Can Photo- and Cathodoluminescence be Regarded as Complementary Techniques?

S. Myhajlenko
Arizona State University

R. A. Puechner
Arizona State University

J. L. Edwards
Arizona State University

D. B. Davito
Epitronics Corporation, Phoenix, Arizona

Follow this and additional works at: https://digitalcommons.usu.edu/microscopy

Part of the Biology Commons

Recommended Citation
Available at: https://digitalcommons.usu.edu/microscopy/vol1995/iss9/19
CAN PHOTO- AND CATHODOLUMINESCENCE BE REGARDED AS COMPLEMENTARY TECHNIQUES?

S. Myhajlenko•, R.A. Puechner, J.L. Edwards and D.B. Davito1
Center for Solid State Electronics, College of Engineering and Applied Sciences, Box 876206, Arizona State University, Tempe, AZ 85287-6206, U.S.A.
1Epitronics Corporation, 21002 N 19th Avenue, Phoenix, AZ 85027, U.S.A.

Abstract

Photoluminescence (PL) usually provides macroscopic, high quality spectroscopic data. Cathodoluminescence (CL), on the other hand, offers the same information with microscopic imaging. However, replicating PL signatures in a CL system is not straightforward since matching experimental conditions, such as temperature and excitation density, is difficult. The matter is further exacerbated by inherent differences in the nature of excitation: electrons versus photons. Our work with high purity semiconductors suggests that CL is generally more sensitive to excitation "circumstance" than PL. For example, electrons can cause sample charging and contamination-related phenomena that dramatically affect CL. Changes in surface attributes (e.g., by chemical passivation) also affect PL and CL signals differently. Here, we extend previous work on GaAs by exploring the role of surface topography (by atomic force microscopy) and temperature (1.8K-100K) on excitonic line-shapes. We find that topographic subtelities strongly influence the character of exciton-polariton luminescence. We interpret these changes in terms of non-classical scattering phenomena derived from microscopic roughness. These microscopic changes also influence the temperature behaviour of excitons in crystals. Specifically, we find that passivated samples are brighter partly because there is a corresponding reduction in the (Arrhenius) activation energy for excitonic processes. In summary, the changes in surface topography and corresponding recombination physics seem well correlated.

Key Words: Atomic force microscopy, cathodoluminescence, exciton-polaritons, microscopic roughness, photoluminescence.

•Contact for correspondence:
Steven Myhajlenko, address as above.
Telephone number: 602 965 2697
FAX number: 602 965 8118

