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Martti Kauranen

Science **342**, 1182 (2013);
DOI: 10.1126/science.1247622

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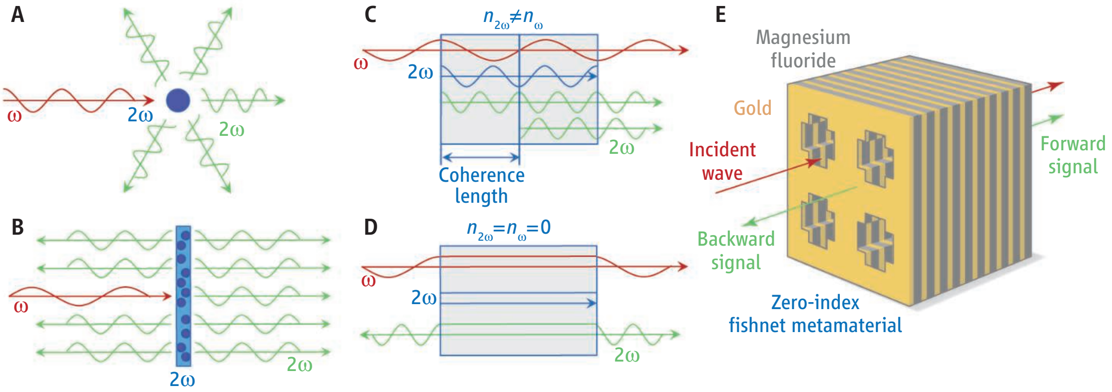
PHYSICS

Freeing Nonlinear Optics from Phase Matching

Optical metamaterials that have a refractive index of zero can boost the efficiency of nonlinear optical processes.

Martti Kauranen

The advent of lasers quickly led to the demonstration of nonlinear optical effects. One of the simplest examples of a nonlinear effect is doubling the input frequency of light with a nonlinear crystal. The output intensity of the frequency-doubled light then grows nonlinearly with the intensity of the laser beam. To maximize macroscopic nonlinear signals, the wavelets emitted by the elementary nonlinear sources need to add up in phase. On page 1223 of this issue, Suchowski *et al.* (*1*) report an interesting approach to phase matching with metamaterials—artificial nanostructures with unusual optical properties created by tailoring their structural features (*2*). The authors avoided the phase-matching problem by relying on a metamaterial that has a refractive index for the interacting wavelengths near zero (keeping in mind that the refractive index of the vacuum is not zero but unity). Furthermore, they achieve equal nonlinear emission in two opposite directions simultaneously, whereas conventional phase-matching techniques maximize the nonlinear signal in only one direction.



Phase matching for second harmonic (SH) generation. The spatial oscillations of the fundamental wave and SH source polarization are shown by red and blue lines, respectively; green lines indicate SH wavelets. (**A**) An elementary SH source emits in all directions. (**B**) A thin film emits symmetrically in the forward and backward directions. (**C**) For thick samples, the phase relation between the SH wavelets is lost after a coherence length, because the source and wavelets oscillate at different rates. (**D**) In a material with refractive index *n* of zero for both wavelengths, the oscillations do not occur and all SH wavelets add up in phase in the forward and backward directions. (**E**) The fishnet structure of Suchowski *et al.*, with zero refractive index, demonstrates equal nonlinear generation in the forward and backward directions for four-wave mixing.

Phase matching is challenging because the wave oscillations in propagation are determined by the refractive index of the material, and the refractive index depends on wavelength. To better understand the phase-matching problem, a closer look at frequency doubling, or second harmonic (SH) generation, is helpful. A wave at the fundamental frequency ω incident on a nonlinear material (such as lithium niobate) gives rise to an oscillating source polarization at the SH frequency 2ω. When the source is a single atom or molecule (see the figure, panel A), SH emission occurs in all directions. For thin-film sources (see the figure, panel B), the SH wavelets add up in phase only in the forward and backward directions, which represents transverse phase matching (perpendicular to the direction of propagation).

For thicker samples (see the figure, panel C), the wave oscillations in propagation become important, and their rate is proportional to the refractive index. The source oscillations follow the fundamental field but occur twice as rapidly. In general, however, the refractive index for the SH wavelets is different because of the higher frequency. The wavelets from different locations then lose their phase relation after a distance known as coherence length. The coherence length for forward SH generation is typically on the order of 10 µm, but is only about 100 nm in the backward process, making it much weaker.

Perfect phase matching in the forward direction occurs when the refractive indices for the two wavelengths are equal. This condition can be achieved in birefringent crystals for different polarizations of the interacting waves. Quasi–phase matching (*3*) relies on micro- or nanostructuring the material so that it reverses the sign of its nonlinearity after every coherence length, thereby restoring the phase relation between the wavelets. This approach is possible for either the forward or the backward process, but not for both at the same time. Finally, if the refractive index can be reduced to zero, the oscillations cease in the material, and all of the wavelets add up in phase in all directions (see the figure, panel D). This case is the one demonstrated by Suchowski *et al.* in a metamaterial.

