

# Raman Spectroscopy—A Versatile Tool for Characterization of Thin Films and Heterostructures of GaAs and $\text{Al}_x\text{Ga}_{1-x}\text{As}$

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**Abstract.** It is shown that Raman spectroscopy can provide useful information on characteristic properties of thin crystalline films of compound semiconductors. Crystal orientation, carrier concentration, scattering times of charge carriers, composition of mixed crystals and depth profiles can be studied in thin layers and heterostructures of GaAs and  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ . The advantages and disadvantages of Raman scattering compared to conventional characterization methods are discussed.

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Thin crystalline films and heterostructures of compound semiconductors become more and more important for devices and device technology. A precise control over thickness and doping profiles of multilayer systems is necessary to achieve, for example, high quality microwave diodes or optoelectronic devices. Liquid phase epitaxy (LPE) [1] and molecular beam epitaxy (MBE) [2–4] are two methods which are used to grow such heterostructures.

Especially MBE has been developed during the past ten years in a way that nowadays one is able to control the uniformity, thickness, doping profiles and composition of thin crystalline layers ( $\lesssim 100 \text{ \AA}$ ) over the whole surface of a given substrate. The relatively low growth temperature, the possibility of abruptly changing the composition, and the low growth rate make MBE a versatile technique for growing heterostructures and superlattices with alternating layers of, for example, GaAs and  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ . The thickness of the individual layers can be kept as thin as a few monolayers [5].

In order to learn something about the growth mechanisms and the characteristic properties of such thin crystalline films, many different characterization methods are employed. The MBE provides the possibility

of in situ characterization of the growing and grown layers by means of, for example, reflection high energy electron diffraction (RHEED), secondary ion mass spectroscopy (SIMS) and Auger electron spectroscopy (AES) [4]<sup>1</sup>. Many of the characteristic properties and the quality of the layer structures have to be checked and studied after growth outside the growing apparatus. Transport and magnetotransport measurements lead to information on carrier concentration, mobility and sometimes also effective masses of the carriers. But often it is not possible to separate the properties of individual layers from each other and from the substrate. Completely different information can be drawn from x-ray diffraction and electron microscopy work. Here one learns something about crystal structure, orientation, and quality. Optical methods like reflection, transmission, and luminescence experiments are employed to characterize single and multiple layers, to learn about recombination mechanisms and the role of interfaces on these mechanisms. In the case of superlattices, most of these measurements show characteristic features because of zone folding effects [5–7].

In this paper we want to show that Raman scattering experiments give additional, new and sometimes more detailed information on the properties and qualities of thin crystalline films and multilayers of GaAs and

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<sup>1</sup> For comprehensive review of the methods see [22].

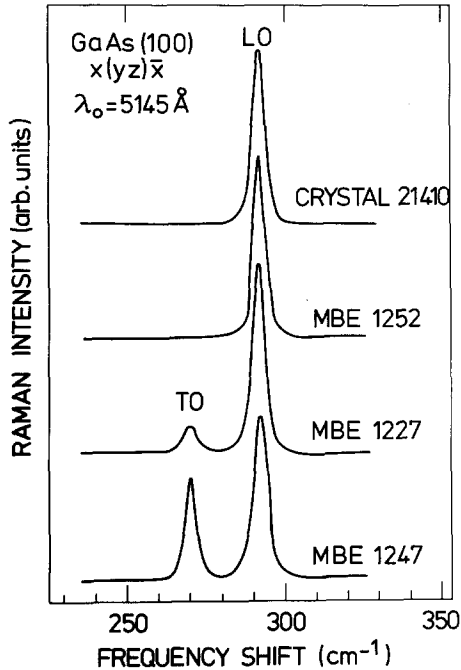


Fig. 1. Examples of Raman spectra obtained in backscattering geometry from a polished and etched (100) surface of GaAs and thin films of GaAs grown with MBE on (100) substrates. In this geometry only scattering by LO phonons is allowed. Some of the MBE grown samples show also a strong TO mode

$\text{Al}_x\text{Ga}_{1-x}\text{As}$ . There are two main reasons why Raman scattering is a useful tool in sample characterization and that one can learn something, where other methods fail often. First, the lattice vibrational spectra of different layers are observed as a superposition of the spectra of each layer. So it is possible to study individual layers in a nondestructive way by using different laser excitation lines with different penetration depths. The second important point is that lattice vibrations are very sensitive to the nearest neighbourhood and therefore probe the crystal structure and quality on an extremely small scale, a scale which is of the order of the lattice spacing.

