

535.375.54

Laser Raman Spectroscopy and Laser Resonance Raman Spectroscopy Investigations of Films $Ga_{1-x}Al_xP$ Grown by Liquid-Phase Epitaxy

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Abstract:

Raman-spectra of a lot of films of (001) and (111) $Ga_{1-x}Al_xP$ of various types grown by liquid phase epitaxy on substrate GaP have been studied, n- and p-type GaP being used as film substrates. This fact was embodied when fixing the Raman spectra. Wavelengths of argon laser 514.5, 488.0, 457.9 nm and helium-cadmium laser 441.6 nm were used to excite the spectra.

Raman-spectra study of $Ga_{1-x}Al_xP$ epitaxial films showed that mixed crystals $Ga_{1-x}Al_xP$ belong to the two-mode behavior systems. Raman scattering study with various wavelengths excitation enabled us to detect the weak resonance Raman-scattering in the proximity of fundamental E_0 direct transition and to estimate epitaxial films thicknesses.

Keywords: Laser Raman scattering, resonance Raman scattering, mixed semiconductor films.

1. Introduction

The mixed $Ga_{1-x}Al_xP$ crystal is an interesting material for semiconductor electronics though less studied. Particular interest in this material is caused by the fact that because of its wide band-gap this material could be used for making the light-emitting devices in visible region of spectrum. For example, on its ground the semiconductor hetero-structural Raman-lasers were made [1]. Optical properties of this compound have not been studied enough while the vibrational dynamics of crystal lattice is discussed in several works [2-9].

A tendency to intensive study of phonon tuning regularities of these mixed semiconductors is seen. Most of these works are performed by Raman scattering. There is a problem in the process of recording the Raman spectra of these films: the frequencies of LO_1 and TO_1 phonons of films $Ga_{1-x}Al_xP$ characterizing vibrations of sub-lattice GaP in films are very close to the frequencies of phonons LO_{GaP} and TO_{GaP} of substrate GaP. Because of this the overlap of the frequencies of phonons of substrate and films $Ga_{1-x}Al_xP$ often takes place when investigating thin films. As a result, it is difficult to separate the frequencies of phonons characterizing the films from that of substrate phonons. Therefore, the exact determination of frequencies of vibrations of sub-lattice GaP of films $Ga_{1-x}Al_xP$ faces certain problems. This fact is especially evident when studying thin films rich of phosphorous.

In [2-3] the infrared spectra of $Ga_{1-x}Al_xP$ are examined and system $Ga_{1-x}Al_xP$ is referred to the semiconductors of two-mode behavior. In other papers the Raman-spectra are studied. In [4] the authors examined films $Ga_{1-x}Al_xP$ synthesized by liquid-phase epitaxy (LPE) with volume excitation of radiation 632.8 nm of helium-neon laser. Therefore these spectra are free of background of substrate GaP. Hence, they determined the frequencies of LO and TO phonons precisely. It is to be regretted that only three compositions rich in phosphorous are studied. They detected an additional structure among the frequencies of phonons LO_1 and TO_1 , the characteristics of vibration of GaP-like sub-lattice. That spectral peak of low intensity almost does not change with the composition. On basis of investigation the authors ascribe system $Ga_{1-x}Al_xP$ to the crystals of

two-mode behavior. In [5] films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ synthesized on (001) GaP substrates from organometallic vapor phase epitaxy (OMVPE) have been studied by argon laser radiation (488.0 nm). In this case radiation 488.0 nm is not absorbed entirely by epitaxial film (especially in case of high composition of aluminum). Accordingly, in Raman-spectra of films the frequencies of phonons LO_{GaP} and TO_{GaP} , the characteristics of substrate GaP, dominate. Because of this the authors grew epitaxial films on substrate GaAs to fix the frequencies of phonons LO_1 and TO_1 , the characteristics of vibration of GaP-like sub-lattice. In [6] films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ grown on GaP (001) substrates by using gas-source molecular-beam epitaxy (GSMBE) have been studied by argon laser radiation 514.5 nm. In that case the effect of frequencies of phonons LO_{GaP} and TO_{GaP} , the characteristics of substrate GaP, upon the Raman-spectra of films is even greater. Therefore the authors of this paper investigated only LO_2 phonon branch, the characteristic of the vibration of AlP-like sub-lattice. In the papers under consideration the Raman-spectra have been recorded at room temperature. In paper [7] films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ on GaP (001) substrates were grown by using metallo-organic vapor phase deposition (MOCVD). The Raman-spectra were studying at 77⁰ K with the aid of krypton laser radiation 482.5 and 476.2 nm. In those experimental conditions too the role of substrate is great, especially for large compositions of aluminum. That fact is seen clearly from the spectra presented in the paper. In [8] the synthesized films on (001) GaP substrates by liquid-phase epitaxy were studied by micro Raman-spectroscopy. The excitation was carried out by He-Ne laser radiation 632.8 nm. The radiation impinged perpendicularly on the film edge so that not to fall within the substrate. The scattering configuration was different. The authors obtained a step-function distribution of film composition. In that paper too $\text{Ga}_{1-x}\text{Al}_x\text{P}$ was referred to the crystals of two-mode behavior.

