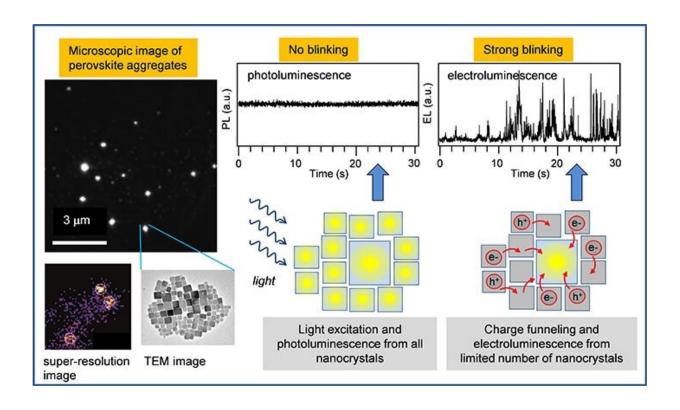


Single-particle spectroscopy of CsPbBr3 perovskite reveals the origin low electrolumine

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Intensity time trace of photoluminescence of one aggregate (middle, top) shows no fluctuations because all nanocrystals within the aggregate emit simulatenously (middle, bottom scheme); Intensity time trace of electroluminescence of one aggregate (right, top) shows strong fluctuations (blinking) because all charges are funneled to one nanocrystal which is emitting (right, bottom scheme). Credit: Tokyo Tech



Researchers from Tokyo Institute of Technology (Tokyo Tech) used single-particle spectroscopy to study electroluminescence in light-emitting devices. They discovered that efficient charge funneling between individual perovskite nanocrystals and the phenomenon of emission blinking are responsible for the low efficiencies of perovskite light-emitting devices.

Metal halide perovskites have recently emerged as an exceptionally promising alternative material for next-generation optoelectronic applications. Nanoscale-sized perovskite structures possess remarkable photophysical properties, such as direct bandgap, color tunability, a large absorption cross-section, and narrow photoluminescence linewidth. Together with their low cost, feasibility for scale-up synthesis, solution processability and compatibility with existing optoelectronic device components, these properties make metal halide perovskite nanocrystals a feasible alternative to other semiconducting materials for a range of light-emitting applications including displays, lighting, lasers, as well as memory devices.

However, while <u>perovskite nanocrystals</u> show very high photoluminescence yield, <u>electroluminescence</u> devices prepared from such nanocrystals have long suffered from low efficiency. Recent efforts have concentrated on device engineering to overcome this problem, but so far, there has been no systematic study on the nanoscale physical origin of the poor efficiencies. Here, the team of prof. Martin Vacha from Tokyo Tech used single-particle microscopic detection and spectroscopy to study the electroluminescence process on the level of individual nanocrystals.

The team used nanocrystals of the perovskite CsPbBr₃ surfacepassivated with oleic acid ligands, dispersed in thin film of a conducting polymer which was used as an emission layer in a light-emitting device (LED). The device was constructed for use on top of an inverted



fluorescence microscope, which enabled comparison of electroluminescence and photoluminescence from the same nanocrystals. The CsPbBr₃ nanocrystals form aggregates within the emission layer, with each aggregate containing tens to hundreds of individual nanocrystals.

The researchers used advanced super-resolution imaging to determine that while in photoluminescence, all the nanocrystals in the aggregate emit light; in electroluminescence, only a small number (typically three to seven) of the nanocrystals are actively emitting (Fig. 1). This is a result of size distribution and the consequent energy landscape within the aggregate. Electrical charges that are injected into the device during the operation are captured on individual nanocrystals and efficiently funneled toward the largest nanocrystals. The largest nanocrystals within the aggregate have the smallest energy bandgap, and their valence and conduction bands work as traps for charges captured originally at the surrounding nanocrystals. The conductive environment present between the nanocrystals enables efficient migration of the charges to these traps from where the electroluminescence takes place, as shown schematically in Fig. 1.

Another important finding is that the intensity of electroluminescence from the actively emitting nanocrystals is not constant, but rather shows strong fluctuations, so-called blinking (Fig. 1). Such blinking is not present in photoluminescence of the same aggregates. The researchers have previously found that the blinking can be caused by the conductive matrix as well as by externally applied electric field. In the LED device, the blinking phenomenon is a crucial factor that contributes to the lower efficiency in electroluminescence. The researchers concluded that electroluminescence efficiency is only about one-third of that of photoluminescence due to the presence of the blinking phenomenon.

The present work points a way toward efficient nanoscale



characterization of electroluminescence of halide perovskite materials for light-emitting applications. One of the keys toward higher efficiency will be surface engineering of the nanocrystals that would suppress the intensity fluctuations.

More information: Dharmendar Kumar Sharma et al, Single-particle electroluminescence of CsPbBr3 perovskite nanocrystals reveals particle-selective recombination and blinking as key efficiency factors, *Nature Communications* (2019). DOI: 10.1038/s41467-019-12512-y

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