Theory turns to reality for nonlinear optical metamaterials

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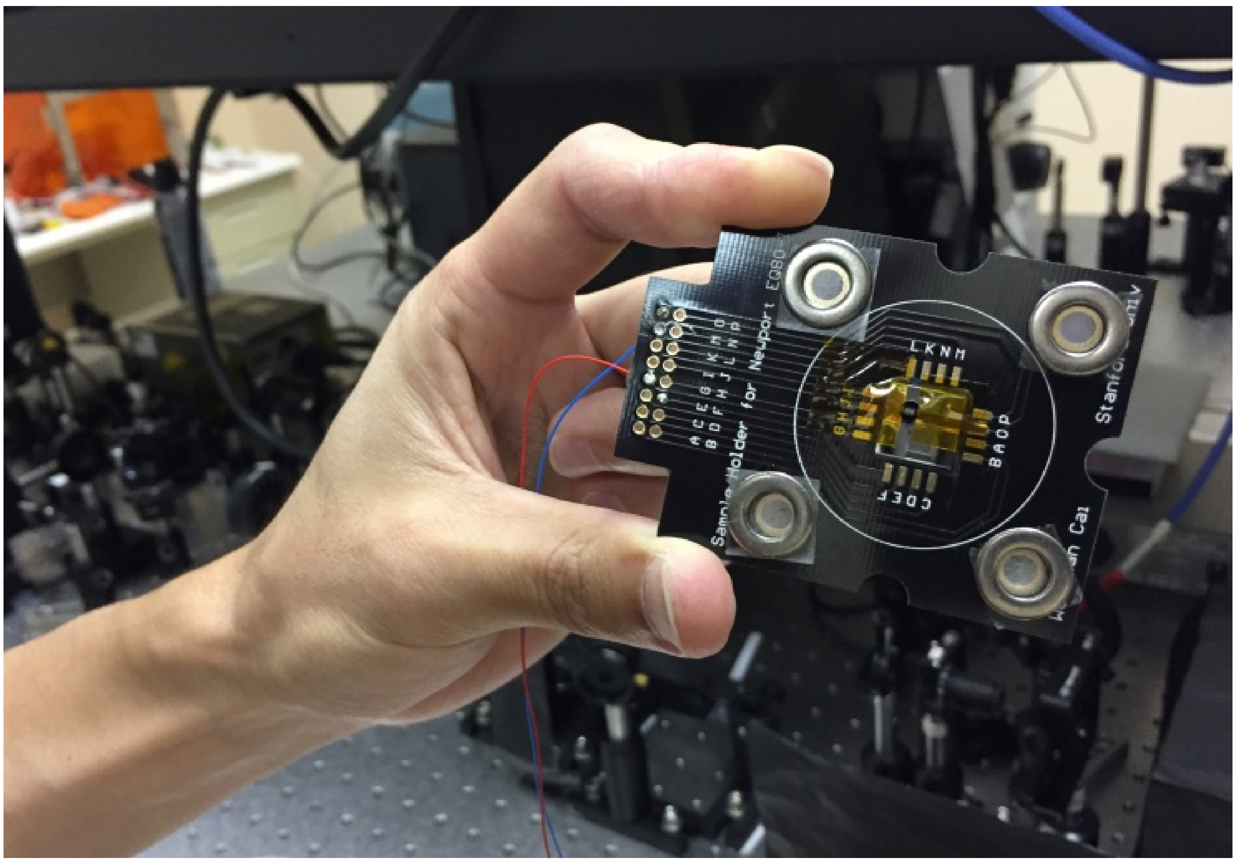

Image shows the metamaterial waveguide located at the center of a silicon chip, wired to an external circuit. A research team at Georgia Tech has created a nonlinear material that has opposite refractive indices at the fundamental and harmonic frequencies of light. Credit: John Toon, Georgia Tech

A research team has realized one of the long-standing theoretical predictions in nonlinear optical metamaterials: creation of a nonlinear material that has opposite refractive indices at the fundamental and harmonic frequencies of light. Such a material, which doesn't exist naturally, had been predicted for nearly a decade.

Observation of "backward phase matching" – a phenomenon also known as the "nonlinear mirror" – provided proof that this new type of metamaterial had been created. Demonstration of the phenomenon was reported by researchers at the Georgia Institute of Technology in a paper published June 15 in the journal *Nature Materials*.

