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Raman Scattering in Resonance with Acceptor-Bound Excitons in GaN

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Introduction Resonance effects in light scattering from solids occur in the range of frequencies of the electronic absorption bands. Resonant Raman scattering, i.e., a strong enhancement of the Raman scattering cross section, can be observed if the energy of the incoming or scattered photons matches real electronic states in the material. This results from the denominator in the Raman scattering cross section tending to zero. One refers to incoming and outgoing resonance, respectively (see, e.g., [1]).

In GaN resonant Raman scattering was reported under excitation slightly above the band gap [2, 3]. The appearance of multiple LO phonons due to Fröhlich interaction indicated the strong polar character of the Ga-N bond. Furthermore, this technique was applied to study compositional non-uniformity in the ternary alloy InGaN [4, 5]. However, resonance Raman scattering at bound excitons as reported for CdS is not known for GaN [6, 7]. To the best of our knowledge we report Raman scattering in resonance with acceptor-bound excitons in GaN for the first time.

Experimental Details The sample under study was a not intentionally doped 220 µm thick GaN film grown by hydride vapor phase epitaxy. Sapphire (0001) with a sputtered ZnO buffer layer was used as substrate [8].

Raman spectra were taken in backscattering geometry $z(..)\underline{z}$ under excitation with the 363.8 nm Ar^+ line. The measurements were carried out with a Dilor UV spectrometer having a resolution of 3 cm^{-1}. In order to study the temperature dependence of the excitonic recombination cathodoluminescence (CL) experiments were performed.

Results and Discussion Figure 1 exhibits a Raman spectrum of a GaN sample at 4 K excited at 3.408 eV, i.e. approximately 100 meV below the GaN band gap. Besides the first-order E_2 mode (569 cm^{-1}) with a low-energy shoulder originating from A_1(TO) and the 1LO mode (739 cm^{-1}) we observe the 2LO (1478 cm^{-1}) and 3LO (2202 cm^{-1}) emission and a sum process due to 2LO phonons plus one E_2 phonon at 2040 cm^{-1}. The LO frequency is somewhat higher than the reported value of 735 cm^{-1} for the A_1(LO) [9] and closer to the E_1(LO) at 742 cm^{-1}. However, since the E_1(LO) mode is not allowed in the present scattering geometry and the value agrees with the A_1(LO) within the experimental accuracy, we assign the mode as A_1(LO) in agreement with [10]. The structure around 1280 cm^{-1} known from second-order Raman spectra [11] probably originates from a combination of A_1(TO) and A_1(LO) phonons.

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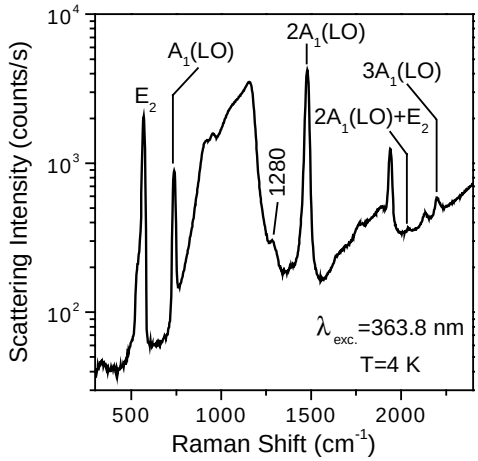

Fig. 1. Resonant Raman spectrum of a GaN sample at 4 K on a logarithmic scale

In Fig. 2 the Raman scattering intensity of the 1LO and 2LO mode as function of the temperature is depicted. The excitation condition was varied by changing the temperature and leaving the excitation energy constant. Starting at 4 K the intensity increases with temperature having a local maximum at 80 K and a main maximum at 130 K. A further temperature increase up to 330 K leads to a rapid intensity reduction. The 1LO and 2LO modes show a similar behaviour with the 2LO mode having a much higher intensity over the whole temperature range.