Background

Photoluminescence (PL) and cathodoluminescence (CL) are regarded as complementary techniques. CL can provide spatial information on the distribution of spectral features detected by non-scanning PL. Under appropriate operating conditions and with suitable sample "physics," sub-micron spatial resolution is possible with bulk samples [31]. Another benefit is that extended crystalline defects are readily visualized by CL if they are electrically and/or optically active. One recent example from our laboratory that highlights the usefulness of complementary PL/CL measurements is imaging of corrugated GaAs quantum wire arrays (QWA). Emission from these wires at room temperature is characterized by a single narrow spectral line. In one sample, PL revealed QWA emission that was considerably broader than expected and at an unexpected wavelength. Room temperature CL imaging at the "wrong" emission wavelength revealed inhomogeneities (Fig. 1). We did not expect such irregularities in the patterned array. This one simple observation suggested that something went wrong during growth and/or processing. The specifics (compositional inter-mixing) are not of concern here. Rather, we note simply that CL imaging can complement PL in a quality control capacity. So, why then do we question the complementary value of the two techniques? Consider a few observations on this matter. In recent years, the need for more detailed spatial information (electrical, optical, physical, etc.) has grown: opto-electronic devices are getting smaller (above example for instance). Also, since no single characterization technique can provide all the answers, use of different methods clearly can help unravel mysteries. The combined use of PL and CL is a natural path to pursue since luminescence fundamentals are (in principle) common. PL yields the macroscopic picture (an average) and CL has microscopic capabilities. However, this information may not be as straightforward to interpret as one thinks, since the techniques may be responding to different sample attributes. For instance, surface topography can influence energy coupling into a crystal. This coupling will be different for electrons and photons (frequency
measurements are more easily performed with CL than CL response. Enhancement or degradation of the CL non-resonant. By comparison, each absorbed photon may come into play is the charged nature of excitation: each absorbed photon produces a single electron-hole pair. Another factor that may come into play is the charged nature of excitation: negatively charged electrons (CL) versus neutral photons (PL). The incoming fast electron represents an impulse of applied charge to the solid. The dielectric response of the solid is fundamental to physical interpretation. For instance, anomalous surface and bulk charging phenomena can occur with poorly conducting materials, especially when bombarded with low energy electrons (< 5 keV). Here, we refer specifically to cross-over voltage phenomena as observed in scanning electron microscopy (SEM). This phenomenon can lead to curious static and dynamic charging effects [7]. Corresponding anomalous behaviour has been reported under low voltage conditions in CL with GaAs [19] and Ce:YAG [10]. Analysis of these effects can be difficult since secondary electron yield and CL emission are coupled to energy loss processes of the same electron beam. Vacuum related contamination is another factor known to influence CL response. Enhancement or degradation of the CL signal from characterized regions can occur with contamination [19, 30]. Use of very high electron energies (> 150 keV) can lead to atomic displacement, where the damage has deleterious effects on CL. Finally, CL can distinguish surface and bulk effects [6]. Depth-resolved measurements are more easily performed with CL than with PL. Electron penetration can be readily varied by changing accelerating voltage. On the other hand, wavelength tuning is severely limited with common argon-ion lasers, though dye lasers and tunable solid state lasers do offer some selectivity. Clearly, CL has a greater number of mitigating circumstances to contend with in comparison to PL. These factors need to be better appreciated in any comparative study.

The luminescence response of high purity GaAs is highly sensitive to surface and prevailing excitation conditions. Our studies have included: the dramatic effects of low voltage charging on CL properties [19]; a depth-resolved CL investigation of surface passivation with (NH₄)₂S [20]; and a combined PL, CL, and photoreflectance (PR) study of charging and passivation effects [21]. We found a number of apparently contradictory observations. The key points are summarized:

1. An apparent contradiction: (NH₄)₂S treatment was found to improve the electrical properties of the epilayer. The mid-gap Fermi-level was unpinned, but PR indicated an increase in the dark surface electric field! The surface recombination velocity on the other hand actually decreased by at least an order of magnitude (as inferred from CL depth-profiles). Thus, the change in surface states character and density was complex.
2. (NH₄)₂S treatment was found to improve the optical quality of the crystal with tremendous enhancement of room temperature PL (> 500X).
3. (NH₄)₂S treatment also modified transport properties of the epilayer. This was inferred from changes in exciton-polariton (notch) spectral behaviour. One notable conclusion (from low voltage CL) was that surface electric fields in themselves were not solely responsible for this change in behaviour. Current sentiment is that no single mechanism can account for all the diverse behaviour reported on this topic!
4. Room temperature CL improvement with treatment was found to be voltage dependent (as expected), but gains were rather modest in contrast with PL (peak gain < 10X). Under operating circumstances designed to emulate PL penetration conditions, the CL response was particularly weak. This difference could not be explained by optical absorption or excitation density differences. In fact, CL dead layer thicknesses were determined (from depth-profiles) to be quite similar for treated and untreated material (another potential contradiction, with the implication of similar residual fields).
5. The dramatic enhancement in CL under low voltage charging conditions was interpreted in terms of (luminescence) dead layer elimination. The sample behaved as if it were under flat band conditions.
6. Although each optical technique yielded different information, no unified PL/CL interpretation was possible because of complex inter-dependencies. This simply

S. Myhajlenko, R.A. Puechner, J.L. Edwards and D.B. Davito
Photo- and cathodoluminescence

Table 1. GaAs epitaxial film parameters.

