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RAMAN ANALYSIS OF $Zn_{1-x}Mg_xSe$ LAYERS GROWN ON GaAs AND ZnTe SUBSTRATES*

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A study of the Raman scattering in $Zn_{1-x}Mg_xSe$ ($0 \leq x \leq 0.4$) epilayers grown by molecular beam epitaxy on (100) GaAs and (111)_{Zn} ZnTe substrates has been performed. Two kinds of longitudinal optical phonon modes (LO_{Zn-Se} and LO_{Mg-Se}) were observed under excitation of the Ar⁺ and Kr⁺ laser lines at room temperature, whose frequencies and intensities depend characteristically on the Mg content.

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1. Introduction

Heterostructures based on a wide gap ternary II-VI alloy have attracted increasing interest in science during last years, mainly due to their suitability for the preparation of lasers emitting at short wavelengths of the visible and UV spectral range. Raman spectroscopy is often applied for the characterization of these epitaxial layers [1-3] because the frequency position and line shapes of LO and TO modes depend on the composition, the crystal quality, and strain in the layers [3]. In such case Raman spectroscopy may be employed to investigate the $Zn_{1-x}Mg_xSe$ layers properties by the specific optical as well as acoustical lattice vibrational modes.

In this paper, we present Raman scattering measurements of $Zn_{1-x}Mg_xSe$ layers with different Mg composition ($0 \leq x \leq 0.4$) grown on GaAs and ZnTe substrates.

2. Experimental

The $Zn_{1-x}Mg_xSe$ epilayers have been grown on semi-insulating (001) GaAs and (111)_{Zn} ZnTe substrates with the Mg contents from 0 to 40% in a MBE system (home made). The growth conditions were described in detail elsewhere [4, 5].

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The layer thicknesses varied from 0.3 μm to 1.2 μm . A mixed epilayer composition was determined by X-ray diffraction methods. All $\text{Zn}_{1-x}\text{Mg}_x\text{Se}/\text{GaAs}$ and $\text{Zn}_{1-x}\text{Mg}_x\text{Se}/\text{ZnTe}$ structures studied had [100] and [111] growth direction, respectively.

Raman spectra were taken at room temperature in backscattering geometry at several exciting energies. Spectra were recorded using a double monochromator equipped with holographic grating and a S20 photomultiplier or CCD detecting system. For the excitation Ar^+ laser lines (476.5 nm, 488 nm and 514.5 nm) and Kr^+ lines (530.9 nm and 647.1 nm) were applied. The powers of exciting lines were varied over more than two orders of magnitude.

3. Discussion

The lattice vibrational frequencies in the optical phonon region of the substrate (GaAs or ZnTe) and the epitaxial $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ ($0 \leq x \leq 0.4$) layers are located between 200 and 350 cm^{-1} . Figure 1 shows a typical example of Raman spectra for $\text{Zn}_{0.95}\text{Mg}_{0.05}\text{Se}$ layer grown on semi-insulating (001) GaAs substrate taken with different powers of the exciting laser line ($\lambda_{\text{exc}} = 488 \text{ nm}$). At the low power (about 2 mW) only two distinct peaks are visible which correspond to scattering by the longitudinal optical phonons of binary ZnSe ($\text{LO}_{\text{ZnSe}} - 253 \text{ cm}^{-1}$) and GaAs ($\text{LO}_{\text{GaAs}} - 292 \text{ cm}^{-1}$) [2, 3, 6]. The intensity of LO_{ZnSe} mode increases proportionally to the power of the incident light. Spectra were normalised to the height of the LO_{ZnSe} peak. As we can see from Fig. 1 the intensity of the LO_{GaAs} peak decreases and broadens and is partly screened at the high incident power. Simultaneously a new peak appears between the LO_{ZnSe} and LO_{GaAs} peaks at

