High harmonic generation with fully tunable polarization by train of linearly polarized pulses

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Abstract

We propose and demonstrate, analytically and numerically, a scheme for generation of high-order harmonics with fully tunable polarization, from circular through elliptic to linear, while barely changing the other properties of the high harmonic radiation and where the ellipticity values of all the harmonic orders essentially coincide. The high harmonics are driven by a train of quasi-monochromatic linearly polarized pulses that are identical except for their polarization angles, which is the tuning knob. This system gives rise to full control over the polarization of the harmonics while largely preserving the single-cycle, single-atom and macroscopic physics of ‘ordinary’ high harmonic generation, where both the driver and high harmonics are linearly polarized.

1. Introduction

Light sources based on high harmonic generation (HHG) are used for a variety of applications, e.g. ultrafast spectroscopy and high-resolution imaging [1–7]. For a long time, the polarization of bright high harmonics was limited to the region of linear polarization. This limitation was generally accepted as a fundamental feature of HHG because it is a recollision process: an electron is first tunnel ionized, then accelerated by the strong laser field, and finally recombines with its parent ion while emitting a high energy photon [8, 9]. The probability for recombination is maximal when the HHG process is driven by a linearly polarized pulse, since in this case the electron’s center of mass motion is one dimensional. The linear trajectory and ‘head on’ recollision yield bright linearly polarized high harmonic radiation. Introducing ellipticity to the laser field can result in elliptically polarized harmonics [10–12]. However, in this case the high harmonic efficiency drops drastically as a result of the electron trajectory, which misses the parent ion upon its return [13, 14]. Nevertheless, over the years there has been significant theoretical [13, 15–23], and experimental [24–36], effort to generate bright high-order harmonics with highly helical polarization, because polarization is a fundamental and useful property of light. Particularly, several techniques were recently demonstrated experimentally. Fleischer et al [27] and others [28, 30, 32, 35, 36] produced circularly and elliptically polarized high harmonics using bi-chromatic circularly polarized (or elliptically polarized) counter-rotating drivers. High harmonics with relatively high elliptical polarization (ellipticity up to 0.8 at photon energy 25 eV) were demonstrated by driving a gas of SF$_6$ molecules with an elliptically polarized pump [29], and also by driving a Ne atomic gas by bi-chromatic co-propagating orthogonally polarized pumps [31]. Circularly polarized high harmonics were also demonstrated by mixing non-collinear circularly polarized counter-rotating pumps [30].

Beyond the generation of bright circularly (and elliptically) polarized high harmonics, the ability to finely tune the polarization is also highly useful. Such control could be useful for probing chiral processes on femtosecond timescales, as well as in coherent control. Also, controlling the ellipticity of the high harmonics would be useful in HHG-based ellipsometry [37], allowing null-ellipsometry, generalized-ellipsometry (used for non-isotropic surfaces), and imaging ellipsometry [38]. For example, the bi-circular method allows full control over the ellipticity of the high harmonics, from circular through elliptic to linear polarization without
compromising the efficiency of the process, by rotating a single waveplate for the pump beam [27, 36].
Nevertheless, the polarization is strongly coupled to other properties of the high harmonic radiation, such as the HHG spectrum, and the polarization of different peaks in the spectrum is not synchronized (i.e., they are different).

Here we present a new scheme to generate elliptically polarized high harmonics, with full control over the ellipticity of all the harmonics, and with a very weak coupling to the other properties of the harmonic spectrum. The driver field is comprised of a train of quasi-monochromatic linearly polarized laser pulses with a varying polarization orientation. The harmonics are circularly polarized pairs with alternating helicity when the field possesses a 3-fold dynamical symmetry (DS), even though the DS period can be much larger than the optical cycle. The ellipticity of all the harmonics can be tuned collectively, from circular through elliptic to linear, by rotating the polarization axes of some of the pulses that comprise the driving field by only one angle. We explore the properties of the generated harmonics using both a simple analytical model and by solving the time dependent Schrödinger equation (TDSE) numerically.