<table>
<thead>
<tr>
<th>Sample Identity</th>
<th>Character Type</th>
<th>Carriers (300K)</th>
<th>Epitaxial Thickness</th>
<th>Mobility (300K)</th>
<th>Mobility (77K)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>cm(^{-3})</td>
<td>(\mu)m</td>
<td>cm(^2)V(^{-1})s(^{-1})</td>
<td>cm(^2)V(^{-1})s(^{-1})</td>
</tr>
<tr>
<td>A</td>
<td>p</td>
<td>4.3x10(^{14})</td>
<td>8.0</td>
<td>390</td>
<td>-</td>
</tr>
<tr>
<td>B</td>
<td>n</td>
<td>8.4x10(^{13})</td>
<td>4.7</td>
<td>6300</td>
<td>87000</td>
</tr>
<tr>
<td>C</td>
<td>n</td>
<td>1.5x10(^{14})</td>
<td>9.1</td>
<td>6900</td>
<td>115000</td>
</tr>
</tbody>
</table>

reflects the nature of the dielectric response.

In this article, we probe further the PL/CL response of GaAs with respect to experimental circumstance, chemical passivation, and low voltage charging. We address some of the unresolved issues from earlier work. We present experimental results that compare PL/CL spectral behaviour. In particular, we continue to direct our attention to excitonic properties. Here, we investigate the temperature and injection level behaviour of exciton-polaritons in chemically passivated and as-grown samples. We also perform some physical characterization to augment the optical approach. Specifically, we explore the influence of surface topography on luminescence response with high resolution SEM and atomic force microscopy (AFM). In particular, we focus on the concept of microscopic roughness. Microscopic roughness relates specifically to scattering phenomena associated with spatial fluctuations in the dielectric constant of the medium. This is non-classical scattering since the dimensions involved are much smaller than the wavelength of light. New interpretations address some of the contradictory observations.

Experimental

The samples used in this study have high quality luminescence signatures [8]. The epitaxial layers are nominally undoped (100) GaAs grown by metal-organic chemical vapour deposition on semi-insulating GaAs substrates. The epilayer characteristics are summarized in Table 1. Sample B is likely to be totally depleted of free carriers.

Our routine PL and CL (Oxford Instruments based) experimental set-ups and procedures have been described elsewhere [21]. High resolution SEM was performed on a Hitachi S-5000 field emission microscope and AFM images were acquired with a Digital Nanoscope III instrument.

We progressively modified our (NH\(_4\))\(_2\)S passivation recipe to optimize room temperature PL response by changing the pH and treatment times [34]. We found that a neutral solution yielded better PL performance than a basic solution. For example, a doubling in the peak PL signal enhancement was observed when the pH was decreased from 10.4 to 7.4 (with same treatment time). Unless specified otherwise, we used the following procedures: (i) ultrasonic degrease with 1-1-1 trichloroethane, acetone and propanol; (ii) oxide removal with ammonium hydroxide (30 seconds); (iii) soak in buffered ammonium sulfide (pH = 7.8, 14 hours); (iv) rinse with de-ionized water (15 MO); and (v) dry with purified nitrogen. Further details can be found elsewhere [21].

Results

We present the results in three sections: (i) experimental conditions; (ii) exciton-polariton temperature behaviour; and (iii) AFM observations. Please note we freely interchange the terms passivated and treated in this article.

Experimental conditions

In this section, we describe two practical aspects of performing CL measurements that are important in PL/CL comparative studies: having a reliable knowledge of the sample temperature; and appreciating specific sample response to operational power conditions. We initially used PL to compare our cold-stage temperature readings with the temperature indicated by CL spectral content. The cold stage thermal sensor was located in close proximity to the liquid helium heat exchanger, and thus, temperature readings were typically too low. We replaced the original sensor with a calibrated glass-carbon resistor that became an integral part of the sample holder (imbedded into Stycast 2850FT/9 thermal cement). This allowed samples to be in good thermal contact with the sensor. We then compared again CL spectra obtained at different temperatures (based on sensor calibration chart) with a PL temperature series obtained from the same sample. This procedure worked well. However, the measured sample temperatures did not completely correlate with CL spectral content. We know that the luminescence response of GaAs is highly sensitive at low temperatures to excitation conditions (a
S. Myhajlenko, R.A. Puechner, J.L. Edwards and D.B. Davito

Figure 1. Room temperature CL spectrum from a GaAs quantum wire array excited by 15 keV electrons and corresponding spectrally resolved (1.663 eV) CL image excited by 3 keV electrons.

