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HIGH RESOLUTION TEM-CL FROM THE CROSS-SECTIONAL SPECIMENS OF GaAs/AlGaAs QWs

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ABSTRACT—One of the advantages of the scanning transmission electron microscope cathodoluminescence (STEM-CL) technique is its higher spatial resolution in comparison with CL performed in a scanning electron microscope (SEM-CL). Our studies of cross-sectional specimens of a GaAs/AlGaAs quantum well structure have clearly demonstrated this. We believe that it is the first time that STEM-CL has been applied successfully to an electron transparent cross-sectional specimen. The aim of this study was to investigate the effect of different growth sequences on QW luminescence and to compare the quality of the successive QW sets. CL spectra were obtained from the cross-sectional samples by directing the electron beam onto each of QWs in turn. We observed marked differences in these spectra. Monochromatic CL images were also generated and they revealed clearly resolved emission from each of the separate sets of QWs in the structure. A spatial resolution of about 50nm was achieved transverse to the QWs in the monochromatic images of this particular structure.

1. Introduction

Cathodoluminescence (CL) techniques have been used extensively in characterizing semiconductor materials. The main advantages of CL analysis are its high spatial resolution and the possibility to correlate the results with the information obtained from other imaging techniques available in an electron microscope. The CL systems are most commonly implemented in scanning electron microscopes (SEM). The applications of SEM-CL have been thoroughly reviewed by a few authors [1-3]. In order to achieve even higher spatial resolution and to correlate the information obtained from CL analyses directly with that derived from diffraction contrast transmission electron microscopy, CL systems have also been implemented in scanning transmission electron microscopes (STEM) in a few laboratories. A review of the performance and applications of a STEM-CL system has been given by Steeds [4]. In this work, we have applied the STEM-CL technique to cross-sectional samples of a specially fabricated GaAs/AlGaAs quantum well (QW) structure. We were able to obtain independent CL spectra from each different set of QWs and investigate the differences observed in these spectra. Monochromatic CL images were also generated and revealed clearly resolved emission from each different set of QWs. The implications of the CL spectra for differences between the sets of QWs are discussed together with the interpretation of the high resolution monochromatic CL images.

2. Experiments

The GaAs/Al_xGa_{1-x}As quantum well (QW) structure studied here was grown by molecular beam epitaxy (MBE) on a (001) GaAs substrate. The layout of the structure is revealed in the cleavage cross-sectional transmission electron micrograph shown in Figure 1. The three sets of QWs, which are labelled as QW1, QW2 and QW3, were notionally identical apart from their proximity to the surface or the substrate and the separation of adjoining sets of QWs by either 3500Å of GaAs or 3500Å of AlGaAs. Each set of QWs contained five individual GaAs quantum wells of nominal

width 51Å with AlGaAs barriers of the same width. The Al composition of all the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layers was $x=0.33$. The cross-sectional specimens were prepared by a standard method/5/ involving the use of epoxy to stick together two samples prior to ion thinning.

Fig.1. TEM image from a wedge-shaped sample shows the layout of the structure studied.

Low temperature and high spatial resolution CL experiments were performed in an extensively modified Philips EM400 Scanning Transmission Electron Microscope. A detailed description of the CL system can be found elsewhere/6,7/. In brief, a retractable ellipsoidal mirror, with a small hole to allow the passage of the electron beam, is used to collect the light generated by the incident electron beam. The beam impinges on the specimen at one focus of the mirror. The light is collected by the mirror, brought out of the microscope, and reflected through quartz optics into a 50cm path length monochromator. An RCA 31034A photomultiplier with a GaAs photocathode was used in these experiments. The acquisition of CL spectra and monochromatic images was controlled by a Link 860 computer. The specimen was cooled to about 30K by mounting it in a cold stage with continuous flow of liquid helium. A 100kV electron beam was used as the excitation source.

