

Managing Light in Nonlinear Disordered Media

Robert Fischer, Dragomir Neshev, Andrey Sukhorukov, Solomon Saltiel, Wieslaw Krolikowski and Yuri Kivshar

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 medium³ 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 ultra-broad 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. ▲

[Robert Fischer, Dragomir Neshev, Andrey Sukhorukov, Yuri Kivshar and Wieslaw Krolikowski (wzk111@rsphysse.anu.edu.au) are with the Center for Ultra-high Bandwidth Devices for Optical Systems and the Research School of Physical Sciences and Engineering at Australian National University, Canberra, Australia. Solomon Saltiel is with the department of quantum electronics at Sofia University, Bulgaria.]

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