Metamaterials first received attention because they can possess a negative refractive index (*4*), which allows imaging with resolution below the diffraction limit (*5*). Metamaterials usually consist of metal-dielectric composites, whose plasmonic resonances (collective oscillations of electrons) can enhance optical effects, including nonlinear ones (*6*). Nonlinear experiments on metamaterials in the optical regime have mainly been limited to thin, surface-type samples (*7*–*9*), where phase matching is not a constraint. However, negative-index bulk materials and their phase-matching properties have been studied in the microwave regime (*10*).

Suchowski *et al.* extend these concepts to a new regime where the refractive index is near zero. They used a "fishnet" metamaterial (*11*, *12*), which consists of alternat-

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ing layers of metal and dielectric perforated with holes (see the figure, panel E). The experiment was performed using four-wave mixing (FWM) instead of the simpler SH generation. The main reason for this is that the structure has a near-zero index only over a narrow wavelength range around 1330 nm. Whereas SH generation involves two very different input and output wavelengths, FWM occurs between nearby wavelengths. Typically, interaction among three beams of nearby frequencies gives a fourth (signal) beam also at a nearby frequency, which is useful for creating multiple frequencies for fiber-optic communications.

Suchowski *et al*. used a laser producing ultrashort-duration (femtosecond) pulses. Because such pulses consist of several frequency components, the FWM process occurs between different frequency components of each pulse, allowing pulse timing and beam alignment difficulties to be avoided. The signal can be detected at a nearby frequency, and all the interacting frequencies are now in the range where the metamaterial has a near-zero index. The forward and backward FWM signals are indeed equally strong, even though the sample cannot be considered thin.

The fishnet metamaterial is highly anisotropic with different properties in the sample plane and along its normal. In the present experiment, the sample has near-zero index only for light propagating along the sample normal, whereas light propagating in the sample plane experiences a very different index (*13*). This anisotropy benefits the experiment because transverse phase matching is enforced in the sample plane, thereby maintaining high directionality of the forward and backward signals. In contrast, an isotropic metamaterial with zero index would emit in all directions, which is undesirable for signal collection.

The present metamaterial has a period of 80 nm in the direction of propagation. The results could potentially arise from a type of quasi–phase matching, but the authors exclude this possibility by estimating that the coherence length for backward FWM is about 460 nm, much longer than the period. They also show (in supplementary material) that essentially identical results are obtained from treating the structure as an effective metamaterial and from accounting for each layer of the structure separately.

It is relatively easy to phase-match FWM between nearby wavelengths in almost any material, albeit only in one direction. Furthermore, the FWM efficiency reported by of Suchowski *et al*. remains low (10^{-5}). Nevertheless, their work demonstrates a completely new principle for overcoming the phase-matching problem. In addition, the possibility of generating equal nonlinear signals in two opposite directions could find use in new types of nonlinear oscillators (*14*). From a more general viewpoint, the report shows that metamaterials can control nonlinear processes in unprecedented ways, opening the path toward nonlinear structures with unconventional emission properties.

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10.1126/science.1247622

DEVELOPMENT

Permission to Proliferate

Epidermal stem cells produce the signals that control tissue homeostasis.

Julia Frede and Philip H. Jones

Maintaining proliferating adult tissues involves a critical balance. Exactly the right number of cells must be produced to replace those lost from the tissue; otherwise, tissue failure or tumor formation will ensue. In mammalian epidermis, differentiated cells are shed from the tissue surface and replaced by dividing stem cells in the basal layer (see the figure) (*1*). On average, each cell division generates one daughter that goes on to divide and produce more such cells, while the other cell differentiates and migrates to the surface of the epidermis (*2*). Cell production must be adjusted in response to surface abrasion or injury, implying that proliferation is regulated by signals from nearby cells. One candidate for this role is the secreted protein Wnt, as mutations that disrupt its downstream cellular signaling pathway result in hairless, thin epidermis (*3*, *4*). Two studies—by Lim *et al*. (*5*) on page 1226 of this issue and by Choi *et al*. (*6*)—elucidate how Wnt functions in the epidermis. They show that Wnt and Wnt inhibitors balance the renewal and differentiation of epidermal stem cells and are both secreted by the stem cells themselves. This suggests autocrine regulation as distinct from the prevailing idea that stem cells are regulated by signals from other cell types in a "niche."

To identify cells receiving Wnt signals, Lim *et al*. and Choi *et al*. generated transgenic mouse strains that allowed them to track cells that express the Wnt target gene *Axin2*. Both studies found Axin2-expressing cells in the proliferative basal layer of the epidermis. Lim *et al*. used a genetic technique to label Axin2-expressing cells in the hind-paw skin of the mouse, which lacks hair follicles that may feed cells into the epidermis under conditions of stress or injury (*7*). The label is transmitted to daughter cells, producing clones (*8*). Using statistical analysis of the clone size distribution, Lim *et al*. found that Axin2-expressing cells constitute a self-maintaining population of functionally equivalent cells. Each cell in the basal layer divides to generate two dividing daughters, two differentiating daughters, or one cell of each type. Although the outcome of individual divisions is unpredictable, the odds are balanced, so homeostasis is achieved across the Axin2-expressing population. Similar balanced outcomes of division leading to "population self-renewal" occur in tail and ear epidermis and the esophageal epithelium (*2*, *9*–*11*).

A question that arises from these observations is how an injury to the paw can be healed. To achieve this, an excess of proliferating cells must be produced. In tail epidermis, this is accomplished by "reserve" cells,

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