one of the two paths seriously envisioned for harnessing fusion energy. In semiconductor physics, polarization-resolved measurements of vertical-cavity surface-emitting lasers have shown bifurcation sequences typical of chaotic behaviour[9], which could be used for secure communications and random number generators. In quantum mechanics, polarization-resolved weak measurements have led to the first direct measurement of the wavefunction and Dirac distributions for polarization states of light[10], which opens perspectives in quantum information and metrology. Coming full circle, Mueller-matrix spectroscopic ellipsometry is used to fully quantify spatial dispersion in optical nanostructured metasurfaces. Ellipsometry thus helps understand the complex relationship between chirality, artificial magnetism and spatial dispersion.

Despite the approximations intrinsic to the design of the metasurfaces, all points on the Poincaré sphere, which gives a parameterization of the relative Stokes parameters (s_1, s_2, s_3), are theoretically reached with a negligible deviation on the order of 10^{-2}. The metagrating and the associated diffraction contrasts thus give an excellent representation of all possible polarization states. It should be noted that, despite being designed for a single wavelength (800 nm), the diffraction contrasts closely represent the Stokes parameters in the wavelength range 700–1,000 nm. However, there will be a compromise between the ability of the grating to measure weak beams and its bandwidth.

To truly benefit a wide array of applications, some of the shortcomings intrinsic to the design used by Pors *et al.*[3] will need to be overcome. Fortunately, some solutions already exist in the literature. For instance, to limit the impact of plasmonic losses, different materials such as dielectrics and transparent conducting oxides can be used as building blocks for the metagrating[11,12]. Besides, one of the limiting factors of the design at present is the grating that splits right-handed and left-handed waves. The geometric phase concept underlying its operation requires a birefringent grating with both a variable optical axis and half-wave-plate behaviour[13]. This latter requirement is not yet fulfilled as acknowledged by the authors themselves. Finally, interweaving the three gratings into one single metagrating significantly degrades their performance due to diffraction in the unwanted *yz* plane. This could be partially remedied by pushing up the diffraction thresholds along that axis by using a non-square lattice such as a triangular lattice.

In recent years, the field of singular optics, which takes advantage of not just spin angular momentum but also orbital angular momentum, has made tremendous strides. Most notably, it offers interesting perspectives for the field of quantum communications and cryptography. To put these notions on a strong theoretical footing, higher-order Stokes parameters, Mueller matrices, and Poincaré spheres have been defined to fully describe the novel polarization states of vector vortex waves[14]. Currently, no specific technique for the measurement of higher-order Stokes parameters has emerged but it would be greatly beneficial for the field of singular optics as a whole. If the metagrating used by Pors and colleagues could be extended to fully characterize waves of any given polarization state, it would provide for a substantial breakthrough. ❐

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References

1. Bohren, C. F. & Huffman, D. R. *Absorption and Scattering of Light by Small Particles* Ch. 2 46–53 (Wiley, 1983).
2. Berry, H. G., Gabrielse, G. & Livingston, A. E. *Appl. Opt.* **16,** 3200–3205 (1977).
3. Pors, A., Nielsen, M. G. & Bozhevolnyi, S. I. *Optica* **2,** 716–723 (2015).
4. Yu, N. *et al. Science* **334,** 333–337 (2011).
5. Ni, X. *et al. Science* **335,** 427 (2012).
6. Hsu, L. Y., Lepetit, T. & Kante, B. *Prog. Electromag. Res.* **152,** 33–40 (2015).
7. Sterzik, M. F., Bagnulo, S. & Palle, E. *Nature* **483,** 64–66 (2012).
8. Mondal, S. *et al. Proc. Natl Acad. Sci. USA* **109,** 8011–8015 (2012).
9. Virte, M., Panajotov, K., Thienpont, H. & Sciamanna, M. *Nature Photon.* **7,** 60–65 (2013).
10. Salvail, J. Z. *et al. Nature Photon.* **7,** 316–321 (2013).
11. Li., D., Fan, P., Hasman, E. & Brongersma, M. L. *Science* **345,** 298–302 (2014).
12. West, P. R. *et al. Laser Photon. Rev.* **4,** 795–808 (2010).
13. Marruci, L., Manzo, C. & Paparo, D. *Phys. Rev. Lett.* **96,** 163905 (2006).
14. Milione, G., Sztul, H. I., Nolan, D. A. & Alfano, R. R. *Phys. Rev. Lett.* **107,** 053601 (2011).

NONLINEAR OPTICS

Twisted high-harmonic generation

Crossing two focused laser beams with opposite circular polarization makes the production and application of circularly polarized light in the extreme ultraviolet and soft X-ray spectral regions considerably easier and more efficient.