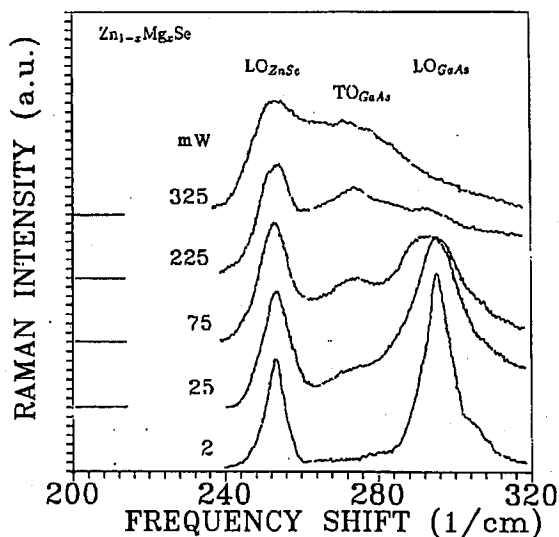


Fig. 1. Raman spectra of $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ layer ($x = 0.05$) grown on GaAs taken with various laser powers. The spectra are normalised to the LO_{ZnSe} intensities.

the position of the TO_{GaAs} phonon at 272 cm^{-1} . Lorentzian oscillators were used to fit the measured curve features leading to a good match between the fit and experimental data.

The appearance of the scattering intensity near the TO_{GaAs} phonon combined with the disappearance of the LO_{GaAs} phonon implies that the creation of photo-excited carriers in the semi-insulating GaAs substrate is responsible for the observed changes and coupled photo-excited plasmon-LO-phonon modes are formed [2, 3, 6, 7]. This coupling occurs due to the macroscopic electric field of the LO phonon and the collective oscillation of free carriers.

Figure 2 shows a typical Raman spectrum measured at room temperature of the $Zn_{1-x}Mg_xSe$ ($x = 0.18$) layer grown on GaAs substrate. Besides the strong contribution of the symmetry allowed LO_{GaAs} phonon at 294 cm^{-1} and corresponding weak symmetry forbidden TO_{GaAs} at 270 cm^{-1} , there are additional features due to scattering by the LO phonon of binary ZnSe at 253 cm^{-1} as well as scattering by three vibrational modes of the ternary $Zn_{1-x}Mg_xSe$, namely the symmetry allowed LO_{Zn-Se} and LO_{Mg-Se} at 250.1 cm^{-1} and 288.4 cm^{-1} , together with the symmetry forbidden TO_{Zn-Se} vibration at 206.2 cm^{-1} . Due

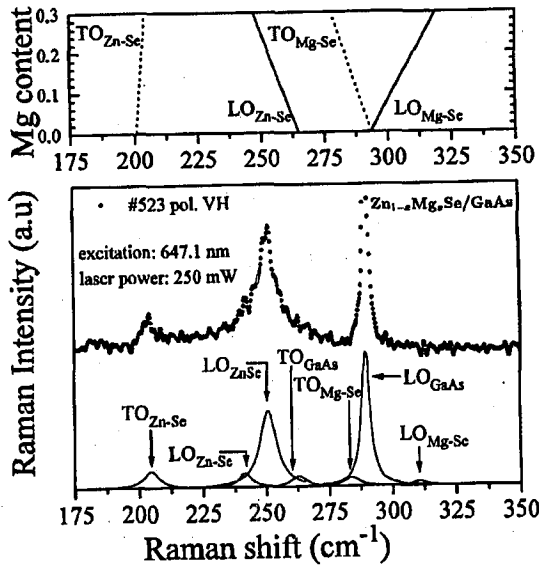


Fig. 2. Raman spectrum (upper curve) and fit (lower curve) of the $Zn_{1-x}Mg_xSe$ layer grown on GaAs ($x = 0.18$) measured at room temperature. The upper part schematically shows the frequency dependence of the vibrational modes on Mg content in the range $0 \leq x \leq 0.3$. LO_{Zn-Se} , LO_{Mg-Se} — the longitudinal optical modes of the ternary $Zn_{1-x}Mg_xSe$ compounds; LO_{ZnSe} , LO_{GaAs} — the longitudinal optical modes of the binary ZnSe and GaAs; TO_{Zn-Se} , TO_{Mg-Se} — the transverse optical modes of the ternary $Zn_{1-x}Mg_xSe$ compounds; LO_{ZnSe} , LO_{GaAs} — the transverse optical modes of the binary ZnSe and GaAs, respectively.