Figure 2. Electron heating effects in CL spectra with sample C as a function of accelerating voltage for constant beam current (3 nA).

The nature of electron beam excitation means that CL is more sensitive than PL to small adjustments in operating conditions. Therefore, the effective (CL spectral) temperature and the measured (sensor) sample temperature will not necessarily correlate when CL is compared with PL data at the same sample (lattice) temperature ($T_L$). We have used the band-acceptor lineshape to extract information on carrier heating effects [9]. This transition is more sensitive than excitons to "low" excitation conditions. Provided we have non-degenerate excitation conditions, this lineshape $I(h\nu)$ may be interpreted in the form:

$$I(h\nu) \propto E^{1/2} \exp(-E/kT_e),$$

where $E = h\nu - (E_g - E_A)$, $E_g$ is the band gap energy, $E_A$ is the acceptor binding energy (in our case, ($e,A^o$)Carbon), and $T_e$ is the electron temperature [18]. The electron temperature provides a useful measure of the injection level since it indicates how the recombination traffic is responding to excitation conditions. When $T_e$ increases relative to $T_L$, single particle transitions typically begin to saturate, such as free-to-bound ($e,A^o$) and donor-acceptor pairs. Excitonic related processes start to take over when $\Delta T_e = T_e - T_L \gg 0$. Luminescence response to power input prior to saturation can be linear in this regime. Further increases in excitation power can lead to exciton and phonon heating. These processes are also described by an effective temperature [25]. Under these circumstances, the luminescence response is non-linear (usually super-linear in the early stages). It was only under relatively low power excitation conditions that we found $T_e \approx T_L$. We endeavoured to perform CL measurements in the linear response regime since this aided subsequent analysis, especially of depth-profiles [33]. Notably, CL output $\propto IV$ is more manageable than the typical response of CL output $\propto$
Table 2. Comparison of photoluminescence and cathodoluminescence exciton behaviour at 15K from sample C under different surface conditions.

<table>
<thead>
<tr>
<th>Surface Condition</th>
<th>Cathodoluminescence</th>
<th>Photoluminescence</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfur Passivated</td>
<td>1.7</td>
<td>1.1</td>
</tr>
<tr>
<td>Native Oxide</td>
<td>1.3-1.5</td>
<td>1.1</td>
</tr>
<tr>
<td>Carbon Coated</td>
<td>1.5</td>
<td>not available</td>
</tr>
<tr>
<td>Beam Charged</td>
<td>1.1</td>
<td>not available</td>
</tr>
</tbody>
</table>

\[ I \text{ beam current, } V \text{ beam voltage, and } n, m \neq 1 \]. Sample C was particularly favourable in this respect since we found excitation regimes where overall CL response to current and voltage was linear.

Figure 2 shows a semi-logarithmic plot of the CL spectral response from n-type material (sample C) as a function of beam voltage (three selected) under constant current conditions \((T_L = 14K)\). Note how the magnitude of the excitonic notch increases as \(T_e\) and power input increases. The effective exciton temperature remained constant in this experiment. The radiative recombination was dominated by non-excitonic transitions. The magnitude of the excitonic notch was similarly excitation dependent in PL, see Figure 3, but was more strongly dependent on temperature and surface passivation (to be described later). In Figure 4, we show more details of the constant current experiment. In particular, we compare integrated band edge and excitonic CL output, and degree of electron heating \((\Delta T_e)\) versus beam voltage. This representation is complicated by the fact that the sample underwent anomalous (CL) charging between 2-4 keV; see offset (plateau) in both integrated and excitonic outputs. Interestingly enough, this charging did not influence electron (recombination) heating, \(\Delta T_e \propto V^{0.75}\) or the linear integrated CL response \((n = 1)\). However, excitonic behaviour was influenced, and above the charging regime exhibited superlinear behaviour \((n = 1.35)\).

