High-harmonic generation: Taking control of polarization

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Polarization is a fundamental property of electromagnetic waves, and it often plays an important role in the way that light interacts with matter. It can be routinely controlled for visible light, whereas it is exceedingly difficult to control in the extreme ultraviolet (XUV) and X-ray regions. Until now, circularly polarized, bright, coherent XUV and X-ray waves could be generated only by using synchrotrons at large facilities. Such chiral light is used to study the chiral properties of matter, ranging from biomolecules to magnetic materials.

Now, in an elegant experiment, Avner Fleischer and his colleagues at Technion in Israel have succeeded in exerting complete control over the polarization of XUV radiation generated using a highly efficient and simple tabletop set-up [1]. They generated the XUV radiation using high-harmonic generation (HHG) – an extremely nonlinear interaction between an intense laser pulse (usually infrared radiation) and a gas of atoms or molecules. The incident pulse can generate harmonics of many tens to many hundreds of (sometimes up to a thousand) times the original laser frequency [2]. Up to 1,000 incident photons combine to produce a single high-energy photon. The basic physics of this remarkable process is sur-prisingly simple [3]: the intense laser pulse liberates an electron from an atom in the gas and then reaccelerates it back towards the parent ion; in the ensuing head-on collision, the electron recombines with the parent ion, emitting its substantial energy in a very short burst of light.

Although HHG is a convenient and popular approach for generating short-wavelength radiation, it usually affords little control over the polarization of the produced harmonics. One might naturally assume that employing elliptically polarized incident light would generate elliptically polarized harmonics, but this idea is sabotaged by the liberated electrons, which miss their parent ions, thereby quenching harmonic emission [4, 5]. Other methods,
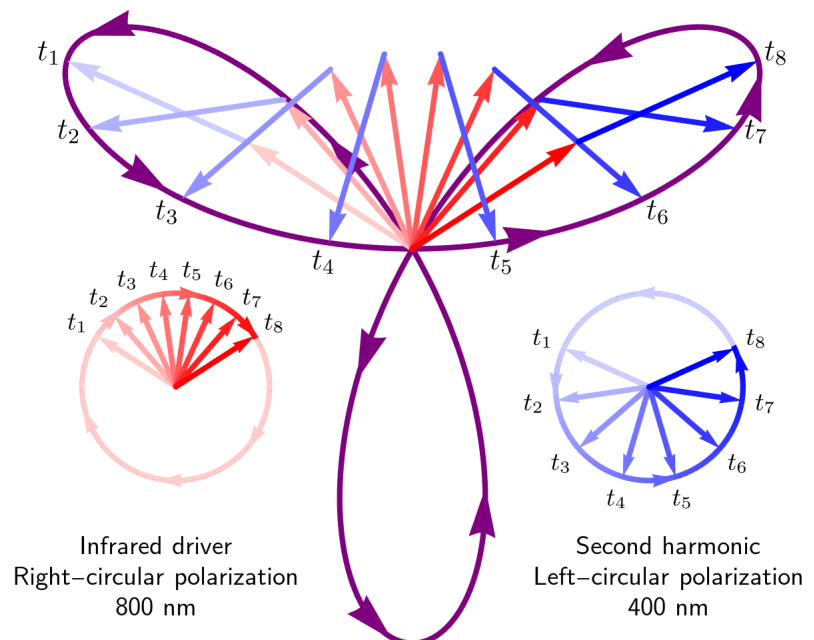

Figure 1: Controlling high-harmonic generation: combining a right-circularly polarized infrared driver (red arrows) with its left-circularly polarized second harmonic (blue arrows) at equal intensities yields a trefoil-shaped electric field (purple curve), which can guide electrons back to their parent ion to generate high harmonics with controllable polarization.

such as using a medium of pre-aligned molecules, have produced emission with relatively high ellipticities [6, 7] – but never with perfectly controlled polarization that is tunable from linear to circular polarization and always at the expense of more complex set-ups.

Inspired by a theoretical proposal by Long et al. [8], Fleischer and his co-workers have finally cracked the problem. Their scheme uses two co-propagating infrared and visible driving fields: a fundamental at a wavelength λ_1 of 800 nm and its (nearly) second harmonic ($\lambda_2 = 410$ nm, frequency $\omega_2 \approx 1.95\omega_1$). Both fields are circularly polarized, but they rotate in opposite directions. Each field separately would not produce any harmonics, but together they perform beautifully. The total electric field traces a trefoil figure (Fig. 1), which guides the liberated electrons away from the parent ion and back again, ensuring that head-on collisions occur from all sides during the laser cycle. The collisions produce a train of short XUV bursts, each with a linear polarization that rotates in space, spun by the driving trefoil. This translates into the circular polarization of the harmonics. In addition, by delicately controlling the polarization states of both drivers, Fleischer et al. have demonstrated full control over the ellipticity of the harmonics without any loss of efficiency in the generation process.

Their technique produces a train of light bursts that contain both left- and right-circularly polarized harmonics. Selecting a particular harmonic will yield a circularly polarized XUV pulse some 10 fs long. The combination of a short pulse duration and perfect control over polarization provides a unique, laboratory-scale light source for a wealth of applications in biology and materials science; this source generates radiation at extreme wavelengths, where it is difficult to engineer and control efficient sources of coherent light.

Crucially, the polarization of the harmonics is independent of the precise intensity ratio of the two drivers, making the experiment very robust. This feature can be easily understood by considering a simple photon picture. Suppose the infrared driver is left-circularly polarized with each photon having spin angular momentum $-\hbar$ (where $\hbar$ is the reduced Planck constant), while the 'blue' photons of the second harmonic each have a spin $+\hbar$. A harmonic photon created from n_1 infrared photons and $n_2 = n_1 \pm 1$ blue photons will have a spin equal to $(n_2 - n_1)\hbar = \pm\hbar$ and will correspondingly be left- or right-circularly polarized, independent of the driver intensity.

