Metals play a crucial role in medicine as a part of therapeutic or diagnostic preparations. However, in the majority of cases, their properties cannot be utilized entirely in free ionic form. Organic molecules capable of chelation are used to open the full potential of the metal. The molecules are called chelators and are the core theme of this thesis. The most important function of these molecules is the chelation and coordination of the metal, but chelators can provide other important functionalities. This work, therefore, focuses on the design, synthesis, and application of such polyfunctional chelators and is divided into two parts:

DO3A-Hyp

This part of the thesis deals with chelators that can be used as amino acids to incorporate lanthanides into peptides. The developed chelators provide a short and rigid connection of the metal to the peptide chain. Tripeptides containing two units of such chelators with a central amino acid bearing a CF_3 group were synthesized to demonstrate the capability of DO3A-Hyp building blocks. Two paramagnetic metals were combined within this tripeptide, and it was shown that such a rigid and locked system could be used for combining their magnetic susceptibility tensors. These magnetic susceptibility tensors were used for manipulation of the ^{19}F NMR shift of the CF_3 reporter group. The combination of two different paramagnetic lanthanides resulted in four clearly distinguishable signals readable by ^{19}F NMR spectroscopy. A system constructed from such a platform and four paramagnetic metals (Dy, Ho, Tb, Tm) was used for 16-bit encoding and decoding of information. To further explore the potential, parallel reading of information by ^{19}F MRI was performed.

PET/MRI bimodal contrast agents

The second part of the thesis explores the area of under-investigated low molecular weight bimodal PET/MRI contrast agents. A suitable structural motif was designed, synthesized, and radiolabeled to prove the concept of a bimodal PET/MRI contrast agent. Cooperation with the Werner Siemens Imaging Center in Germany was set up to probe the properties and perspectives of prepared compounds. Within this cooperation, radiolabeling conditions were optimized and developed for radiolabeling on an automated radiosynthesis module. Two contrast agents were prepared. First, based on the DO3A motif, was prepared to serve as a perfusion agent. The second, structurally related to DO2A, was synthesized to serve as a lactate-responsive contrast agent. Properties such as kinetic inertness, relaxivity, and cytotoxicity were determined for these compounds *in vitro*. The perfusion PET/MRI contrast agent was synthesized, and phantoms thus prepared were measured simultaneously in a combined PET/MRI scanner to prove the capability for the intended use. As the last step, *in vivo* experiments were performed on mice, resulting in data that confidently confirmed the suitability of this molecule as a perfusion bimodal PET/MRI contrast agent.