3.Results and Discussion

3.1 CL spectra

It is well known that in high quality unintentionally doped GaAs/AlGaAs QW structures the luminescence spectra appear to be essentially intrinsic, in contrast with bulk GaAs emission/8-10/. The simpler QW luminescence is probably due to the localization of excitons at interface defects and the decrease of the exciton lifetime which results from the increase overlap of the electron and hole wavefunction in QW. The former prevents the excitons from reaching the impurities. The latter increases the exciton recombination efficiency and implies that unless the impurity separation in the QW is comparable with the free exciton diameter, the free exciton recombines before it is captured by a impurity. However experimental observations of acceptor related features in PL spectra of un-intentionally doped MBE grown GaAs/AlGaAs QWs have been reported/11-16/. It has been proposed that carbon could provide an explanation of the origin of these effect. Carbon is less soluble in AlGaAs than in GaAs and is a well known background impurity in MBE grown GaAs/17/. Thus, as the AlGaAs layers grow, an ever increasing amount of carbon floats to the AlGaAs-vacuum interface. After the growth of a thick AlGaAs layer, a considerable number of these impurities is trapped in the QWs grown immediately after the termination of AlGaAs growth. The GaAs/AlGaAs QW structure examined here was designed to investigate the effect of different growth sequences on extrinsic luminescence properties of QWs.

A typical low temperature CL spectrum of the thin cross-sectional sample, including both the bulk AlGaAs emission at 1.965eV and the bulk GaAs emission at 1.516eV, as shown in Figure 2. The luminescence peak at 1.617eV in the figure corresponds to an $n=1$ electron-heavy hole (e-hh) exciton transition from the GaAs QWs. We

Fig.2 CL spectrum obtained from the cross-sectional sample of the structure.

note that in the energy region immediately below the QW emission there is additional very broad luminescence, marked as I. The emission peak energy is about 30 meV below the QW intrinsic emission. We also observed that this emission saturated at high electron beam currents i.e. high excitation levels. The energy shifts, the breadth of the emission band and the saturation of the emission at high excitation levels all lead us to conclude that this additional emission is impurity related. Comparison with the theoretical predictions/18-20/ and the experimental results of deliberately introduced shallow impurities in QWs reported by other researchers/11,21/ and the work on unintentionally doped samples cited above leads us to conclude that the extrinsic emission observed here results from the incorporation of acceptor-like impurities, probably carbon. As these QWs were grown immediately on top of a thick AlGaAs layer a high level of carbon incorporation is expected.

To determine the distribution of impurities among the different sets of QWs, we obtained three CL spectra by pointing a focused electron beam at each set in turn. The resulting difference in intensity of the impurity related emission is clearly shown in Figure 3. Spectrum(a) corresponds to the CL emission acquired from QW3 which was grown immediately after the GaAs buffer layer. It corresponds to a low impurity emission level. The origin of these impurities may be residual impurities in the growth chamber or on the substrate surface.

Fig.3 CL spectra obtained from the cross-sectional sample of the structure. Emission spectra are shown for QW3(a), QW2(b) and QW1(c).

Spectrum(b) and spectrum(c) in Figure 3 correspond respectively to the CL emission acquired by directing the electron beam at QW2, which was grown immediately after the GaAs separation layer, and to QW1, which was grown immediately after a thick AlGaAs separation layer. A substantial increase in the intensity of impurity emission was observed from both sets of QWs and was even higher for QW1 than for QW2. Since these two sets of QWs were separated by a thick AlGaAs layer, where the carriers have a higher potential energy, some of the excess carriers

created when the electron beam was incident on the AlGaAs will diffuse into the adjoining QWs. The finite beam diameter (approx. 100nm) and subsequent beam spreading in the rather thick regions (>300nm) used for these experiments means that even when the beam was directed at QW1 or QW2 some overspill into the AlGaAs layer was inevitable. The excitation subsequently diffusing to the adjoining sets of QWs emerges as QW emission. Considering this effect, which will be discussed in more detail below in connection with CL image analysis, it is conceivable that a considerable proportion of the impurity emission apparent in spectrum(b) from QW2 is in fact a spill-over of AlGaAs carriers diffusing to QW1. Therefore, we may conclude that the impurity related emission mainly occurs in QW1. Assuming that the strength of the impurity related emission depended on the impurity concentration in the QWs our observation that QW contain a high impurity concentration is consistent with the idea, previously explained, that QWs grown after the thick AlGaAs layer are likely to be rather impure. However, the fact that the set QW1 was situated near to the sample surface with only a thin barrier layer means that the possibility of impurity diffusion from the sample surface into the QW1 cannot be ruled out.

