Highlights

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Role of Polar Phonons in the Photo Excited State of Metal Halide Perovskites

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In the last years metal halide perovskites have come up as very promising solar cell materials. They show the potential of becoming competitive with current silicon based solar cells mainly because of their high photovoltaic efficiency about 20 % reached in very short time. The most frequently studied material is $MAPbI_3$ which show a large range of exciton binding energies as reported by various experiments. In this work we present advanced relativistic calculations capable to directly calculate the excitonic properties from first principles. We apply this method to different halide perovskites. At low temperatures, this method predicts large exciton binding energies in agreement with the experimental values ranging from 5 to 55 meV. For $MAPbI_3$, phonon modes present in this frequency range have a negligible contribution to the ionic screening. Furthermore, we show that at room temperature this situation does not change. Therefore, we exclude ionic screening as a possible explanation for the observed reduction of the exciton binding energy at room temperature. Here we suggest that polarons formation may have a dominant role for exciton binding energy reduction a higher temperature. Our study suggests that electrons and holes separate after optical excitation forming two individual polarons, lowering the fundamental gap by 42 meV. This scenario would offer an intriguing new possibility for designing novel polaronic solal cell materials, if validated experimentally.

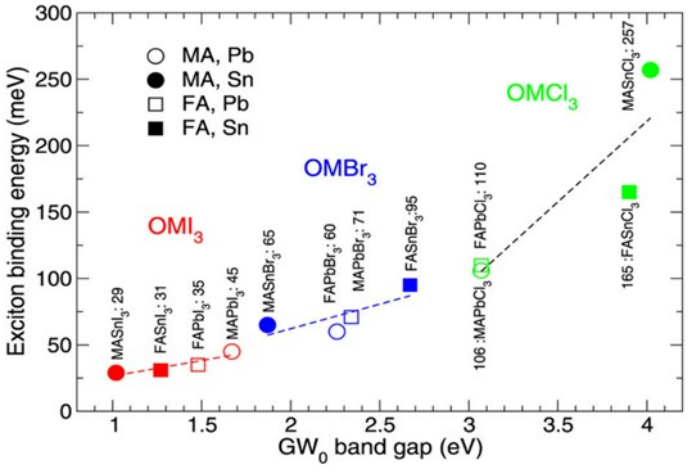

Fig.1: Calculated exciton binding energies and GW0 band gaps of twelve metal halide perovskites (OMX_3, {O=MA,FA,M=Pb,Sn,X=I,Br,Cl}. MA is methylammonium, FA is formamidinium).