In the following sections we describe various properties of thin layers which can be studied by Raman spectroscopy. Examples are shown for GaAs and  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  crystals. At the beginning we want to mention a few experimental details.

## 1. Experimental Aspects

Because all of our Raman scattering experiments are performed with incident laser light in the frequency range where GaAs is opaque, we use backscattering geometry. The wavevector of the excited phonon is therefore perpendicular to the sample surface. The laser beam is focussed on to the sample with a

cylindrical lens. The sample itself is mounted either at room temperature or on a cold finger of a liquid nitrogen dewar. The inelastically backscattered light is collected to the entrance slit of the spectrometer (both J rrel Ash and Spex double monochromators are used). The collected light is analyzed and stored in a multichannel analyzer using an appropriate photomultiplier tube and conventional pulse counting electronics for signal detection. Both  $\text{Ar}^+$  and  $\text{Kr}^+$  ion lasers are used as light sources.

The studied samples are single crystals of GaAs with various doping concentrations (Te, Sn, Se) and thin crystalline films and multilayers of GaAs and  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  grown with LPE and MBE. The surfaces of the bulk single crystals are polished and etched with an aqueous solution of NaCl. The MBE and LPE samples are measured as grown.

In order to get information from different depths one varies the laser frequency. The penetration depth into GaAs changes from about  $1\ \mu\text{m}$  at  $\lambda_0 = 8000\ \text{ }$  to  $\sim 150\ \text{ }$  at  $\lambda_0 = 4000\ \text{ }$  [8]. In the mixed crystal  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  the direct bandgap increases with increasing Al content. At  $x \sim 0.40$  the conduction bands at the  $\Gamma$  and  $X$  point of the Brillouin zone cross, we have a semiconductor with an indirect bandgap. Because of the shift of the bandgap to higher energies, the penetration depth into  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  gets larger. For example at  $x \sim 0.40$  the bandgap is already 2.0 eV, which means the crystal is transparent for the red  $\text{Kr}^+$  ion laser lines while in the blue the penetration depth is still very short. Thus also in the mixed crystal system a depth profiling using different laser lines is possible.

## 2. Possibilities, Results, and Discussion

### 2.1. Crystal Orientation

From the selection rules we learn that under certain scattering configurations not all lattice vibrations can be observed in a Raman scattering experiment. Let us consider only first-order scattering (phonons at the  $\Gamma$ -point). Following Loudon's description of the Raman tensors [9] we find for crystals with diamond structure that for backscattering from a (100) surface, only the LO phonon can be observed. From (110) surfaces only TO phonons are allowed and for (111) surfaces both TO and LO lattice vibrations should be observable.

Figure 1 shows on the top a Raman spectrum in true backscattering geometry from a (100) surface of a slightly doped  $n$ -GaAs crystal ( $n = 1.8 \times 10^{16}\ \text{cm}^{-3}$ ). The polarization of the incident light is along (010) and the scattered light along (001). We write this scattering configuration in the following way:  $x(yz)\bar{x}$ . As expected, only the LO phonon mode is observed in the Raman spectrum.

We have also studied thin films of GaAs grown with MBE and LPE on GaAs substrates with orientation  $\langle 100 \rangle$  perpendicular to the surface. In Fig. 1 we show typical examples of observed Raman spectra of such layers. Some of the samples show a strong TO mode. In very thin layers it is difficult to learn something about the crystal structure from x-ray work because of the large information depth. Sometimes electron and x-ray diffraction under very small angle of incidence can provide insight into the crystal orientation and surface smoothness.

On the other hand, one can easily see deviations from the Raman scattering (Fig. 1) selection rules.

The strong TO mode which is observed in some MBE films grown on (100) GaAs substrates demonstrates that the orientation of at least parts of the films are different from the expected (100) direction. Comparing different MBE samples and the way of their fabrication we realize that the TO mode always appears when the initial substrate surface seems to be slightly contaminated by carbon, even after chemical etching in H<sub>2</sub>O<sub>2</sub>/H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O. We have detected this contamination in situ by AES [4] prior to epitaxial growth.

