We recently observed\(^1\) a striking form of optical pattern formation (see Fig. 1) in which, in passing through a sodium vapor cell, a single laser beam without feedback breaks up into a stable, regularly structured beam. Pattern formation in optical systems\(^2\) is an area of widespread interest, both from the conceptual point of view of understanding how regular patterns can emerge from uniform or randomly structured input fields, and from the practical point of view of using such patterns in image formation and manipulation. Spontaneous pattern formation has been studied previously in a variety of material systems,\(^2\) including atomic vapors, liquid crystals, photorefractive crystals, organic liquids, glasses, semiconductors and bacteriorhodopsin. Most previous observations of regular pattern formation were obtained in systems in which optical feedback plays an important role. This feedback can be produced by the use of optical resonators and/or by the use of counter-propagating beams. In contrast, the patterns that we observe occur when a single laser beam (i.e., with no feedback mechanism) passes through an atomic sodium vapor. Pattern formation of this type appears to have been previously unreported, although pattern formation has been observed in sodium vapor under somewhat different experimental conditions.\(^2\)

In our experiment, we inject a 150-mW collimated laser beam with a diameter of 220 μm into a 6-cm-long sodium vapor cell containing \(8 \times 10^{12}\) atoms/cm\(^3\). The laser is tuned 1.5 GHz to the blue side of the D\(_2\) resonance line. In passing through the cell, the beam is found to break into multiple components as a consequence of self-focusing effects. Under certain input conditions, the beam breaks up into three components of comparable power, the positions of which form an equilateral triangle. An example of such an arrangement is shown in the left panel of Fig. 1. The far-field pattern of this emission has the symmetric honeycomb pattern shown in the right panel of the figure. We find both the near- and far-field patterns to be stable for tens of minutes. The highly structured yet stable beam of the sort we have observed may constitute a system in which to study quantum statistical effects such as quantum images.\(^8\)

In summary, we have observed a dramatic example of optical pattern formation in which a single laser beam propagating without feedback through atomic sodium vapor develops a stable, regular, transverse structure. In particular, a three-filament near-field pattern leading to a honeycomb far-field pattern occurs at intensities near the saturation intensity and at powers larger than (but of the order of magnitude of) the critical power for self-focusing. The three-filament pattern has a uniform phase profile and strongly correlated power fluctuations, which suggest that it is perhaps a quantum image. These observations were also found to be in good agreement with numerical simulations of filamentation in a two-level atomic medium.\(^1\)

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**Figure 1** (a) Experimental setup used to study optical pattern formation; patterns were recorded both in the near and far fields. (b) Example of pattern formation as observed in the near field (left) and far field (right).
Precise Retardation Measurements Through Nonlinear Optics
Stefano Cattaneo and Martti Kauranen

Polarization is one of the most important characteristics of optical beams because many processes involving light depend on its polarization. Polarization is also extremely sensitive to the optical properties of materials. The fact that the optical properties of materials may affect polarization offers unique opportunities in areas including materials science, chemistry, biology and physics, but can also lead to detrimental effects. Polarization-mode dispersion, for example, can severely limit transmission speed in optical fibers. For this reason, in all these applications it is important to precisely determine the state of polarization.

The state of polarization can be specified by the relative amplitudes of two orthogonal polarization components and the phase difference (retardation) between them. Polarization in general and retardation in particular are very difficult to measure precisely. For example, the retardation of common birefringent elements such as wave plates is usually measured at most to \( \lambda/1000 \). Considerable effort has been put into refining existing measurement techniques, often at the expense of complicated experimental arrangements or data analysis. Therefore, any real advances in retardation measurements must be based on novel physical principles rather than on incremental improvements of traditional techniques.

Second-order nonlinear optical processes are remarkably sensitive to material symmetries and light polarization. These features have been successfully employed for detailed characterization of materials of low symmetries. We have recently shown that the situation can be reversed: measuring the second-order response of highly symmetric materials permits precise determination of the polarization state of light. In the initial demonstration of the technique using a simple setup, we have already measured the true retardation of a nominal half-wave plate to a precision and repeatability better than \( \lambda/10000 \).

Our technique is based on second-harmonic generation from thin films and relies on the fundamental symmetry properties of the nonlinear interaction. Two beams at the fundamental frequency \( w \) are applied on a thin film with in-plane isotropy [Fig. 1(a)]. The polarization of the target beam (wave vector \( k_1 \)) is to be determined. To do this, a probe beam (wave vector \( k_2 \)) with a controllable polarization is used and coherent second-harmonic light is detected in the sum direction \( k_1 + k_2 \). An arbitrary target polarization comprises linear and circular polarization components whose relative magnitudes depend on the target retardation. The circular component results in different second-harmonic intensities for left- and right-hand circular probe polarizations [see Fig. 1(b)]. This allows a precise determination of the target retardation.

