therefore, the HHG yield and spectral characteristics. This is because the laser field's polarization distresses the tunneling rate of the electrons, which determines the ionization rate and, ultimately, the HHG yield. Polarization is a fundamental feature of electromagnetic waves and often acts as a significant role in the interaction of light with matter. It is particularly investigated that using circularly polarized short EUV pulses for generating harmonics which are ultra-fast spin dynamics, circular dichroism, chirality allocation magnetic microscopy, and so forth. Various theoretical approaches have been proposed for generating circularly polarized harmonics (Husakou, Kelkensberg, Herrmann, and Vrakking, 2011; D. B. Milošević, Becker, and Kopold, 2000; Yuan and Bandrauk, 2011). For instance, recent studies on the very strong dependence on polarization ellipticity in high harmonic generation require that numerical approximation to intense field ionization to go beyond the current models based on linear polarization (Budil, Salières, L'Huillier, Ditmire, and Perry, 1993; Liang, Angst, Ammosov, and Lazarescu, 1995).

The polarization is not only important to study light-matter interactions over basic optical views, but also a time-varying polarization state, which forms the basis of numerous spectroscopic and harmonics control techniques related to HHG in soft X-ray and XUV spectrum in Attoscience (Brixner et al., 2004; Kerbstadt, Englert, Bayer, and Wollenhaupt, 2017). There are many experimental and theoretical scientific studies used in the generation of harmonics obtained by circular polarization laser field interacting with the matter, for instance, non-adiabatic tunneling by spin-polarized electrons, producing vortex-shaped photoelectron momentum distributions, attosecond control by spin-dissolved recoil dynamics, and examining atto-clock techniques via angular streaking using cold target recoil ion momentum spectroscopy form (Ayuso, Jiménez-Galán, Morales, Ivanov, and Smirnova, 2017; Barth and Smirnova, 2011; Hartung et al., 2016; Herath, Yan, Lee, and Li, 2012; D. Milošević, 2016).

In this study, consequently, the event of interacted circularly polarized beam with an atom that is defined cut-off region in the conventional harmonic spectrum will be shown by reference (Emilio Pisanty et al., 2018) to a different numerical method for generating harmonics.

Here, a proposed simple method will be used to obtain forward ellipticity in the magnetic fields presence that can move along with Lorentz force in the same direction, so that the harmonic emission is

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allowed to equal intensity and wavelength using two non-collinear opposite circular polarized beams.

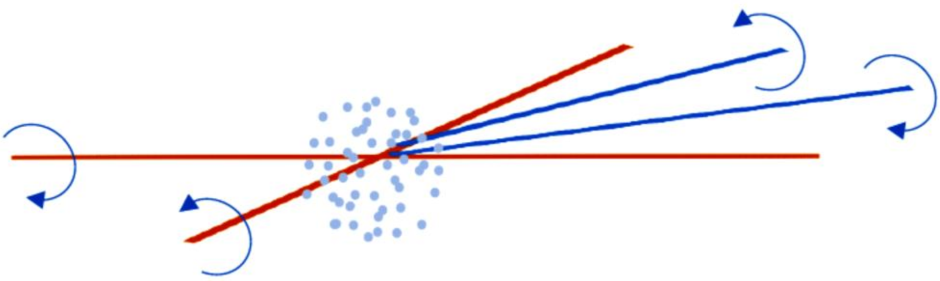

Fig 1. Scheme for two co-linear circular polarized beams generating high harmonics (Hernández-García et al., 2016).

MATERIAL AND METHODS

To analyze the HHG process in the circularly polarized laser field's presence, it is essential to use a model which accounts for the time-dependent nature of the Lorentz force. One such model is the strong field approximation (SFA), which is built on the assumption that the ionized electron experiences a constant acceleration in the laser field and can be defined by a classical trajectory. The SFA has successfully reproduced the experimental HHG spectra for a wide range of atomic and molecular systems.