In the all discussed papers without exception, only the epitaxial films of orientation (001) has been studied.

We have been studying this system by Raman-spectroscopy earlier [9]. But now we are publishing new results. In this case we pay special attention to the methods of separating in films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ the spectral band of LO_1 phonon, the characteristic of vibrations of sub-lattice GaP, from the spectral bands of the phonons of substrate GaP.

We regularly study with Raman-spectroscopy the semiconductors modified by ion implantation, especially the complex semiconductors synthesized by ion implantation [10-11]. Simultaneously, as the result of Raman scattering study the same semiconductors synthesized by ordinary chemical methods, for example, by liquid phase epitaxy, we receive the standards for qualitative identification of ion synthesized films and their quantitative estimation. As is known the ion synthesis is a non-equilibrium process. Therefore physical-chemical properties of new compounds synthesized with the aid of ion synthesis, partially optical characteristics, will be slightly different from the same compounds synthesized by usual chemical equilibrium reactions. Respectively, the Raman-spectra of the same compounds synthesized by these two different technologies cannot be identical. Hence, the concentration graphs received by Raman-spectroscopy of $\text{Ga}_{1-x}\text{Al}_x\text{P}$ synthesized by liquid-phase epitaxy cannot be the first-order standards for exact determining the compositions of the same compounds obtained by ion synthesis. These standards are valid only for evaluation of approximate values of unknown concentrations. At the same time by comparison of magnitudes of compositions of same compounds $\text{Ga}_{1-x}\text{Al}_x\text{P}$ received by ion synthesis, estimated by the mentioned standards with theoretical values, the role of defects induced during ion implantation synthesis may be cleared up.

The above-mentioned papers devoted to this aim.

2. Experiment

In the present work Raman scattering has been studied in a lot of n- and p-type $\text{Ga}_{1-x}\text{Al}_x\text{P}$ epitaxial films. We studied undoped films as well. The mentioned films were grown by liquid-phase epitaxy on GaP substrates with (001) and (111) orientations. The film thicknesses were in the range of 5-18 mmk. The epitaxial films were grown on substrates GaP of n- and p-type.

The Raman-spectra were recorded on the home-made laser Raman-spectrometer constructed on base of double monochromator DFS-24 with 800 mm focal length. Gratings with 1200 gr/mm worked in the first order of the spectrum and caused maximal concentration of light in the blue-green region of the spectrum. Argon laser emission of 514.5, 488.0, 457.9 nm and helium-cadmium laser emission of 441.6 nm wavelengths respectively served as excitation source. The Raman system experimental equipment is described in details in [12].

3. Results and discussions

Fig.1 shows the Raman-spectra of (111) GaP crystal with excitation 488.0 nm. It is seen in Fig. 1, that $LO_{GaP}=402\text{ cm}^{-1}$ and $TO_{GaP}=363\text{ cm}^{-1}$. Fig. 2 shows the Raman-spectra of mixed