2. The driving pulse train

In this section, we present the train of linearly polarized pulses for generation of high harmonics with fully controlled polarization. We first discuss the syntheses of such a train that produces circularly polarized high harmonics, and then introduce an adjustment knob in the train that controls the harmonics polarization. Recall the \( \omega - 2\omega \) bi-circular method that was shown to yield bright circularly polarized high harmonics [27]. In that scheme, the driving field exhibits the following 3-fold DS [28, 39, 40]

\[
\vec{E} \left( t + \frac{T_3}{3} \right) = \hat{R} \left( \frac{2\pi}{3} \right) \vec{E} \left( t \right),
\]

where \( \vec{E} \left( t \right) \) is the two-dimensional (2D) vectorial driver field, \( T_3 \) is the cycle time of the DS, \( \hat{R} \left( 2\pi/3 \right) \) is a 2D rotation operator at an angle of \( 2\pi/3 \), and the electric field is periodic with an overall period of \( T_3 \). The high harmonics generated upon the interaction of driving fields that exhibit this DS with isotropic media are circularly polarized. Notably, the circular polarization property is due to the 3-fold DS and is otherwise insensitive to the structure of the driving field. Thus, we explore here HHG driven by a train of quasi-monochromatic pulses that overall exhibits the 3-fold DS whereas each pulse is polarized linearly, hence largely preserving the familiar physics of HHG driven by linearly polarized drivers. In our train, the pulses are identical except for their polarization direction, which in order to attain the 3-fold DS, rotates by \( 2\pi/3 \) from one pulse to the next (figure 1). In order to produce elliptically polarized high harmonics, the 3-fold DS needs to be broken. As will be shown below, we found that rotating the polarization axes of some of the pulses can be used as a fine knob for controlling the ellipticity values of all the high harmonics simultaneously (i.e. all the harmonics have the same ellipticity) while barely influencing the HHG power spectrum. Specifically, in every group of three consecutive pulses, the polarization axis of one pulse is maintained fixed while the other two pulses are rotated by \( \pm \Delta \alpha \) (inset of figure 1).

Following the discussion above, the driving field that we consider in this work takes the form:

\[
\vec{E}_{\text{tot}} \left( t \right) = \sum_{m=-\left(M-1\right)/2}^{\left(M-1\right)/2} \vec{E}_{\text{UC}} \left( t - mT_3 \right),
\]

where the total field contains integer \( M \) repetitions of the unit cell train, \( \vec{E}_{\text{UC}} \), which is \( T_3 \) long and consists of three pulses:

\[
\vec{E}_{\text{UC}} \left( t \right) = \vec{E}_{\text{p} \left( t; 0 \right)} + \vec{E}_{\text{p} \left( t - \frac{T_3}{3}; \frac{2\pi}{3} + \Delta \alpha \right)} + \vec{E}_{\text{p} \left( t - \frac{2T_3}{3}; \frac{4\pi}{3} - \Delta \alpha \right)}
\]

here \( \vec{E}_{\text{p}} \left( t; \theta \right) \) represents a linearly polarized pulse at polarization angle \( \theta \):

\[
\vec{E}_{\text{p}} \left( t; \theta \right) = E_0 \left( t \right) A \left( t \right) \sin \left( \omega_0 t \right) \left( \cos(\theta) \hat{x} + \sin(\theta) \hat{y} \right),
\]

where \( E_0 \) is the field maximum amplitude, \( \omega_0 \) is the fundamental laser frequency, and \( A \left( t \right) \) is the real pulse envelope (for simplicity, we assume that the pulses are not chirped). Furthermore, we assume that each pulse contains \( \left(N_p + 1\right) \) optical cycles and that there is a gap between consecutive pulses with duration \( \tau \). Thus, the period of the total field (i.e. the duration of the unit cell train) is \( T_3 = 3(N_p + 1)T + 3\tau \), where \( T = 2\pi/\omega_0 \) is the optical cycle duration. An example of such a driving field with \( M = 1, N_p = 4, \tau = 0, \Delta \alpha = 0 \), and a trapezoidal envelope function is shown in figure 1.
3. Analytical model

