Computational design of novel semiconducting perovskites and fundamental electronic properties.

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Over the last seven years we have witnessed the rise of lead-halide perovskites for optoelectronic applications such as photovoltaics, sensors and light-emitting diodes. In this talk I will briefly showcase recent efforts towards new materials that are alternatives to traditional lead-halide perovskites, for which computational design approaches from first-principles have been extensively successful and revealed a series of new compounds within the so-called halide double perovskites family [1-3]. Among these, $Cs_2BiAgBr_6$ has the narrower indirect band gap of 1.9 eV, and $Cs_2InAgCl_6$ is the only direct band gap semiconductor, yet with a large gap of 3.3 e. All of them exhibit low carrier effective masses and consequently, are prominent candidates for a range of opto-electronic applications such as photovoltaics, light-emitting devices, sensors, and photo-catalysts. Here, we will outline the computational design strategy that lead to the synthesis of these compounds, and particularly focus on the insights we can get from first-principles calculations in order to facilitate the synthesis, improve their opto-electronic properties and the in-silico identification of compounds with properties that are similar to the lead-halide perovskites.

Oxide perovskites have a much longer history and are pivotal in a wide range of technological applications. Yet, a rational connection between these two important classes of materials is missing. Here, I will employ a computational design strategy to explore this missing link and demonstrate that for each halide perovskite there are several lookalike oxide perovskites with similar optoelectronic properties-analogs. Our new concept of analogs led us to identify a new oxide double perovskite semiconductor, Ba_2AgIO_6, which exhibits an electronic band structure remarkably similar to that of our recently discovered halide double perovskite $Cs_2AgInCl_6$, but with a band gap in the visible range at 1.9 eV. [4] I will show results on the successful synthesis of Ba_2AgIO_6 by solution process and its crystallographic and optical characterization. Ba_2AgIO_6 and $Cs_2AgInCl_6$ are both analogs of the well-known transparent conductor $BaSnO_3$, but the significantly lower band-gap of Ba_2AgIO_6 makes this new compound much more promising for oxide-based optoelectronics and for novel monolithic halide/oxide devices [4].

In the last part, I will focus on the intrinsic properties of the well-known photovoltaic material $FAPbI_3$. I will show how first-principles calculations can be employed to interpret oscillations that appear on the absorption spectrum of $FAPbI_3$ thin films [5].

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