It has already been pointed out previously [2] that a carbon impurity of  $>0.2$  monolayer of the initial surface does cause surface faceting and twinning in the growing film. We have checked this phenomenon on a large number of MBE samples by Raman scattering and we always find the same result: The appearance of the "forbidden" TO phonon line depends strongly on the carbon contamination of the initial surface. There is found no TO phonon only in the case when the residual carbon on the initial surface is less than 0.1 monolayer, which is the detection sensitivity of our AES measurements. The required purity of the substrate surface can be achieved [2] by either an extremely careful chemical treatment, which might vary from sample to sample, or in a more reproducible manner by an additional Ar<sup>+</sup> ion sputter cleaning and annealing prior to epitaxial growth.

We have strong evidence that the strength of the TO phonon line is directly correlated with the amount of carbon on the initial surface. Starting with a contamination  $>0.3$  monolayer carbon, a twinning of the growing GaAs film can be observed even in the RHEED pattern and the sample shows a strong "forbidden" TO phonon, with an intensity about 2/3 of that of the LO phonon (Fig. 1, spectrum at the bottom). When thin films ( $<200$  Å) grown on these contaminated substrate surfaces are examined by scanning electron microscopy, a three-dimensional nucleation yielding irregular three-dimensional centers and discrete three-dimensional crystallographically shaped growth centers are observed. For a carbon contamination  $<0.2$  monolayer there is no observable

effect in our RHEED experiments, but there is still a small "forbidden" TO line in the Raman spectrum (Fig. 1).

The nature of the misorientation is not yet clear. It is difficult to get information on this question from Raman spectroscopy. Therefore further x-ray and electron diffraction work is necessary to get insight into the problem what actually happens during the growth. Here we just want to show that Raman scattering experiments can be used to study the crystal orientation of thin films on a microscopic scale. The effect just described has been discovered only because we are using, besides other methods, also Raman spectroscopy to characterize the MBE and LPE grown samples.

## 2.2. Carrier Concentration and Scattering Time in *n*-GaAs

Several years ago Mooradian and coworkers [10] have shown that it is possible to observe also coupled LO phonon plasmon modes in *n*-GaAs using Raman spectroscopy. The frequencies of the coupled modes are essentially given by the zeros of the real part of the following sum: the longitudinal dielectric function of the electron gas  $\epsilon_e(q, \omega)$  plus the one of the lattice vibration. The early experiments have been performed with incident laser light  $\lambda_0 = 1.06$  μm. In this frequency region GaAs is transparent and the scattering wavevector  $k = 4\pi \text{Re}\eta/\lambda_0$  ( $\eta$  is the refractive index) involved in Raman scattering is very small. The observed frequencies of the coupled modes (labelled  $L_+$  and  $L_-$ ) agree well with the calculated roots of the equation  $\text{Re}\{\epsilon(q \rightarrow 0, \omega)\} = 0$ .

Recently it has been demonstrated that it is also possible to study the wavevector dependence of  $L_+$  and  $L_-$  using incident laser light in the visible ( $6764 \leq \lambda_0 \leq 4416$  Å) [11]. Using the Drude expression for the dielectric function of the electron system the frequency of the  $L_+$  mode can be written as [11]

$$\omega_+ = \left\{ \frac{\omega_{\text{LO}}^2 + \omega_p^2(q)}{2} + \left[ \frac{[\omega_{\text{LO}}^2 + \omega_p^2(q)]^2}{4} - \omega_p^2(q)\omega_{\text{TO}}^2 \right]^{1/2} \right\}^{1/2}, \quad (1)$$

where  $\omega_{\text{LO}}$  and  $\omega_{\text{TO}}$  are the frequencies of the LO and TO phonons at the  $\Gamma$ -point, respectively, and  $\omega_p(q)$  is the wavevector dependent plasma frequency [12]

$$\omega_p^2(q) = \omega_p^2 + 3/5(qv_F)^2 \quad (2)$$

with

$$\omega_p^2 = 4\pi n e^2 / \epsilon_\infty m^*; \quad (3)$$

$n$  is the carrier concentration,  $v_F$  the Fermi velocity, and  $m^*$  is the effective mass of the carriers.