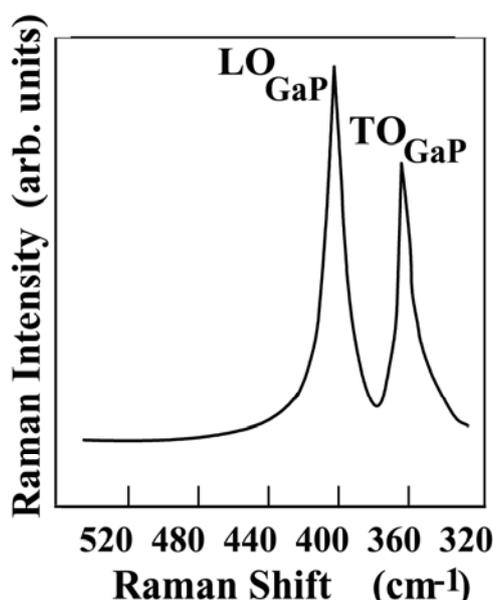


Fig. 1 Raman-spectrum of (111) GaP, $\lambda_L=488.0\text{ nm}$

$Ga_{1-x}Al_xP$ crystals of different compositions, recorded by 488.0 nm wavelength excitation. The film orientation in that case is (001). As it is clear from the picture, the peaks corresponding to LO_{GaP} and TO_{GaP} phonons at 402 and 363 cm^{-1} frequencies typical for GaP substrate are observed constantly in the Raman spectra of all three compositions. These peaks are observed always when the exciting wavelength is not absorbed completely by film $Ga_{1-x}Al_xP$ and because of this the signal excitation from the substrate takes place. Besides it should be noted that according to the selection rules the peak corresponding to TO_{GaP} phonon must not be observed from GaP surface of (001) orientation. That is, one could assume that at film growth on the substrate, the substrate surface orientation change takes place, which may be caused by some strain. It is seen from Fig. 1 and Fig. 2, that the main characteristics of mixed semiconductor films $Ga_{1-x}Al_xP$ with (001) orientation are two new peaks in Raman spectrum formed at a frequency distinguished from that of the peak corresponding to LO_{GaP} phonon characterizing (001) GaP substrate. One of these peaks corresponds to LO_2 phonon vibration of AlP sublattice, while the other corresponds to LO_1 phonon vibration of GaP sublattice. The frequencies corresponding to the two peaks are changing by a certain law depending on compositions of mixed crystals. When studying many films it turned out that the Raman-spectra of some (001) $Ga_{1-x}Al_xP$ films show a forbidden peak corresponding to TO_2 phonon, which perhaps indicates

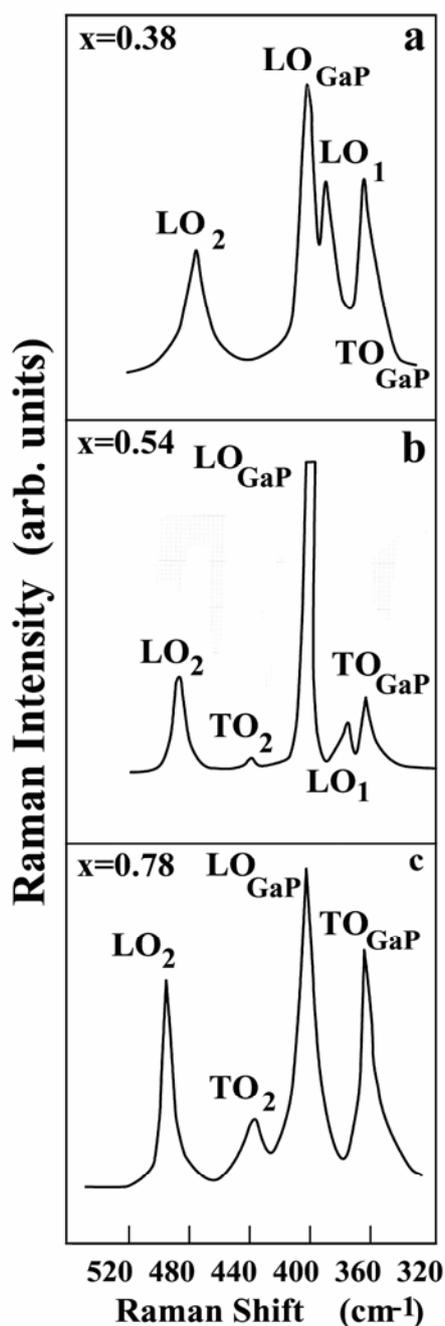


Fig. 2 Raman-spectra of mixed $\text{Ga}_{1-x}\text{Al}_x\text{P}$ crystals of different compositions; orientation (001); $\lambda_L = 488.0$ nm; a – $x=0.38$; b – $x=0.54$; c – $x=0.78$

also at the film surface deviation from (001) orientation. The phonons LO_2 and TO_2 characterize the phonon vibration of AlP sublattice in mixed $\text{Ga}_{1-x}\text{Al}_x\text{P}$ films. Thus with the aid of Raman-spectra it is possible to control technological processes of growth and discuss both film and substrate quality.