The technique is remarkably insensitive to possible sources of error and can also be applied to measure low-level residual birefringence in optical components. We are investigating ways to extend the technique to arbitrary values of retardation and to further improve its precision.

References

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Figure 1. (a) Geometry of nonlinear retardation measurements. Two beams (target and probe) intersect in a poled thin film and the intensity of the second-harmonic signal (SHG) is measured. (b) SHG signal as a function of the rotation angle of the probe quarter-wave plate (QWP). The rotation angles of \(-45°\) and \(+45°\) correspond to left-hand circularly (LHC) and right-hand circularly (RHC) polarized probe beams. The determined target retardation was 178.45 ± 0.03°.
Conical Harmonic Generation In Isotropic Materials
Kevin D. Moll, Doug C. Homoelle, Alexander L. Gaeta and Robert W. Boyd

Nonlinear optical harmonic generation is the process by which light at frequency $\omega$ is converted to light at frequency $n\omega$, where $n$ is an integer. In order for this process to occur efficiently, two conditions must typically be met: the light-material interaction must produce a nonzero polarization component oscillating at the harmonic frequency; and the phase velocities of the fundamental and harmonic waves must be equalized (i.e., phase matched). For an isotropic medium, the nonlinear susceptibility tensor elements $\chi^{(n)}$ vanish for even values of $n$, which results in the commonly held belief that generation and amplification of light at even multiples of the harmonic frequency are not possible. Furthermore, dispersion of the refractive index inhibits phase matching in isotropic media, and the generation of the odd-order harmonics occurs with very low conversion efficiency.

We have recently investigated a novel process by which the problems associated with phase matching and the symmetry conditions in isotropic materials can be overcome. Unlike the typical scheme of generating the $n$th harmonic through a $\chi^{(n)}$ interaction, we used a higher-order $\chi^{(2n+1)}$ process. As an example, the photon-energy diagram associated with the $\chi^{(3)}$ process responsible for second-harmonic generation is depicted in Fig. 1(a). The consequence of such an interaction is two-fold. First, a nonvanishing odd-order susceptibility is responsible for the conversion, and therefore even-order harmonic generation is allowed in an isotropic material. Second, most materials exhibit normal dispersion in the frequency range in which they are transparent. However, the added degrees of freedom inherent in a higher order process allow for phase matching by emission of the harmonic radiation at an angle relative to the input beam, as shown in Fig. 1(b), which—when revolved around the input beam—results in a cone.

Unlike the conventional harmonic generation process, a seed beam must be present at the harmonic wavelength for the conical-emission process to occur. The seed beam may result from quantum background radiation, be induced by surface-nonlinear effects, or (in the case of odd-harmonic generation) may grow from the phase-mismatched $\chi^{(2)}$ process. We performed simulations of third-harmonic generation through a phase-mismatched $\chi^{(2)}$ process and a higher order $\chi^{(3)}$ process from a focused input beam. As seen in the inset of Fig. 1(c), the third harmonic is generated in the form of a cone. As a proof of principle, we observed this third-harmonic process experimentally in sapphire [see Fig. 1(d)].

Even though higher order interactions are typically weak, we expect this class of processes to play a role whenever intense electric fields are used. For example, in the case of high-harmonic generation, the perturbative expansion is no longer satisfied, and higher order processes may occur with the same probability as the lower order ones with the additional attribute that the phase-matching condition would be automatically satisfied.

References

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Figure 1. (a) Photon-energy picture of the higher order nonlinear optical process responsible for second-harmonic generation in an isotropic material and (b) corresponding phase-matching diagram. (c) Results from simulations that model third-harmonic generation seeded by the phase-mismatched $\chi^{(2)}$ process. Inset: Emitted radiation is in the form of a cone. (d) Far-field image of the third-harmonic cone emitted from a sapphire sample using tightly focused femtosecond pulses at 1350 nm.
Surfactant Enhanced Optical Reorientation in Azo-Dye-Doped Liquid Crystals
Artem Petrossian and Stefania Residori

Azo-dye-doped liquid crystals are attractive candidates for photonic devices because, at extremely low input light intensities, they are characterized by high degrees of birefringence, optical selectivity and photorefractivity. The huge optical nonlinearity they exhibit has been attributed to light-induced modifications of the anchoring strength at the bounding plates of the cell. We have conducted a systematic investigation of the influence of the liquid-crystal anchoring-layer interface on the optical reorientation phenomena. Homeotropic anchoring of the liquid crystals was obtained by use of different types of alignment agents. We have shown that the high degree of optical nonlinearity is mediated by the specific anchoring agent and that it is extraordinarily large only when the confining plates have been coated with an ionic surfactant, such as hexadecyl trimethyl ammonium bromide (HTAB) or hexadecyl trimethyl ammonium chloride (HTAC).

