Interface luminescence of GaAs/Ga$_{1-x}$Al$_x$As heterostructures: Threshold effect of the interface formation conditions

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Photoluminescence of single GaAs/Ga$_{1-x}$Al$_x$As heterostructures was studied as a function of the interface formation conditions (epitaxy temperature, layer growth rate, etc.). Photoluminescence spectra contained, apart from the bulk luminescence bands of GaAs and Ga$_{1-x}$Al$_x$As, a band related to the interface (interface luminescence band). It has been shown that the emergence of interface luminescence critically depends on the interface formation temperature T^F: it can be observed for temperatures above T_{cr}^F and vanishes below this threshold. This temperature threshold decreases with decreasing value of the layer growth rate and initial supersaturation and depends on perfection and/or doping of the heterostructure narrow-gap region (GaAs). This sharp, threshold-type emergence and disappearance of the interface luminescence has been accounted for by invoking two models. The first model is that of the interface luminescence as due to annihilation of the heterodimensional interface exciton formed by a two-dimensional (2D) electron (hole) at the interface and a 3D hole (electron) in the bulk of GaAs; the second model assumes that the roughening phase transition at the substrate surface produces a drastic change in the interface morphology (at the microscopic level) causing a sharp change in conditions required for the existence of the heterodimensional interface exciton.

I. INTRODUCTION

It is known that in photoluminescence spectra of single GaAs/Ga$_{1-x}$Al$_x$As heterostructures alongside with peaks characteristic of bulk GaAs and Ga$_{1-x}$Al$_x$As there is observed a specific band relating to the interface or the interface luminescence band.[1-4]

Properties of the interface luminescence differ essentially from those of the bulk luminescence from GaAs and Ga$_{1-x}$Al$_x$As. The most important difference is the high-energy shift of the interface luminescence peak with excitation intensity.[1-4]

A number of explanations for the origin of the interface luminescence has been proposed. One of these postulates that electrons (holes) in two-dimensional (2D) quantum notches recombine with 3D holes (electrons) in the bulk GaAs.[1,2] An alternative interpretation is based on an analogy with donor-acceptor recombination.[4] However, authors of more recent studies[5-7] seem to prefer the first interpretation. Besides, in Refs. 8–10 the interface luminescence was presumed to be of an excitonic origin.

The interface luminescence has been observed in GaAs/Ga$_{1-x}$Al$_x$As structures grown by different techniques: molecular-beam epitaxy,[4,5] metal-organic chemical-vapor deposition,[6] and liquid-phase epitaxy.[1,3] It as been observed in In$_{1-x}$Ga$_x$As/InP heterostructures as well.[11] These observations warrant a conclusion that the interface luminescence is an intrinsic property of the interface.

Conditions of the heterojunction formation are known to affect considerably its structure. The epitaxy temperature and both the layer growth rate and initial supersaturation determine the extent of the transition region between the layers and its morphology.[12,13] Besides, the growth mechanism of epitaxial GaAs was found to be changed at some temperature as determined from surface morphology studies.[14] Yet, in research on the nature of the interface luminescence no attention has been paid so far to its relation to the interface formation conditions.

The present study aims at finding out the relation between the characteristics of the interface luminescence and formation conditions of the GaAs/Ga$_{1-x}$Al$_x$As interface. One of the major results of the study is the observation that the emergence of the interface luminescence critically depends on the interface formation temperature. It has been established that this kind of luminescence also critically depends on other interface formation conditions and the imperfection and/or doping (as derived from the half-width of the bound exciton line in GaAs) of the heterostructure narrow-gap region.

II. EXPERIMENT

Chemically polished GaAs wafers were used with their planes off the (100) face by 0.15°–0.3° toward the [110] axis. On these wafers, GaAs layers of a thickness 5–10 μm and various conductivity types, with the intrinsic doping level $n,p \cong 10^{13} - 10^{17}$ cm^{-3} were first grown using gas transport epitaxy. These structures were used as substrates for a subsequent liquid-phase epitaxy process, which was used to grow thin (≈ 500–5000 Å) Ga$_{1-x}$Al$_x$As ($x = 0.1$–0.3) layers both N (Te doped) and P type (Ge doped), with the doping level $N,P \cong 10^{17} - 10^{18}$ cm^{-3}.