One important difference between PL and CL is the power density used for excitation. Power densities used in CL measurements typically vary from mW cm\(^{-2}\) to MW cm\(^{-2}\). From this perspective, PL can be viewed as a flood illumination version of CL. In our CL experiments, we have used the range mW cm\(^{-2}\) to W cm\(^{-2}\) to best match PL excitation. In Figure 5, we show the effect of SEM electron probe focus on the CL spectral response of p-type material (sample A). Defocussing of the probe (effectively flood illumination) caused the recombination to move from a situation dominated by excitons to one dominated by donor-acceptors (DA). As a result of defocussing, the electron temperature decreased by 6K. This behaviour is commensurate with a significant reduction in excitation density (injection level). The 0.7 meV red shift observed in the DA peak is consistent with this. A corresponding reduction in the magnitude of the excitonic notch was observed with the decrease in excitation density (and beam voltage). The notch is barely discernible in the 0.5 keV spectrum (Fig. 2). In addition, n-type material of comparable doping to sample A (sample C, for instance), exhibited even greater sensitivity to beam focus and power density.

Exciton-polariton behaviour

In our previous study, we noted the sensitivity of exciton-related luminescence (spectral character and signal strength) to injection level, surface passivation, and charging effects [21]. CL exhibited a greater diversity in response than did PL. Exciton injection level behaviour versus surface condition is summarized in Table 2. The low voltage charged CL response of (carbon-coated) sample C is identical to PL for sulfur passivated and non-passivated (native oxide) cases. The carbon coating experiments (thickness < 10 nm) were performed to investigate possible effects from hydrocarbon contamination. In fact, carbon-coated and oxide-coated samples exhibited no measurable differences in behaviour; that is, CL depth-profiles, CL spectral content, etc., were all found to be similar in character and magnitude. With respect to exciton-polariton properties, non-passivated (oxide-coated) samples exhibited the notch effect in both PL and CL. Sulfur passivated samples, on the other hand, typically exhibited a shoulder at the exciton-resonance energy. In the case of PL, exciton injection level dependence was identical for passivated and non-passivated cases \((n = 1.1)\), whereas CL exciton signal behaviour was more varied \((n = 1.1-1.7)\). Yet, exciton-polariton spectral behaviour was the same with PL and CL with respect to oxide-coated (notch) and passivation (shoulder) situations. This suggests a mechanism that affects PL and CL response to power input differently (viz. spectral content and signal strength). For instance, the above differences with passivation may
S. Myhajlenko, R.A. Puechner, J.L. Edwards and D.B. Davito

Figure 3. PL spectra showing the effect of excitation power on excitonic behaviour for sample C. PL laser power increases from 0.03, 0.1 to 0.5 W/cm², respectively.

Figure 4. Electron heating $\Delta T_e$ (see linear scale on left, squares) and integrated CL intensities of band edge (dots) and excitonic (triangles) emissions (see logarithmic scale on right) as a function of accelerating voltage under constant current conditions.

Figure 5. Effect of focus and defocus on CL spectral output from sample A (25 keV).

In Figure 6, we show the temperature dependence of PL spectra from sulfur-passivated and untreated (oxide-coated) samples (C). Note, the strong dependence on temperature of the exciton-polariton notch in untreated material. The most dramatic effect is seen at 30K (see Fig. 7). In passivated material, a shoulder is observed at the resonance energy (around 1.515 eV). Also, it is interesting to note that the notch phenomenon disappears in untreated material as the shallow acceptors ionize with increasing temperature ($T > 60K$). This implies the notch phenomenon is tied to a recombination (mechanism) center within the crystal. A comparison of (Arrhenius) thermal activation energies for excitonic-related processes in the temperature range 25K-100K (decreasing notch strength) yielded $\approx 15$ meV difference in energies between treated and untreated material: untreated samples had the higher activation energy. From a kinetics perspective, this would explain why treated samples are "brighter" in this temperature regime: production and/or escape of exciton-related light is easier.

Surface topography

In Figure 8, we show AFM images and linescans of treated and untreated material (sample B). The image depicts growth terracing (or steps) with spatial frequency in the range (100 nm⁻¹)-(150 nm⁻¹) with some higher frequency corrugation superimposed (7 nm⁻¹)-(10 nm⁻¹). The primary difference between the two images was an increase in the amplitude of the random height corruga-
Photo- and cathodoluminescence

Figure 6. PL temperature dependence of untreated (a) and treated (b) sample C. Laser power $\approx 1 \text{ W/cm}^2$.

