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Light amplification by stimulated emission from electrically driven colloidal quantum dots finally achieved

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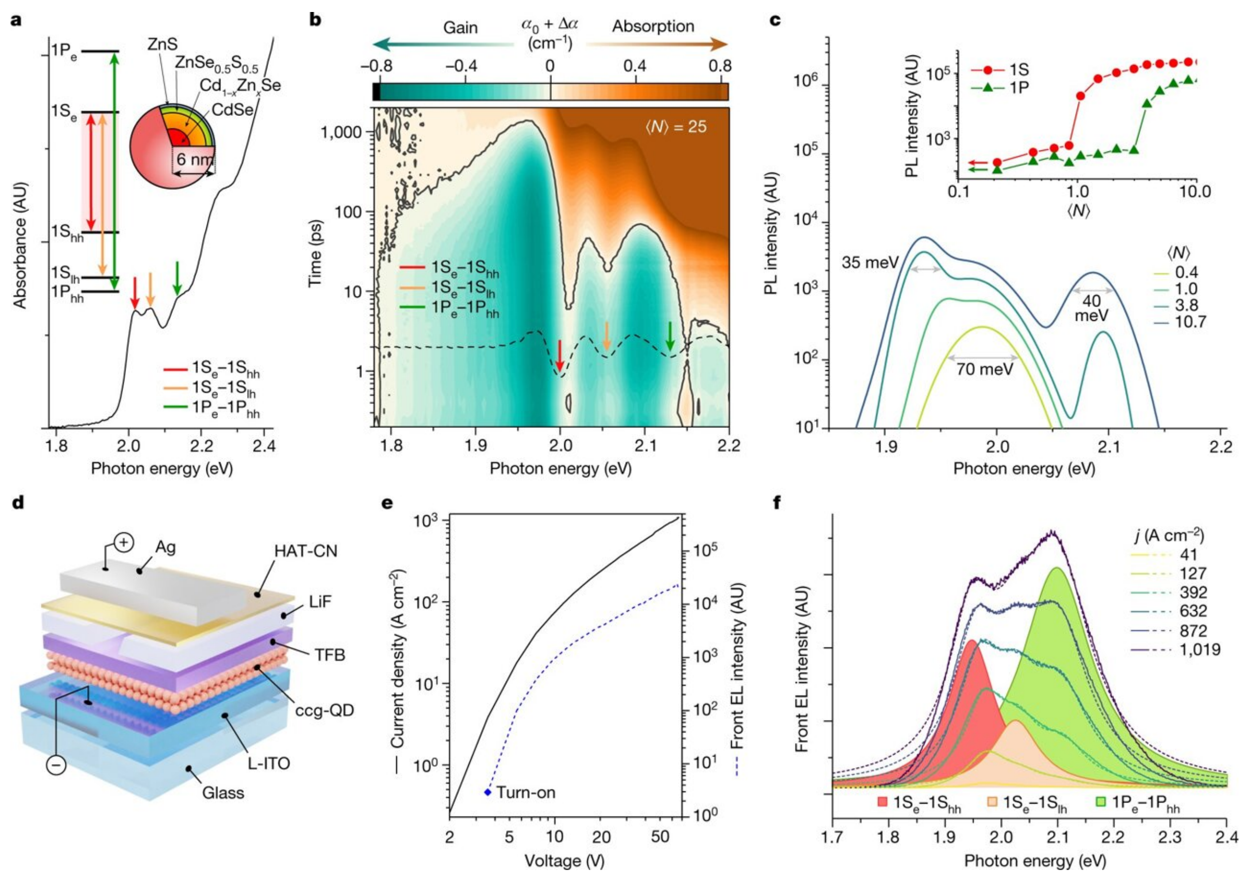

Optical and EL properties of ccg-QDs. Credit: *Nature* (2023). DOI: 10.1038/s41586-023-05855-6

In a result decades in the making, Los Alamos scientists have achieved light amplification with electrically driven devices based on solution-cast semiconductor nanocrystals—tiny specs of semiconductor matter made via chemical synthesis and often called colloidal quantum dots.

This demonstration, reported in the journal *Nature*, opens the door to a completely new class of electrically pumped lasing devices—highly flexible, solution-processable laser diodes that can be prepared on any crystalline or non-crystalline substrate without the need for sophisticated vacuum-based growth techniques or a highly controlled clean-room environment.

"The capabilities to attain light amplification with electrically driven colloidal quantum dots have emerged from decades of our previous research into syntheses of nanocrystals, their photophysical properties and optical and electrical design of quantum dot devices," said Victor Klimov, Laboratory Fellow and leader of the quantum dot research initiative.

"Our novel, 'compositionally graded' quantum dots exhibit long optical gain lifetimes, large gain coefficients and low lasing thresholds—properties that make them a perfect lasing material. The developed approaches for achieving electrically driven light amplification with solution-cast nanocrystals might help resolve a long-standing challenge of integrating photonic and electronic circuits on the same silicon chip and is poised to advance many other fields ranging from lighting and displays to quantum information, medical diagnostics and chemical sensing."