Circular polarization is a type of polarization in which the electric field of the electromagnetic wave rotates around the direction of propagation with a constant angular frequency. In this case, the Lorentz force will also rotate around the direction of propagation with a constant angular frequency. This will result in an HHG spectrum that is more complex, with contributions from both dipole and non-dipole transitions. Non-dipole transitions occur when the ionized electron recollides with the atomic core at an angle relative to the direction of the electric field. In HHG, a strong laser field is used to ionize an atom and generate high-energy photons through the recombination of the ionized electron with the atomic core. In the laser field, the circular polarization can affect the HHG process by modifying the Lorentz force experienced by the ionized electron.

Here we take into account the harmonics produced in Argon gas by two opposite circular polarization beams within propagating plane.

RESULTS AND DISCUSSION

Using Strong Field Approximation with Lewenstein Model

In this paper, we use a non-dipole and non-relativistic Strong Field Approximation (SFA) model within Lorentz force $\vec{F} = \vec{V}/c \times \vec{B}$ which is reduced photon emission (Brennecke and Lein, 2018; D. B. Milošević, Hu, and Becker, 2000). Firstly, we noted that the Hamiltonian is

$$H = \left(\frac{1}{2} \left[-i\nabla + \vec{A}(\vec{r},t) \right]^2 + V_0 \right) \tag{1}$$

Considering the HHG in noble gas by two opposite circular polarized propagation wave vectors are

$$\vec{k} = k\left(\sin(\theta), 0, \cos(\theta)\right) \tag{2}$$

Then, defined vector potential and determined initial conditions $\left(z=0, kx\sin(\theta)=\pi/2\right)$ (Emilio Pisanty et al., 2018).

$$\vec{A}(\vec{r},t) = \sum_{\pm} \frac{F}{2\omega} \begin{pmatrix} \cos(\theta)\cos\left(\vec{k}\cdot\vec{r}-\omega t\right) \\ \pm\sin\left(\vec{k}\cdot\vec{r}-\omega t\right) \\ \sin(\theta)\cos\left(\vec{k}\cdot\vec{r}-\omega t\right) \end{pmatrix}$$
$$= \frac{F}{\omega} \begin{pmatrix} \cos(\theta)\cos\left(kz\cos(\theta)-\omega t\right)\cos\left(kx\sin(\theta)\right) \\ \cos\left(kz\cos(\theta)-\omega t\right)\sin\left(kx\sin(\theta)\right) \\ -\sin(\theta)\sin\left(kz\cos(\theta)-\omega t\right)\sin\left(kx\sin(\theta)\right) \end{pmatrix} \tag{3}$$

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$$\vec{A}(\vec{r},t) \approx \frac{F}{\omega}\begin{pmatrix} \cos(kx\sin(\theta)) \\ \sin(kx\sin(\theta)) \\ 0 \end{pmatrix}\cos(\omega t) \tag{4}$$

$$\vec{A}(\vec{r},t) = \frac{F}{2\omega}\begin{pmatrix} 0 \\ \cos(\omega t) \\ \sin(\theta)\sin(\omega t) \end{pmatrix} \tag{5}$$

Transforming length gauge and here define Hamiltonian

$$H = \frac{1}{2}\left[-i\nabla + \vec{A}(0,t)\right]^2 + \frac{1}{c}\hat{k}\cdot\hat{r}\left[-i\nabla + \vec{A}(0,t)\right]\cdot\vec{F}(t) + V_0 \tag{6}$$

$$H = \frac{1}{2}\left[\hat{p} + (\hat{r}\cdot\vec{\nabla})\vec{A}(0,t)\right]^2 + \hat{r}\cdot\vec{F}(t) + V_0 \tag{7}$$

Then neglected $\left[(\hat{r}\cdot\vec{\nabla})\vec{A}(0,t)\right]^2$ terms in Equation (7) yield final Hamiltonian (Emilio Pisanty et al., 2018)

$$H = \frac{\hat{p}^2}{2} + \hat{r}\cdot\vec{\nabla}\vec{A}(t)\cdot\hat{p} + \hat{r}\cdot\vec{F}(t) + V_0 \tag{8}$$

The harmonic emission calculation due to the above Hamiltonian is as conventional as in the case of dipole and the continuous wave function must be changed to express the non-dipole term. The Equation (9) solution can be stated as a time-dependent Lippmann-Schwinger equation containing the Green function related to $H(t)$ (Cohen, DuMond, Bethe, and Salpeter, 1957; Reiss, 1980). Lewenstein and co-workers using the SFA approach neglecting dipole moment and the full Green function is replaced by the non-dipole and non-relativistic Volkov state with the Hamiltonian which is described that free electron laser field interacted with ionic core (Lewenstein, Balcou, Ivanov, L'huillier, and Corkum, 1994; Salieres, L'huillier, Antoine, and Lewenstein, 1997).

