

Summary

Over the past decade, experimental breakthroughs have revealed superconductivity in hydrogen-rich compounds under high pressure at near-room temperature, renewing interest in the role of hydrogen in this phase of matter. Indeed, incorporating such an element into materials can substantially alter their electronic properties, sometimes giving rise to new superconducting phases or modifying existing ones. It can also influence with competing electronic orders, such as spin and charge density waves, providing valuable insights into the mechanisms behind superconductivity and its interplay with other ordered states.

Given this potential, hydrogen could, in principle, also serve as a promising tool in the search for topological superconductors. These materials are predicted to host exotic quasiparticles capable of encoding quantum information in a way intrinsically protected from local noise and decoherence, offering a potential route toward fault-tolerant quantum computation.

Within this perspective, this thesis ultimately aims to intercalate hydrogen into two layered materials (Bi_2Se_3 and $\text{FeSe}_{1-x}\text{Te}_x$) that are promising candidates for hosting topological superconductivity. The purpose is to investigate whether the incorporation of such an element induces effects that provide deeper insight into the actively pursued quantum state these systems are predicted to host. To achieve this, a multistep approach is employed.

The first step focuses on the technique used for hydrogen intercalation: the ionic gating-induced protonation. Although this method, based on ionic gating, has demonstrated considerable potential for modifying the electronic properties of materials, it remains relatively underexplored, leaving opportunities for optimization towards efficient and reliable hydrogen loading. Accordingly, the initial part of this thesis investigates ionic gating-induced protonation using a deep eutectic solvent, an electrolyte of a class not previously explored for this purpose. Specifically, the study examines how tuning the process parameters can maximize hydrogen loading in palladium, a material chosen for its highly suitable properties. The results reveal a clear positive correlation between the applied gate voltage and the hydrogen concentration achieved. Notably, the concentrations attained exceed those typically attained with conventional ionic liquids, which are commonly employed in

this technique. These findings demonstrate that deep eutectic solvents can outperform traditional electrolytes used in ionic gating-induced protonation and provide a potential framework for maximizing hydrogen loading in various materials, though material-specific adjustments may be required.

Next, hydrogen intercalation via ionic gating-induced protonation is performed on the layered transition metal dichalcogenide TiSe_2 . While this material has recently been shown to exhibit superconductivity upon hydrogen intercalation, a detailed characterization of its superconducting state remains lacking. Its study is particularly compelling, as it offers insight into the interplay between superconductivity and the intrinsic charge density wave order of the pristine material. Through magnetotransport measurements, point-contact Andreev reflection spectroscopy, and tunneling spectroscopy, the superconducting phase is found to exhibit a nontrivial superconducting order parameter with two nodeless gaps. The temperature dependence of the gaps and of the upper critical magnetic field are consistent with both Bardeen-Cooper-Schrieffer theory and the Eliashberg formalism for a two-gap superconductor. This study not only clarifies the superconducting properties of H_xTiSe_2 but also provides hands-on experience with ionic gating-induced protonation and point-contact Andreev reflection spectroscopy on layered materials, offering transferable expertise for the topological candidates explored later in this thesis.

Building on this expertise, hydrogen intercalation is then performed on the layered topological superconductor candidates Bi_2Se_3 and $\text{FeSe}_{1-x}\text{Te}_x$. In Bi_2Se_3 , no superconducting transition is observed in samples treated with ionic gating-induced protonation across a wide range of parameters, even when following the original protocol previously claimed to induce superconductivity. In contrast, in $\text{FeSe}_{1-x}\text{Te}_x$ with $x \approx 0.5$, ionic gating-induced protonation successfully tunes both the superconducting critical temperature and the superconducting gaps, as measured by point-contact Andreev reflection spectroscopy performed directly under gating – a measurement configuration not previously explored, since ionic gating was considered to be incompatible with such a technique. In rare cases, signatures of potential unconventional surface pairing, consistent with topological surface superconductivity, are also observed both in pristine and H-intercalated $\text{FeSe}_{0.5}\text{Te}_{0.5}$.