PET and MRI provide complementary information in the study of the human brain. Simultaneous PET/MRI data acquisition allows the spatial and temporal correlation of the measured signals, creating opportunities impossible to realize using stand-alone instruments. This paper reviews the methodologic improvements and potential neurologic and psychiatric applications of this novel technology. We first present methods for improving the performance and information content of each modality by using the information provided by the other technique. On the PET side, we discuss methods that use the simultaneously acquired MRI data to improve the PET data quantification. On the MRI side, we present how improved PET quantification can be used to validate several MRI techniques. Finally, we describe promising research, translational, and clinical applications that can benefit from these advanced tools.