The effect of interface formation conditions on the interface luminescence was studied. To study the effect of epitaxy temperature, Ga$_{1-x}$Al$_x$As layers were grown at several temperatures in the range $600 \leq T \leq 920$ °C. The

interface formation rate was varied by changing either the initial melt supersaturation or the growth rate.

Layer growth at various supersaturation values was performed from supercooled molten solutions in thermal equilibrium. In the growth process, after the $Ga_{1-x}Al_xAs$ layer has been formed at one area of the substrate and the molten solution essentially depleted, the substrate was shifted so that the growth on another substrate area from the same molten solution proceeded at a lower initial supersaturation. The layer growth from the same melt at various rates was carried out as in Ref. 15.

Photoluminescence spectra of grown GaAs/$Ga_{1-x}Al_xAs$ structures were investigated under excitation with an He-Ne laser ($h\nu = 1.95$ eV) at liquid-helium temperatures using the photon-counting technique.

III. EXPERIMENTAL RESULTS

In photoluminescence spectra of GaAs/$Ga_xAl_{1-x}As$ structures grown under certain conditions, alongside with bands due to the bulk donor-acceptor recombination (DA) and bulk bound excitation annihilation (BE), the interface luminescence band (H) can be observed (Fig. 1).
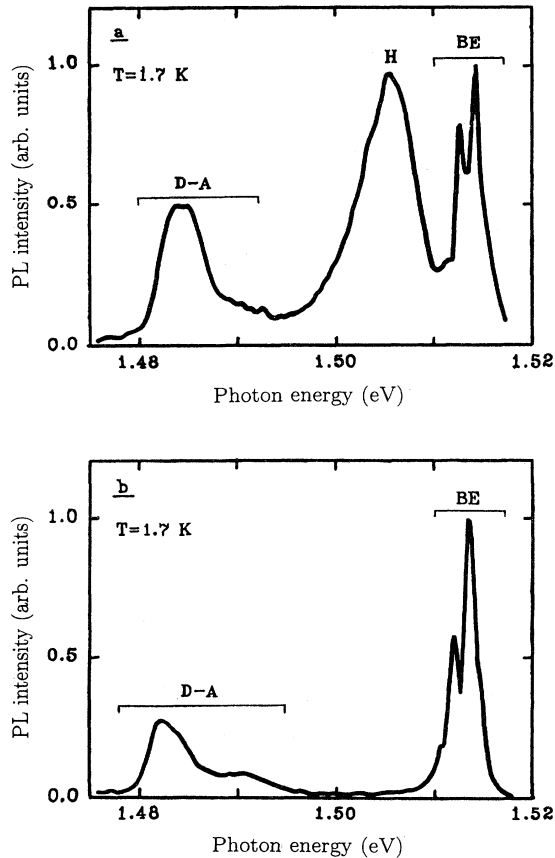
The interface luminescence was observed both in n-type GaAs/N,P-type $Ga_{1-x}Al_xAs$ and p-type GaAs/N, P-type $Ga_{1-x}Al_xAs$ heterostructures (Fig. 2). Characteristics of the interface luminescence in the four structure types are practically identical. The spectral band of the interface luminescence has a half-width of 5–10 meV and shifts towards higher energy with increasing excitation level (by 3–5 meV per decade change in the excitation intensity). The upper limit for the shift of the H band corresponds to the peak position of the exciton bound to a neutral acceptor in GaAs (1.512 eV). The intensity of the H band relative to that of the DA band and BE band is decreased with temperature and at 20 K the interface luminescence could not be detected.