Kjeld S. E. Eikema

Circularly polarized light in the extreme ultraviolet (XUV) (wavelength less than 100 nm) and soft X-ray regions is highly interesting for a number of applications, in particular for spectroscopy and as a probe of chiral matter. An example is magnetic circular dichroism, which enables the identification of differently magnetized domains due to a reflectivity difference when probed with either left- or right-handed circularly polarized light at short wavelengths[1].

Synchrotrons and free-electron lasers can provide the required circularly polarized XUV light and soft X-rays, but there is a need for systems on a smaller scale. In particular, the time resolution that the big facilities can provide (typically 100 fs) is far worse than the sub-femtosecond time resolution that has been demonstrated with ultrafast lasers[2]. Interestingly, the same process — high-harmonic generation (HHG) — that enables a high time resolution with lasers also generates short wavelengths. When strong laser pulses are focused in a gas target to an intensity of 10^{14} W cm^{-2} or more, sub-femtosecond bursts of light are generated with photon energies that are odd multiples (harmonics) of the incoming fundamental photons. In this manner even soft X-ray light has been generated[3] using far-infrared laser pulses. However, one cannot simply make circularly polarized light by HHG using a circularly polarized fundamental beam. Conservation of spin angular momentum strongly suppresses HHG in this case.

As they report in *Nature Photonics*, Daniel Hickstein and colleagues[4] experimentally demonstrate an elegant way around this problem, with surprising

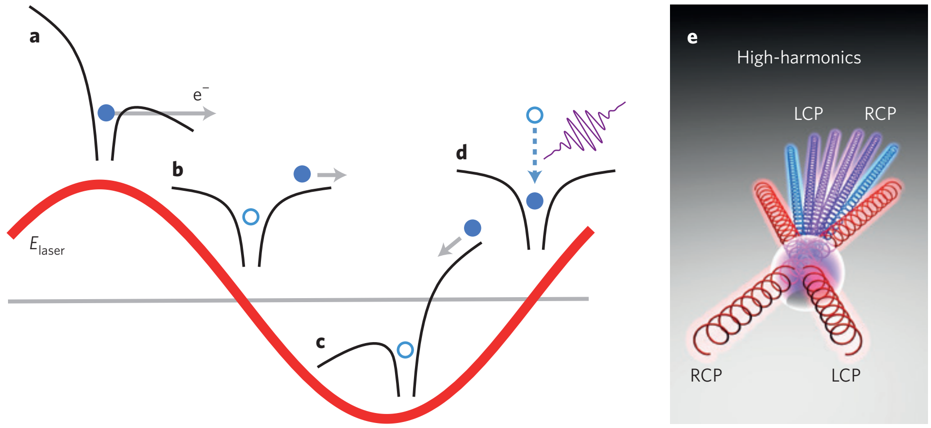

Figure 1 | Three-step model of high-harmonic generation with linear polarization in a gas, and the nonlinear geometry to generate circularly polarized light. **a**, Distortion of the Coulomb potential due to the electric field of the focused laser (E_{laser}), inducing tunnel-ionization of an electron. **b**, The electron wavepacket is accelerated away from the atom. **c**, Due to electric field reversal the electron wavepacket is accelerated back. **d**, The electron recombines with the parent ion when the field is nearly zero, leading to the emission of extreme ultraviolet radiation. **e**, By combining left-handed (LCP) and right-handed (RCP) circularly polarized fundamental beams in a non-collinear fashion, both LCP and RCP high-harmonics are produced, separated from the fundamental beams and at unique propagation directions.

consequences. Instead of using one single circularly polarized fundamental beam, two beams with opposite circular polarization (helicity) are focused at an angle in a gas jet. On first sight this seems like a bad idea. For example, the overlap region between the two beams in the focus is shortened compared with that of a single beam of light. This leads to a reduced interaction region, lowering the HHG yield. However, in the history of HHG, this is not the first time that a seemingly bad idea actually proves to be a very good one.

The crossed-beam geometry changes the phase-matching condition of the HHG process so that a higher gas pressure can be used, restoring the HHG efficiency. More importantly, it also leads to the generation of two separate groups of oppositely circularly polarized harmonic beams (Fig. 1e). If the angle between the fundamental beams is made large enough, then the generated XUV light is separated from the fundamental beams, and each harmonic has its own unique propagation angle. Therefore no grating or other lossy method has to be employed to separate the generated harmonics from the fundamental infrared beams. Effectively this leads to a much higher yield than earlier demonstrations of circularly (or elliptically) polarized HHG based on different methods[5–8]. Another unique feature is that both right- and left-handed circularly polarized light are available at the same time for each harmonic, which is also very helpful for accurate measurements of magnetic circular dichroism.