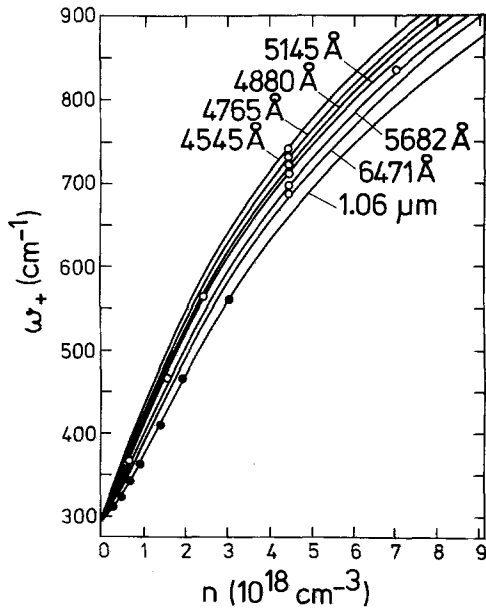


Fig. 2. The dependence of the frequency of the  $L_+$ -mode on the carrier concentration is shown for different laser lines. Experimentally obtained values of  $\omega_+$  (open circles) agree well with the theoretical curves. The closed circles are results taken from [10]

The position of the high-frequency coupled mode depends on the carrier concentration  $n$ . Therefore it can be used to determine  $n$  of highly doped crystals and thin layers. We have calculated  $\omega_+(n)$  for the strongest lines of  $\text{Ar}^+$  and  $\text{Kr}^+$  ion lasers and for the  $1.06\ \mu\text{m}$   $\text{Nd}^{3+}$  YAG laser line used in the work of [10]. This is necessary because the refractive index  $\eta$  of GaAs depends on the wavelength [8] so that one gets a variation of the scattering wavevector from  $0.3 \times 10^6\ \text{cm}^{-1}$  to  $1.3 \times 10^6\ \text{cm}^{-1}$  for the used laser lines ( $1.06\ \mu\text{m} \leq \lambda_0 \leq 4545\ \text{\AA}$ ). The results are shown in Fig. 2. One can clearly see the wavevector dependence of  $\omega_+$ . At fixed  $n$ ,  $\omega_+$  shifts to higher frequencies when  $\lambda_0$  is changed from red to blue. This wavevector dependence was discussed extensively in [11] and [13]. We have not yet mentioned that the conduction band of GaAs is nonparabolic and therefore the effective mass  $m^*$  depends on the carrier concentration. This has to be taken into account using a mass variation of  $m^* = 0.070m_0$  for  $n = 1 \times 10^{17}\ \text{cm}^{-3}$  to  $m^* = 0.096m_0$  for  $n = 1 \times 10^{19}\ \text{cm}^{-3}$  [14]. The other values used in the calculations are:  $\epsilon_\infty = 11.1$ ,  $\omega_{\text{LO}} = 295\ \text{cm}^{-1}$ ,  $\omega_{\text{TO}} = 270\ \text{cm}^{-1}$ . The  $\omega_+(n)$  curves shown in Fig. 2 can be used to determine the carrier concentration in the region  $n > 1 \times 10^{17}\ \text{cm}^{-3}$ .

So far we have discussed only the theoretical part without considering the experimental situation. At a real surface of  $n$ -GaAs there normally exists a depletion space charge region with a barrier height of  $\sim 1\ \text{eV}$ . The width of this carrier free layer varies from

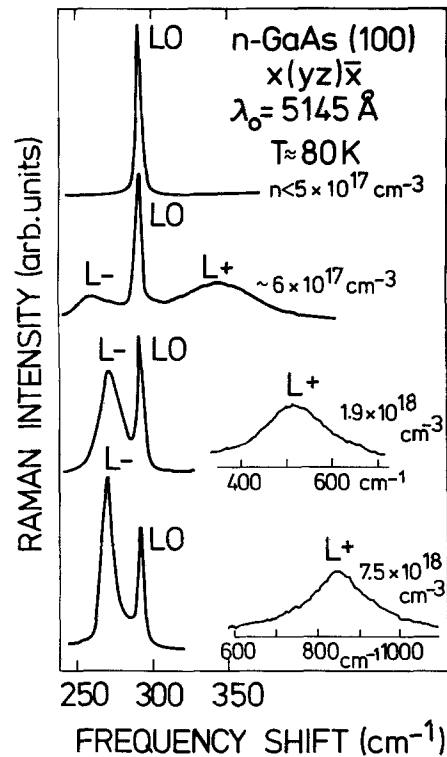


Fig. 3. Typical Raman spectra of thin MBE layers of  $n$ -GaAs with different donor concentrations. Both the LO phonon mode from the depletion layer and the coupled LO phonon-plasmon modes  $L_+$  and  $L_-$  are observed

