Managing Light in Nonlinear Disordered Media

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It is well known that efficient energy transfer in nonlinear parametric optical processes such as second harmonic generation (SHG) requires phase-matching between the interacting waves. In order to fulfill this condition, researchers have developed various methods, including the quasi-phase-matching (QPM) technique,[1] which relies on the spatially periodic modulation of the medium's nonlinear properties.

In the case of ferroelectric crystals, this is typically achieved by inducing a periodic anti-parallel ferroelectric domain structure, which leads to periodic reversal of the sign of quadratic nonlinearity. However, as the modulation period critically depends on the wavelengths of interacting waves, once fabricated the structure is only effective for the particular choice of the wavelength, hence limiting its practical applicability. To overcome this restriction and broaden the useful range of the wavelengths, one needs to fabricate costly multi-period or chirped structures.

Interestingly enough, nature itself offers another solution. We have recently achieved broadband SHG in strontium barium niobate (SBN) crystal using femtosecond pulses,[2] where, for the input infrared beam propagating along the direction of the domains, the second harmonic wave is emitted in a form of a cone, as shown in part (a) of the figure.

This effect is based on the fact that naturally grown ferroelectric crystals already exhibit multi-domain structure, with domains having random distribution of size and orientation. Such a disordered nonlinear medium[3] is equivalent to an effective QPM system with an almost infinite set of reciprocal wave vectors, enabling quasi-phase-matching for any parametric process—for example, SHG or sum-frequency mixing in an ultrabroad frequency range, which is limited only by the transparency window of the material. In the case of SBN crystals, the anti-parallel domains are tens of microns long with a typical diameter from one to a few microns.[4]

The ability to achieve SHG for almost arbitrary wavelength suggests an immediate application for short-pulse monitoring. We demonstrate this concept by realizing a simple autocorrelator with counter-propagating femtosecond pulses,[5] as shown in (b). When pulses propagate in the direction perpendicular to the domains, the second-harmonic signal consists of two components—first, one generated by each individual pulse, and, second, one created by the mixing of photons from both pulses. The latter is the autocorrelation signal while the former forms a background.

Parts (c) and (d) depict typical autocorrelation traces as seen with the CCD camera for pulses with straight and tilted pulse fronts, respectively. The signal to background of this simple device can be greatly enhanced by propagating both pulses along the optical axis; this coincides with the domain's long dimension. In this case, the transversely mounted CCD camera will record the second-harmonic signal generated only by counter-propagating pulses in their overlapping region, leading to a background-free autocorrelation trace.

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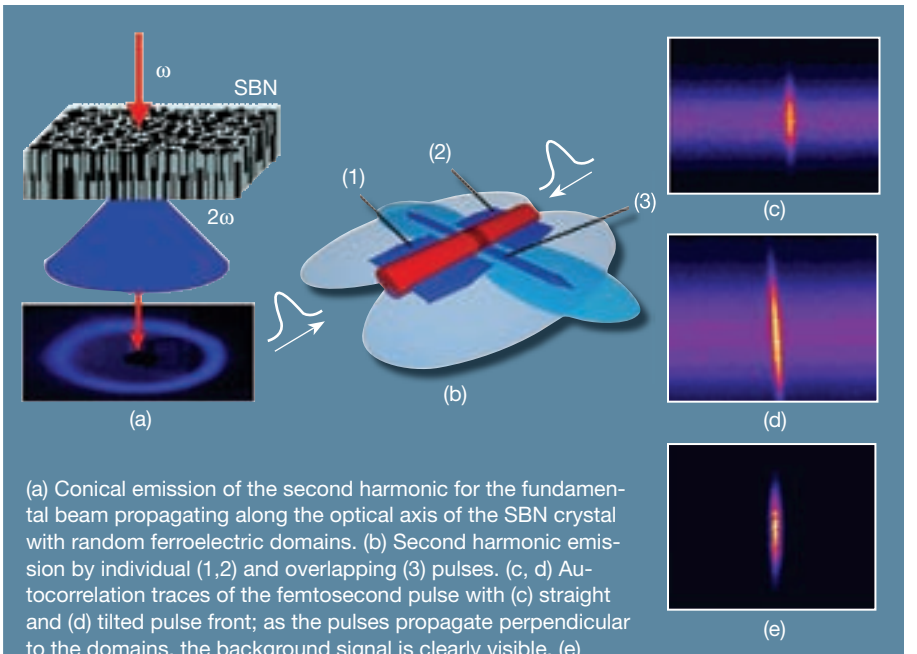

(a) Conical emission of the second harmonic for the fundamental beam propagating along the optical axis of the SBN crystal with random ferroelectric domains. (b) Second harmonic emission by individual (1,2) and overlapping (3) pulses. (c, d) Autocorrelation traces of the femtosecond pulse with (c) straight and (d) tilted pulse front; as the pulses propagate perpendicular to the domains, the background signal is clearly visible. (e) Background-free autocorrelation trace for pulses propagating along the domains.

