to the excess energy during the oscillations? To answer this question, we notice that our system is coupled to an external laser, whose field is considered classical. This means that it has an unlimited energy reservoir, which can accommodate the cyclic variations of the cavity-stored energy. Remarkably, the classical external field does so with no detectable change in its own state, which allows one to separate it from the state of the rest of the system and to treat the latter as pure in the quantum-mechanical sense. It is intriguing to think about how the system analysis and its actual behaviour would change if the pump field was in a quantum state sensitive to the super-Rabi oscillations inside the cavity.

The presence of super-Rabi oscillations means that the cavity may contain N photons or none — all other possibilities are strongly suppressed. Each photon in a cavity has a chance to leave it during any cycle of the oscillations. The probability of such an event is determined by the oscillation period and the cavity decay rate. This is a subtle point. Whereas super-Rabi oscillations are a coherent collective process involving all N photons, leaving or staying in the cavity is a stochastic 'decision' that each photon makes on its own. However, if even one photon leaves the cavity (Fig. 1b), the oscillations will be disrupted: there is not enough energy remaining in the cavity to get to the $|e, N\rangle$ state again, and all other possible states are off resonance. The remaining photons have no other choice but to also leave the cavity (Fig. 1c). Thus, following the emission of the first photon, the entire N-bundle is radiated within the cavity decay time, while the external laser is delivering the energy necessary to restart the oscillations. The relative rate of these two concurrent processes determines whether the source operates as a randomly firing 'N-photon gun' or as a continuous-wave 'N-photon laser'.

Sánchez Muñoz and colleagues distinguish their N-photon bundle from a Fock state $|N\rangle$ based on the presence of a large vacuum component and on the photon correlation in the former. The vacuum component corresponds to a bundle that is not emitted. The correlation is suggested by the exponential distribution of the time intervals between the photons within a bundle, such that the earlier detected photons are more likely to be found closer together than those detected later. It is interesting to observe that such an exponential distribution for time intervals is typical for independent events governed by Poisson statistics; it may therefore be expected from the time-interval measurements in a Fock-state wave packet, provided that the measurement resolution is much better than the wave-packet length. For $N = 2$, such an exponential distribution has been experimentally observed[6].

This suggests that a photon bundle could be used as a flying Fock state that is conditional on the first detection. The ramifications of such an achievement for both fundamental and applied quantum optics are significant. For applied quantum science, the generation of Fock states is a long-awaited enabling technology for high-resolution imaging, lithography and metrology, as well as for quantum information processing. Even though the proposed source has yet to be demonstrated in a laboratory, many of its key components are already available, including tunable high-Q cavities[6] and microcavities integrated with quantum dots[7,8]. Various quantum emitters implemented as trapped ions or atoms[9] or superconducting quantum circuits[10] are also available. This makes the researchers confident that the N-photon quantum light source will be built in the near future. And in terms of fundamental science, the concept of light has received yet another interesting perspective. ❐

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HIGH-HARMONIC GENERATION

Taking control of polarization

The ability to control the polarization of short-wavelength radiation generated by high-harmonic generation is useful not only for applications but also for testing conservation laws in physics.

Misha Ivanov and Emilio Pisanty

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 surprisingly 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, 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 (λ_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.

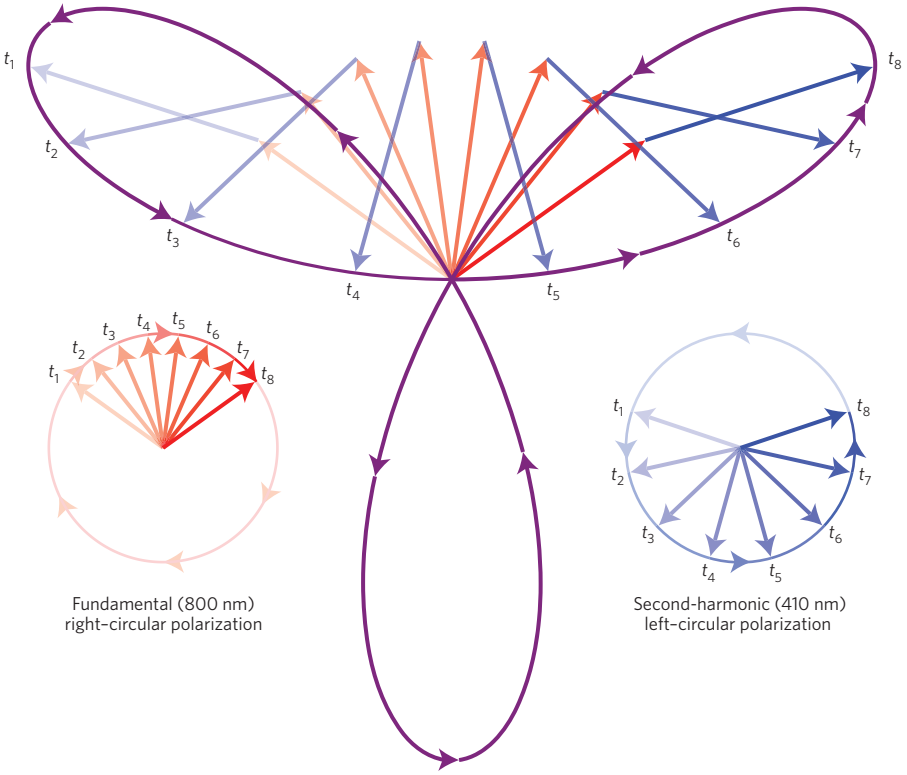

Figure 1 | Controlling high-harmonic generation: combining a right-circularly polarized infrared driver (red arrows) with its left-circularly polarized second harmonic (blue arrows) at equal intensities yields a trefoil-shaped electric field (purple curve), which can guide electrons back to their parent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

In one particularly ingenious twist of their experiment, Fleischer and co-workers 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 $\hbar\omega_2 = 2\hbar\omega_1$ ($\sigma_2 = +1$), 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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VIEW FROM... MRS SPRING

Materials pushing solar

Rapid developments in perovskite solar cells, photocatalysis and transparent conductors were showcased at the 2014 MRS Spring Meeting.

David Pile

San Francisco hosted the Materials Research Society's Spring Meeting (MRS Spring) during April 21–25 2014. Although the meeting had a strong materials focus, it overlapped significantly with photonics in the areas of detectors, sensors and solar energy. A particularly hot topic in the area of solar energy was cells incorporating perovskites.

Nam-Gyu Park of Sungkyunkwan University in Suwon, Korea, shared new results on his group's perovskite solar cell, which is based on methylammonium lead iodide (MALI) and formamidinium lead iodide (FALI) and exhibits a high conversion efficiency. Park explained to *Nature Photonics* that by controlling the crystal growth rate they prepared MALI samples of various crystal sizes. They found that increasing the MALI size above 200 nm improved the power conversion efficiency (PCE) due to enhanced charge carrier extraction and light harvesting efficiency. They also showed that FALI is photostable, does not exhibit hysteresis under an electric field and can enable record efficiencies.

"We achieved a best PCE of 17% and an average PCE of 16.4%. MALI showed a tetragonal-to-cubic phase transition at a temperature of ~57 °C, which may impair the photovoltaic performance, as the phase-transition temperature lies within the operating temperature of the solar cell," Park explained. "As FALI shows no phase transitions near the operating temperature,