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Hyper-Raman Scattering on GaN and CdS

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Introduction. In the recent years hyper-Raman (HR) scattering has become an important tool besides Raman scattering to obtain information about structural phase transitions or infrared and Raman-inactive (so-called silent) vibrational modes (for a review see, e.g., Ref. [1]). To the best of our knowledge HR measurements on wurtzite crystals were only performed on CdS [2]. We report on results of HR experiments on GaN and compare them with CdS.

Experimental Details. The samples under investigation were a nominally undoped 200 μm thick hexagonal GaN layer grown on sapphire by hydride vapor phase epitaxy (HVPE) and a bulk crystal of CdS. The experiments were carried out in 90° scattering geometry. The HR spectra were excited by the 1064 nm line of a mode-coupled Nd : YAG laser with pulse repetition rate of 76 MHz and pulse width of 100 ps. Focusing the laser light on a spot with diameter of 100 to 300 μm led to a peak power density of about 25 MW/cm^2. We mounted the samples on a water-cooled copper block in order to avoid overheating and burning off the crystals due to defects or surface imperfectness. The scattered light was analyzed by a Dilor triple monochromator and detected by a charge-coupled device (CCD) camera.

Theoretical Background. HR scattering is a nonlinear process. In wurtzite crystals (point group C_{6v}) incident light can lead to the generation of the second harmonic (SHG). However, the exciting light can be scattered by phonons leading to a HR shift relative to the SHG. For the point group C_{6v} two lattice vibrations at the Γ point of the Brillouin zone ($\mathbf{k} = \mathbf{0}$), the B_1(low) and the B_1(high) mode, are silent but HR active. So in principle HR experiments facilitate the observation of these lattice vibrations. The other optical modes of the wurtzite structure at $\mathbf{k} = \mathbf{0}$ are Raman and HR active.

Both the SHG and the HR-active modes obey selection rules (see Refs. [3] and [1], respectively). For example the intensity of the fully symmetric A_1 mode fulfills the following relation:

$$I_s \propto \left| \mathbf{e}_s \cdot \begin{pmatrix} & & \\ & & \\ b & b & a \end{pmatrix} \cdot \mathbf{e}_i \right|^2 , \tag{1}$$

where $\mathbf{e}_i$ and $\mathbf{e}_s$ refer to the polarizations of the two exciting photons and the scattered photon, respectively.

Discussion. Fig. 1a shows a HR spectrum of GaN taken in $x(z\text{-})z$ geometry. Both the SHG at 0 cm^{-1} and the longitudinal-optical (LO) E_1 mode at 735 cm^{-1} are detected. The A_1(LO) mode cannot be seen because of the HR selection rules. Neither transverse-optical (TO) nor silent modes could be detected here or in other scattering configurations. From that we deduce that mainly Fröhlich interaction contributes to the scattering mechanism [2].

Care has to be taken to exclude cascade processes caused by the SHG. However, the SHG was too weak to excite a Raman signal with the observed intensity. Furthermore the selection rules for the SHG should lead to Raman scattering in $x(z\text{-})z$ geometry. In this configuration the Raman-active E_1(TO) mode should be observed with a comparable intensity as the E_1(LO) mode (Fig. 1a, inset), which is not the case. Also the occurrence of so-called quasi modes, e.g. the mixture of E_1(LO) and A_1(LO) modes, can be excluded because the latter one is forbidden due to relation (1). Although GaN is transparent in the spectral region in which we performed our measurements we explain the softening of the E_1(LO) mode normally observed at 742 cm^{-1} by a heating up of the sample due to the absorption of the incident light. In spite of the taken precautions to avoid such effect it seems that heating up of the sample due to imperfectness and intrinsic defects can not be excluded.

CdS exhibits a similar behavior (Fig. 1b). Beside the SHG we only observe the E_1(LO) mode at 303 cm^{-1}. That indicates a similarly strong Fröhlich interaction in HR scattering as in GaN [2]. Note that in contrast to Raman scattering the disappearance of the luminescence background and the second-order peak at 606 cm^{-1} (Fig. 1b, inset) exclude a cascade process caused by the SHG. The disappearance of the second-order peak can be explained by the difference of the parity of the quantum-mechanical states, which are involved in Raman and HR scattering [2].

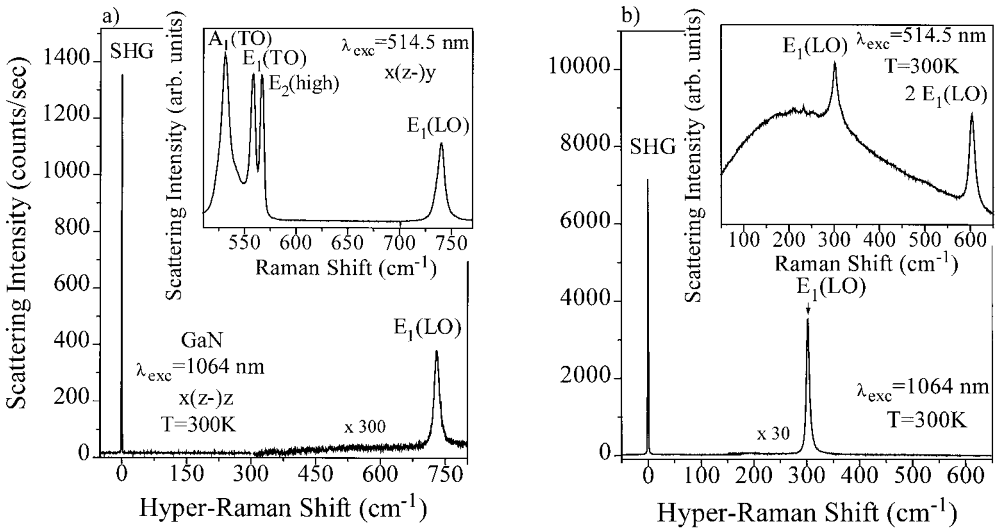

Fig. 1. Room-temperature HR spectra of a) GaN and b) CdS. The insets show Raman spectra taken at room temperature for comparison

Conclusions. We were able to detect unambiguously HR scattering in wurtzite GaN and CdS. The lack of the TO and silent modes in the HR spectra point to the weak contribution of the deformation potential to the scattering mechanism. The observed LO modes indicate, that HR scattering on GaN and CdS is dominated by Fröhlich interaction.

References

[1] V.N. Denisov, B.N. Mavrin, and V.B. Podobedov, Phys. Rep. **151**, 1 (1987).
[2] L.E. Zubkova, K.K. Ondriash, Yu.N. Polivanov, and K.A. Prokhorov, JETP Lett. **57**, 348 (1993).
[3] N. Bloembergen, Nonlinear Optics, W.A. Benjamin, Inc., New York 1977.