$\sim 3000$  to  $\sim 150\ \text{\AA}$  with donor concentrations in the range  $2 \times 10^{17} \lesssim n \lesssim 5 \times 10^{18}\ \text{cm}^{-3}$ . On the other hand, using laser lines covering the range  $6471\text{--}4545\ \text{\AA}$ , the penetration depth into GaAs changes from  $\sim 3000$  to  $\sim 300\ \text{\AA}$ . This has been discussed already in the experimental part. Thus it is possible to observe both the coupled LO phonon-plasmon modes from the bulk and the unscreened LO phonons from the surface depletion region. The relative intensities can be used to study the surface band bending and the barrier heights of free and covered GaAs surfaces [15].

Here we concentrate on the signals from the bulk. In Fig. 3 some typical spectra are shown for different carrier concentrations. One can see that the strength of the bulk signal ( $L_+$  and  $L_-$ ) decreases compared to the surface signal (LO-phonon) with decreasing carrier concentration. This is due to the increased depletion width, while the penetration depth of  $\sim 1000\ \text{\AA}$  at  $\lambda_0 = 5145\ \text{\AA}$  remains unchanged. Therefore the observability of  $L_+$  and  $L_-$  is limited to the region  $n \gtrsim 5 \times 10^{17}\ \text{cm}^{-3}$ . In this region the frequency of the  $L_-$  mode depends only very weakly on the carrier concentration, contrary to the  $L_+$ -mode which shifts quite strongly to higher energies if  $n$  is increased.

In Fig. 2 the experimental results of  $\omega_+$  obtained with  $\lambda_0 = 5145 \text{ \AA}$  on samples with known carrier concentration are shown (open circles). The closed circles are results taken from [10] ( $\lambda_0 = 1.06 \mu\text{m}$ ). For a sample with  $n = 4.5 \times 10^{18} \text{ cm}^{-3}$  we also show the results obtained with other laser lines. The agreement between theory and experiment is very good. Therefore we believe that the measurement of the  $L_+$  mode gives a very accurate value of the carrier concentration of  $n$ -GaAs in the region  $n \gtrsim 5 \times 10^{17} \text{ cm}^{-3}$ .

If one has the possibility to use a Raman spectrometer which works with infrared frequencies, one can extend this region to  $n \gtrsim 1 \times 10^{16} \text{ cm}^{-3}$  by studying the frequency of the  $L_-$ -mode, which depends strongly on  $n$  in the range  $1 \times 10^{16} \lesssim n \lesssim 5 \times 10^{17} \text{ cm}^{-3}$ . Then the depletion layer is no problem, because GaAs is transparent in the infrared.  $\omega_-(n)$  can be taken, for example, from [10].

We have studied the  $\omega_+$  mode of various thin crystals of  $n$ -GaAs grown with MBE and LPE and could so determine the carrier concentration. The results agree well with those obtained from capacitance versus voltage and Hall effect measurements. The advantages of the Raman spectroscopy are that no contacts and no Schottky-barriers are necessary, that the substrate does not influence the results, and that the carrier concentration can be determined within very small areas (focus of the laser beam down to  $4 \mu\text{m}^2$ ).

If the samples are very thin, the penetration depth of the laser light can be large enough to reach the interface to the substrate. Depending on the substrate, the scattering volume with carrier density  $n$  gets smaller. In very thin samples the entire layer can be depleted. Such effects result in an increase of the intensity of the unscreened LO mode compared to the coupled modes. We have observed this behaviour on MBE layers of various thicknesses.

Apart from the position of the  $L_+$  mode one also gets useful information from the linewidth and the lineshape. At high carrier concentration ( $n > 10^{18} \text{ cm}^{-3}$ ) the  $L_+$  mode is plasmon-like and the damping should essentially be determined by the scattering time of the charge carriers. Using the electron damping  $1/\tau = e/m^* \mu$  deduced from the mobility  $\mu$ , we find on the other hand that the actually observed linewidth of  $L_+$  is always larger than expected [13]. This indicates that additional damping mechanisms play an important role. Such contributions to the broadening of the plasmon-like mode may arise from an increased scattering rate caused by the presence of the surface. Sometimes one also gets an inhomogeneous broadening because of a spatial variation of the donor distribution. When the penetration depth exceeds the depletion layer only very little, we observe an asymmetric lineshape of the  $L_+$  mode, because at the interface

between the bulk and the depletion region, one always has a gradient in the carrier concentration.