The process of HHG and the role of polarization can be understood by the elegant three-step model[9,10]. The first step is that the strong electric field of the focused laser beam distorts the Coulomb potential of the atoms (or molecules), which act as a nonlinear medium in the focus. On one side the Coulomb potential is raised, but on the other side the potential is lowered and an electron wavepacket can tunnel out (Fig. 1a). The field of the laser then accelerates the electron away from the parent-ion (Fig. 1b). When the field direction reverses after half an optical cycle, the electron wavepacket is accelerated back to the ion (Fig. 1c). The maximum kinetic energy of the returning electron wavepacket is given by its ponderomotive energy U_p, which depends on the fundamental laser frequency (ω) and laser intensity (I) through the relation $U_p \sim I/\omega^2$. Close to a zero crossing of the electric field of the light (assuming linearly polarized light), the electron wavepacket can recombine with the ion to radiate its excess energy in the form of high-energy photons (Fig. 1d). The maximum generated photon energy, E_{max}, depends on the ionization energy, I_p, and U_p through the relation $E_{max} = I_p + 3.17U_p$. This process can take place every half-cycle of the fundamental wavelength, leading to a series of XUV light flashes, each typically only a few hundred attoseconds long. The spectrum of this emission looks like a series of harmonics positioned at odd multiples of the fundamental photon energy. From the scaling laws above it is clear that a longer fundamental wavelength generates higher-energy photons. However, it also leads to a dramatically reduced efficiency per atom due to quantum diffusion of the electron wavepacket. This reduction can be compensated by proper phase matching using a higher density by confining the gas in a capillary[11].

The three-step HHG model is simple, yet powerful. It can predict all the basic properties of HHG, and is valid over a wide range of fundamental and generated wavelengths[11]. It also explains why linear polarization is normally required for HHG. In that case the returning electron wave packet stays in the same plane as the ion and can recombine. However, for a circularly polarized fundamental wave the electron will also move sideways, and can miss the ion, leading to ionization instead of harmonic generation.

Several approaches have been demonstrated recently to overcome this constraint. Elliptical vacuum ultraviolet radiation has been demonstrated using resonant HHG[8], or by using HHG with aligned molecules as a medium[5]. Full circular polarization was probably first achieved[12] by a pioneering experiment in 1995. In this case an ultrafast fundamental laser beam at 773 nm and its second harmonic at 386 nm were combined collinearly with opposite circular polarizations. Although efficient HHG was demonstrated in this case, the polarization state of the generated light was not analysed. Recently two experiments[6,7] confirmed that circularly polarized harmonics can indeed be generated in this manner, and also explained the mechanism. The combined influence of the two beams produces a complex rotating electric field with three-fold symmetry. Electron wavepackets can therefore return to the ion three times at angles of 120 degrees. Circularly polarized light is then generated by the recombining electron, while each subsequent harmonic is of opposite helicity.

However, Hickstein and co-workers have taken a new approach[4]. As mentioned before, two equal-intensity, but opposite circularly polarized ultrafast laser beams are crossed at an angle to generate harmonics. The combined electric field in the focus has linear polarization everywhere, so that the physics of standard HHG applies. Efficient HHG is therefore possible, in principle even to high photon energy, but why is the emerging HHG light then circularly polarized? For each point in space the phase is different between the two fundamental beams. When both beams combine to linearly polarized light, this phase leads to a polarization rotation as a function of the transverse position in the focus. The harmonics follow this spatial polarization pattern, which after the focus in the far field is equivalent to two beams of opposite circular polarization, even though locally the fundamental and harmonic lights in the focus are linearly polarized everywhere.

Many aspects of this non-collinearly circularly polarized high-harmonic generation (NCP-HHG) method are described by Hickstein *et al.*[4] in both the wave and photon picture. In the photon picture one has to consider that the HHG process should conserve parity, linear momentum and spin angular momentum. This means that only certain combinations of odd and even numbers of photons from both beams are possible. Emission of the generated light takes place in the direction given by the sum of the wave vectors of the interacting fundamental photons. As each harmonic can be produced by two different sets of combinations, it leads to emission in two opposite and unique directions, and with opposite circular polarization (Fig. 1e). This automatic separation from the fundamental beams and the separation in left- and right-handed circularly polarized light are much better and more efficient than any other method demonstrated so far. To make their point about this aspect, Hickstein *et al.* show in the Supplementary Information of their paper that a human hair can be put in the high-harmonic beams only 1 cm away from the focus where the harmonics are generated without inducing damage to the hair, even when the angle between the two high-power laser beams is only 30 mrad. The power of NCP-HHG is particularly demonstrated with an actual XUV (30–45 nm) magnetic circular dichroism (MCD) measurement of an iron film. An MCD asymmetry at the per cent level is readily detected in a matter of minutes. Moreover, simulations were performed predicting that NCP-HHG could possibly also be used to generate single circularly polarized attosecond pulses.

