Abstract: Highly mismatched “designer” alloys are materials that contain chemical elements with very different atomic sizes and abilities to attract nearby electrons. When a few atoms with larger or smaller atomic sizes are added to a host material, its electrical and optical properties often change dramatically. For example, the incorporation of dilute fractions of nitrogen and bismuth solute atoms into III-V semiconductors induces significant energy bandgap narrowing; thus, emerging dilute-nitride-bismuthide alloys are of significant interest for long-wavelength applications ranging from temperature-insensitive laser diodes to ultra-high efficiency multijunction photovoltaic cells. While the properties of emerging dilute-nitride-bismuthide alloys are highly sensitive to local atomic environments, the solute incorporation mechanisms are not well understood. In this talk, we present combined computational-experimental studies which enabled our pioneering epitaxy and band structure engineering of GaAs(N):Bi alloys. In addition to describing recent advances in surface reconstruction-driven control of solute incorporation [1,2] and atomic-ordering [3], we present a new “magic ratio” for lattice matching of GaAsBi with GaAs substrates [4]. We also present a strategy for the synthesis and tailored electronic structure of III-V bismuthides [5] for integration with III-V based electronics.

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