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Title: Li-Doping and Ag-Alloying Interplay Shows the Pathway for Kesterite Solar Cells with Efficiency Over 14%

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Abstract: Kesterite photovoltaic technologies are critical for the deployment of light-harvesting devices in buildings and products, enabling energy sustainable buildings, and households. The recent improvements in kesterite power conversion efficiencies have focused on improving solution-based precursors by improving the material phase purity, grain quality, and grain boundaries with many extrinsic doping and alloying agents (Ag, Cd, Ge...). The reported progress for solution-based precursors has been achieved due to a grain growth in more electronically intrinsic conditions. However, the kesterite device performance is dependent on the majority carrier density and sub-optimal carrier concentrations of 10^{14} – 10^{15} cm^{-3} have been consistently reported. Increasing the majority carrier density by one order of magnitude would increase the efficiency ceiling of kesterite solar cells, making the 20% target much more realistic. In this work, LiClO_4 is introduced as a highly soluble and highly thermally stable Li precursor salt which leads to optimal ($>10^{16}$ cm^{-3}) carrier concentration without a significant impact in other relevant optoelectronic properties. The findings presented in this work demonstrate that the interplay between Li-doping and Ag-alloying enables a reproducible and statistically significant improvement in the device performance leading to efficiencies up to 14.1%.



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