If we neglect inhomogeneous broadening, which is reasonable for symmetric lineshapes, we can define a scattering rate for the electrons observed in Raman scattering

$$\frac{1}{\tau} = \frac{1}{\tau_i} + \frac{1}{\tau_p} + \frac{1}{\tau_s}. \quad (4)$$

$1/\tau_i$  is the scattering rate by impurities,  $1/\tau_p$  by phonons, and  $1/\tau_s$  describes surface scattering. If one measures the bulk mobility, only impurities and phonons contribute to the scattering rate and at high donor concentration  $1/\tau_i$  dominates. Using, however, the width of the  $L_+$  mode to determine  $1/\tau$  or  $\mu$  we have to consider also surface scattering, because only electrons close to the surface depletion layer contribute to the Raman signal. The actual surface which one should consider here, is the interface between depletion layer and bulk.

Experimentally we find for  $n = 7 \times 10^{17} \text{ cm}^{-3}$  the scattering rate deduced from the  $L_+$  mode to be about two times larger than expected from the bulk mobility. For higher carrier concentrations the difference gets smaller and smaller. This can be explained qualitatively with surface scattering. First of all the impurity scattering rate increases with increasing donor concentration. Second, with higher carrier concentration, the depletion width gets smaller so that at a fixed laser frequency the region containing electrons which contribute to the signal gets larger. On the average the carriers are further away from the interface. Therefore surface scattering should be less important when  $n$  is very large.

Despite of these problems we believe that especially for high carrier concentrations ( $n > 10^{18} \text{ cm}^{-3}$ ) we can use the width of the  $L_+$  to estimate the scattering rate and the mobility of the charge carriers in new crystals or thin layers by comparing the results with the values of known samples.

### 2.3. Mixed Crystals

The frequencies of the LO and TO modes of the system Al<sub>x</sub>Ga<sub>1-x</sub>As have been studied previously by Ilegems and Pearson [16] from a Kramers-Kronig analysis of infrared reflectance spectra and by Tsu and coworkers [17] using Raman spectroscopy. In the entire alloy composition range, the spectra show essentially two distinct bands, one at frequencies close to the modes of pure GaAs and the other close to pure AlAs. In Fig. 4 we show the dependence of the vibrational modes on the molar fraction  $x$  of Al in GaAs [16, 17]. The frequency variation of these modes can be used to

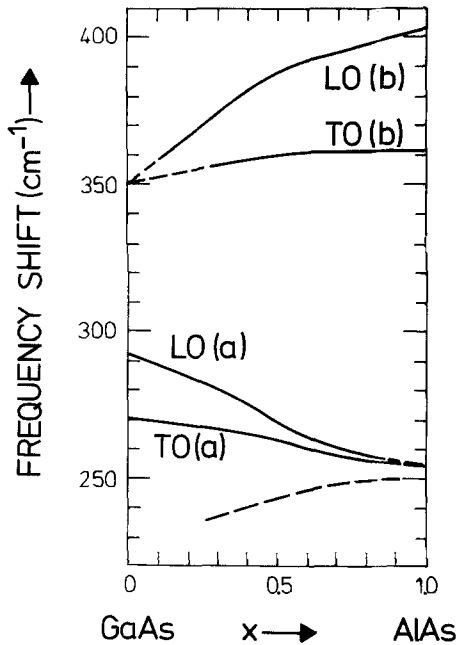


Fig. 4. The variation of the mode frequencies of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  with  $x$  is shown [16, 17]. This plot is used to determine  $x$  in unknown MBE and LPE layers of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$

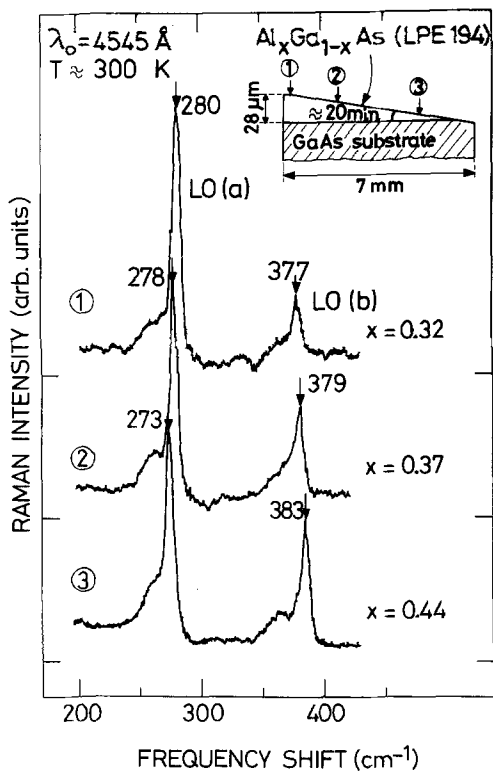


Fig. 5. Some Raman spectra of a  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  LPE sample which has been wedged in order to study the depth profile of the Al concentration. The sample cross section is shown schematically in the insert