3.1. Circular HHG

We begin by formulating a simple analytical model for HHG driven by the field in equations (2)–(4), with $\Delta \alpha = 0$, i.e. when the field exhibits a 3-fold DS. We assume that the field interacts with an isotropic medium, hence the DS of the field is transferred to the electronic dipole response (the Hamiltonian is invariant under the DS operator [39]). For simplicity of the analytical analysis, we assume that the pulses have a square envelope, each half cycle generates a delta-like attosecond emitted field (with the sign of the optical half cycle). In addition, we assume that $N_p$ and $M$ are even and odd integers, respectively, resulting with a temporally symmetric train. Under these assumptions, and denoting $a(t) = a_x(t) \hat{x} + a_y(t) \hat{y}$ as the vectorial dipole acceleration which is proportional to the emitted HHG field, we get:

$$a_x(t) = \sum_{m=-(M-1)/2}^{(M-1)/2} \left\{ f(t - mT_3) - f\left(t - mT_3 - \frac{T_3}{3}\right) - \frac{1}{2} f\left(t - mT_3 - \frac{2T_3}{3}\right) \right\},$$

$$a_y(t) = \frac{\sqrt{3}}{2} \sum_{m=-(M-1)/2}^{(M-1)/2} \left\{ f\left(t - mT_3 - \frac{T_3}{3}\right) - f\left(t - mT_3 - \frac{2T_3}{3}\right) \right\},$$

where

$$f(t) = \sum_{n=-N_p/2}^{N_p/2} \delta(t - nT) - \sum_{n=-N_p/2}^{N_p/2} \delta(t - nT - T/2)$$

is the polarization acceleration due to a single linearly polarized pulse and $\delta(t)$ is the Dirac delta function. Fourier transforming the dipole acceleration gives the emitted spectrum. We project the spectrum onto left and right circular components through:

$$\tilde{a}_{\pm,\pm}(\Omega) = \mathcal{F}[a_{\pm}(t) \pm ia_{\pm}(t)] = \tilde{a}_{\pm}(\Omega) \pm i\tilde{a}_{\pm}(\Omega),$$

Figure 1. Exemplary vectorial driver field consisting of a train of quasi-monochromatic linearly polarized pulses with different polarization axes. The driver is set according to equation (2), with the parameters: $M = 1$, $N_p = 4$, $\tau = 0$, and a trapezoidal envelope function. The inset shows a schematic front view of the train, with blue arrows representing the linear-polarization axes of the composing pulses when the field exhibits a 3-fold DS. In this case, the polarization axis rotation angle between consecutive pulses is $120^\circ$ (black arrows). The red numbers represent the order of linear pulses in the train. Dashed arrows represent a case of broken DS, where the orientation of the linear pulses in the train is shifted from the symmetric $120^\circ$ separation by $\pm \Delta \alpha$. 

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where $\mathcal{F}$ represents a Fourier transform. The spectral power of the left and right helical components is retrieved as the absolute power of equation (8):

$$I_{\pm}^\pm(\Omega) = |\tilde{a}_{\pm}^\pm(\Omega)|^2.$$  

Plugging equations (5)–(7) into equations (8)–(9), leads to

$$I_{\pm}^\pm(\Omega) = \frac{8}{\pi} \sin^2\left(\frac{\pi}{2}\frac{\Omega}{\omega_0}\right) \times \left(1 + 2 \sum_{n=1}^{N_p/2} \cos\left(2\pi n \frac{\Omega}{\omega_0}\right)\right)^2 \times \sin^2\left(\frac{\Omega T_f}{6}\right) \times \left[1 + 2 \sum_{m=1}^{(M-1)/2} \cos(mT_f\Omega)\right] \times \left\{2 + \cos\left(\frac{\Omega T_f}{3}\right) \mp \sqrt{3} \sin\left(\frac{\Omega T_f}{3}\right)\right\}.$$  

(10)

The obtained expression in equation (10) is comprised of five terms: the first two are responsible for a stronger emission of radiation near odd order harmonics of the fundamental. This feature can be regarded as a result of the linear–polarization nature of the recollisions in each pulse in the train. However, in contrast to the ‘common’ HHG scheme in which HHG is driven by linearly polarized quasi-monochromatic drivers, and where only odd-order harmonics of the pump pulse are obtained, here the emission at exact odd harmonics is forbidden, as can be seen from the last three term, which forbid any integer harmonic from appearance. Specifically, the second term dictates the width around each odd–harmonic peak, which is narrowed with increasing duration of the pulse (increasing $N_p$), again in accordance with the ‘common’ HHG scheme. The last three terms dictate the ‘new’ selection rules due to the 3-fold DS of the pulse train with a period of $T_p$. The third term forbids the appearance of harmonics (of $2\pi/T_f$) which are integer multiples of 3, that is, forbids the appearance of harmonics $6\pi q/T_f$ where $q$ is an integer. Hence, the allowed radiation is found in the frequency pairs:

$$\Omega_{\pm} = \frac{2\pi}{T_f}(3q \pm 1), \quad q \in \mathbb{Z}.$$  

(11)