Self-Healing and Laser Hardening of Nonlinear-Optical Materials

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All materials degrade in the presence of light. Paper yellows, fabrics fade and light sources burn out. Materials that generate, manipulate, transmit or record light suffer from some degree of photodegradation, especially at high intensities or prolonged exposure. The design of more robust materials is an important prerequisite for developing new photonic technologies.

The top portion of the figure shows red amplified spontaneous emission (ASE) generated from a poly (methyl methacrylate) (PMMA) polymer optical fiber (green) doped with Disperse Orange 11 (DO11) dye that is pumped with 532 nm light. ASE light exiting the fiber from the left illuminates a white screen, while a passive fiber guides ASE light that is emitted to the right.

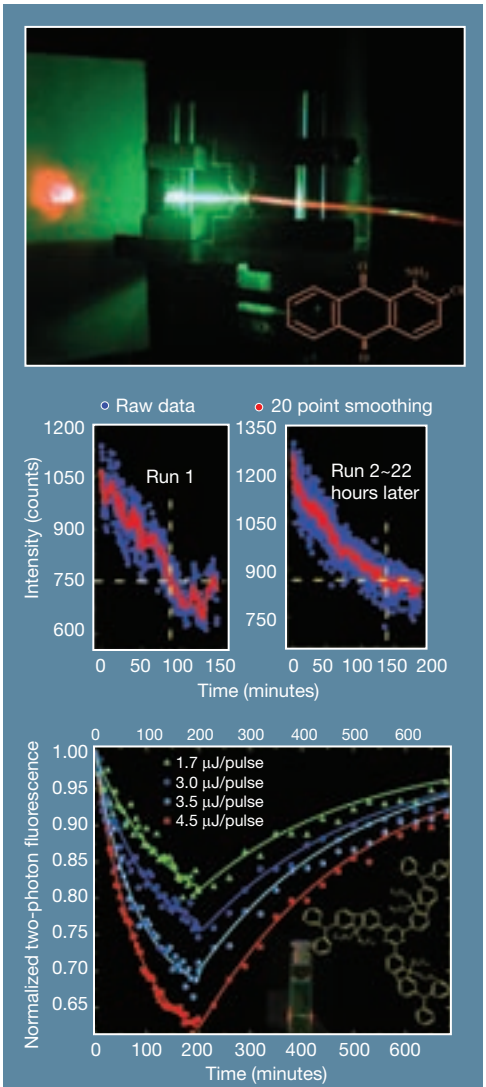
Surprisingly, while the DO11 dye degrades irreversibly in liquid solution,[1] in a polymer host, a degraded sample recovers in the dark. The central part of the figure shows two degradation cycles separated by about 22 hours of rest.[2] After the first cycle, the ASE efficiency increases by 20 percent and the photodegradation half-life increases by almost a factor of two. At the start of the third cycle, the efficiency increases by another 20 percent (not shown), suggesting that the polymer can be hardened to subsequent photodegradation and made more efficient by laser cycling.

When excited, DO11 relaxes nonradiatively into an excited state of its tautomer, which is formed when a proton jumps from the NH_2 group to the oxygen. ASE light is emitted upon de-excitation to the tautomer ground state.[2] The tautomer either converts back to the DO11 ground state, where it can again be excited and emit ASE, or the strongly dipolar tautomer can form a dimer with a nearby DO11 molecule. Fluorescence and ASE are quenched in the dimer.[3] The material self-heals in the dark when the dimers dissociate into single molecules.

Given the complex nature of light generation, photodegradation and self-healing, it was believed that the DO11/PMMA material was unique. Recently, a very different chromophore, AF455 (shown in the inset of the bottom portion of the figure) was discovered to photodegrade during two-photon fluorescence and self-heal after about an 8-hour rest.[4] So, the phenomenon of photodegradation and self-healing appears to be a more universal one than previously thought: Two very different optical processes and two dissimilar molecules show the same behavior. However, the mechanisms are likely quite different. (The AF455 molecule is not believed to tautomerize, and cannot form a dimer because it is too big to move appreciably through the polymer.)

The polymer host appears to mediate the interaction of light with the guest molecules in a way that provides a new degradation pathway that is reversible. This new pathway is favored over the irreversible one that is typical for molecules in solution.

Our finding suggests the exciting possibility that more robust materials can be developed by using combinations of existing chromophores and polymers, then hardening them by laser cycling. If this phenomenon can be harnessed in a broad range of materials, it would impact all technologies that generate and manipulate light.



