The neutron, one of the most common building blocks of matter, is also a unique probe for studying materials and fundamental interactions. The only electrically-neutral nucleus, the neutron passes through most materials with ease, even at the lowest energies. Neutrons are particularly powerful at characterizing magnetic structures. In order to extend the applications of neutron physics as quantum probes/sensors, we have developed methods based on Quantum Information Processing for preparing structured waves such as orbital and spin-orbit states of neutrons. Such states may be prepared for the beam as a whole, or with reference to the coherence length of the neutron wave packet, which is much smaller than the width of useful beams. I will show how we can control the wave packet momentum distribution by various phase gratings and will describe experiments to prepare and characterize neutron beams with specific orbital and spin-orbit structure based on neutron interferometry. Finally, I will show how we can apply these methods to other types of probes such as light.