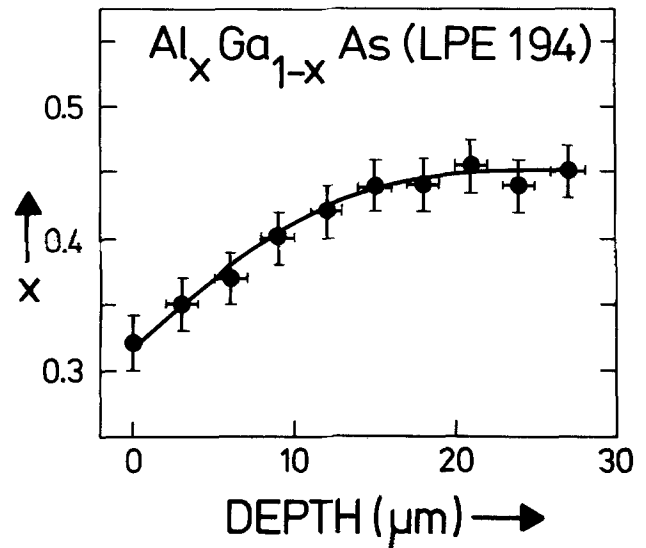


Fig. 6. Molar fraction of Al in the LPE sample described in Fig. 5 as a function of the distance from the surface

determine the value of  $x$  of unknown samples. We have used this method to characterize thin  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  layers grown by MBE and LPE.

As discussed before, a depth profiling can be obtained, using laser excitation lines with various penetration depths. For a gradually varying Al concentration, however, this method is not very accurate. One observes only asymmetric lines caused by the slowly changing Al content (inhomogeneous broadening). Therefore we use a different method to study the Al profile of LPE layers of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  on GaAs substrate. The studied LPE samples have a thickness of about  $30\ \mu\text{m}$ . We have polished and etched the samples under an angle of  $\sim 20^\circ$  tilted with respect to the surface parallel. With a sample length of  $0.7\ \text{cm}$  we get on one side the original sample surface while on the opposite side the LPE layer is polished away. Thus when moving the laser beam over the surface, it is possible to observe Raman spectra from different depths of the grown layer. The cross section of the sample is shown schematically in Fig. 5. The experiments are performed with  $\lambda_0 = 4545\ \text{\AA}$   $\text{Ar}^+$  laser line, a wavelength with a very short penetration depth. In Fig. 5 we also show some spectra from different parts of the surface. One clearly observes a shift of the mode frequencies which is caused by a variation of the Al concentration towards the surface. Together with the frequency shift of the Raman lines we notice also an increasing intensity of the band originating from the pure AlAs when we measure closer to the substrate. The intensity ratio of the two different bands might also be used to determine the Al concentration. In Fig. 6 we show the dependence of the Al content on the

distance from the surface. Raman scattering experiments are accurate to about  $\pm 2\%$  for determining the Al concentration in the region  $x=0.05$  to  $0.90$ .

This method of determination of the composition of mixed crystals can be extended to other systems. The mode frequencies of many III-V alloy systems have been studied during the past ten years both with Raman and infrared spectroscopy and the frequency shifts with composition can be found in the literature.

#### 2.4. Multilayer Structures

So far we have only discussed properties of single layers of GaAs or  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ . Now we want to show that Raman spectroscopy is also a useful tool to characterize multilayer systems. During the past few years some Raman studies of periodic systems or superlattices have been performed [18–21]. These experiments show interesting features due to zone folding effects in periodic structures. We concentrate on multilayer systems which have no special periodicity so that no zone folding occurs.

