Intermetallic phases are solid state compounds formed between metals that exhibit a wide structural variety and a myriad of important properties such as magnetism, catalysis, and superconductivity. Developing their properties requires an ability to control or guide their crystal structures, which can be wildly elaborate, involving thousands of atoms per unit cell. One principle for approaching this problem is the recognition that many of the most complex phases encountered in intermetallics can be understood in terms of fragments of simpler structures. The challenge of tailoring of intermetallics then lies in understanding the driving forces for the insertion of interfaces into simple structures to build complexity. We present the synthesis and the crystal structure of a new phase that sheds light on this issue, GdFe$_{0.69}$Si$_{1.94}$. It shares structural motifs with two neighboring phases in the Gd-Fe-Si system with common structure types, GdSi$_2$ (AlB$_2$-type) and GdFe$_2$Si$_2$ (ThCr$_2$Si$_2$-type). GdFe$_{0.69}$Si$_{1.94}$ arises as the hexagonal nets of GdSi$_2$ are cut perpendicular to the a axis by square nets, forming distinct slabs. The surroundings of the Fe square nets locally resemble slabs of the GdFe$_2$Si$_2$ structure, but the Fe content of the nets varies, depending on the presence of an Fe/Si mixed site. GdFe$_{0.69}$Si$_{1.94}$ thus represents an intermediate point between the GdSi$_2$ and GdFe$_2$Si$_2$ phases. This viewpoint offers the possibility of a series of crystal structures linking GdSi$_2$ and GdFe$_2$Si$_2$, representing a continuum of GdFe$_x$Si$_2$ phases with varying degrees of Fe incorporation, which we are now exploring synthetically.

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