Though by itself the discovery may have few immediate practical applications, realization of a material that had been predicted by theorists is a milestone that could lead to new areas of study, and prompt a re-evaluation of the fundamental rules governing nonlinear optics.

"Nonlinear optics is critically important to controlling light for information processing, sensing and signal generation," said Wenshan Cai, an associate professor in Georgia Tech's School of Electrical and Computer Engineering who led the research team. "Our effort substantially expands the scope of nonlinear light-matter interactions in artificially structured media with engineered, unconventional linear and high-order material parameters."

Engineered metamaterials offer unique properties not available in natural materials. This is especially useful in nonlinear optics, where materials with unconventional properties could make a difference anywhere light must be actively controlled. Researchers at multiple institutions have already created optical metamaterials that could be used to produce more efficient solar cells, faster computer chips, improved sensors – and even cloaking to permit invisibility.

"The linear responses of metamaterials have substantially augmented the linear properties available from naturally-occurring materials," noted Cai, who also holds a faculty position in the Georgia Tech School of Materials Science and Engineering. "In the same way, the studies of nonlinear metamaterials may have a revolutionary impact on the field of nonlinear optics. The unconventional electromagnetic parameters made possible by metamaterials will provoke us to rethink and re-evaluate many of the established rules of nonlinear optics."

Metamaterials obtain their properties from a repeated unit structure rather than the constituent materials. At the frequency range between visible and infrared light, subwavelength metallic structures can serve as building blocks – essentially "meta-atoms" – to create optical materials with properties that have not been available in the past.

Experimentally, the researchers struggled to create a negative index material – which has been a holy grail for metamaterial research – with a sample size large enough for testing. They had to tailor the refractive indices at both the fundamental and harmonic frequencies simultaneously.

The research team was able to demonstrate backward phase-matching by exploiting two distinct modes in a nonlinear plasmonic waveguide, which was built with a thin dielectric spacer of relatively high refractive index sandwiched between two flat silver films. A large dielectric constant in the gap enabled a pronounced separation of the surface plasmon and the bulk plasmon frequencies, while a narrow gap pushed the operating point away from the surface plasmon frequency and helped balance the magnitudes of the refractive indices of the two modes.

The waveguide operated with the real part of the mode refractive indices at 3.4 and -3.4 for the fundamental and harmonic waves, respectively. The observed peak conversion efficiency at the excitation wavelength of about 780 nanometers indicated fulfillment of the phase matching condition in which the coherent harmonic wave emerged along a direction opposite to that of the incoming fundamental light, the researchers reported in their paper.

The research team made a comprehensive set of measurements to experimentally confirm the phenomenon of backward phase-matching in the waveguide.

"We proposed a smart scheme that allowed us to experimentally achieve the backward phase-matching condition in a realistic way," Cai explained. "Experimental demonstration of backward phase-matching in negative index metamaterials was considered to be extremely challenging, which is why it took nearly ten years to be realized."

Phase matching is a standard technique to achieve efficient frequency generation using nonlinear optics, usually achieved by fine-tuning the orientation or temperature of a nonlinear crystal, Cai noted. In second harmonic generation – a nonlinear process to double the frequency of light – phase matching requires that the refractive index of the initial (fundamental) and the doubled (harmonic) frequencies be identical. That way, the fundamental light will gradually be converted to its harmonic along the direction of propagation, and the output harmonic light will co-propagate along the same direction.

In addition to Cai, the research team included first author Shoufeng Lan, a graduate student in the School of Electrical and Computer Engineering. Other co-authors are Lei Kang, Sean Rodrigues and Yonghao Cui from Georgia Tech, and David Schoen and Mark Brongersma from the Geballe Laboratory for Advanced Materials at Stanford University.

What's ahead for research in this area?

"Beyond the backward phase-matching, there are many other intriguing phenomena in nonlinear metamaterials awaiting exploration," Cai said. "The ability to design custom, nonlinear materials, which is possible in metamaterials, is bound to open entirely new outlooks for nonlinear light-matter interactions."

More information: "Backward phase-matching for nonlinear optical generation in negative-index materials," *Nature Materials* 2015. dx.doi.org/10.1038/nmat4324

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