(Invited) Raman Spectroscopy for Epitaxial Silicon-Germanium-Tin Alloys

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ECS Meeting Abstracts, Volume MA2024-02, G03: SiGe, Ge, and Related Compounds: Materials, Processing, and Devices 11: In Memory of Qizhi Liu

Citation Davide Spirito *et al* 2024 *Meet. Abstr.* **MA2024-02** 2349

DOI 10.1149/MA2024-02322349mtgabs

Abstract

Raman spectroscopy is well established as a metrology tool and as a method for the investigation of materials. It is an all-optical method that requires little or no sample preparation. It measures the energy and strength of vibrational modes, connected to structural features such as crystal phase, strain, and disorder. By tuning the experimental parameters such as laser excitation wavelength and polarization, sample orientation, and temperature, it provides a deep understanding of sample properties that can be fruitfully complemented with other experimental and theoretical methods. In addition, sub-µm resolution mapping is possible to gain insight into devices.

In this work, we present several facets of Raman spectroscopy for the characterization of SiGeSn alloys, focusing on the case of epitaxial layers of technological interest. The SiGeSn material system holds great promise for CMOS-compatible applications in electronics, optoelectronics, photonics, and thermoelectrics. Together with challenges in crystal growth and device design, reliable and informative characterization methods, such as Raman spectroscopy, are a prerequisite in the development of this technology.[1]

The Raman spectrum of SiGeSn consists experimentally of several features, that have been associated with vibrations of pairs of the constituent atoms in a cubic crystal with random alloy ordering. The use of polarization-dependent Raman spectroscopy reveals a more complex landscape that can be successfully compared with atomistic simulations.[2] In GeSn, Ge-Ge pairs give rise to two vibrational modes with different energy and same symmetry, associated with the proximity of the pair to regions of locally higher or lower Sn concentration. This suggests that a short-range order configuration, different from a completely random alloy, dominates even at a low Sn content of around 10 at.%. In addition, modes with different symmetry can be attributed to the disorder rather than to specific pairs of atoms. Finally, this method allows a calibration of the linear dependence of the modes energy on the composition.

Composition and epitaxial strain are the strongest factors in shifting the modes energy at a given temperature. As the temperature is varied, an additional shift occurs because of anharmonic coupling and differential thermal expansion in epitaxial layers.[3] Thus, the use of Raman spectroscopy for strain metrology needs to be reassessed. We present an analysis of these temperature-dependent mechanisms. Comparison with experimental data shows that the anharmonicity in GeSn with Sn content up to 15 at.% is not different from Ge, and that a reliable calibration can be obtained to measure strain in epitaxial GeSn at various temperatures. In this way, the strain distribution in microdevices can be obtained even at the cryogenic operating temperature.

Finally, Raman spectroscopy for measurement of thermal properties is presented. [4] Knowing the dependence of the modes on temperature, the local heating of the sample surface by the laser

excitation can be estimated. In combination with a model for heat conduction in layered structures, the thermal conductivity of GeSn layers grown on Ge is thus calculated. The conductivity decreases with increasing Sn content, and is controlled by alloy scattering. The measured low values of thermal conductivity, together with the knowledge of the band structure and electrical transport, allows an estimation of the thermoelectric figure of merit, indicating that GeSn may be successfully applied to energy harvesting around room temperature.

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