The occurrence of the interface luminescence critically depends on the interface formation temperature T^F: it can be observed in heterojunctions formed at temperatures above T_{cr}^F, while for formation temperatures below T_{cr}^F its intensity drops dramatically (by nearly two orders
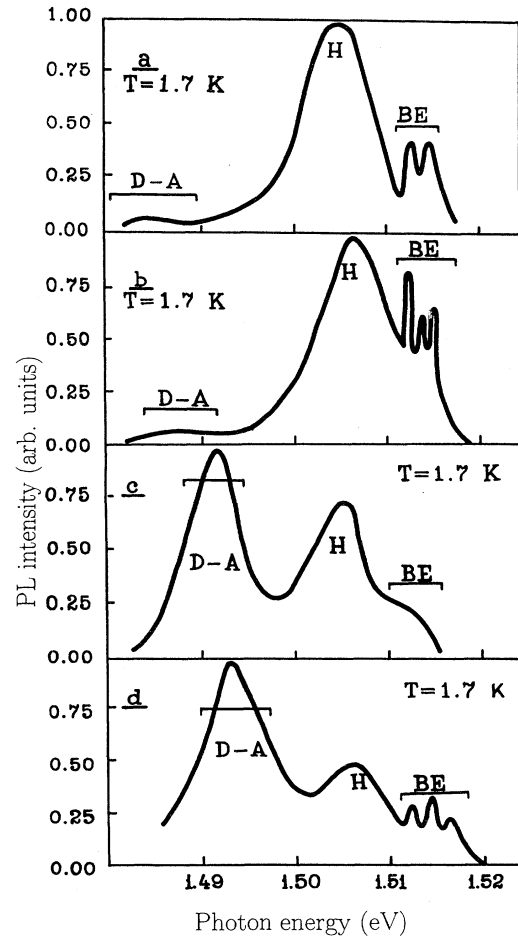

FIG. 1. Photoluminescence spectra of two n-type GaAs/N-type $Ga_{0.8}Al_{0.2}As$ ($n = 2 \times 10^{15}$ cm^{-3}; $N = 10^{18}$ cm^{-3}) heterostructures grown on the same substrate [$\Delta(h\nu) = 1.7$ meV] and from the same molten solution at two different temperatures T^F: (a) $T^F = 850$ °C; (b) $T^F = 825$ °C.

FIG. 2. Photoluminescence spectra of the four types of heterostructures: (a) n-type GaAs/N-type $Ga_{0.8}Al_{0.2}As$ ($n = 2 \times 10^{15}$ cm^{-3}; $N = 5 \times 10^{17}$ cm^{-3}); (b) n-type GaAs/P-type $Ga_{0.75}Al_{0.25}As$ ($n = 2 \times 10^{15}$ cm^{-3}; $P = 10^{18}$ cm^{-3}); (c) p-type GaAs/N-type $Ga_{0.8}Al_{0.2}As$ ($p = 4 \times 10^{15}$ cm^{-3}; $N = 5 \times 10^{17}$ cm^{-3}); (d) p-type GaAs/P-type $Ga_{0.75}Al_{0.25}As$ ($p = 4 \times 10^{15}$ cm^{-3}; $P = 5 \times 10^{17}$ cm^{-3}).

of magnitude at the least), to values below those that could not be detected with the instrumentation used (Fig. 3).

The critical temperature T_{cr}^F for the forming of the interface depends markedly on the interface formation rate. For example, higher initial supercooling ΔT of the molten solution causes an increase in T_{cr}^F (Fig. 4). On the other hand, increasing the layer growth rate V_g while the interface is being formed beyond a certain value with temperature fixed at $T^F \approx T_{cr}^F$ results in the abrupt disappearance of the interface luminescence band from the spectra (Fig. 5).

The critical temperature depends on the perfection and/or doping of the GaAs substrate, which can be estimated by the half-width $[\Delta(h\nu)]$ of the BE band in n-type GaAs. So, for example, $T_{cr}^F = 850\,°C$ for the substrate with $\Delta(h\nu) = 1.7$ meV ($n = 2 \times 10^{15}$ cm^{-3}) and T_{cr}^F is in the range 770–800 °C for the substrates with $\Delta(h\nu) = 0.4$–0.8 meV ($n = 10^{13} - 10^{14}$ cm^{-3}).

