Nonribosomal peptide synthetases (NRPSs) are biosynthetic enzymes that produce diverse secondary metabolites, including antibiotics and other therapeutics. They are arranged as an assembly line of modules where each module incorporates one aminoacyl monomer into the nascent peptide. Little is known about the interplay between modules within multimodular NRPSs. We determined five x-ray crystal structures from the first two modules of linear gramicidin synthetase, including the full core dimodular structure showing trans-module delivery of the peptide intermediate during the condensation reaction. The structures, along with small-angle x-ray scattering data, show that adjacent modules undergo massive conformational rearrangements and that relative module positions are not governed by the catalytic cycle. Using the structures and covariation analysis, we bioengineered a module-swapped dimodular NRPS with improved catalytic abilities.