The SFA approach definition of dipole moment in the strong laser field is

$$d(t) \approx \int_{-\infty}^{t} dt' \int drdr' \phi_0^*(r,t) r G_V^+(r,t;r',t') H(t)\phi(r',t') + c.c \tag{9}$$

where the $\phi(r,t) = \phi_0(r)\exp(I_p t)$ is initial eigenstate of Hamiltonian, (I_p) is ionization potential and $G_V^+(r,t;r',t')$ is green function. In the dipole approximation Equation (9) has been neglected the ground state depletion (Salieres et al., 1997; Tempea, Geissler, and Brabec, 1999) to account for the component of magnetic field generalized easily by Volkov state is expressed as:

$$\left|\psi_p^V(t)\right\rangle = e^{-\frac{1}{2}\int \pi(\vec{p},\tau)^2 d\tau}\left|\pi(\vec{p},t)\right\rangle \tag{10}$$

where $\pi(\vec{p},t)$ momentum at the plane-wave

$$\pi(\vec{p},t) = p + A(t) + \frac{1}{c}\left[\vec{p}\cdot\vec{A}(t) + \frac{1}{2}A^2(t)\right]\hat{k}\,. \tag{11}$$

Equation (11) modified monochromatic field and using
$\int \vec{\nabla}\vec{A}d\tau \sim \frac{k}{\omega}A = \frac{1}{c}A,$

$$\pi(\vec{p},t) = p + A(t) + \int \vec{\nabla}\vec{A}(\tau)\cdot(\vec{p} + \vec{A}(\tau))d\tau\,. \tag{12}$$

Then harmonic emission can be calculated with the SFA approach by using a non-dipole Volkov state wave function that states dipole in the form of:

$$d(t) \simeq \int_{t_0}^{t} dt' \int dp d(\pi(p,t)) e^{iS(p,t,t')} F(t')\, d\left(\pi(p,t')\right) + c.c. \tag{13}$$

where $S(p,t,t')$ is

$$S(p,t,t') = I_p(t-t') + \frac{1}{2}\int_{t'}^{t} \pi(p,t)^2 d\tau\,. \tag{14}$$

In practical terms from the theoretical perspective, the non-dipole Volkov states we employ the basis of Schrödinger equation solution by choosing circular polarization which tends to act in the opposite direction intense laser field. Calculating the harmonic generation by this method is known as a 'bicircular' laser field that is superposed of two opposite circular polarization (Long, Becker, and McIver, 1995; D. B. Milošević, Becker, et al., 2000; Emilio Pisanty et al., 2018). This is a general outline of how to model the circular polarization effect on HHG in the SFA using the Lewenstein model and a

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mathematical approach. There are many details and subtleties that would need to be accounted for a more comprehensive treatment, such as the specific form of the atomic potential, the choice of boundary conditions, and the method used to resolve the time-dependent Schrödinger equation (TDSE).

Argon is a heavier atom with a more complex electronic structure compared to lighter atoms such as hydrogen or helium. As a result, argon atoms are more likely to exhibit stronger non-dipole effects in strong laser fields.

The dipole and non-dipole effects for argon atoms can also vary depending on the laser wavelength and intensity. For example, at longer wavelengths or lower intensities, the dipole approximation may be more accurate, whereas at shorter wavelengths or higher intensities, the non-dipole effects can become more pronounced.

In addition, the argon's ionization potential is relatively high (15.76 eV), meaning that it requires a high energy input to ionize an argon atom. This can result in a complex ionization dynamics for argon in strong laser fields, which can further contribute to the non-dipole effects (Wiehle, 2005).

Overall, the non-dipole and dipole effects for argon atoms can be significant in intense laser fields, especially at shorter wavelengths or higher intensities. Understanding these effects is important for accurately modeling and predicting the actions of argon atoms in intense laser fields.

