Most intelligent transportation systems use a combination of radar sensors and cameras for robust vehicle perception. The calibration of these heterogeneous sensor types in an automatic fashion during system operation is challenging due to differing physical measurement principles and the high sparsity of traffic radars. We propose - to the best of our knowledge - the first data-driven method for automatic rotational radar-camera calibration without dedicated calibration targets. Our approach is based on a coarse and a fine convolutional neural network. We employ a boosting-inspired training algorithm, where we train the fine network on the residual error of the coarse network. Due to the unavailability of public datasets combining radar and camera measurements, we recorded our own real-world data. We demonstrate that our method is able to reach precise and robust sensor registration and show its generalization capabilities to different sensor alignments and perspectives.