# 'Harmful' effects paradoxically enhance solar cell efficiency

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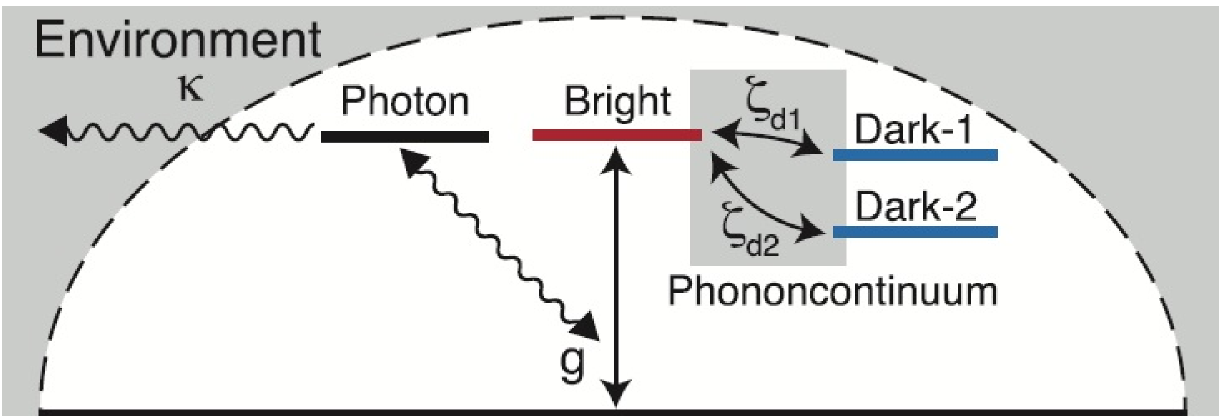


A model of energy conversion in a semiconducting carbon nanotube, showing that bright excitons are transformed into dark excitons, which have longer lifetimes and so can undergo charge separation without recombining back into photons. Credit: Yamada, et al. ©2015 American Physical Society

(Phys.org)—Dissipation and decoherence are typically considered harmful to solar cell efficiency, but in a new paper scientists have shown that these effects paradoxically make the exciton lifetime in semiconducting carbon nanotubes 50 times longer than before, which leads to a higher overall efficiency. The results provide new guidelines for exploring new photovoltaic materials that may offer unexpectedly high efficiencies.

The scientists, Yasuhiro Yamada, Youhei Yamaji, and Masatoshi Imada at The University of Tokyo (Yamada is currently at Osaka University), have published a paper on the counterintuitive exciton lifetime enhancement in a recent issue of *Physical Review Letters*.

"The principle of better efficiency by energy dissipation and decoherence has already been inferred by the photosynthesis process at the chloroplasts," authors told *Phys.org*. "However, it has remained as speculation until now."

As the researchers explain, understanding this enhancement requires a microscopic understanding of that way that energy is converted from sunlight into electricity—or, in terms of particles, from photons into excitons, the latter of which are bound states of an electron and an electron hole.

In this energy conversion process, there is usually a tradeoff when it comes to the photon absorption rate of the photovoltaic material. A high absorption rate is beneficial for the first step when excitons are generated from incoming photons, but harmful in a later step when the electrons and electron holes must be separated at different electrodes. Unfortunately, before this charge separation can occur, the high absorption rate causes more of the excitons to quickly recombine back into photons, which are emitted back into the environment.

In the new study, the scientists showed that there is a way to reap the benefits of a high absorption rate without paying the price later on because exciton recombination can be suppressed by—quite surprisingly—dissipation and decoherence. Normally, these two effects are considered harmful to photovoltaic efficiency: energy dissipation means that some energy is lost to the environment; and due to decoherence, the quantum coherence between photons and excitons that helps promote exciton generation loses its quantumness and becomes classical.

Despite these drawbacks, the researchers here showed that a certain amount of dissipation, combined with an optimal coexistence of coherence and decoherence, can increase the exciton lifetime so that it has enough time to separate into an electron and hole before recombination occurs.

"Normally, the separation process takes much longer time than the recombination process," authors explained. "Therefore, we need to elongate the exciton lifetime to wait until the separation process works."

To do this, the mechanism transforms short-lived "bright excitons" into longer-lived "dark excitons," which live long enough to be separated into an electron and a hole without succumbing to recombination. Key to this transformation is that dissipation and decoherence impose a desirable quantum-to-classical transformation that makes this process irreversible: a dark exciton cannot be transformed back into a bright exciton. As the researchers explained, figuring this out was not easy to do.

"The quantum-to-classical crossover process accompanied by dissipation is at the heart of difficult non-equilibrium many-body problems, and solving it requires developing an efficient computational tool with a new theoretical formulation," authors said. "After solving the formulated quantum master equation, the principle of optimizing the decoherence and dissipation for the better efficiency has been established in the present work. It overturned the common sense notion that better efficiency should be pursued in materials with better 'quantum yield' that have a higher photoluminescence rate. It provides us with new guidelines."

As the scientists explained, part of the reason why the recombination suppression benefit of dissipation and decoherence has gone unnoticed until now is that the mechanism paradoxically causes a decrease in the photoluminescence, or light emission, so that a material with these effects would at first glance appear unpromising as a photovoltaic device. However, the decrease in photoluminescence is a result of the fact that the mechanism converts bright excitons (which emit light) into dark excitons (which do not). So even though more dark excitons make the material appear dark, they are what enable the material to convert light into electricity with a high efficiency.

"In the next step, we urgently need to clarify how the separation of the exciton into an electron and a hole occurs with the transport of them to the opposite electrodes to generate an electromotive force, provided that their recombination to a photon is suppressed in the present mechanism," authors said. "This requires a much longer and complex time evolution process. Another direction of research is of course to design a solar cell with better efficiency by utilizing the present principles and guidelines. This may be done for new candidates of materials."

**More information:** Yasuhiro Yamada, et al. "Exciton Lifetime Paradoxically Enhanced by Dissipation and Decoherence: Toward Efficient Energy Conversion of a Solar Cell." *Physical Review Letters*. DOI: 10.1103/PhysRevLett.115.197701



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