(Top) Amplified spontaneous emission (ASE) from DO11 chromophore (inset) doped in PMMA polymer fiber; (center) decay and recovery of ASE; and, (bottom) decay and recovery of two-photon fluorescence from the AF455 chromophore in PMMA; and chromophore in solution (inset).

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All-Optical Quasi-Phase Matching in Extreme Nonlinear Optics

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In regimes where conventional phase matching is not possible, quasi-phase matching (QPM) techniques permit significant conversion efficiency in parametric nonlinear processes. QPM is especially critical for extreme nonlinear optical processes, such as high harmonic generation (HHG), where phase matching is restricted to photon energies <130 eV. In HHG, near-IR light is coherently upconverted into the extreme ultraviolet (EUV) and soft X-ray regions of the spectrum.

However, during HHG, the medium is ionized, and the associated strong plasma dispersion precludes phase matching at high energies. Previous implementations of QPM to enhance HHG used hollow waveguides with a modulated diameter to modulate the intensity of the driving laser, perturbing the HHG process to implement QPM. This work enhanced the conversion efficiency by >10×, limited by variations in phase matching conditions within the medium.

We demonstrated that counterpropagating beams of light can be used implement quasi-phase matching of high harmonic generation over a broad energy range from the EUV to > keV.[1-4] The presence of the counterpropagating field perturbs the phase of the radiating electron, which in this case is in the continuum. This perturbed phase is directly mapped onto the coherent X-ray phase, providing a means for implementing QPM.

We first used a single counterpropagating pulse to map *in situ* the coherence length for the HHG frequency conversion process.[1] By scanning the collision point between an intense forward-propagating laser pulse that generates the harmonics, and a weak counterpropagating pulse that modulates their phase, one can observe large modulations in the output. These oscillations correspond to the coherence length of the HHG process, which varies over the length of the gas-filled hollow waveguide.

To implement QPM, one must use a light pattern that matches the coherence length oscillations. Using three counterpropagating pulses in sequence, we enhanced the flux of a single isolated harmonic by suppressing harmonic emission from consecutive out-of-phase zones.[2] Selective enhancement of >300× at photon energies around 65 eV (41st harmonic) was observed in argon in a regime that cannot be phase matched using conventional approaches. In the future, by adding more pulses, and optimization of their shape,[3] we should obtain even greater enhancement and spectral selectivity. This technique has also been extended to higher photon energies in helium, showing >100× enhancement at the 89th harmonic order (140 eV).

In theoretical work, we demonstrated that all-optical QPM can be extended to very high energies (keV) using a scheme that is intermediate between QPM and true phase matching. Instead of using a sequence of pulses, one can use the oscillating field of a quasi-continuous infrared laser to adjust the phase of the emitted X-rays. This technique appears feasible even for the generation of coherent hard X-rays.[4] All-optical quasi-phase matching is promising for extending efficient coherent X-ray generation to high energies and for applications in atomic and molecular dynamics, nano- and bio-imaging, and materials science.

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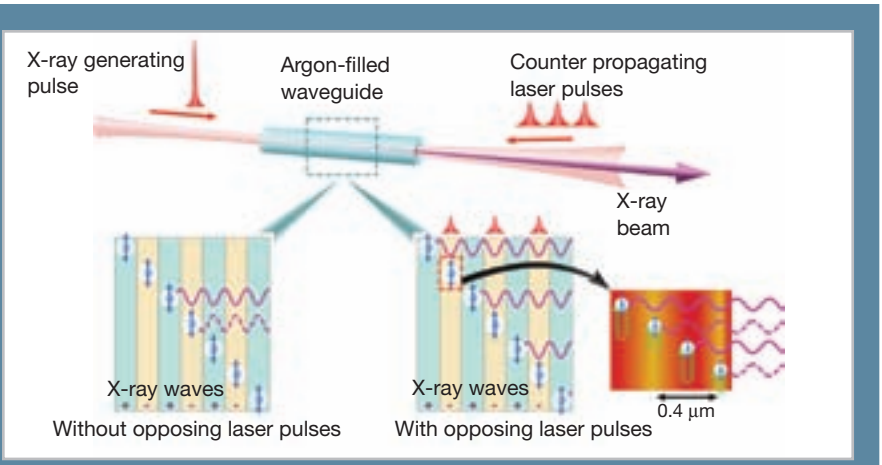

Light patterns generated by a counterpropagating pulse-train in a hollow waveguide allow quasi-phase matching of high-order harmonic generation by manipulating the phase of the harmonic emission. Under conditions of poor phase matching (bottom left), blue and orange coherence zones destructively interfere due to the phase slip between the propagating harmonic and driving fundamental fields. When counterpropagating pulses overlap with the driving pulse in the orange regions (bottom right), the harmonic emission is suppressed by scrambling the electron and X-ray phase (inset), allowing emission from the blue regions to interfere constructively to generate a bright EUV output.