As the fourth term in equation (10) implies, the harmonics peak intensity and width are approximately proportional to $M^2$ and $1/\sqrt{M}$, respectively. The last term indicates the polarization of the allowed peaks, which is left or right circularly polarized. Notably, at the peak frequency of a given helicity emission, the intensity of the other helicity is small, and is precisely zero at the peak’s maxima (which can be seen in figure 2, and by substituting the allowed frequencies in equation (11) into (10)). Hence, the polarization at the peak’s maxima is purely circular. Figures 2(a) and (b) show the power spectra of the right and left rotating circular harmonics in the region of an odd harmonic order (the power spectrum is perfectly repetitive because of the modeled delta function pulses) for $M = 1$ and $M = 3$, respectively. As the number of unit cells increases, the polarization becomes circular throughout the entire support of each peak, and is not limited to the maxima. We note that the resulting harmonic spectrum is strongly influenced by the existence of two dominant time-scales in the driver ($T$ and $T_f$). The limiting factor of spectral resolution is the free spectral range: $2\pi/T_f$.

### 3.2. Tunable ellipticity

Next, we analyze HHG driven by the pulse train in equations (1)–(4) with $\Delta \alpha = 0$, i.e. when the 3-fold DS is broken. In this case (following the same procedure that led to equation (10)), equation (10) is replaced by:

$$I_{\pm}(\Omega, \Delta \alpha) = \frac{2}{\pi} \sin^2\left(\frac{\pi}{2}\frac{\Omega}{\omega_0}\right) \times \left(1 + 2 \sum_{n=1}^{N_p/2} \cos\left(2\pi n \frac{\Omega}{\omega_0}\right)\right)^2 \times \left[1 + 2 \sum_{m=1}^{(M-1)/2} \cos(mT_f\Omega)\right] \times \left\{3 - 2 \sin\left(\frac{\pi}{6} + \frac{T_f \Omega}{3} + \Delta \alpha\right) - 2 \sin\left(\frac{\pi}{6} + \frac{T_f \Omega}{3} - 2\Delta \alpha\right) - 2 \sin\left(\frac{\pi}{6} + \frac{2T_f \Omega}{3} + \Delta \alpha\right)\right\},$$  

(12)

where equation (12) degenerates back to (10) for $\Delta \alpha = 0$. The ellipticity, $\varepsilon$, of the emitted harmonics at their peaks’ maxima is the same for all emitted harmonics, and is given by:

$$\varepsilon = \frac{\sqrt{3} \cos\left(\frac{\pi}{6} + \Delta \alpha\right)}{1 + \sin\left(\frac{\pi}{6} + \Delta \alpha\right)}.$$  

(13)

Figure 2(c) shows the ellipticity as a function of $\Delta \alpha$. As shown, the ellipticity decreases almost linearly from 1 at $\Delta \alpha = 0$ to 0 at $\Delta \alpha = 60^\circ$ (where all pulses are aligned along the same axis). The intensity variation of the emitted harmonics as a function of $\Delta \alpha$ is given by:
As shown in figure 2 (d), equations (13) and (14) predict that the harmonics intensity decreases by only 11% as the polarization is tuned from circular to linear. At this point, it is instructive to discuss the origin for the relatively weak coupling between the ellipticity and the intensity of the emitted harmonics. Since tuning the relative angle $\alpha$ does not change the periodicity, the emitted harmonic frequencies are unaltered. However, breaking the circular 3-fold DS allows emission of previously forbidden harmonics at frequencies: $\frac{p}{\rho q T}$ for integer $q$. These harmonics radiate with a relatively weak intensity, which increases as the driver is taken further away from the circular case. The appearance of these new peaks reduces the intensity in the original helical harmonics.

4. Quantum numerical calculations

We explored the generation of circularly and elliptically polarized high harmonics by solving numerically the TDSE. We first describe the numerical model, and then present the results. We also compare between the numerical quantum and analytical models.