As Fig. 1 and Fig. 2 show, for the whole composition of films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ there are no characteristic frequencies of any substrate phonon overlapping it near the frequencies of phonons LO_2 and TO_2 . Thus, exact fixing of the frequencies of phonons LO_2 and TO_2 for all compositions of epitaxial films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ in no problem. Therefore, the graph of the phonon branch of sub-lattice AlP is a good calibration curve to estimate unknown compositions (for example, for films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ synthesized by ion implantation). Fig. 1 and Fig. 2 also show that there is the counterposition when fixing LO_1 phonons of films. In that case phonon LO_1 characterizing the film

is in the small interval of frequencies of two intense phonons LO_{GaP} and TO_{GaP} of substrate GaP. Therefore it is difficult to fix a well-defined spectral peak corresponding to phonon LO_1 of the film. Besides, for some compositions, for instance, for high content of aluminum, phonon LO_1 is not fixed at all (Fig. 2c).

Two methods were used to solve the problem. The first method consists in selecting the exciting wavelength, for which the optical distance αd is less or equal to the film thickness, d . Here α is the absorption coefficient of the film. At least it is possible the film thickness to be a bit less than αd . These conditions are kept approximately when emission 441.6 nm of helium-cadmium laser serves as exciting source. Fig. 3 shows the Raman-spectra of $Ga_{1-x}Al_xP$ of various composition with excitation. 441.6 nm. The spectra from Fig. 3 show that the substrate effect on the Raman-spectra recorded in such experimental conditions is either eliminated at all or is negligible. Hence, the peaks corresponding to phonons LO_{GaP} and TO_{GaP} are either not observed or are small.

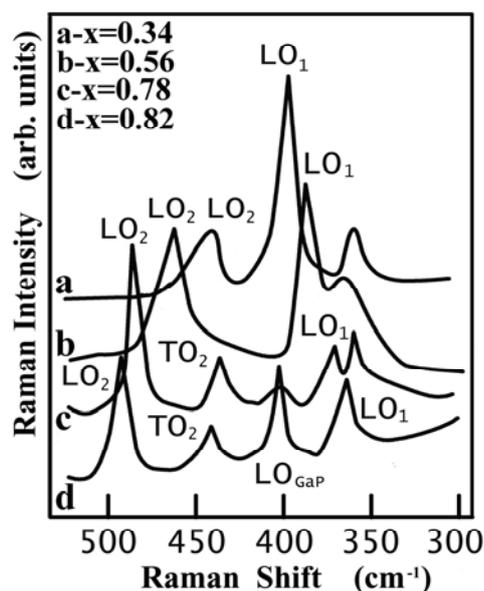


Fig.3 Raman-spectra of mixed $Ga_{1-x}Al_xP$ crystals of different compositions; orientation (001); $\lambda_L = 441.6$ nm; a – $x=0.34$; b – $x=0.56$; c – $x=0.78$; d – $x=0.82$

By the second method we tried to study films $Ga_{1-x}Al_xP$ grown on n-GaP and not on p-GaP. Here the idea is to decrease to minimum the effect of plasmon-LO phonon interaction of substrate GaP. As is known from [13-14], and we too have shown in [15], that in n-GaP the half-width of spectral band corresponding to the effect of plasmon-LO phonon interaction is far less than the half-width of the same spectral band of p-GaP. In this case it is not expectable that the long-wave wing of LO_{GaP} phonon of substrate n-GaP, more exactly, that of the spectral band of plasmon-phonon interaction, overlaps the low intense spectral band of LO_1 phonon of sub-lattice GaP of film $Ga_xAl_{1-x}P$; especially, for those compositions rich in phosphorous, the LO_1 phonon frequencies of which are shifted a little from frequencies LO_{GaP} . Our Raman-spectra proved that these two methods required the proper and appropriate use. It turned out that recording the spectra by the second method is especially important when very thin films are being studied by relatively long-wave excitation. For some compositions (relatively thick films) we received the Raman-spectra of high quality (with good resolution) even for 514.5 nm and 488.0 nm wavelengths (Fig. 4).

Fig. 5 shows the Raman-spectra for $Ga_{0.22}Al_{0.78}P$ composition when exciting by various wavelengths of laser. It is clear from the picture that when we use short waves to get a spectrum, the intensity of LO_{GaP} phonon of GaP substrate at 402 cm^{-1} diminishes gradually and when carrying out excitation with helium-cadmium laser radiation with 441.6 nm wavelength, the peak characterizing

LO_{GaP} phonon vanishes at all. Simultaneously the intensity of LO_2 phonon increases at 487 cm^{-1} which characterizes vibration of sublattice similar to AIP in $Ga_{0.22}Al_{0.78}P$ system.