IV. ORIGIN OF THE INTERFACE LUMINESCENCE: APPROACH TO A MODEL

Our interpretation of the critical behavior of the interface luminescence is based on the use of two models modified so as to make them mutually consistent. The first model is that of the heterodimensional interface exciton (which happens to be sensitive to the interface micromorphology); the second model presumes that a jumpwise change in the interface morphology at the microscopic level is produced as a result of the roughening phase transition at the substrate surface. This drastic change in the micromorphology, in its turn, brings about a sharp change in conditions required for the existence of the heterodimensional interface exciton.
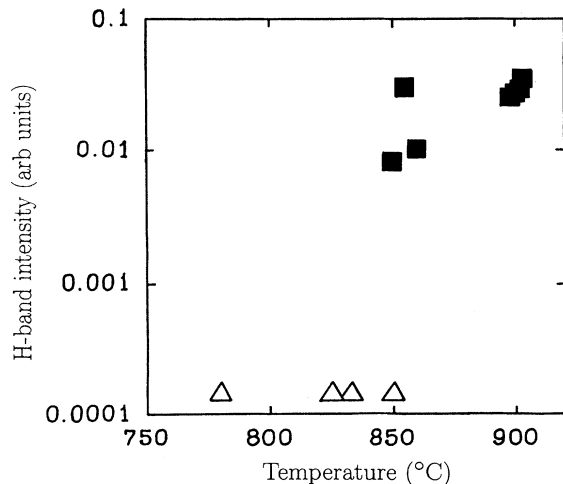
FIG. 3. Dependence of the H-band intensity on the interface formation temperature T^F for the n-type GaAs/N-type Ga$_{1-x}$Al$_x$As ($n = 2 \times 10^{15}$ cm^{-3}; $N = 10^{18}$ cm^{-3}) heterostructure [$\Delta(h\nu) = 1.7$ meV] at the excitation intensity $I = 4$ W/cm^2 (triangles represent the H-band intensity below threshold; squares represent the H-band intensity above threshold; the dashed line is the signal detection threshold).
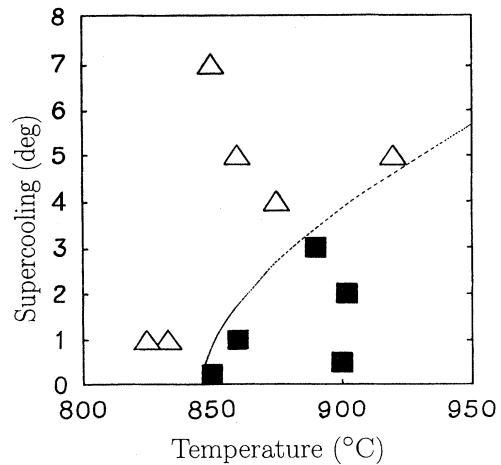
FIG. 4. Schematic diagram of the occurrence of the interface luminescence in n-type GaAs/N-type Ga$_{1-x}$Al$_x$As ($n = 2 \times 10^{15}$ cm^{-3}; $N = 10^{18}$ cm^{-3}) heterostructures [$\Delta(h\nu) = 1.7$ meV] grown at various values of T^F and the initial melt supercooling ΔT (triangles represent the H-band intensity below threshold; squares represent the H-band intensity above threshold; the dashed line is the approximate boundary between these two regions).