4.1. Quantum model

We numerically solve the 2D TDSE in the length gauge, within the single active electron approximation, and the dipole approximation. In atomic units the TDSE is given by:

$$\frac{i}{\hbar} \frac{\partial}{\partial t} |\psi(t)\rangle = \left( -\frac{1}{2} \nabla^2 + V_{\text{atom}}(\vec{r}) + \vec{r} \cdot E_{\text{pulse}}(t) \right) |\psi(t)\rangle,$$

where $|\psi(t)\rangle$ is the time dependent wave function of the single electron, and $V_{\text{atom}}(\vec{r})$ is the atomic potential. We use a model of a spherical atomic potential, where the electron is initially in the 1s orbital ground state, found by complex time propagation. In case the atom has valence p states (known to cause asymmetry in circular HHG [22, 35, 41]), our approach is still valid provided that the angular momentum of the total electronic valence states is zero (because the driver is quasi-linearly polarized). The Coulomb interaction is softened at the origin with one free parameter set to describe the ionization potential of the Ne atom ($I_p = 0.793$ a.u.). We set the atomic potential according to [42]:

$$\frac{\tilde{I}_1/\left(\Omega_{\alpha}, \Delta\alpha = 0\right)}{\tilde{I}_1/\left(\Omega_{\alpha}, \Delta\alpha = 0\right)} = \frac{2}{9} \left( 3 + \cos\left( \frac{\pi}{3} + 2\Delta\alpha \right) + 2 \sin\left( \frac{\pi}{6} + \Delta\alpha \right) \right).$$

(14)

Figure 2. Results of the analytical model—(a) harmonic spectrum around the 21st order for the circular HHG case (i.e. $\Delta\alpha = 0$) according to equation (10) with $N_p = 4, \tau = 0$, and $M = 1$. (b) Same as (a) but with $M = 3$. (c) Ellipticity at the peak emission’s maxima as a function of angle variation $\Delta\alpha$ according to equation (13). (d) Normalized intensity at the harmonic peak emission’s maxima as a function of angle variation $\Delta\alpha$ according to equation (14). Blue and red colors represent right and left circular emission, respectively.
Numerical results. Parameters of the driving pulse: \( N_p = 4 \), \( \tau = 0 \), \( M = 1 \), with \( I_0 = 3.36 \times 10^{14} \) W cm\(^{-2} \) and \( \lambda = 800 \) nm. (a) Harmonic spectrum for \( \Delta \alpha = 0 \). (b) Magnification of (a) around the 23rd harmonic order in the plateau. (c) Calculated ellipticity–helicity product for several representative harmonics as a function of \( \Delta \alpha \). The predicted line according to the analytical model (equation (13)) is shown in black. (d) Intensity variation of same representative harmonics as a function of \( \Delta \alpha \).

The harmonic spectra was calculated as the Fourier transform of equation (13)

\[
\int_{-\infty}^{\infty} \psi(x,t) \psi^*(x,t) e^{i k x} \, dx, \quad k = n a \frac{\lambda}{\Delta \alpha},
\]

where \( n a \) is a linear knob for the harmonic orders 22 and 24 are shown in figures 3 and 4, respectively. The HHG power spectra with similar parameters for the driving fields \( N_p = 4 \), \( \tau = 0 \), \( M = 1 \), and \( M = 3 \) in figures 3 and 4, respectively. The HHG power spectra with \( \Delta \alpha = 0 \) are shown in figures 3(a) and 4(a). The spectra between harmonic orders 22 and 24 are shown in figures 3(b) and 4(b), respectively. Comparing figures 3(a) and (b) with figures 2(a) and (b), respectively, shows that the harmonics in the quantum numerical results correspond to the analytical model selection rules. The average ellipticity of the harmonics (averaged over all the harmonics in the plateau region) was calculated numerically, and found to be \( \Delta \alpha \approx 0.98 \), i.e. very close to circular. Figures 3(c) and 4(c) present the calculated helicity–ellipticity product of several exemplary harmonics as a function of \( \Delta \alpha \). Clearly, all the harmonics closely follow the predicted (approximately linear) curve by the analytical model, from circular through elliptic to linear polarization. Thus, rotation of \( \Delta \alpha \) is a linear knob for the harmonic orders.
controlling the ellipticity of all the harmonics, collectively. Figures 3(d) and 4(d) show the peak intensity of several exemplary harmonics as a function of \( \Delta \alpha \). The intensities are fairly constant, varying by up to 25%, considerably less than in other techniques in which polarization control was demonstrated [27, 29].