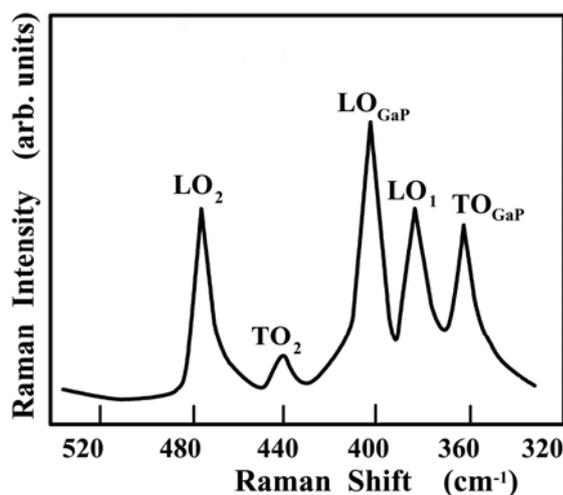


Fig. 4 Raman-spectrum of $Ga_{0.46}Al_{0.54}P$; orientation (001); $\lambda_L = 488.0\text{ nm}$

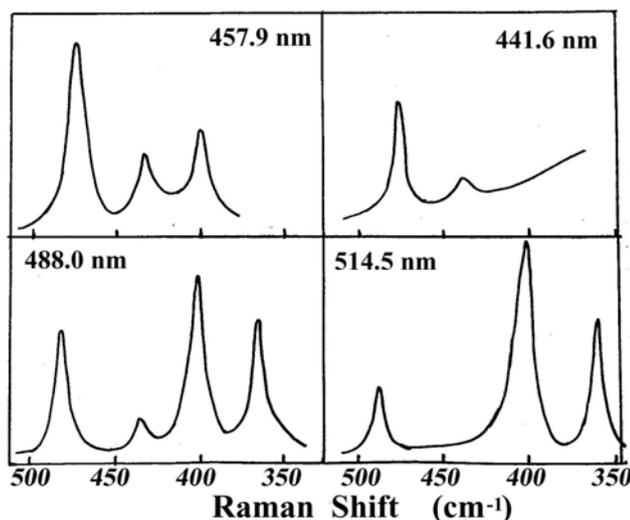


Fig. 5 Raman-spectra of $Ga_{0.3}Al_{0.7}P$ excited by various wavelengths of laser

Fig. 6 shows two Raman spectra of epitaxial film $Ga_{0.65}Al_{0.35}P$ we have recorded. The only difference between the two spectra is that the spectrum in Fig. 6a gives the Raman scattering for $Ga_{0.65}Al_{0.35}P$ grown on substrate n-GaP and the spectrum in Fig. 6b gives the Raman-spectra for the same composition grown on substrate p-GaP. The comparison of the two spectra shows clearly that it is advantageous to study Raman-spectra of system $Ga_{1-x}Al_xP$ with epitaxy this mixed semiconductor on substrate n-GaP. Fig. 6a shows clearly LO_1 phonon of film $Ga_{0.65}Al_{0.35}P$ which is separated sharply from the narrow band of plasmon- LO_{GaP} phonon interaction of substrate n-GaP. The same cannot be said on the Raman-spectra in Fig 6b. In this case it is seen distinctly that the long-wave wing of wider spectral band of plasmon- LO_{GaP} phonon interaction overlaps the most part of LO_1 phonon band of film $Ga_{0.65}Al_{0.35}P$. Therefore in this case the exact evaluation of the frequency of phonon LO_1 is impossible.

As mentioned above, for some “convenient” compositions of thicker epitaxial films we have recorded the Raman-spectra of good quality with wholly satisfactory resolution and 514.5 nm and 488.0 nm excitations. In this case when the distance of penetration of laser beam in the