More than two decades of research

Research over more than two decades has sought to achieve colloidal quantum dot lasing with electrical pumping, a prerequisite for its widespread use in practical technologies. Traditional laser diodes, ubiquitous in modern technologies, produce highly monochromatic, coherent light under electrical excitation. But they have deficiencies: challenges with scalability, gaps in the range of accessible wavelengths, and, importantly an incompatibility with silicon technologies that limits their use in microelectronics. Those problems have spurred the search for alternatives in the realm of highly flexible and easily scalable solution-processable materials.

Chemically prepared colloidal quantum dots are especially attractive for implementing solution-processable laser diodes. In addition to being compatible with inexpensive and readily scalable chemical techniques, they offer the advantages of a size-tunable emission wavelength, low-optical gain thresholds and high-temperature stability of lasing characteristics.

However, multiple challenges have hindered the technology's development, including fast Auger recombination of gain-active multicarrier states, poor stability of nanocrystal films at high current densities required for lasing, and the difficulty of obtaining net optical gain in a complex electrically driven device wherein a thin electroluminescent nanocrystal layer is combined with various optically-lossy, charge-conducting layers that tends to absorb light emitted by the nanocrystals.

Solutions for colloidal quantum dot laser diode challenges

A number of technical challenges needed to be solved to realize electrically driven colloidal quantum dot lasing. Quantum dots not only need to emit light, they need to multiply generated photons via stimulated emission. That effect can be turned into laser oscillations, or lasing, by combining the quantum dots with an optical resonator that would circulate the emitted light through the gain medium. Solve that, and you have electrically driven quantum dot lasing.

In quantum dots, stimulated emission competes with very fast nonradiative Auger recombination, the primary impediment of lasing in these materials. The Los Alamos team developed a highly effective approach to suppress nonradiative Auger decay by introducing carefully engineered compositional gradients into the quantum dot interior.

Very high current densities are also required for attaining the lasing regime. That current, though, can doom a device.

"A typical quantum dot light-emitting diode operates at current densities that do not exceed about 1 ampere per square centimeter," said Namyoung Ahn, a Los Alamos Director's Postdoctoral Fellow and the lead device design expert for the project. "However, the realization of lasing requires tens to hundreds of amperes per square centimeter, which would normally lead to device breakdown due to overheating. This has been a key problem hindering realization of lasing with electrical pumping."

To resolve the overheating problem, the team confined the electric current in spatial and temporal domains, ultimately reducing the amount of generated heat and simultaneously improving heat exchange with a surrounding medium. To implement these ideas, the researchers incorporated an insulating interlayer with a small, current-focusing aperture into a device stack and used short electrical pulses (about 1 microsecond duration) to drive the LEDs.

The developed devices were able to reach unprecedented current densities of up to approximately 2,000 amperes per square centimeter, sufficient to generate strong, broad-band optical gain spanning across multiple quantum dot optical transitions.

"A further challenge is to achieve a favorable balance between optical gain and optical losses in a complete LED device stack containing various charge conducting layers that can exhibit strong light absorption," said Laboratory postdoctoral researcher Clément Livache, who coordinated the spectroscopic component of this project. "To tackle this problem, we added a stack of dielectric bi-layers, forming a so-called distributed Bragg reflector."

Using a Bragg reflector as an underlying substrate, the researchers were able to control a spatial distribution of an electric field across the device and shape it so as to reduce field intensity in optically lossy charge conductive layers and to enhance the field in the quantum-dot gain medium.

With those innovations, the team demonstrated an effect pursued by the research community for decades: bright amplified spontaneous emission (ASE) realized with electrically pumped colloidal quantum dots. In the ASE process, "seed photons" produced by spontaneous emission launch a "photon avalanche" driven by stimulated emission from the excited quantum dots. This boosts the intensity of the emitted light, increases its directionality and enhances coherence. ASE can be considered as a precursor of lasing, the effect which emerges when an ASE-capable medium is combined with an optical resonator.

The ASE-type quantum dot LEDs represent considerable practical utility as sources of highly directional, narrow-band light for applications in consumer products (for example, displays and projectors), metrology, imaging and scientific instrumentation. Interesting opportunities are also associated with the prospective use of these structures in electronics and photonics, traditional and quantum, where they can help realize spectrally tunable on-chip optical amplifiers integrated with various types of optical interconnects and photonic structures.

What's next?

Presently, the team is working on realizing laser oscillations with electrically pumped quantum dots. In one approach, they incorporate into the devices a so-called "distributed feedback grating," a periodic structure that acts as an optical resonator circulating light in the quantum dot medium. The team also aims to extend spectral coverage of their devices, with a focus on demonstrating electrically driven light-amplification in the range of infrared wavelengths.

Infrared, solution-processable optical-gain devices could be of great utility in silicon technologies, communications, imaging and sensing.

More information: Namyoung Ahn et al, Electrically driven amplified spontaneous emission from colloidal quantum dots, *Nature* (2023). DOI: 10.1038/s41586-023-05855-6

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