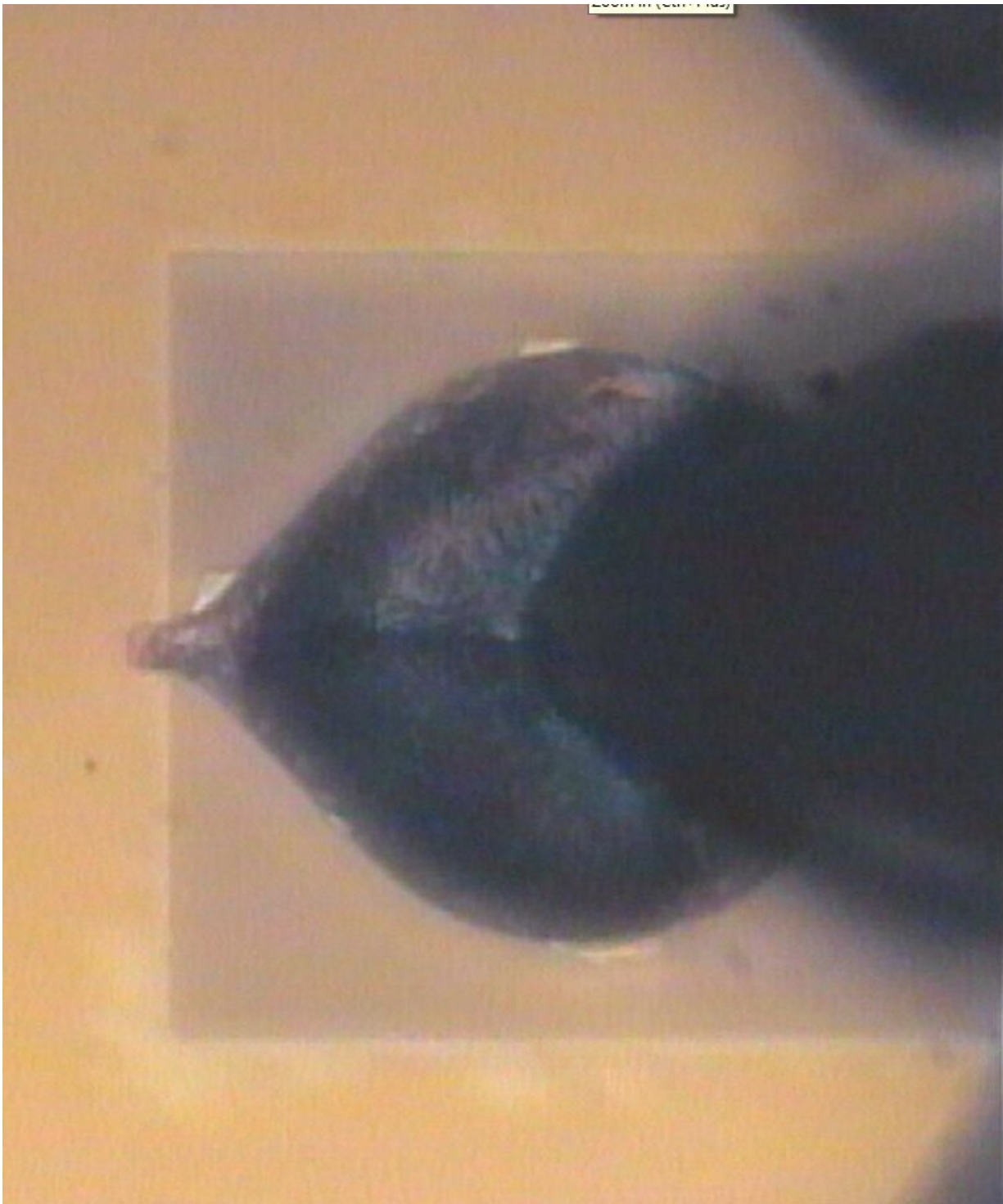


Perovskites reveal huge resettable single-photon response

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Micrograph of a probed device. Credit: ACS Nano Letters

Despite the progress in perovskite device efficiencies, these systems are not fully understood, in particular the frequency- and power-dependence of their response to light. Yu-Hwa Lo and colleagues at the University of California in San Diego (UCSD) now report on systematic investigations of how these devices respond to light for frequencies varying over eight orders of magnitude and power ranging from millions to single photons.