To investigate the origin of this pronounced resonance, temperature-dependent CL experiments were performed. The resulting spectra are shown in Fig. 3 including the excitation energy marked by a dotted line. According to [12] we assign the luminescence at 3.454 eV as recombination from acceptor-bound excitons (A^0X). The luminescence maximum is below the excitation wavelength at 18 K, crosses the dotted line and is at higher wavelength for temperatures above 160 K. A closer inspection of this luminescence requires three peaks to fit the

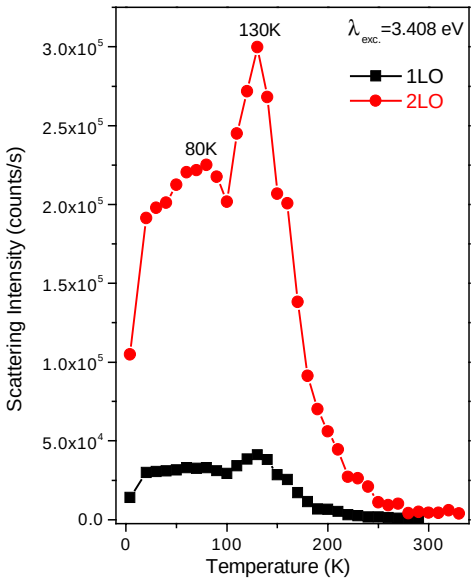

Fig. 2. Normalised scattering intensity of the 1LO and 2LO mode as function of the temperature excited at 363.8 nm

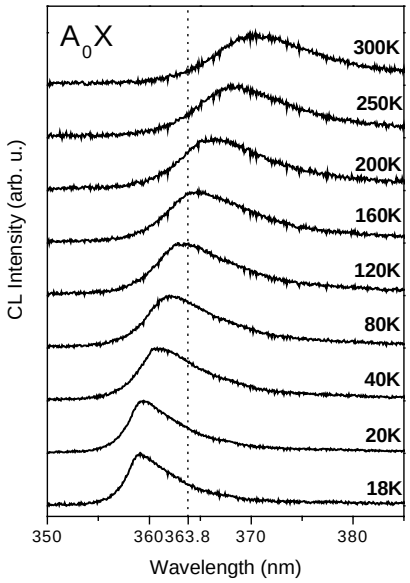

Fig. 3. CL spectra of GaN taken at various temperatures

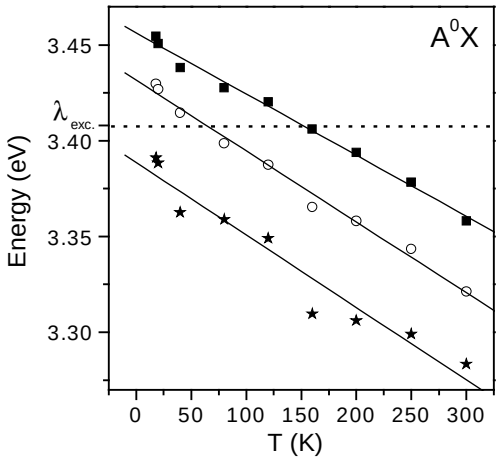

Fig. 4. Peak positions of the three A^0X peaks vs. temperature. Solid lines: linear fit, dotted line: excitation energy

lineshape. The peak positions of these three peaks are shown in Fig. 4. Here, the solid lines represent linear fits. The solid lines are crossing the dotted line at 60 K and 140 K. This means that the energy of the incoming photons matches the energy of the A^0X luminescence at these positions. Within the experimental accuracy the crossing points fit the temperatures where we find the maxima in the LO scattering intensity. This evidences that we have an incoming resonance at acceptor-bound excitons in GaN. The acceptor-bound exciton line consisting of three peaks may originate from three different acceptors. However, we rather think that we observe just one acceptor with a splitting of the exciton state for the reasons explained in [13]. The chemical nature of the acceptor was suggested to be Zn, which can diffuse from the buffer layer at the high growth temperatures [14].

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