It is clear that NCP-HHG has a lot of potential, but there are also still some downsides. Although respectable HHG yields on the order of 10^8 photons per pulse are reported, this is mostly achieved when short fundamental wavelengths, such as 400 nm, are used. Considering the scaling law for the maximum photon energy as described earlier, one would also like to go to longer fundamental wavelengths, but the resulting reduction in HHG efficiency cannot currently be fully compensated by increasing the gas pressure in the focus for technical reasons. Also the spatial separation of the harmonics, from each other and the fundamental beams, works best for short wavelength fundamental beams because of the higher photon linear momentum.

However, there is no doubt that NCP-HHG provides an exciting new tool for generating circularly polarized harmonics, and it is expected that it will find many applications in fields such as (time-dependent) magnetic circular dichroism, circularly polarized attosecond pulse generation and spectroscopy. ❐

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References

1. Schütz, G., Knülle, M. & Ebert, H. *Phys. Scr.* **T49,** 302–306 (1993).
2. Krausz, F. & Ivanov, M. *Rev. Mod. Phys.* **81,** 163–234 (2009)
3. Popmintchev, T. *et al. Science* **336,** 1287–1291 (2012).
4. Hickstein, D. D. *et al. Nature Photon.* **9,** 743–750 (2015).
5. Smirnova, D. *et al. Phys. Rev. Lett.* **102,** 063601 (2009).
6. Fleischer, A., Kfir, O., Diskin, T., Sidorenko, P. & Cohen, O. *Nature Photon.* **8,** 543–549 (2014).
7. Kfir, O. *et al. Nature Photon.* **9,** 99–105 (2015).
8. Ferré, A. *et al. Nature Photon.* **9,** 93–98 (2015).
9. Corkum, P. B. *Phys. Rev. Lett.* **71,** 1994–1997 (1993).
10. Lewenstein, M. *et al. Phys. Rev. A* **49,** 2117–2131 (1994).
11. Popmintchev, T. *et al. Proc. Natl Acad. Sci. USA* **106,** 10516–10521 (2009).
12. Eichmann, H. *et al. Phys. Rev. A* **51,** R3414–R3417 (1995).

OPTICAL MEMORY

Phase-change memory

Integrated nano-optical memories may help overcome the limitations of communication speeds and energy costs in electronic chips. Now, using nanoscale phase-change materials researchers have realized the first multi-bit all-optical non-volatile memories with a very small footprint.

Eiichi Kuramochi and Masaya Notomi

The deployment of CMOS-based processors has led to a communication bottleneck because the energy cost of electrical signal communication in a chip has become increasingly dominant in electronic circuits. Research is now under way to introduce optical communication in a chip[1] because the energy cost is low, even for high-bit-rate communications. Among various possibilities to pursue this, the introduction of optical bit memories is vitally important as communication between electronic memories and processors is one of the most difficult bottlenecks to overcome in a chip. However, optical memories are also considered to be one of the most difficult optical counterparts of electronic devices in a chip, because we need to realize high-bit-rate operation, large-scale integrability and low power consumption simultaneously. Against this background, there have been a number of studies designed to realize integrable optical bit memories in a chip. Most of these memories employ optical bistability, which relies on the optical nonlinearity of materials. For example, optical RAMs realized in photonic crystal nanocavities that can operate with 40 Gbits s^{-1} optical signals have been reported[2]. As a result of tight light confinement, the power consumption is reduced to 40 nW (recently, it was further reduced by more than an order of magnitude). The required writing pulse is only a few fJ. In addition, over-100-bit wavelength-addressable optical memories have been demonstrated by integrating nanocavities with various resonant wavelengths[3]. However, these optical bistable memories need some bias power input to sustain bistable states. In that sense, they are volatile memories, similar to DRAM. Some applications certainly require non-volatile memories, which are hard to realize using optical bistability. Now, as they report in *Nature Photonics*, Carlos Rios *et al.*[4] have solved this problem, and have demonstrated multi-bit non-volatile all-optical memories by combining phase-change materials (PCMs) and nanophotonics.

PCMs are widely employed for large-capacity rewritable optical storage media, such as rewritable DVDs and Blu-ray disks[5]. Most established PCMs for this purpose are made of $Ge_2Sb_2Te_5$ (GST), whose phase can be changed at will between crystalline and amorphous via appropriate heating processes. There are two important facts regarding this phase change in GST. First,