Anharmonicity of the E_2(high) and A_1(LO) phonons in GaN studied by temperature-dependent Raman spectroscopy

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Currently, the wide band gap semiconductor GaN is intensively investigated in view of its potential for opto-electronic devices as well as for high-frequency and high-temperature electronics. Commonly, wurtzite (α-) GaN layers are grown on Al_2O_3 and 6H-SiC substrates. However, the strong impact of the heteroepitaxy, e.g., on the vibrational properties of the GaN lattice, is mostly neglected.

We study the intrinsic anharmonicity of the lattice vibrations in α-GaN as well as the strong influence of the heteroepitaxy by using temperature-dependent Raman spectroscopy. We focus on the E_2(high) and the A_1(LO) phonon lines, which are the strongest ones for backscattering along the c-axis among the six Raman-active vibrational modes. The measured temperature dependence of the widths and shifts of these lines for layers grown on sapphire agree well with recently published values [1]. Detailed data for films grown on 6H-SiC have not published. From measurements of layers grown on both substrates, we obtain the matrix elements for three- and four-phonon scattering of the E_2(high) and A_1(LO) phonons, which differ remarkably from those obtained in Ref. [1]. For heteroepitaxial grown GaN layers, the intrinsic anharmonicity of the GaN lattice vibrations is predominantly reflected by the temperature-dependent part of the linewidth, whereas the temperature-dependent Raman shift mainly reflects the strong influence of the substrate on the GaN lattice.

With regard to the total linewidths we find that the temperature-dependent contribution to the linewidth is the same for all samples. This part is due to the intrinsic phonon-phonon scattering in α-GaN and does not depend on the substrate. However, the temperature-independent part of the linewidth depends on the substrate. Its value, which can be precisely determined at low temperatures, increases with the defect concentration and the inhomogeneous strain in the layer. For high-quality α-GaN layers, we obtain at low temperatures a linewidth of 0.63 and 1.7 cm^{-1} for the E_2(high) and A_1(LO) modes, respectively. The small linewidth for the E_2(high) phonons is due to the rather small two-phonon density of states, which is about three times smaller compared to that of the A_1(LO) phonons at the corresponding phonon energies.

The temperature-dependent shift of the E_2(high) phonon frequency is dominated by the thermal lattice expansion of the GaN layer and not by the rather small contribution of the three-phonon scattering. The anisotropic lattice expansion in the GaN layer is determined by the intrinsic thermal properties of α-GaN as well as by the mismatch between the in-plane thermal lattice expansion coefficients of GaN and the substrate, which causes an additional small biaxial stress in the layer. Both effects can be well described by the Grüneisen approach, if the true strain of the GaN layer within its plane and parallel to the c-axis are taken into account. We estimate the bare phonon frequency to be 569.7 and 740 cm^{-1} for the E_2(high) and A_1(LO) phonons, respectively, and compare these values with published data.

[1] W.S. Li, Z.X. Shen, Z.C. Feng, and S.J. Chura, J. Appl. Phys. **87**, 3332 (2000).