In one particularly ingenious twist of their experiment, Fleischer and coworkers managed to dig into this photon picture by introducing a slight redshift Δ to the second harmonic, causing it to be produced at 410 nm instead of 400 nm. This generates a slight redshift in the harmonics produced, enabling one to count the number n_2 of blue photons. For example, the combination of four infrared photons with three blue photons yields a frequency $\Omega(4, 3) = 4\omega_1 + 3(2\omega_1 - \Delta) = 10\omega_1 - 3\Delta$. A small but non-zero Δ allows for unambiguous identification of the net number n_2 of blue photons that 'went in', and the net number n_1 of red photons can also be determined from energy conservation.

With this information in hand, the researchers proceeded to test the conservation of spin angular momentum in their experiment – the first such attempt in HHG studies. Conventionally, harmonic generation is viewed as a parametric process: the atoms convert a certain number of driver photons into a harmonic photon of higher energy without changing the final atomic state. In this case, the generation process should preserve the angular momentum of light, including its spin angular momentum, which is associated with the polarization state. One can thus (i) change the amount of spin angular momentum that goes into the process by varying the ellipticity of the drivers from circular through elliptical to linear polarization; (ii) measure the spin angular momentum of the outgoing harmonics; and (iii) see whether the results match the expected conservation law.

Here, the experimental data are clean and clear and agree well with the numerical simulations performed by the researchers, but their interpretation is difficult. In their analysis, Fleischer et al. use the relationship between the expectation value for the spin angular momentum σ and the light ellipticity ε, $\langle\sigma\rangle = 2\varepsilon/(1 + \varepsilon^2)$, where the sign of ε depends on the helicity of the light. After measuring the ellipticity $\varepsilon(n_1, n_2)$ of the harmonic line that corresponds to the net absorption of n_1 infrared and n_2 blue photons, the researchers checked whether the corresponding spin angular momentum $\sigma(n_1, n_2)$ matches the conservation law $\sigma(n_1, n_2) = n_1\sigma_1 + n_2\sigma_2$.

They found that it does not. As soon as one of the driving fields became elliptic, the researchers observed a non-zero mismatch

$\delta(n_1, n_2) = \sigma(n_1, n_2) - (n_1\sigma_1 + n_2\sigma_2)$. However, when the mismatches of several lines (sometimes just two adjacent ones) were added, they perfectly cancelled each other. A tantalizing possibility emerges: for certain pairs of harmonics, the generation process conserves angular momentum only as a joint process, as if the photons were emitted together in correlated photon pairs. In other regimes, however, the angular momentum appears not to be conserved as a whole, even for the entire harmonic generation process. This would indicate that the generation process is not closed, and should be accompanied by some other excitation. The researchers speculate that the liberated electron carries away some angular momentum.

On the one hand, the model of Fleischer et al. appears to provide a good qualitative explanation for the selection rules observed in the experiment, and harmonic channels are born and disappear with ellipticities consistent with the model. On the other hand, the suggestion that the harmonic photons are sometimes emitted in pairs or that the generation process might not even be closed contrasts starkly with the standard view.

Certainly, the model proposed by Fleischer et al. has some puzzling features. For example, without the empirical correction $\delta(n_1, n_2)$, it would predict that certain channels are allowed only for perfectly circular drivers. Even infinitesimally small deviations yield grave consequences, not only for the high harmonics, but also for the conventional lower-order processes, such as four-wave mixing. Consider, for example, the $n_1 = 2$ and $n_2 = 1$ channel, which corresponds to the absorption of two left-circularly polarized photons $\hbar\omega_1$ ($\sigma_1 = -1$) and one right-circularly polarized photon $\hbar\omega_2 = 2\hbar\omega_2$ ($\sigma_2 = +1$), followed by the emission of the left-circularly polarized photon $\hbar\Omega = 2\hbar\omega_1 + \hbar\omega_2 = 4\hbar\omega_1$: $\sigma(2,1) = 2\sigma_1 + \sigma_2 = 2(-1) + 1 = -1$. However, as soon as the second harmonic is not perfectly circular, this emission becomes forbidden: for $\sigma_1 = -1$ and $\sigma_2 < 1$ one finds $\sigma(2,1) = 2\sigma_1 + \sigma_2 < -1$, which is simply not possible.

One potential weakness of the model developed by Fleischer and co-workers is that it tacitly assumes that the spin angular momentum will be conserved for the expectation values of the photon spins. This need not be the case. Elliptically polarized light can be viewed as a combination of two counter-rotating fields of different intensities. The channel $(2, 1)$ is still allowed for the elliptically polarized blue

driver – only its intensity is lower because there are fewer right-circularly polarized photons available for the process. Similar arguments apply to the higher-order processes [9].

Irrespective of how the theoretical discussion plays out, two points must be stressed. First, even if the harmonic photons are not emitted in quantum-correlated biphoton or multiphoton bunches, their emission is correlated in the classical sense: adjacent emission lines result from the same underlying electronic motion. Second, let there be light in every laboratory that needs it: coherent, bright, short pulses of XUV light with tunable polarization. Fleischer et al. have shown how to realize the proposal of Long and colleagues [8]. In their latest work [10], the team shows that it is even possible to implement phase matching to select emission lines of a specific chirality – an important step in converting individual circularly polarized emission lines into a circularly polarized attosecond pulse.

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