Strong interaction between localized surface plasmons and nanoscale objects has led to the development of highly sensitive biochemical sensing in planar metallic nanostructures with sensing performances mainly dependent on the interaction volume and the local electric field. However, the sensitivity and the interaction volume of these planar structures have been limited by the achievable aspect ratios based on standard lift-off process. Here, we propose combining electron beam lithography (EBL) for template definition with pulsed electrodeposition directly onto indium tin oxide-coated glass for structure growth. We first show the advantages of pulsed electrodeposition over that using direct current. We demonstrate how this can be used to fabricate arrays of high quality gold dimer structures with high aspect ratio sub-10-nm gaps. We also compare the sensitivity of the proposed high aspect ratio structures with that of their planar counterparts, and investigate the feasibility of using them as a practical biosensing substrate. We also demonstrate the process of using negative resist with electrodeposition to fabricate inverse structures.