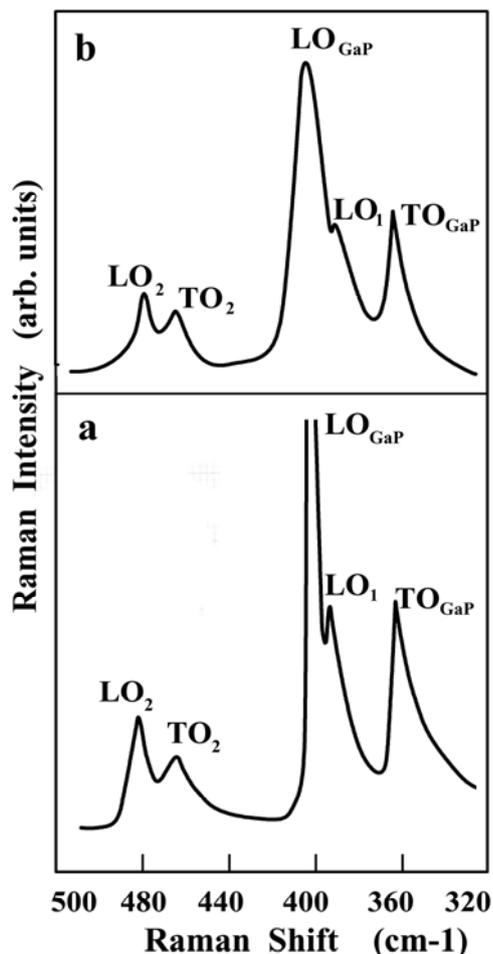


Fig. 6 Raman spectra of epitaxial film $\text{Ga}_{0.3}\text{Al}_{0.7}\text{P}$; orientation (001); $\lambda_L = 488.0$ nm; a - on substrate n-GaP; b - on substrate p-GaP

substrate is big enough, nevertheless, we received the band of LO_1 phonon of film $\text{Ga}_{0.46}\text{Al}_{0.54}\text{P}$ sharply separated from the substrate influence. Fixing such good-quality spectra is especially easy in case of thick films. Here too is necessary to keep the proper ratio between the excitation wavelength and αd magnitude. This case is given in Fig. 4.

Fig. 7 shows the Raman-spectrum of epitaxial film $\text{Ga}_{0.4}\text{Al}_{0.6}\text{P}$ of orientation (111) we have recorded. The excitation was carried out by 488.0 nm emission of argon laser. It is seen from the comparison of this spectrum with the spectra in Fig. 2 that the Raman-spectra of (111) films fix distinctly TO_2 phonon corresponding to the vibration of the AlP-like sub-lattice. According to the selection rules, at this configuration both LO_2 and TO_2 phonons are seen in the Raman-spectra. In this case TO_2 phonon is far intensive than in case of (001) film. The ratios of intensities LO_2/TO_2 also differ. Fig. 7 also shows that the very intense TO_{GaP} phonon of the substrate almost entirely covers LO_1 and TO_1 phonons corresponding to the vibration of the GaP-like sub-lattice. In this case these phonons become apparent as small structures. In papers of other authors we have cited, the Raman-spectra of epitaxial (111) films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ are not studied.

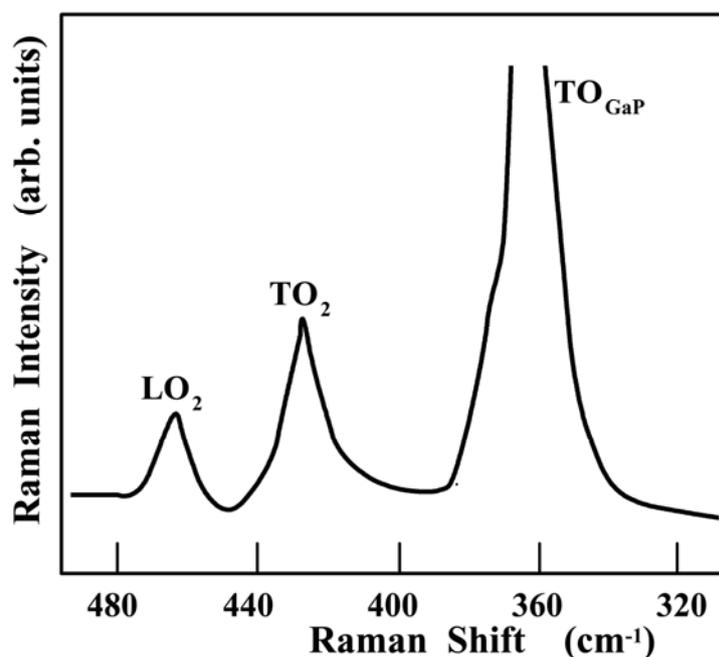


Fig. 7 Raman-spectrum of epitaxial film $\text{Ga}_{0.4}\text{Al}_{0.6}\text{P}$ of orientation (111); $\lambda_L = 488.0 \text{ nm}$

Thus, by studying the Raman-spectra of epitaxial (001) and (111) films of $\text{Ga}_{1-x}\text{Al}_x\text{P}$ we have fixed clearly the frequencies of LO_2 and TO_2 phonons corresponding to the vibration of the AIP-like sub-lattice and LO_1 phonon corresponding to the vibration of the GaP-like sub-lattice. These frequencies are given in Table 1.