The results reveal different response regimes, including the first observation of a quasi-persistent resettable single-[photon](#) response that cannot be explained by existing physical models for the material. The results may find use in several new applications of perovskites, such as analogue memory for neuromorphic computing.

Erroneous assumptions

"There is a misconception in photodetection for perovskites," Lo tells phys.org, as he explains a tendency among the [research community](#) during this kind of study. Often, researchers take measurements in low-frequency, (quasi) DC conditions for the power-dependent responsivity, that is, the amount of electrical output per optical input. However, they then assume the same DC responsivity applies when testing at [high frequencies](#) for responsiveness, that is, how long a system takes to respond to an impulse.

For their study, the UCSD researchers used the perovskite MAPbI_3 , where M is methyl CH_3 and A is ammonium NH_3 , as it is well understood and relatively easy to process. It also conveniently has a bandgap of ~ 1.58 eV so that it is sensitive to visible light.

In contrast to previous studies, Lo and colleagues measured the response as the current difference before and after a pulse, and the responsivity by dividing the photocurrent by the absorbed optical power at frequencies down to 0.1 Hz. Their study revealed that the response was,

in fact, very slow at low quasi-DC frequencies, taking around 10 seconds for the current to rise. Greater surprises were to come.

Regime change

The researchers found that the photoresponse was essentially frequency-independent, but with an apparent regime change. They identified an inversely proportional relationship between the responsivity and the power raised to the power of a factor β , which remained unchanged over a frequency range from 5 Hz to 800 MHz. However, below 5 Hz, the value of β changed from -0.4 to -0.9. This gives a maximum internal responsivity of 1.7×10^7 A/W at 10 aW, which decreases rapidly with increasing power.

Their explanation for the change in exponent is that at higher frequencies, electrons and holes form, whereas at lower frequencies, ions and ion vacancies are mobilized. They also observed that the photoresponse persisted, that is, it did not return to the dark level current until reset with the bias voltage. The researchers explain the quasi-persistent change in the material's conductivity in terms of the redistribution of ions and charged vacancies, which effectively change the material's properties. Reflectivity measurements, which revealed peak shifts in this regime, supported this explanation.

The real surprise came as they brought the power down below 10 aW, where just 10 photons are incident on the device at a time. At this point, the slope plateaued, a condition in which the value of β is zero, the output photocurrent depends linearly on the number of photons absorbed, and the responsivity is independent of the power value right down to the single-photon level. These observations suggest that a single photon was capable of mobilizing as many as 10^8 ion-vacancy pairs. Previously reported results had assumed just one pair mobilized per photon.

Unexplained physics

"When we decreased the absorbed photon numbers (to around 10 photons), the quasi-persistent photoresponse almost stayed the same," says Lo. "We were surprised by this observation, especially when it entered the single-digit photon range, since there wasn't an available physical model to explain this. Ion migration is nothing new in perovskite, but the internal signal amplification mechanism is."

The researchers suggest that there may be some avalanche effect behind the phenomenon, such that under a bias, an iodide ion mobilized by an incident photon could knock another iodide and so on. Beyond 10 incident photons, all the ion-vacancy pairs that can move have been mobilized, and the net photoresponse becomes almost independent of the incident photon number, or in other words, the responsivity per incident photon becomes inversely proportional to the incident power. They also have an explanation for the marked decrease of the effect without a sufficient bias, as the ions would then need to travel a longer distance before they have enough energy to trigger another ion-vacancy pair, so that this is less likely to happen before the ion runs into a charge trap.

As well as analogue memories for neuromorphic computing, Lo and colleagues suggest the effect may present further opportunities for exploiting perovskites in energy harvesting, high capacity memory and optical switches. They are interested in designing a device that would be able to inject a small number of electrons that would achieve a similar effect to the quasi-persistent single-photon response. However, they also remain curious to better understand the physical mechanism behind the phenomenon, perhaps in collaboration with a theory group in computational condensed matter physics.

More information: Zihan Xu et al. Frequency- and power-dependent

photoresponse of a perovskite photodetector down to the single-photon level *Nano Letters* (2020).

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