

Hyper expanded molecule intercalated iron selenides with robust superconducting response

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Iron-chalcogenides along with the copper oxides constitute two families of 2D materials that exhibit high superconducting critical temperature (T_c). In these, magnetism and superconductivity typically coexist, and the superconducting state emerges when antiferromagnetic (AFM) order is suppressed by carrier doping, structural modifications under external pressure, or chemical pressure via isovalent substitutions. Since the parent materials of Fe-based superconductors have layered structures, doping by intercalation offers a facile avenue to suppress the AFM state with concomitant enhancement of the T_c . Electron donor molecules co-intercalated with alkalis in the β -FeSe (Fig. 1b) allow to study the impact on the electronic structure of the host, as the intercalation increases the interlayer separation and leads to a five-fold rise of the T_c (44 K). We have developed low-T solvothermal routes, for such high- T_c $\text{Li}_x(\text{C}_5\text{H}_5\text{N})_y\text{Fe}_{2-z}\text{Se}_2$ (Fig. 1a) [1] in order to study their structure-property relations by high-resolution synchrotron-based tools. Element-specific (Fe & Se K-edge) X-ray absorption (XAS) and emission (XES) spectroscopies were utilised to provide valuable insights on how the interlayer guests [Li-C₅H₅N] impact the evolution of (i) the FeSe₄ building blocks in the FeSe electronically active layers, including the Fe local moments, and (ii) the magnitude of T_c . The near edge region of the XAS confirmed doping-mediated local atomic rearrangements and progressive filling of orbital states, with concomitant reduction of empty levels near the E_F with respect to β -FeSe. The $K\beta$ XES point that the intercalated compounds, carry a low-spin state, as well as a somewhat reduced Fe local magnetic moment. Local structure assessments, based on modeling the EXAFS oscillations, suggest that while the Fe-Se bond remains stiff and covalent in nature, the Fe-Fe bond evolves to become softer for high Li content (x), due to raised Fe-site deficiency (Fig. 1c). The findings provide insights on the desirable combination of local electronic and structural parameters that tune the T_c in such strongly correlated electron systems, possibly offering an avenue to engineer robust superconducting state at elevated temperatures.

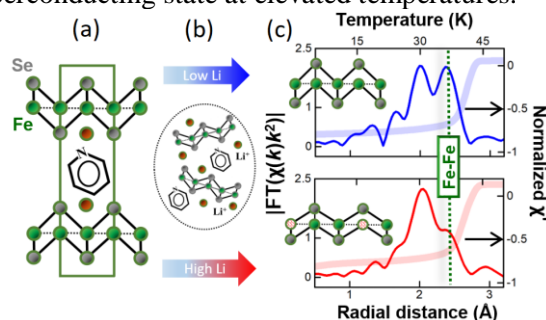


Figure 1: 2D iron-selenides, with large interlayer separation, probed by synchrotron XAS, bear Fe-vacancies with increased Li-content, but retain a robust superconducting state.

References

[1] Berdiell I. C. *et al.*, *Inorg. Chem.* **61**, 10 (2022).

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