A. Heterodimensional interface exciton

Properties of the interface luminescence are quite consistent with a viewpoint that it is due to annihilation of the heterodimensional interface excitons formed by 2D electrons (for the n,p-type GaAs/N-type Ga$_{1-x}$Al$_x$As interface) confined in a notch at the interface and 3D holes in the GaAs bulk near the interface [Figs. 6(a) and 6(c)]. With the n,p-type GaAs/P-type Ga$_{1-x}$Al$_x$As interface, the respective heterodimensional interface excitons will be formed by 2D holes and 3D electrons [Figs. 6(b) and 6(d)]. In what follows, we will refer to the interface excitons formed by carriers of different dimension (2D and 3D) as the heterodimensional excitons. This model is similar to that of the surface exciton proposed to account
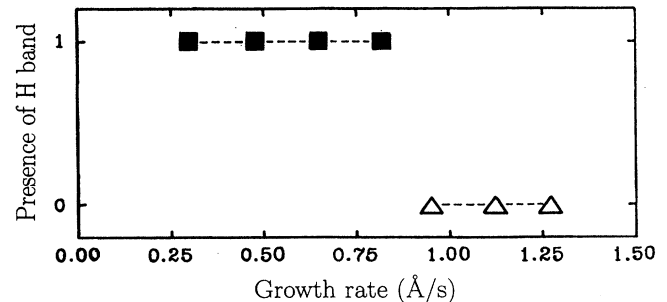
FIG. 5. Schematic diagram of the presence of the interface luminescence in n-type GaAs/N-type Ga$_{1-x}$Al$_x$As ($n = 5 \times 10^{16}$ cm^{-3}; $N = 10^{18}$ cm^{-3}) heterostructures [$\Delta(h\nu) = 2.0$ meV; $T^F = 900\,°C$] for various values of the layer growth rate V_g at the excitation intensity $I = 4$ W/cm^2 (triangles represent the H-band intensity below threshold; squares represent the H-band intensity above threshold).
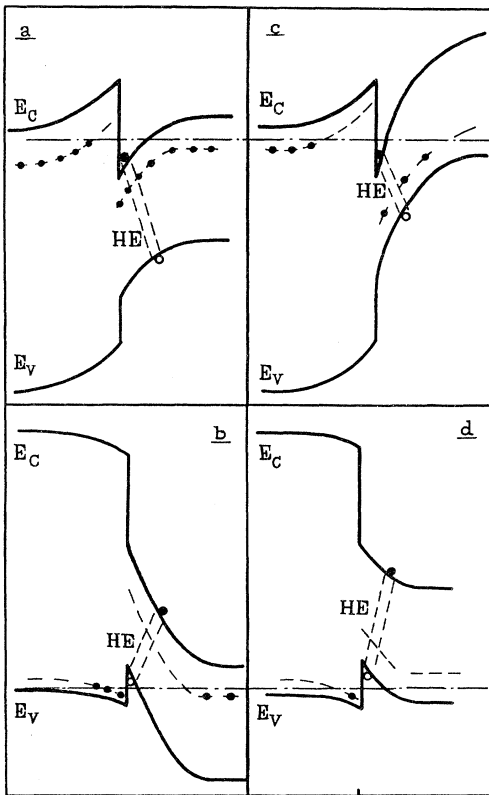
FIG. 6. Schematic band diagram of the four types of heterojunctions and the heterodimensional interface exciton: (a) n-type GaAs/N-type GaAlAs; (b) n-type GaAs/P-type $Ga_{1-x}Al_xAs$; (c) p-type GaAs/N-type $Ga_{1-x}Al_xAs$; (d) p-type GaAs/P-type $Ga_{1-x}Al_xAs$.

for the surface luminescence in CdS (Ref. 16) and silicon metal-insulator-semiconductor (MIS) structures.[17] This model was also suggested for the interface luminescence in $In_{1-x}Ga_xAs/InP$ structures.[11]

Such a model complies with the main feature of the interface luminescence: the large shift of the spectral band to higher energies with rising excitation intensity. Indeed, an increase in the intensity results in lower interface band-bending value in GaAs and, consequently, in larger energy photons being generated as exciton annihilates. Then, merging of the interface luminescence band and the bulk bound exciton band observed at very high excitation intensities corresponds to nearly complete leveling out of the interfacial barrier in GaAs.

Interpretation of the interface luminescence as due to annihilation of the heterodimensional interface exciton supplements the model by Yuan et al.[1] by providing a detailed mechanism for the trapping of the 3D charge carriers near the interface (e-h Coulomb interaction). This refinement of Yuan's model leaves intact all its consequences, on the one hand, and removes objections as to the seemingly low concentration of 3D carriers near the interface, on the other.

According to the model of the heterodimensional interface exciton, the 3D electron (or hole) cannot move everywhere but only in a half-space, so its wave function

should turn zero in the heterojunction plane (zero boundary condition). If, for simplicity, we consider the 2D hole (or electron) as being immobile [an analog of the hydrogenlike donor (acceptor)] then the ground state of the exciton will be one of the p states (p_0 or p_z, if the z axis is taken to be perpendicular to the heterointerface) rather than an s state. The s state cannot occur as it does not meet the zero boundary condition in the heterojunction plane. That is the reason why the heterodimensional interface exciton at the boundary plane has a lower binding energy compared with the bulk exciton. The lower binding energy of the heterodimensional interface exciton agrees with the temperature behavior of the H band (disappears at approximately 20 K).[1] As distinct from the ordinary 3D exciton, the linear Stark effect can be observed with this kind of exciton due to the fact that the contributions from the two half-spaces do not cancel each other as is the case with the ordinary bulk exciton. Qualitatively, the linear Stark effect for this exciton agrees with the observed shift of the H band in GaAs when the bands flattening occurs as a result of increasing excitation intensity.