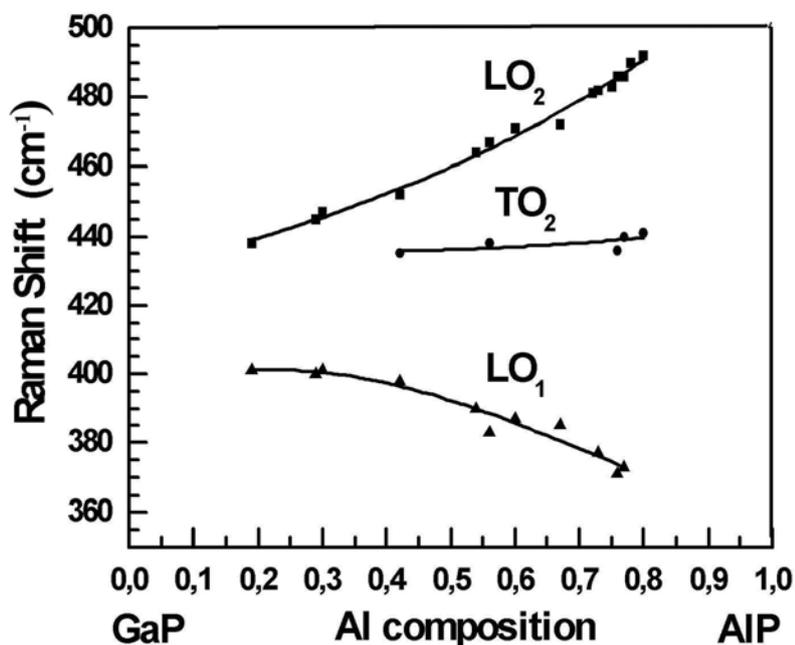


Fig. 8 Dependence of LO_1 , LO_2 , TO_2 , phonons frequencies upon the concentration x of mixed crystals $\text{Ga}_x\text{Al}_{1-x}\text{P}$

Fig. 8 shows the dependence of frequencies of LO_1 , LO_2 , TO_2 , phonons upon the concentration x of mixed crystals. This dependence indicates that the system $\text{Ga}_{1-x}\text{Al}_x\text{P}$ belongs to the two-mode behavior crystals.

We were the first to have studied the resonance Raman-scattering (RRS) for one of compositions of system $\text{Ga}_x\text{Al}_{1-x}\text{P}$, partially for $\text{Ga}_{0.22}\text{Al}_{0.78}\text{P}$. In the limited interval of energies of discrete exciting quanta of argon and helium-cadmium lasers has been investigated the RRS. The

resonance for LO₂ phonon of film Ga_{0.22}Al_{0.78}P has been studied. The frequency of this phonon is equal to 487 cm⁻¹. These results are given in Fig. 9.

Fig. 9 shows for Ga_{0.22}Al_{0.78}P the dependence of ratio of LO₂ phonons intensities at 487 cm⁻¹ and LO_{GaP} phonons intensities at 402 cm⁻¹ frequencies respectively upon the energy of exciting photons.

The direct band-gap E₀ of system Ga_{0.22}Al_{0.78}P is 3.5 eV [16]. Fig. 9 shows that when the exciting quantum energy approaches to the amount of direct band of Ga_{0.22}Al_{0.78}P, the increase of LO₁ phonon intensity is observed; the RRS takes place. The mentioned does not give completely the picture of resonance Raman-scattering because exciting quanta energies are sufficiently far from direct band width. We shall have the complete picture if we are able to use shorter wavelengths of the laser for excitation. Besides it would be more correct if we take the ratio of intensity of LO₄₈₇ to phonon vibration intensity of standard crystal CaF₂. The fact that we study the system-GaP substrate with Ga_{1-x}Al_xP film-by various wavelength of laser emission, enables us to estimate rough thicknesses of epitaxial layers by Raman-spectroscopy.

