COMPENSATING FOR STRESS FROM DOPANTS IN THE GROWTH OF ROOM-TEMPERATURE FERROMAGNETIC SEMICONDUCTORS

Significant provide the science and application of electronics based on electron spin and charge—is a burgeoning field with vast potential applications. But the full realization of its potential depends on the development of semiconductors that are ferromagnetic at room temperature and also compatible with traditional silicon-based circuits. Such semiconductors have proven devilishly difficult to create, however, because doping at the high levels needed for room-temperature ferromagnetism induces too much epitaxial strain, which can lead to disorder and phase separation in the crystal lattices. How is it possible to achieve the high doping percentage required while preserving the semiconductor? A team of researchers from the University of North Carolina and Argonne National Laboratory, using beamlines MHATT/XOR 7-ID-C, XOR 2-BM, and XOR 2-ID-E, has found a possible answer to this question. Employing two different elements as dopants, the experimenters were able to compensate for the epitaxial strain and suppress the phase separation effects manifested at higher doping levels.

The team grew crystals highly doped with Co and Mn in sequence with 2 Å intervals of Ge(001) by using combinatorial molecular beam epitaxy. The total concentration (x) of dopants was varied, while the relative concentration (a/b) between Co and Mn was maintained. By using real-time *in situ* scanning reflection high-energy electron diffraction (RHEED), x-ray diffraction (XRD), and x-ray fluorescence spectroscopy (XFS) from 7-ID-C, 2-BM, and 2-ID-E, the epitaxial growth process and the structural evolution and composition of the samples were studied.

Above a doping concentration x, a roughening transition occurs, which is confirmed by RHEED and XRD and also by complementary scanning probe microscopy (SPM) and high-resolution transmission electron microscopy. Around this transition, as the atomically smooth two-dimensional (2-D) RHEED intensity diminishes and peak width increases, rough three-dimensional (3-D) RHEED features begin to appear, turning completely 3-D at higher x values. As x increases further, growth becomes still more disordered. An epitaxial phase diagram derived from these measurements shows that this transition depends on the relative doping concentration. With $a/b \sim 3$ up to maximum doping of ~ 14 at. %, there is coherent 2-D epitaxial growth. Within this window, promising Ge magnetic semiconductors have been discovered with ferromagnetic transition temperature T_c near room temperature. As a/b deviates from ~3, lattice strain increases and growth becomes unstable, with the films exhibiting lattice relaxation, surface roughening, and phase separation. The differing tetrahedral covalent radii of Mn and Co compared to Ge (Mn is larger, Co smaller) make it



Fig. 1. Evolution of structure, magnetism, and electronic transport of highly doped Ge(001) epitaxial films using combinatorial approach. a. Composition profile versus position on the sample measured by XFS (blue for Co and red for Mn). b. Surface morphology measured by RHEED: the 0th order specular intensity in log scale versus q perpendicular to the surface (vertical) and composition. c. X-ray diffraction intensity in log scale versus reciprocal lattice vector L in the [11L] direction of Ge (vertical) and composition. d. Conductivity in log scale and Curie temperature (points) versus temperature and composition. The structure exhibits a transition from smooth coherent epitaxy to rough/disordered growth at doping concentration ~ 10 at. % (b and c) that correlates with a semiconductor to metal transition (d) and an anomaly in Curie temperature. Within the ordered semiconducting regime, the correlation between the doping profile (a) and epitaxial strain as indicated by the fringes in (c) demonstrates the strain compensation effect due to the two dopants in Ge lattice.

possible for Co and Mn to compensate for each other's strain effects. Specifically, the expansion caused by Mn doping can be offset by the compression resulting from Co doping.

Further studies show that the onset of the roughening transition also depends on the film thickness. The observed thickness dependence indicates that phase separation, along with strain, is a major factor in controlling the epitaxial growth. In other words, when compared to systems with low solubility alone, the added lattice strain can lead to earlier onset of defective and inhomogeneous crystal growth as doping concentration increases.

The research team has shown that using complementary transition metal dopants, such as Mn and Co, can reduce internal stress and thus resist phase separation and stabilize epitaxial growth at the higher doping levels needed for the production of room-temperature ferromagnetic semiconductors. Because not all potential dopants possess the complementary characteristics to compensate for each other's individual strain profiles, it is possible to narrow the search for appropriate materials based on known structural and chemical characteristics. Moreover, the team's work demonstrates the practicality of engineering the semiconducting materials with tailored electronic and magnetic properties, thus bringing the great promise of spintronics technology much closer to realization. — *Mark Wolverton*

See: F. Tsui¹, L. He¹, A. Tkachuk², S. Vogt², and Y.S. Chu², "Evidence for Strain Compensation in Stabilizing Epitaxial Growth of Highly Doped Germanium," Phys. Rev. B **69**, R081304-1 (2004).

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