In the case of a heterointerface which, though chemically abrupt, abounds in bumps and pits (referred to below as rugged heterointerface) the heterodimensional interface exciton will have still lower binding energy compared to the exciton at the flat heterointerface because the ground states will have higher energies. For example, in the case of the zero boundary condition being applied to a cone having the angle of taper equal to $\arccos(1/\sqrt{3}) \approx 55°$ and with the immobilized hole located in the vertex of the cone, the ground state will be one of the d states, namely d_0 or d_{z^2}. Corresponding wave functions are from Ref. 18.

At the rugged interface, the exciton is more likely to be destroyed by the interface electric field or thermal fluctuations, because of the lower binding energy of the heterodimensional interface exciton. Such destruction of the heterodimensional interface exciton should take place when the irregularities research the size of the exciton.

B. Threshold-type dependence of the heterointerface micromorphology on the forming temperature

It is known that the threshold-type (jumpwise) dependence of some property of the material on temperature is evidence of a phase transition. A jump in the quantum efficiency was observed only for the interface luminescence and did not take place for the bulk luminescence (Figs. 1 and 3); the phase transition in question should take place at the boundary between GaAs (substrate) and Ga-Al-As melt. The state in which this boundary is found prior to forming of the GaAs/$Ga_{1-x}Al_xAs$ interface will be determining the micromorphology (imperfection) of the latter. Imperfection of the chemically sharp GaAs/$Ga_{1-x}Al_xAs$ heterointerface is generally understood[19] as its ruggedness.

The roughening transition at the GaAs substrate surface is suggested as the phase transition responsible for the jumpwise change in the ruggedness of the interface.

It has been found[14] that in epitaxial growth of GaAs

the use of temperatures above a certain critical value T_{cr} resulted in a dramatic change of the state of the growing surface called the roughening transition. At lower temperatures ($T < T_{cr}$), the growth surface is smooth on the atomic scale and the growth proceeds as the movement of a succession of monoatomic steps, i.e., according to the stepwise growth mechanism. At temperatures above T_{cr}, the steps on the growing surface become smeared and eventually disappear as the stepwise growth mechanism gives place to the continuous growth.

When the interface is formed through the stepwise mechanism of growth, Ga and Al atoms will be captured from the molten solution by the moving steps. Because of fluctuations of Ga and Al concentrations in the molten solution and nonuniform distribution of the growth steps over the substrate surface, both nonuniform incorporation of Al atoms into the interface[20] and fluctuations in the position of the heterointerface are possible and can result in formation of an imperfect (rugged) heterointerface.

When the interface is formed through the continuous growth mechanism, incorporation of Ga and Al atoms from the melt will occur at the kinks, which are present in great numbers on the substrate surface at $T > T_{cr}$. At low enough layer growth rates (lower or at least comparable to the diffusion rate of Ga and Al atoms in the near-surface region of the solid phase) for the equilibrium Al concentration to be established at the growing surface, there will take place nearly complete homogenizing in the plane of the interface, which should result in substantially less irregular heterointerface as compared with the stepwise growth mechanism of the interface.

So, for temperatures lower than the critical interface formation temperature, and heterointerface should be strongly rugged; while for temperatures above the critical temperature, it should be considerably less rugged.

Thus, the interface luminescence should be observed in epitaxial GaAs/Ga$_{1-x}$Al$_x$As heterostructures grown at temperatures above T_{cr} (mechanism of continuous growth). In structures formed at temperatures lower than T_{cr} (stepwise mechanism of growth), the interface luminescence cannot be observed, the interface heterodimensional exciton being quenched due to imperfection (ruggedness) of the heterointerface.

V. DISCUSSION

Similarity of the properties of the interface luminescence in all four structure types, that is in structures with notches for electrons (n,p-type GaAs/N-type Ga$_{1-x}$Al$_x$As) and for holes (n,p-type GaAs/P-type Ga$_{1-x}$Al$_x$As) warrants a conclusion that it is due to intrinsic properties of the heterointerface. This suggests an excitonic nature of the H band because it is the property of an exciton to be symmetric relative to the type of conductivity. Indeed, the reduced mass of the heterodimensional interface exciton is essentially independent of the kind of carrier confined in the near-interface notch.