All our experiments were performed with 10-μm-thick, homeotropically aligned cells filled with the same mixture of liquid-crystal 4-cyano-4-pentylbiphenyl (5CB) and azo-dye Methyl Red (0.3 wt.%). For all the cells, we saw a photovoltaic effect, but only for the cells treated with HTAB or HTAC did we observe an extremely large optical reorientation. When one of these cells was illuminated by two interfering Ar⁺ laser beams (total input intensity \( I_{\text{in}} \approx 100 \mu \text{W/cm}^2 \), normal incidence), strong diffraction of the probe He-Ne laser beam was obtained. Diffraction efficiency is at maximum when the probe polarization is orthogonal to the polarization of the pumps. The diffraction pattern observed for a HTAC-coated cell is shown in the figure, together with the measured diffraction efficiency; \( I_0 \) and \( I_{+1} \) are the intensities of the 0 and +1 order, respectively.

By use of the usual formula for Raman-Nath diffraction and by expressing the refractive index change as \( \Delta n = n_2 I_{\text{in}} \) [Ref. 1], we estimate the nonlinear coefficient to be as large as \( n_2 \approx 100 \text{ cm}^2/\text{W} \), which is the largest value observed up to now for any nonlinear optical medium. By testing different concentrations for the ionic surfactant, we have been able to identify the one that optimizes the optical reorientation and hence the nonlinear response of the cell. We have also shown that the reorientation takes place even when the cell is illuminated by only one pump, that is, the process does not require a grating. Indeed, once started at the surface, reorientation propagates in the bulk of the cell, where it is sustained by the trans-cis photoisomerization of the azo-dye dopants.

References

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Figure 1. Top: Typical diffraction pattern observed for a HTAC coated cell. Bottom: Diffraction efficiency measured as a function of the input intensity.
All-Optical Switching in a Coherent Atomic Medium
Min Xiao, Hai Wang and David Goorskey

Electromagnetically induced transparency (EIT) is interesting in its own right and also possesses certain qualities—namely reduced absorption and large dispersion near resonance—that make it promising for future device applications. Enhanced optical nonlinearity is present near EIT conditions, which allows for many useful and interesting applications. One of the most important elements of all-optical computation and information processing is fast, all-optical switching. With the increased Kerr nonlinearity that exists near EIT conditions, we have recently demonstrated that all-optical switching can be realized in an atomic medium within an optical ring cavity.

Kerr nonlinearity is a phenomenological process by which the index of refraction of a medium is modified by the intensity of the light passing through the medium. We have directly measured the Kerr nonlinear index of refraction as a function of frequency detuning from resonance of a probe beam in rubidium vapor and showed that EIT significantly modifies the Kerr nonlinear index near resonance [Fig. 1(a)]. We find the Kerr nonlinear index depends strongly on several experimentally controllable parameters: the probe and coupling laser frequency detunings and the coupling optical power. With only small frequency detunings (<10 MHz) the Kerr nonlinear index can change from a large positive value of ~7 x 10^6 cm^2/W to almost nothing, or it can change sign. This kind of sensitive control in nonlinearity allowed us to observe bistability and dynamic instability at low thresholds.

With such controllable nonlinearity, we also achieved all-optical switching in a three-level atomic medium (in a vapor cell) by switching between two distinct steady-state intensities inside an optical cavity. A probe beam enters the cavity through a concave mirror and a coupling beam enters via a polarizing beam splitter. As the probe beam intensity is scanned up and down using an electro-optic modulator, the cavity output undergoes a clear hysteresis pattern [see Fig. 1(c)] indicative of optical bistability, because of the EIT-enhanced Kerr nonlinearity. Because the nonlinearity is controllable, we can switch between low and high steady-state intensities at the cavity input power of 0.39 mW. We accomplish this by switching the frequency detuning of the coupling beam between two values (24 MHz separation) through electro-optic modulation [Fig. 1(e)] near EIT resonance conditions. The cavity output power is switched back and forth between low and high steady-state values with a switching ratio of ~30:1 as the frequency of the coupling laser is modulated [Fig. 1(d)]. This is the first experimental demonstration of all-optical frequency-to-amplitude switching in such a system.

The current switching speed is limited by the speed of our frequency modulation (~200 kHz).

We have developed an efficient, all-optical switching device by taking advantage of the sensitive controllability of EIT-enhanced Kerr nonlinearity near resonance in three-level rubidium atoms. While the current device has not been optimized, we see it as a proof-of-principle demonstration that will enable greater advances in the field of all-optical communication and information processing. With the advent of EIT in solid-state materials, it will be possible to realize the practical applications of such all-optical switching.

References

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