To further analyze the effect of circular polarization on the HHG process in an argon atom illuminated by a laser field with an intensity of 10^{15} W/cm^2 and wavelength of 800 nm, it is useful to compare the HHG spectra obtained for different circular polarizations. This can be done using a mathematical software program such as Mathematica.

To visualize the HHG spectrum for an argon atom irradiated by non-collinear beams with opposite circular polarizations at 800 nm, we could employ Mathematica to plot the harmonic spectrum as the n^{th} harmonic order function. The non-dipole and dipole regimes can be identified by comparing the results for different values of n.

Primally, we demonstrated how to function non-collinear beams with reverse circular polarization with Mathematica program codes. The program run into non-dipole approximation in the non-collinear strong magnetic field that its implementation is available at references 40 and 41. Fig. 2 shows that harmonic spectrum in non-dipole regime at 800 nm drivers for Argon atom and proposed □ $10^{15}\, W/cm^2$ (Hernández-García et al., 2016) intensities with ionization potential $15.7\,eV$ (D. B. Milošević, Becker, et al., 2000) using $\theta = 4°$ half-angle beam. Finally, z-polarized harmonics eliminated because the region is arbitrary.

For characterizing the several solutions of saddle point approximation to calculate the momentum integral harmonic emission from arbitrary linear beam (D. Milošević and Becker, 2002; E Pisanty, 2016; Emilio Pisanty et al., 2018), in the present paper, we will calculating the harmonic emission rate attained by the saddle-point approximation that are showed in Fig.3. This is of crucial importance of classification of quantum orbit where explained the cut-off and plateau region from our analysis. This numerical analysis data is based on of HHG and ATI (Above Threshold Ionization) processes. In the high intensity show that Fig. 3, the attendance of non-dipole effects origins the intensity to a decrease so re-enables most of the harmonic emission with the effect of circular polarization (at $\theta = 2°$) that causes changing cut-off region.

The concepts of saddle points and turning points are related to the discussion of dipole and non-dipole effects in circularly polarized light and above-threshold ionization (ATI) spectra but in slightly different ways.

In the context of ATI spectra, the dipole and non-dipole effects refer to the different mechanisms by which high-energy photoelectrons are generated in circularly polarized light. The dipole effect stems from the interaction of the laser field with the electric dipole moment of the atom or molecule, while the non-dipole effect arises from higher-order multipole moments, such as the electric quadrupole moment. These effects can give rise to different patterns in the distribution of high-energy photoelectrons in the ATI spectrum.

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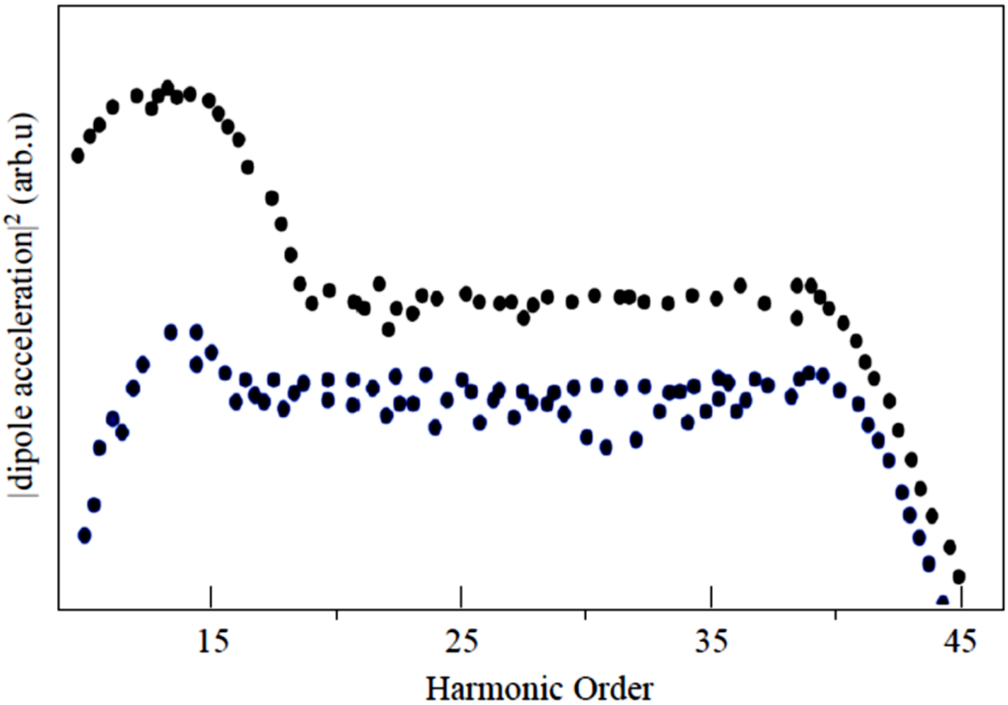

