

# Order-parameter evolution in the Fulde-Ferrell-Larkin-Ovchinnikov phase probed by $^{13}\text{C}$ NMR spectroscopy

**H. Kühne,<sup>1</sup> S. Molatta,<sup>1</sup> T. Kotte,<sup>1</sup> D. Opherden,<sup>1</sup> G. Koutroulakis,<sup>2</sup> J. A. Schlueter,<sup>3,4</sup>  
G. Zwicknagl,<sup>5</sup> S. E. Brown<sup>6</sup> and J. Wosnitzer<sup>1,7</sup>**

<sup>1</sup>*Hochfeld-Magnetlabor Dresden (HLD-EMFL) and Würzburg-Dresden Cluster of Excellence ct.qmat, Helmholtz-Zentrum Dresden-Rossendorf, Germany, e-mail: h.kuehne@hzdr.de*

<sup>2</sup>*Department of Physics, UCSB, USA*

<sup>3</sup>*Materials Science Division, Argonne National Laboratory, USA*

<sup>4</sup>*Division of Materials Research, National Science Foundation, Alexandria, USA*

<sup>5</sup>*Institute for Mathematical Physics, Technische Universität Braunschweig, Germany*

<sup>6</sup>*Department of Physics and Astronomy, UCLA, USA*

<sup>7</sup>*Institut für Festkörper- und Materialphysik, TU Dresden, Germany*

The Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state represents a general concept of pairing in multi-component Fermi liquids with strong population imbalance. Clear experimental realizations of FFLO superconductivity require stringent conditions, such as an orbital critical field much larger than the Pauli paramagnetic limit, and a mean free path larger than the coherence length. In the FFLO state, the spin polarization varies as the modulus of the spatially modulated superconducting gap amplitude, and, thus, its modulation amplitude represents the order parameter. Indeed, NMR experiments of the organic superconductors  $\beta''$ -(ET)<sub>2</sub>SF<sub>5</sub>CH<sub>2</sub>CF<sub>2</sub>SO<sub>3</sub> and  $\kappa$ -(ET)<sub>2</sub>Cu(NCS)<sub>2</sub> showed the emergence of inhomogeneous line broadening, which stems from the spatial modulation of the spin polarization [1-3].

We report on our experimental studies of the temperature-dependent order parameter of the superconducting FFLO state. For that, we utilize  $^{13}\text{C}$  NMR spectroscopy studied in  $\beta''$ -(ET)<sub>2</sub>SF<sub>5</sub>CH<sub>2</sub>CF<sub>2</sub>SO<sub>3</sub>. From a comparison of our experimental results to a comprehensive modeling of the  $^{13}\text{C}$  NMR spectra, we determine the evolution of the local spin-polarization modulation amplitude  $\Delta K_{\text{spin}}$  upon condensation of the FFLO state. In the experimentally probed temperature regime,  $\Delta K_{\text{spin}}$  yields a mean-field-type evolution of the order parameter. Further, the modeling of the spectra in the superconducting phase allows to quantify the decrease of the average spin polarization, stemming from the spin-singlet coupling of the superconducting electron pairs in the FFLO state of  $\beta''$ -(ET)<sub>2</sub>SF<sub>5</sub>CH<sub>2</sub>CF<sub>2</sub>SO<sub>3</sub>.

[1] J. A. Wright *et al.*, Phys. Rev. Lett. **107**, 087002 (2011).

[2] H. Mayaffre *et al.*, Nat. Phys. **10**, 928 (2014).

[3] G. Koutroulakis *et al.*, Phys. Rev. Lett. **116**, 067003 (2016).