

## FunPhotoSource

### Fundamental characterization and photoemission properties of multi-alkali antimonides for ultra-bright electron sources

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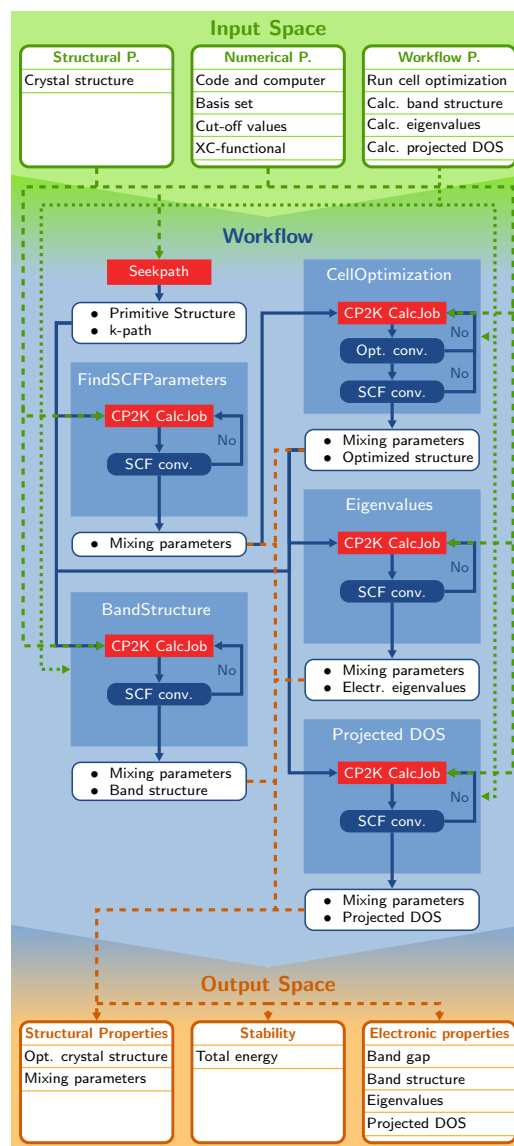
#### In Short

- Development of novel materials used as electron sources in particle accelerators.
- Theoretical screening of surfaces and point defects.
- *High-throughput* study within the framework of density functional theory.

Advances in the generation of ultra-bright electron beams have benefited from the ongoing developments of laser-driven photocathode radio-frequency injectors and, in turn, have enabled the development of new experimental techniques, such as ultra-fast electron diffraction and time-resolved electron microscopy, that offer unprecedented opportunities for material characterization following the dynamics of charge carriers in their native space and time scales. In current state-of-the-art photoinjectors, electron beam emittances are largely dominated by the thermal emittance at the cathode. To overcome these problems, alkali antimonide and telluride semiconductors have emerged as new classes of photocathode materials and are largely employed in many particle accelerators worldwide [1–3].

In spite of their potential as ultra-bright electron sources, these classes of compounds present issues in terms of stability, lifetime, and reproducibility of the growth procedure. Hence, controlled growth of multi-alkali antimonides with well-defined composition, stoichiometry, and crystal structure is far from trivial. The experimental exploration of a manifold of different stoichiometries and configurations is irrevocably connected with tremendous efforts and is therefore hardly feasible without a proper guidance from theory.

*Ab initio* methods such as density functional theory (DFT) and many-body perturbation theory have been applied to unravel the electronic structure of Cs-based antimonides with unprecedented insight and accuracy [4–7]. Embedding first-principles simulations in automatized high-throughput workflows is currently the state-of-the-art method to efficiently explore configurational spaces composed of hundreds or even thousands of compounds [8]. Recently, we have integrated in AiIDA [9], one of the



**Figure 1:** Schematic overview of the developed computational workflow for bulk crystals. The green area on the top represents all relevant input parameters in the input space; the blue area in the middle includes the steps of the workflow, where all relevant sub-processes are indicated within rectangular fields; the orange area at the bottom shows calculated properties that are stored as output nodes in the database. Green and orange dashed arrows show the data transfer from the input space into the workflow and from the workflow to the output space, respectively. Dark blue arrows denote transfer of data between the sub-processes defining dependencies among them. Green dotted arrows show the transfer of control parameters. Figure taken from Ref. [10]

most established platforms for this type of calculations, routines to perform high-throughput screening of materials for targeted applications, such as photocathodes. The success of this approach has been demonstrated by exploring the configurational space of Cs-Te compounds, and in particular the stability and the electronic structure of a number of crystalline stoichiometries and polymorphs [10]. The workflow implemented and adopted to perform these calculations is shown in Figure 1. In parallel, we have benchmarked the performance of the most popular approximations for the exchange-correlation potentials in DFT on the binary systems Cs<sub>2</sub>Te and Cs<sub>3</sub>Sb, checking that the SCAN functional offers the best trade-off between computational costs and accuracy in reproducing band gaps [11]. Equipped with this knowledge, we have moved on investigating surface properties in CsK<sub>2</sub>Sb slabs. In that work, we have examined the stability and the electronic structure of the surfaces with Miller indices 0 and 1 for all possible atomic termination, identifying both metallic and semiconducting systems [12]. This characteristic evidently has a strong impact on the work function, which is one of the key properties to determine the photoemission performance of the system.

With this knowledge, we will extend our investigation on the entire spectrum of (multi-)alkali antimonides including binary and ternary compounds as part of the project “FunPhotoSource”, recently funded by the DFG. In the first phase, we will explore the stability of all binary and ternary materials that arise from combining one or two alkali species, respectively, with Sb; in this analysis, we will consider both bulk crystals as well as surfaces. Especially for the ternary systems, each slab may have many different terminations, which have to be characterized explicitly, as they can substantially impact the electronic properties of the system [12]. Furthermore, we will study the impact of different kinds of defect on the electronic properties of multi-alkali antimonides. In the atom deposition procedures adopted experimentally for sample growth, the formation of vacancies and substitutional defects is very likely and can affect the macroscopic characteristic of the materials dramatically. To complement measurements, we will explore from first principles how favorably can these defect form, and whether and how the nature of such defects and their concentration in the samples modifies the work function and the electronic structure of the materials. The information obtained from these calculations will be essential to predict the photoemission behavior of the cathodes using the three-step model and to connect the corresponding outcomes to the experimental findings. These activities will be carried out in the second and third year of the project funding period.

## WWW

<https://uol.de/en/est>

## More Information

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## Project Partners

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## DFG Subject Area

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