...tion (discernible from the linescan profiles) by a factor of 2 to 3 in treated material. This translates to height modulations in the range 0.5-0.75 nm. The treated sample thus appears rougher than untreated (oxide coated) material. The surface terracing was also visible in high resolution SEM ($> 100,000X$) where the contrast consisted of a regular array of wavy dark-bands with repeat wavelength of $\approx 110 \text{ nm}$. This spatial frequency was consistent with the AFM results. In Figure 9, we show much lower magnification ($\approx 10X$) panchromatic room temperature CL images from the same samples. As expected (from previous studies), the treated sample (right-hand side) has higher luminescence efficiency ($\approx 6X$ higher light output) than untreated material (left-hand side). This improvement in luminescence efficiency with GaAs allows the use lower excitation powers in imaging experiments. Under said excitation conditions, the spatial inhomogeneities in the epilayer, replication of substrate cellular structure, are clearly visible in the treated sample. This contrast was not associated with the passivation process, since untreated material exhibited the same features but required higher excitation conditions. Influence of the substrate on subsequent epitaxial quality has also been seen in material grown by molecular beam epitaxy (MBE) and metal-organic vapour phase [29].

Discussion

We focus our discussion on exciton-polariton properties. We consider the significance of the injection level, temperature and surface topography results in light of our previous work [21]. To begin with, we review some recent thoughts and observations on the origin of exciton-polariton notches. From the literature, scattering based mechanisms are currently favoured with polariton descriptions: "Any mechanism that affects the initial distribution either in space or in energy, the transport of polaritons in the crystal, or the nature of the crystal boundary itself, can modify the experimentally observed polariton spectrum. Thus no single mechanism can explain the huge variety of observed polariton spectra in different crystals." [26] We agree with this sentiment.

The reader should be aware of following interpretations of polariton behaviour: (i) optical re-absorption (implicit in the polariton picture) [13]; (ii) spatial dispersion (the dielectric function dependence on wavevector k) yielding upper and lower branches of polariton...
states \[14, 24\]; (iii) differences in surface crystal chemistry \[11\]; (iv) elastic scattering by neutral impurities \[16\]; (v) variations in epitaxial thickness giving rise to different exciton-polariton scattering lengths \[5\]; (vi) polariton group velocity dependence on energy that influences transport to the surface \[26\]; (vii) sub-surface damage \[17\]; (viii) temperature dependence of the exciton-polariton coupling interaction \[35\]; and (ix) enhanced exciton-electron scattering associated with high level crystal excitation \[1\]; other correlations include; (x) presence of dislocations in cell walls \[28\]; (xi) passivation by hydrogenation \[27\]; (xii) passivation with polyimide \[15\]; and (xiii) presence of surface electric fields \[23\].

To conceptualize a unified theory that accounts for all the above observations is not a straightforward matter. To integrate our PL/CL results into such a model, clearly, is beyond the scope of this present work. However, we feel that based upon our observations to date, scattering related theories do currently represent the best approach. We now consider another potential scattering mechanism in light of the AFM results: optical roughness. Optical roughness is localized non-uniformities in structure defined in terms of micro- and macro-scopic effects. Microscopic roughness refers to spatial perturbations in the dielectric constant of the medium. The dimensions of the physical perturbations are by defini-
Photo- and cathodoluminescence

Figure 9. Room temperature panchromatic CL images of untreated (left) and treated (right) samples used in AFM measurements in previous figure (25 keV, 40 nA).

Figure 10. A schematic representation of how microscopic roughness can produce an effective change in the surface layer dielectric function. The surface terracing and higher spatial frequency components as seen in the AFM images are depicted in 1-D slice (a) and 3-D cases (b). The random vertical undulations in the high frequency component cause the volumetric fraction to differ, and hence the effective dielectric constant, from the untreated to treated sample. See text for further details.