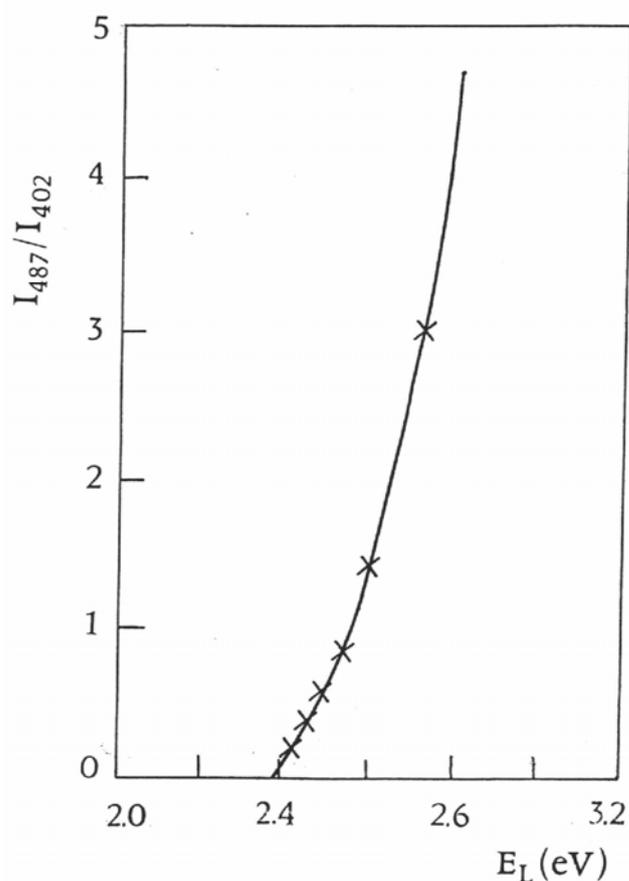


Fig. 9 RRS for LO₂ phonon of film Ga_{0.3}Al_{0.7}P

According to [17] the Raman-scattering intensity

$$I = (I_0 R / 2\alpha) (1 - \exp(-2\alpha d)) \quad (1)$$

where I_0 is the intensity of impinged radiation, R_0 – scattering cross-section, α —absorption coefficient, d –film thickness .

If we suppose film Ga_{1-x}Al_xP grown on GaP substrate with steep front, then it is possible to assume, that the scattering intensity equals to the sum of intensities of substrate and film Raman scattering.

$$I_{\text{Total}}=I_f+I_{\text{subst}}\exp(-2\alpha_f d) \quad (2)$$

$$I_f=(I_0 R_f/2\alpha_f)[1-\exp(-2\alpha_f d)] \quad (3)$$

$$I_{\text{subst.}}=(I_0 R_{\text{subst.}}/2\alpha_{\text{subst.}})\exp(-2\alpha_f d) \quad (4)$$

If we take the ratio of intensities, it will be possible to calculate the film thickness.

$$I_{\text{subst.}}/I_f=(R_{\text{subst.}}\alpha_f/R_f\alpha_{\text{subst.}})\times[\exp(-2\alpha_f d)]/[1-\exp(-2\alpha_f d)] \quad (5)$$

By the above mentioned method we estimated the thickness of film of composition $\text{Ga}_{0.22}\text{Al}_{0.78}\text{P}$ and got $d=12.5\text{mmk}$. The same film thickness measured by the ordinary microscopic method gives $d=12\text{mmk}$.

4. Conclusion

Thus, we have studied by Raman-spectroscopy a large amount of epitaxial films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ with excitation of various wavelengths of laser. To fix good-quality Raman-spectra, shortwavelength 441.6 nm excitation of He-Cd laser has been used. As a result, in most cases the influence of parasitic background of the substrate was entirely eliminated. It has been shown that to receive the good-quality Raman spectra of epitaxial films $\text{Ga}_x\text{Al}_{1-x}\text{P}$, it is far more advantageous to investigate films grown on the substrate n-GaP and not on that of p-GaP. Generally, to get the good-quality Raman-spectra it is necessary to select the corresponding optimal ratio between exciting wavelengths and magnitudes αd of films. By studying the Raman-spectra of epitaxial films $\text{Ga}_{1-x}\text{Al}_x\text{P}$ of two different orientations (001) and (111), both LO_2 and TO_2 phonons have been fixed clearly. We have established that the mentioned mixed semiconductor belongs to the two mode behavior crystals. We have found out in this system the resonance Raman scattering in proximity of E_0 direct band and estimated epitaxial film thicknesses with the Raman-spectra.

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Article received: 2009-12-23