3.2 CL Images and Line-scans

The spatial resolution of CL images is related to three factors; the electron beam probe size, the diameter of the electron-hole pair generation volume, and the carrier diffusion length. In practice the spatial resolution of CL may be determined by any one of these factors or by a combination of them depending on the material under study, the electron beam energy and the specimen thickness. For a bulk sample all these factors will be effective. For a thin, electron transparent specimen, the electron beam spreading (i.e. the generation volume) is reduced as the film thickness decreases. The high surface non-radiative recombination velocity of a thin film also reduces the effective carrier diffusion length. Therefore, the spatial resolution of CL images may be increased by studying regions of reduced specimen thickness. However, this improvement is accompanied by a reduction in CL intensity for a homogeneous semiconductor thin film. In the case of semiconductor multilayer structures band structure modification of the material leads to modification of the excess carrier distribution. The transfer of excess carriers from the barrier layers to the well layers and the effect of carrier confinement in the wells leads to a great enhancement of the electron-hole recombination rate in the quantum wells. Further the carrier diffusion length is reduced within the quantum wells to a distance dependent on the pinning provided by interface steps. The restriction of carrier movement transverse to the quantum wells causes greatly enhanced spatial resolution in CL images from cross-sections of multilayer structures as is shown in the following experiments.

A monochromatic CL image acquired from a rather thick area of a cross-sectional sample at the energy corresponding to QW exciton emission is shown in Figure 4(a) together with a TEM image (Figure 4(b)) of thinner region of the same sample for comparison. As a result of carrier confinement in the separate sets of quantum wells, which is an essential aspect of the high transverse resolution achieved, the three sets of QWs are clearly resolved. As is shown in the line scan parallel to the growth direction of integrated QW CL intensity in Figure 4(c), QW2 and QW3 are very well resolved since they are separated by a thick GaAs layer (3500Å) and the intensity almost reached zero at the middle of the layer. In this case, the CL image resolution was determined only by the electron beam probe size and beam broadening. Carrier diffusion does not make a significant contribution, since the band gap of bulk GaAs is less than that of the QWs. By contrast as QW1 and QW2 were separated by a thick AlGaAs layer, a certain level of QW emission was detected even when the electron beam was directed at the middle of the separation layer; this is evidence of the spilling over effect mentioned above. The CL image resolution was clearly affected by the (unknown) carrier diffusion length in the AlGaAs in this cases. The CL image acquired from the impurity related emission is shown in Figure 4(d) and consists of an unresolved emission band located near the sample surface. This result as well as the spilling effect support our assumption of that the larger proportion of impurity emission seen in the QW2 luminescence spectrum actually originated in QW1. We also note that the apparent distance between QW1 and QW2 in the CL image is less than that between QW2 and QW3 although they were in fact equally separated as is evident from the TEM image. This apparent discrepancy is a consequence

Fig.4 Monochromatic CL images, obtained at QW intrinsic emission(a), Impurity related emission(d), bulk AlGaAs emission(e) and bulk GaAs emission(f) from a cross-section sample of the structure, are shown with TEM image(b) of the same sample and monochromatic CL line-scan profile(c) of the image (a).

of the carrier diffusion from the AlGaAs layer into the adjacent QWs and their subsequent recombination. An intimately related effect is apparent in the shrinkage of the CL image of the AlGaAs separation layer (see Figure 4(e)) in comparison with that of the GaAs separation layer (see Figure 4(f)). The weak intensity of QW1 in comparison with QW2 and QW3 in the CL image(Fig.4(a)) is probably due to its high impurity related emission.

4. Conclusions

CL spectra and monochromatic images have been generated from an electron transparent cross-section of a GaAs/AlGaAs QW structure. CL spectra obtained from each of the three sets of QWs present in the structure revealed clear differences which we attribute to the effect of the growth sequence. The monochromatic CL images revealed clearly resolved emission from each of the three sets of QWs. A one dimensional spatial resolution of about 50nm was achieved in one of the CL images as a result of carrier confinement in the QWs.

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