Fig 2. Harmonic spectrum (the black dot is odd, and blue is even harmonics) produced in Argon atom at 800 nm in the non-dipole regime for monochromatic laser field.

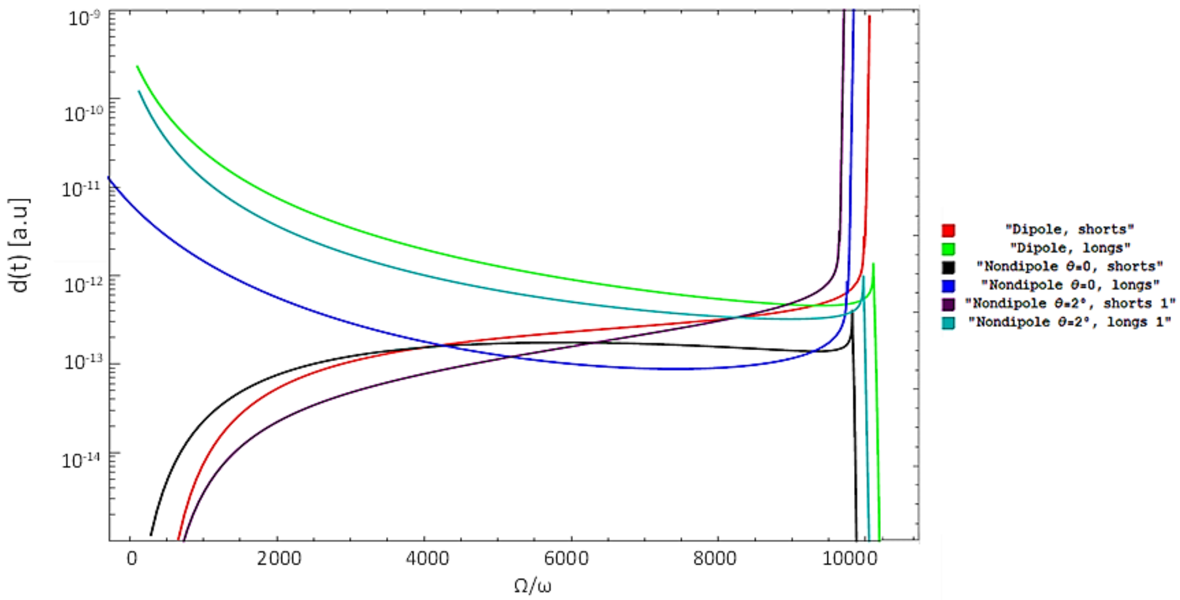

Fig 3. Saddle Points trajectory for an Ar in 800 nm field of I = 10^{15} W/cm^2 intensity, computed in dipole and non-dipole effect show that cut-off region.

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On the other hand, saddle points are critical points for the system's potential energy surface. In the context of ATI spectra, saddle points can be important for understanding the behavior of the system near the ionization threshold. Specifically, near the ionization threshold, the potential energy surface has a saddle point where the system is in a state of unstable equilibrium. At this point, the system can either ionize or return to the ground state, depending on the dynamics of the system.

The non-dipole and dipole effects can influence the behavior of the system near the saddle point by affecting the shape of the potential energy surface. In particular, the dipole effect tends to lower the energy of the saddle point, making it easier for the system to ionize, while the non-dipole effect tends to increase the energy of the saddle point, making it harder for the system to ionize.

Overall, the interplay between dipole and non-dipole effects, and the influence of these effects on the potential energy surface and the behavior of the system near the saddle point, can be vital for considering the dynamics of ionization in circularly polarized light and ATI spectra (E Pisanty, 2016; Emilio Pisanty et al., 2018).