Luminescence from the GaAs/Ga$_{1-x}$Al$_x$As heterointerface as an excitonic effect was treated in Refs. 8–10. In Refs. 8 and 9, a quasi-2D exciton was considered local-

ized as a whole at the interface because its annihilation energy is lower compared with the bulk exciton on account of the quadratic Stark effect. Such an exciton is similar to the exciton localized in a quantum well and can form even in the case of flat bands.

The heterodimensional interface exciton differs from the quasi-2D exciton in that only one carrier (e.g., hole) is localized at the interface notch. Despite its lower binding energy, such an exciton can exist in the presence of the interface electric field, as shown theoretically for the case of the Si/SiO$_2$ heterointerface.[17]

It is to be noted that an exciton in which the electron is associated with the gas of 2D electrons and the hole is in the valence band has been identified in a study of the interface luminescence in magnetic fields.[10]

The rather large half-width of the H band is due, possibly, to the e-e interaction because in the potential notch at the heterointerface, the 2D electron (hole) gas is degenerate. In contrast to the ordinary bulk exciton, which is suppressed when inside the degenerate gas, the screening of the heterodimensional interface exciton does not occur because the hole (electron) is found outside the channel filled with the degenerate gas of electrons (holes).

The fact that a minor change in growth conditions decides whether or not the interface luminescence will take place suggests that a phase transition occurs while the interface is being formed.

If the interface formation temperature is raised above some critical value, a roughening transition takes place at the surface of the GaAs substrate. Correspondingly, the interface forming mechanism should change from stepwise to continuous, resulting in a drastic reduction of the interface ruggedness and favoring the existence of the heterodimensional interface exciton (Fig. 3). Note that the temperature of the roughening phase transition at liquid-phase epitaxy of GaAs (Ref. 14) ($T_{cr} = 765\,°$C) is essentially the same temperature as T_{cr}^F for structures grown on substrates having $\Delta(h\nu) = 0.4$ meV ($T_{cr}^F = 770\,°$C). It should be noted that in the work by Yuan et al.[1] where the observation of the interface luminescence band was first reported the epitaxy temperature was high ($T = 850\,°$C) and the occurrence of the interface luminescence in such structures corresponds to our experimental data and model.

With increasing layer growth rate or the initial melt supersaturation, the critical temperature for the emergence of the interface luminescence should increase because equilibrium conditions will not be reached[20] and the interface will be rugged. Indeed, the interface luminescence is observed from interfaces formed at small values of the layer growth rate and initial supercooling; it is absent when these values are large (Figs. 4 and 5).

One of the interesting conclusions to be drawn is that the higher quality interface (from the viewpoint of optoelectronic properties) will be formed at temperatures above the critical temperature; yet one has to bear in mind that the temperature should not be too high so as not to cause generation of defects, which might lead to degradation of the heterostructure properties. In our opinion, the interface luminescence can serve as an indicator of the quality of the interface.

It should be noted also that our interpretation of the emergence and disappearance of the interface luminescence can be extended to interfaces prepared by methods of molecular-beam epitaxy and metal-organic chemical-vapor deposition. In these heterostructures as well, the emergence of the interface luminescence is possible if interface ruggedness do not exceed some limiting value. However, the character of variation of the interface ruggedness can be determined by other factors beside the change of the growth mechanism.

VI. CONCLUSION

It has been demonstrated that a minor variation of the interface formation conditions during liquid-phase epitaxy of $GaAs/Ga_{1-x}Al_xAs$ might be a critical factor determining whether or not the interface luminescence can be observed, which is explained by the change of the interface formation mechanisms occurring as a result of the roughening phase transition at the GaAs substrate surface.

The interface luminescence in these structures is observed in those cases when the interface formation temperature is higher than some critical temperature T_{cr}^F, which in its turn depends on the layer growth rate, the initial melt supercooling, and the perfection and/or doping of the GaAs substrate.

The interface luminescence is due to the annihilation of the heterodimensional interface exciton, which cannot be formed if the heterointerface is rugged. This ruggedness is small in continuous growth and large in stepwise growth.

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