In Raman scattering experiments one observes the vibrational spectra of each individual layer. This is a great advantage compared to other optical characterization methods. In reflection or transmission experiments one always gets information averaged over the studied layers, for example, the refractive index or the absorption coefficient. Raman scattering can be used to study the composition of individual layers. From the dependence of signal strength of the different layers on the thickness and on the wavelength one can also obtain information on the absorption coefficient of the different layers.

In Fig. 7 we show the Raman spectra of a three layer system consisting of  $1000\text{ \AA}$   $\text{Al}_x\text{Ga}_{1-x}\text{As}$ ,  $\sim 200\text{ \AA}$  GaAs,  $\sim 2000\text{ \AA}$   $\text{Al}_x\text{Ga}_{1-x}\text{As}$  on a GaAs substrate. One can clearly separate the different spectra of the layers. In the spectrum obtained with the laser excitation line  $\lambda_0=4545\text{ \AA}$ , we see only the spectrum of the top single  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  layer. For this layer we find  $x=0.15$ . The second spectrum with  $\lambda_0=5682\text{ \AA}$  shows also the LO mode of the GaAs layer ( $294\text{ cm}^{-1}$ ) and a stronger asymmetry of the  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  lines, which indicates that the deeper  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  layer has a higher Al concentration. The Raman lines of the two deeper layers are observed because of the larger penetration depth of the  $\lambda_0=5682\text{ \AA}$  laser line.

If the Al content is higher, the separation of the different spectra is more clear. This can be seen in the lowest spectrum of Fig. 7, which has been obtained with a  $0.5\text{ }\mu\text{m}$  thick  $\text{Al}_{0.85}\text{Ga}_{0.15}\text{As}$  layer on a GaAs substrate using  $\lambda_0=5145\text{ \AA}$ .

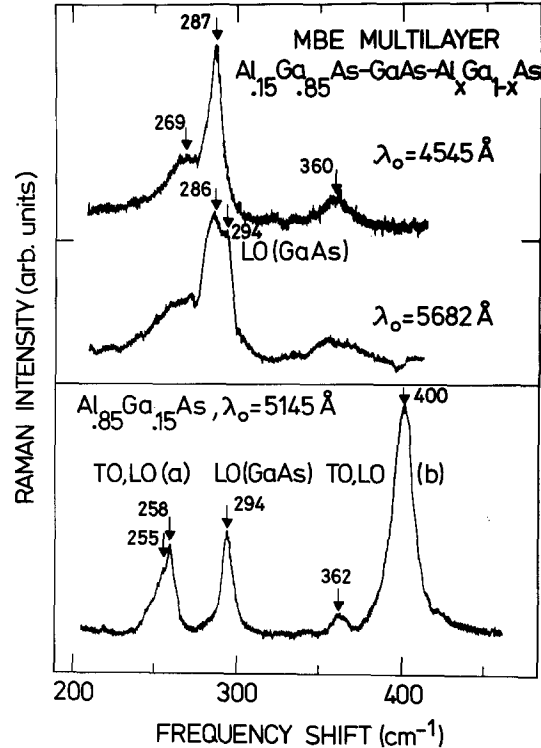


Fig. 7. Raman spectra of a three layer system  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ —GaAs— $\text{Al}_x\text{Ga}_{1-x}\text{As}$  obtained with two different laser excitation lines. One clearly can separate the spectra of the different layers. As shown in the lowest trace, the frequency separation is larger for a sample with higher Al concentration. This spectrum has been obtained of a thin  $\text{Al}_{0.85}\text{Ga}_{0.15}\text{As}$  layer on a GaAs substrate

#### Concluding Remarks

It has been shown that Raman spectroscopy can be used as a powerful technique to characterize thin films. Even depth profiling can be done in a non-destructive way. A detailed lineshape analysis might lead to further information, for example, on defects or inhomogeneous distributions of dopants and impurities.

We want to emphasize that Raman scattering experiments can be performed on very small spots on the sample surface. Thus it is also possible to study all the properties discussed in this work along the whole surface with a spatial resolution of the diameter of the focussed laser beam (a few  $\mu\text{m}^2$ ). This might be of interest for technological applications, because it is often necessary to check the homogeneity of thin films or layered structures for devices over the whole surface area. One can also study, for example, specially grown layers for opto-electronic couplers with variations in composition, or the quality and/or carrier concentrations over large areas of samples used in semiconductor device technology. Therefore, Raman spectroscopy, a technique so far mainly used in pure research,

is also a versatile tool for sample characterization and thus a useful technique in related technological areas.

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