Comparing the Influence of Rubidium and Cesium Dopants on Carrier Mobility and Lifetime in Hybrid Perovskite Thin Films

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Introduction

What are hybrid perovskites?

- Hybrid perovskites, in general, are compounds with a cubic crystal structure of the form ABX_3, where A is typically an organic cation, B is an inorganic cation, and X is a halide anion

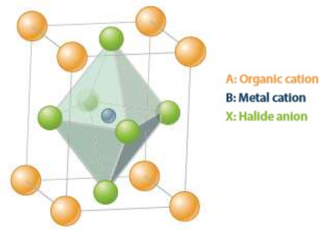

Figure 1. Unit cell of a hybrid perovskite[1]

What makes perovskites promising photovoltaic materials?

- Efficient at absorbing light and transporting charges
- Derived from convenient solution-based techniques
- Able to reach solar conversion efficiencies above 20%
- Can come in a wide variety of compositions
- Have tunable optical and electrical properties

Where do rubidium and cesium come into play?

- The ability to accurately determine solar cell efficiency suffers from *hysteresis*, or variation in efficiency depending on how voltage across a solar cell device is applied
- Research has shown that incorporating small amounts of either rubidium or cesium—two alkali metal cations—boosts device efficiency, but only rubidium reduces hysteresis
- We know from prior experiments that rubidium improves carrier mobility, but the same effect for cesium (in the same type of perovskite) has remained unclear
- If the mobilities for cesium-doped samples are significantly lower than those for rubidium-doped analogs, then carrier mobility could act as a probe into how to eliminate hysteresis from perovskite solar cells

[1]Figure adapted from Herz, L. M. *Annu. Rev. Phys. Chem.* **2016**, *67*, 65–89.

Perovskites of Interest

- Control (no Rb nor Cs added)
- Rb series: 1% Rb, 5% Rb
- Cs series: 1% Cs, 5% Cs
- RbCs series: 1% Rb + 1% Cs, 2.5% Rb + 2.5% Cs

Figure 2. Spectrum of perovskite thin film samples

Experimental Set-Up

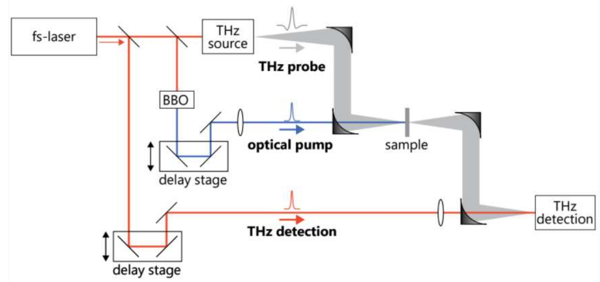

Figure 3. Time Resolved Terahertz (THz) Spectroscopy Set-up

Main Steps

a) 400 nm pump hits the sample a certain amount of time before the THz probe arrives
b) Based on the number of free carriers generated by the pump and the sample's carrier mobility, the THz probe signal will be absorbed proportionately
c) As time between the pump and probe increases, free carriers recombine—leading to smaller absorption of THz

Trends to Look For

- Samples with high mobilities show high initial THz absorption
- Samples with long lifetimes show little decay in THz absorption

Results and Discussion

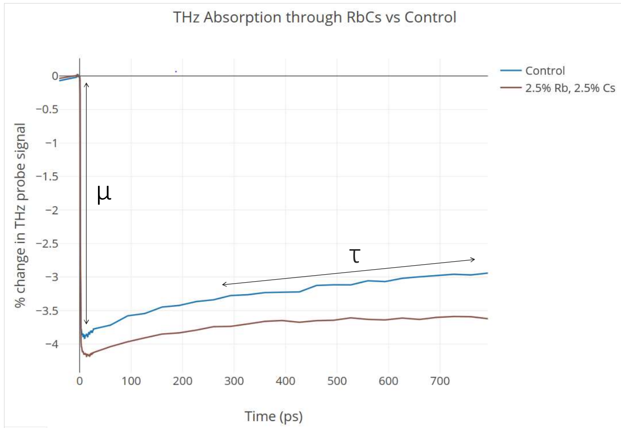

Figure 4. Initial THz probe absorption is larger for RbCs than for the control, indicating higher carrier mobility (μ). Additionally, the decay in THz absorption over time is slower for RbCs, signaling better retention of excited carriers or lifetime (τ).

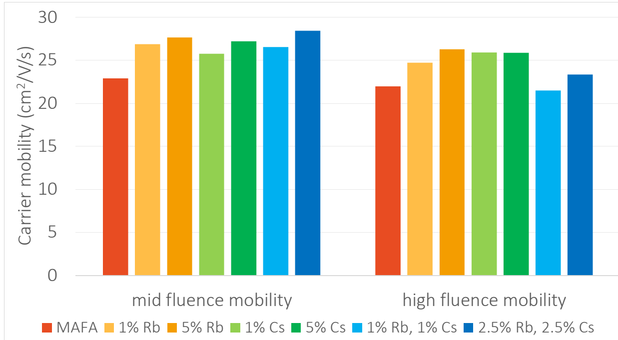

Figure 5. Addition of Rb, Cs, and RbCs all increase mobility for both 1% and 5% incorporation. The extent to which Rb and Cs increase mobility is comparable. RbCs samples exhibit surprisingly lower mobilities at high fluence, compared to Rb and Cs samples. A possible explanation for this drop in mobility could be quicker degradation from light or oxygen exposure.

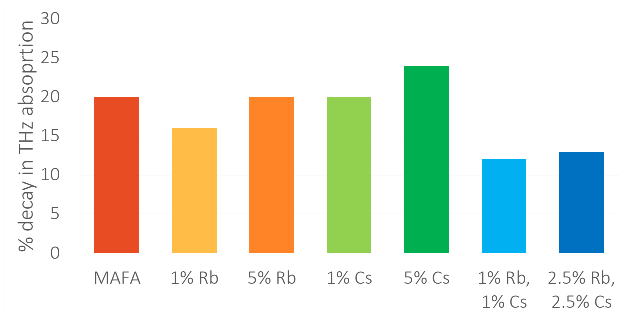

Figure 6. Rb-doped samples exhibit better lifetime than Cs-doped analogs, while RbCs samples exhibit the best lifetimes overall—in contrast to RbCs mobility trends.

Conclusions and Future Work

- The equivalent improvement in carrier mobility for Rb-doped and Cs-doped samples indicates that a direct correlation between carrier mobility and hysteresis is unlikely
- The drop in mobility for the RbCs samples is surprising, given they exhibit the best lifetimes and are known to yield the highest solar cell efficiencies for these materials
- Looking at diffusion length (which is proportional to $\sqrt{\mu\tau}$) would offer a glimpse into whether the RbCs samples still offer the best balance for charge transport
- Photoluminescence measurements can quantify lifetime, which can then be combined with THz mobility data

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