Having established that the ellipticity of the high harmonics can be tuned finely by rotating the polarization axes of some of the pulses comprising the train of the driver, it is important to explore the sensitivity of this scheme to deviations of the pulse parameters, specifically to deviations in the intensity and temporal delays between the pulses. To this end, we calculated the ellipticity of the high harmonics using the field used in figure 3 with the following deviations: we reduce the intensity of the first of the three linearly polarized pulses in the train with respect to the other two by \( \Delta I \), or increase the delay between the first two pulses by \( \Delta \tau \). The deviation in ellipticity \( \Delta e \) is defined as the difference between the numerically calculated ellipticity and the analytically predicted ellipticity as given in equation (13). Figure 5 presents the ellipticity deviations of the 21st harmonic (figure 5(a)) and 13th harmonic (figure 5(b)) as a function of the normalized intensity deviation, \( \Delta I/\bar{I} \), and delay deviation, \( \Delta \tau/T \), when \( \Delta \alpha = 0 \) and \( \Delta \alpha = 30^\circ \). As shown, the ellipticity is quite sensitive to deviations in the driving pulse. For example, to insure \( \Delta e < 0.2 \) with target ellipticity \( \varepsilon = 1 \) for the 21st harmonic, \( \tau/T \) should be smaller than 0.5%, and \( \Delta I/\bar{I} \) should not exceed 1%. Notably, the sensitivity decreases for a smaller target ellipticity (compare in figure 5 between the deviations with \( \Delta \alpha = 0 \) and \( \Delta \alpha = 30^\circ \)), and also for decreasing harmonic order (compare figures 5(a) and (b)). Importantly, since the harmonic’s intrinsic phase depends on the intensity, so does the sensitivity to \( \Delta I \).

**Figure 4.** Same as figure 3, except that here \( M = 3 \) instead of \( M = 1 \), and the peak intensity of the driver is changed to \( I_0 = 2.68 \times 10^{14} \text{ W cm}^{-2} \).

**Figure 5.** Sensitivity of harmonic’s ellipticity due to deviations from the ideal pulse train which was used in figure 3. Deviation \( \Delta I \) represents intensity reduction of only the first linearly polarized pulse and deviation \( \Delta \tau \) represents additional delay between only the first two pulses in the train. (a) Ellipticity deviation of the 21st harmonic as a function of intensity and delay deviations with \( \Delta \alpha = 0^\circ \), \( \Delta \alpha = 30^\circ \). (b) Same as plot (a) but for the 13th harmonic order.
5. Conclusions

We propose and explore theoretically a new scheme to produce circularly and elliptically polarized high harmonics. The scheme is based on driving the HHG process by a train of quasi-monochromatic linearly polarized pulses that are identical except for their polarization axes. Circularly polarization high harmonics are generated when the train of pulses exhibits a 3-fold DS, i.e. when the polarization axis of each pulse is rotated by 120° with respect to the previous pulse in the train. Breaking the 3-fold DS by rotating the pulse’s polarization axes leads to high harmonics with fully controlled ellipticity, from circular through elliptic to linear. The scheme can be experimentally realized by a three-arm Mach–Zehnder like geometry, or through pulse shaping [46]. A probably more favorite and stable geometry would be fully in line [36]. The new technique exhibits several attractive features. First, the ellipticity of all the high-order harmonics vary evenly through the entire emitted spectrum (collectively), and also predictively according to an analytic model. Second, while tuning the ellipticity of the harmonics (by a single knob), the intensities of the harmonics vary by a relatively small amount. Third, this method largely preserves the single-cycle single-atom and macroscopic physics of the ‘standard’ and largely optimized scheme of HHG, where both the driver and high harmonics are linearly polarized. Forth, the allowed harmonic frequencies can be fine-tuned simply by changing the delays within the train. We believe these advantages will facilitate new opportunities in the research and applications of helically polarized high harmonics, including extreme UV coherent control, ellipsometry, and imaging ellipsometry.

Acknowledgements

This work was supported by the Israel Science Foundation (grant no. 1225/14), the Israeli Center of Research Excellence ’Circle of Light’ supported by the I-CORE Program of the Planning and Budgeting Committee and the Israel Science Foundation (grant no. 1802/12), and the Wolfson foundation.

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