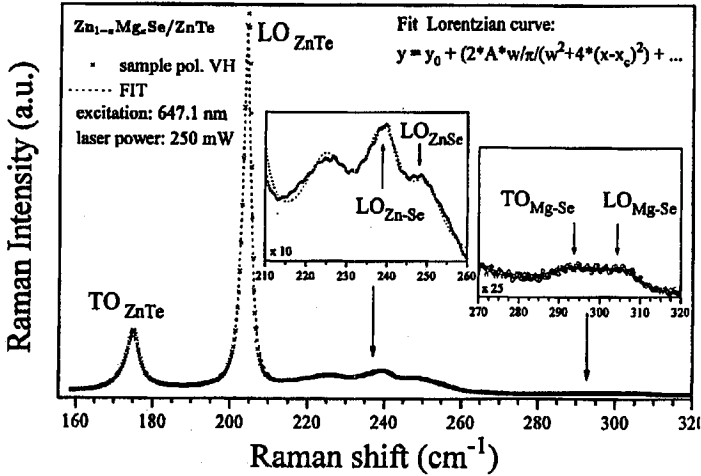


Fig. 3. Raman spectrum of $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ layer ($x = 0.18$) grown on $(111)_{\text{Zn}}$ ZnTe substrate measured at room temperature. List of symbols — see Fig. 2.

to the relative low Mg content, the $\text{LO}_{\text{Mg-Se}}$ peak is much weaker than that of $\text{LO}_{\text{Zn-Se}}$. The eigenfrequencies of these vibrational modes depend systematically on the magnesium composition as sketched in the upper part of Fig. 2 for Mg contents in the range $0 \leq x \leq 0.3$. It can be employed to determine the Mg content of the $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ layers from the Raman peak positions. The full width at half maximum (FWHM) of these $\text{LO}_{\text{Zn-Se}}$ and $\text{LO}_{\text{Mg-Se}}$ peaks broadens with increasing Mg content in the $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ layer but simultaneously the intensity ratio $I_{\text{LO}}/I_{\text{TO}}$ decreases.

For $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ layers grown on $(111)_{\text{Zn}}$ ZnTe substrate the analysis is somewhat complicated by the occurrence of three-mode phonon branches, the $\text{LO}_{\text{Zn-Se}}$, $\text{LO}_{\text{Mg-Se}}$ and $\text{TO}_{\text{Zn-Se}}$ vibration, whose frequencies depend characteristically on the Mg content and the stress (Fig. 3) [3, 8, 9]. The occurrence of both the $\text{LO}_{\text{Zn-Se}}$ and $\text{LO}_{\text{Mg-Se}}$ vibrational modes is typical for the two-mode behaviour of the $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ crystals. Since the energy E_g of $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ also depends on the Mg content in the layer [5, 8, 9], the choice of the excitation energy is of crucial importance in order to obtain the optimum signal intensity. Moreover, for strained layers grown at different temperatures possible additional frequency shifts have to be taken into account.

4. Conclusions

The multimode behaviour of the zone centre optical phonons in $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ layers with different Mg content ($0 \leq x \leq 0.4$) grown on GaAs and ZnTe substrates was investigated using Raman spectroscopy. The zone centre optical modes of these layers exhibit a two-mode behaviour with distinct $\text{LO}_{\text{Zn-Se}}$, $\text{LO}_{\text{Mg-Se}}$, $\text{TO}_{\text{Zn-Se}}$ and $\text{TO}_{\text{Zn-Mg}}$ modes across the entire composition range. It can be employed to determine the Mg content of the $\text{Zn}_{1-x}\text{Mg}_x\text{Se}$ layers from the Raman peak positions.

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