By changing the laser beams' intensity and analyzing the ellipticity effect, it is probable to gain a better understanding of how the laser intensity affects the HHG process in an argon atom. This can provide insight into the optimal intensity for producing high-energy harmonics with a specific degree of circular polarization. In the saddle point trajectory, the dipole approximation assumes that the electron trajectory is in the same direction as the electric field. However, in reality, the electron trajectory can deviate from the direction of the electric field due to the presence of the Coulomb potential of the ion. This deviation results in a shift in the cutoff energy of the spectrum towards higher energies, known as the non-dipole effect.

The non-dipole effect is more pronounced for higher laser intensities, and at shorter laser wavelengths. For an 800 nm laser field with an intensity of 10^{15} W/cm^2, the non-dipole effect would result in a shift of the cutoff energy towards higher energies, and a broadening of the spectrum. This effect can be accounted for using the modified saddle point approximation, which accounts for the deviation of the electron trajectory from the direction of the electric field.

At an intensity of 10^{15} W/cm^2 and 800 nm wavelength, the non-dipole effects in the spectrum's cut-off region for an argon atom are expected to be significant. The laser field's circular polarization can affect the non-dipole effects by altering the electric field direction and the phase of the laser field. If the circular polarizations of the beams are opposite and the phase angle is 2, the non-dipole effects may be reduced, leading to a decrease in the spectrum in the cut-off region. However, it is important to note that the dipole effects, which are proportional to the laser field's intensity, may also contribute to the spectrum at these intensities. The effect of circular polarization on the spectrum in the cut-off region will be based on the relative strengths of the dipole and non-dipole effects, which can vary depending on the angle between the beams and other factors. Furthermore, in the dipole approximation, the spectrum exhibits a strong angular dependence, with a narrow angular distribution of emitted electrons along the laser field's polarization direction. However, in the non-dipole approximation, the angular distribution of emitted electrons becomes broader, with a significant contribution from electrons emitted at angles away from the laser's field polarization direction (Duesterer et al., 2013).

CONCLUSION

It is also useful to analyze the cut-off region of the HHG spectrum, which corresponds to the maximum energy that can be reached by the ionized electron during the collisional process. The cut-off energy can be calculated using the SFA by evaluating the ionized electron's kinetic energy at the moment of recollision. To do this, it is necessary to solve the classical equation of motion for the ionized electron in the laser field and determine the maximum kinetic energy that can be reached. This can be done using Mathematica by defining the classical equations of motion and solving them numerically. The cut-off energy can then be indicated on the HHG spectrum plot using a vertical line. In summary, circular polarization can significantly affect the HHG process in an argon atom exposed to a laser field with 800 nm wavelength and 10^{15} W/cm^2 intensity.

We compared the results obtained using the non-collinear beams with opposite circular polarizations with those obtained using a single circularly polarized beam or a linearly polarized beam. This can help to assess the beam configuration's influence on the

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HHG process. By performing these analyses, you can gain a deeper understanding of the role of opposite circularly polarized non-collinear beams in HHG and the underlying physical mechanisms at play in the process. By exploring these and other factors, we can gain a better understanding of the HHG process and the influencing factors when using opposite circularly polarized non-collinear beams.

In recent breakthroughs, HHG techniques offer a lot of methods containing that are most commonly used non-collinear beams which are opposite circularly polarized extreme ultraviolet driving laser light. Furthermore, all techniques proposed in the literature for the production of HHG are based on Attosecond science. We summarize the mechanisms of non-collinear HHG and evaluate the HHG spectrum for Argon atom interacted with 800 nm circular intense laser field. The plateau and cut-off regions change dependence on the non-dipole effect. In the plateau regions, polarization calculation remains more smoothly connected to the field intensity of the harmonic phase, while the cut-off region maintains perfectly in the HHG spectrum. To conclude, we note that the results of numerical calculation propose and analyze different schemes of HHG.

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CONFLICT OF INTEREST

The Authors report no conflict of interest relevant to this article.

RESEARCH AND PUBLICATION ETHICS STATEMENT

The authors declare that this study complies with research and publication ethics.

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