Researchers approach quantum limit in third-order nonlinear light-light interaction

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Unprecedented nonlinear optical efficiency could make small organic molecules useful for optical computing, data processing and telecommunication.

Researchers from Lehigh University and the Swiss Federal Institute of Technology in Zurich (ETH) have reported unprecedented nonlinear optical efficiency in some small organic molecules that makes the molecules potentially useful for optical computing, optical data processing, and optical telecommunication.

In an article to be published in the journal *Optics Letters*, the researchers say that the optical nonlinearities of the molecules are "extraordinarily large relative to the small molecular mass of the molecules and are within a factor of 50 from the fundamental quantum limit."

"We have found that donor-substituted cyanoethynylethene molecules…show one of the strongest [nonlinear optical responses], if not the strongest nonlinear response observed to date, when expressed both in terms of [their] proximity to the fundamental limit and in terms of their specific third-order polarizability.

"These facts, combined with the compatibility of these molecules with vapor- deposition methods, make them a very interesting system [for] the development of efficient and flexible elements for integrated nonlinear optics."

This development, the researchers say, is necessary for optical computing and for the direct processing of information in an all-optical network. Such a network requires efficient "optical transistors" that increase bandwidth and avoid the time-consuming and inefficient conversions of optical signals to electronic signals and back that are now necessary in the Internet and other networks.

The article is titled "Highly Efficient Third-Order Optical Nonlinearities in Donor-Substituted Cyanoethynylethene Molecules."

A nonlinear optical response occurs in a material when the intensity of light alters the properties of the material through which light is passing, affecting, in turn, the manner in which the light propagates.

A "third order" optical nonlinearity is a measure of how well matter can mediate the interaction of different light waves. The fundamental limit, or maximum possible nonlinear-optical susceptibility of molecules allowed by quantum mechanics, was calculated by Washington State University physicist Mark Kuzyk in 2000 and is known as the Kuzyk Limit.

Ivan Biaggio, professor of physics and member of Lehigh's Center for Optical Technologies, said the nonlinear susceptibility of the CEE molecules investigated by his group approaches the Kuzyk Limit.

"These molecules, in contrast to many that have been previously reported, come very close to the Kuzyk Limit," said Biaggio. "They come within a factor of 50, and it is astonishing to get that close in a real system, given that the fundamental limit is obtained assuming that all molecular properties are ideal at the same time."

Third-order optical nonlinearities occur when three photons, or light packets, enter a material and interact, producing a fourth photon that may have a different wavelength and color, or a different propagation direction. Materials with third-order nonlinear responses are required for all-optical networks and devices in which light waves, not electronics, perform switching, routing, amplification and other functions. These developments could lead to much more efficient ways to route signals between fibers and communication channels, which in turn could speed up, by orders of magnitude, the rate at which information is transmitted and processed.

Biaggio said the ETH researchers have synthesized several variants of donor substituted cyanoethynylethene (CEE) molecules and the Lehigh researchers have conducted physical experiments to determine the efficiency with which the molecules lead to multi-photon interactions.

In their experiments, the researchers applied a tool called Degenerate Four-Wave Mixing at wavelengths ranging from the visible to the near and far infrared to determine how the CEE molecule interacts simultaneously with three photons to generate a fourth photon.

The optical nonlinearity of organic molecules can be influenced by, among other things, the manner in which atomic groups that act as electron acceptors or electron donors are arranged around a molecular backbone containing delocalized electrons. The researchers varied the geometrical arrangement of donors around the X-shaped core of the CEE molecule.

"We have experimented with different geometrical arrangements of donors in order to better understand how the nonlinearity arises," said Biaggio, who holds a doctorate from ETH and is a former team leader in ETH's Nonlinear Optics Laboratory.

"It was like building a variety of figures with the same few Lego blocks. Thanks to our experiments, we have improved our ability to predict the effect of different configurations of these blocks on the nonlinearity of the overall molecule."

Joshua May, a graduate student in Biaggio's group, used a tunable laser which is capable of emitting short light pulses along the spectrum of visible and invisible light, to determine the nonlinear optical properties of the CEE molecules, Biaggio said.

"We have measured the nonlinear response of the molecules along the spectrum of wavelengths ranging from 500 nanometers to 1.6 microns," he said. "The tunable laser enables us to go to the physically meaningful regions of the lightwave spectrum, to move to wherever we need to be, in order to study things like two-photon absorption or the so-called 'non-resonant' molecular response that occurs only at sufficiently long wavelengths in the infrared.

"This makes it possible to compare the different molecules that are studied in various research groups and determine which ones have the best nonlinear susceptibility, because material properties change along the wavelength spectrum."

The family of donor-substituted CEE molecules is characterized by small size and by high density of nonlinearity - important properties, said Biaggio, that may make it easier to assemble the molecules into useful materials.

"It is not enough if a molecule has a high optical nonlinearity. It must also be possible to assemble it efficiently into a useful solid-state material. The small size and robustness of these new molecules that we studied enables us to use various interesting technologies to assemble them. For example, they can be evaporated into a gas and vapor-deposited, making it easy to process them into a stable solid-state materials which we can expect to have extremely high bulk nonlinear optical susceptibility."

Source: Lehigh University