The AFM images in Figure 8 clearly indicate that the surfaces of passivated and untreated GaAs are microscopically rough: compare the dimensions of the corrugation with those of free excitons (~30 nm) and photons (~818 nm). In Figure 10, we illustrate qualitatively how the AFM results can translate into layers of different dielectric properties, where we use the effective medium representation [3]. In our case, we depict a 1-D slice of the surface layer, though the real circumstance is a 3-D problem. In Figure 10, ε and ε' represent the dielectric function (composed of real ε and imaginary ε' parts) of the sample and vacuum, respectively: ε is primarily sensitive to roughness. An average effective dielectric function can be defined based on the volume fraction changes in the vertical distribution of microscopic roughness. This value will be different for the treated sample compared with the untreated case. For simplicity, we have ignored the effects of the oxide overlayer (in untreated) and sulfide overlayer (in treated without oxide) in order to illustrate the effect of microscopic roughness. The real picture is clearly more complex when these other factors are considered in the overall optical response of the solid. The passivated surface appears rougher because of the increased amplitude of the higher spatial frequency corrugation. This change in roughness is difficult to define since Fourier analysis is required over many orders of magnitude of spatial dimensions. In fact, in some instances, surface roughness has been studied by fractal geometry [22]. Here, we wish simply to point out that a correlation exists between the change in exciton-polariton behaviour with treatment (notch to shoulder) and a change in surface microroughness. This observation is pertinent to the controversy over whether luminescence is useful for characterizing the interface roughness of quantum-well structures [4, 32].

In one study of undoped MBE grown (100) GaAs, a changeover from a double peak (notch) to a single peak (shoulder) was correlated with scattering center concentration and sample thickness [5]. However, in that study, the authors used some undefined etch to reduce the thicknesses of the epilayers. We point out that microscopic roughness of (100) GaAs surfaces is strongly dependent on the pH of etchant solutions [2]. It is conceivable that the above authors generated similar
passivation was observed to correlate with a dramatic change in exciton-polariton behaviour. A corresponding increase in high spatial frequency corrugation with surface microroughness at the Si(100)-SiO₂ interface. The effects of microscopic roughness should be considered in luminescence interpretation, especially with reference to excitons and polaritons. Namely, a significant effect on incoming electrons, then from the CL perspective, the CL increase comes only from reduced surface recombination. Any changes in optical extraction with passivation should be similar with PL and CL. In our case, the primary difference in PL/CL response is determined by relative excitation power coupling.

Conclusions

The temperature behaviour of the excitonic notch in untreated GaAs is consistent with previous observations [35]. However, we are not sure how the passivated data (excitonic-shoulder) fits into a model based on exciton-acoustic phonon scattering. In this approach, at some critical temperature (20K-30K), the polariton picture (strong exciton-photon coupling) changes over to a situation where independent excitons and photons exist (weak interaction). The previous study did not investigate passivated surfaces. How the difference in activation energies for excitonic emission in passivated and untreated material reflects on acoustic-phonons is unclear at present, especially since we have correlated the disappearance of the notch/shoulder exciton spectral feature with acceptor ionization.

Microscopic roughness will influence differently the coupling of various excitations into and out of a solid. This is relevant to PL and CL comparative studies for reasons mentioned earlier. Our previous PR measurements and the observed differences in room temperature luminescence enhancement between PL (> 500X) and CL (< 10X) with treatment [21] could be explained in terms of microroughness. We have inferred that a change in surface roughness influences polariton luminescence at low temperatures. Here is a qualitative argument for the room temperature observations. The magnitude of the PR signal is consistently larger with treatment [21]. In light of the AFM observations, this suggests that more of the laser (light) power is coupled into the crystal relative to the untreated (oxide coated) case. The subsequent increase in generated excess carriers, coupled with the inferred reduction in surface recombination velocity, will clearly produce more PL. If we assume that this change in microroughness has no significant effect on incoming electrons, then from the CL perspective, the CL increase comes only from reduced surface recombination. Any changes in optical extraction with passivation should be similar with PL and CL. In our case, the primary difference in PL/CL response is determined by relative excitation power coupling.

Acknowledgements

This work was made possible by an ASU Research Incentive Award. The authors express sincere thanks to H. Song and D.R. Allee for the AFM images, B.J. Skromme for the quantum wire array sample, and Nissei Sangyo America Ltd for high resolution SEM.

References

Photo- and cathodoluminescence

8171–8186.


