Journal of Luminescence 7 (1973) 284–309. North-Holland Publishing Company

OPTICAL GAIN IN SEMICONDUCTORS

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Direct measurements of optical gain in semiconductors give information on the stimulated recombination process that cannot be easily obtained from an oscillating semiconductor laser. The effects of various material parameters on the gain and saturation factors can be easily studied. The temperature dependence of the stimulated recombination in high purity GaAs will be presented as an example. The pertinence of the gain and saturation factors to the threshold and efficiency of semiconductors laser oscillators will be discussed. The significance of gain measurements on GaP doped with isoelectronic traps will be considered in terms of the observed gain and saturation properties.

1. Introduction

We describe a technique for studying optical gain spectra in semiconductors which involves the measurement of the single-pass, amplified spontaneous emission [1–4]. Optical gain has long been recognized as an important parameter in evaluating the possible utility of laser systems [5], yet in most semiconductor laser systems, historically, gain parameters have been determined from threshold condition and only after successful laser oscillators have been fabricated [5]. Such measurements of gain are made after the fact, and so are not particularly useful for evaluating potential laser materials. Further, the gain is determined from laser threshold, so that information is obtained on only one point in the gain spectrum. Since it is generally observed [2, 6] that semiconductors have broad-band gain, it is important to have information on the entire spectrum. In addition to this advantage as a gain measuring technique the method described here also provides information on the spontaneous emission, loss and saturation characteristics of the material. Hence, its usefulness is not limited to evaluating potential new laser materials [7–10], and it can be used for optimizing the material parameters (e.g., doping, alloy composition, etc.) of known semiconductor laser materials. Moreover, knowledge of the properties of the gain spectrum can lead to an understanding of the physics of the gain processes [10], which in turn, can suggest ways of improving laser operating characteristics.

In this paper, we first discuss the experimental technique. Then we present data to show the range of conditions over which measurements can be made and to demonstrate the various types of effects that can be observed. Following this we anal-

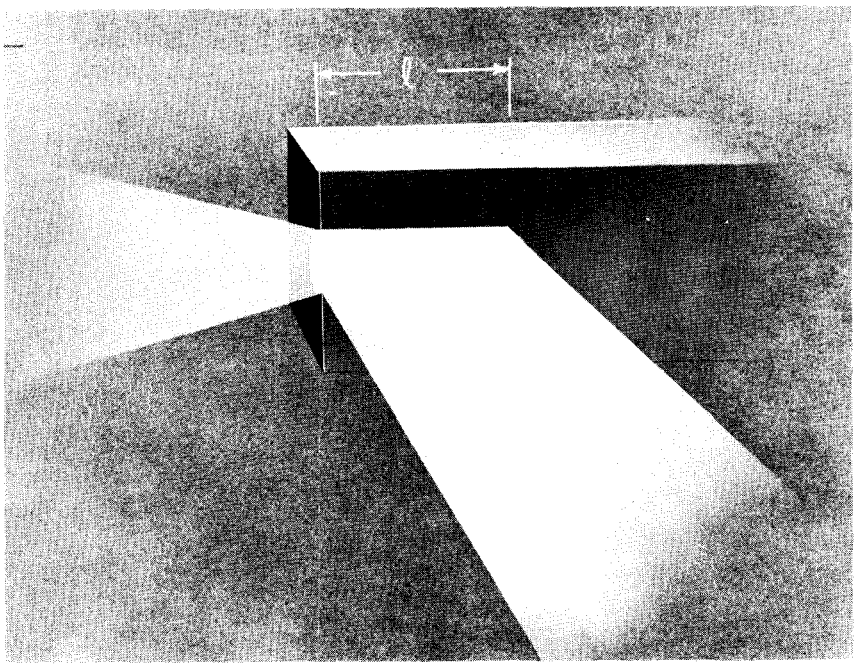

Fig. 1. Schematic of excitation geometry for stimulated emission measurements in semiconductors. A beam of light from the right excites a region of lenght ℓ on the sample. The stimulated emission passes out the edge of the sample to the left.

yze the highly excited (gain) region of the semiconductors as a parameterized optical amplifier. The model chosen is quite elementary, but it provides a good description of the experimental observations. Finally, we illustrate the significance of these measurements to the design and operation of semiconductor lasers.

2. Gain measurement technique

In conventional laser systems, optical gain is usually measured by injecting a probe signal and observing the build-up of this signal as it passes through the amplifying medium [1]. This approach is not particularly applicable to semiconductors for two reasons: (1) the gain is usually confined in a thin region of the material and hence, it is geometrically difficult to match the injected signal to the amplifying region, (2) since semiconductors are typically high gain devices, the super-radiant (amplified spontaneous) emission may overwhelm the injected signal. However, it is possible to make use of the super-radiant emission to determine the gain, as illustrated in fig. 1. Here we show a semiconductor being excited by a beam from a pump source at the right. For the present discussion, the important dimension is the

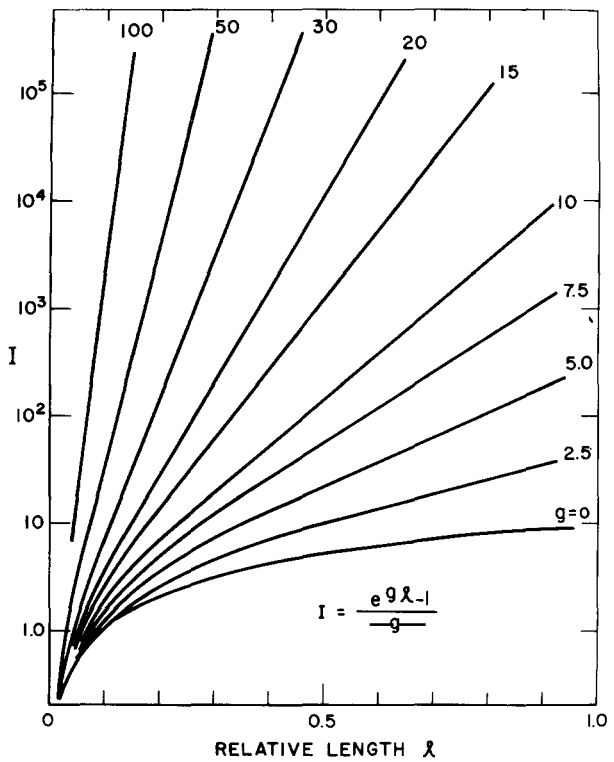

Fig. 2. Expected variation of the amplified luminescence intensity with excitation length for different values of the gain.

length ℓ of the excited region. We consider that the resulting excitations may decay via spontaneous emission with power density J_s (W/cm^3 steradian) or via stimulated emission which gives rise to an optical gain g(cm^{-1}). Let us consider the spontaneous emission into the small solid angle Ω along the axis of the length of the excited region. It is easily shown [3] that the intensity of the amplified spontaneous emission passing out the end of an excited region of length ℓ is given by

$$I(\mathrm{W/cm^2\ steradian}) = \frac{J_s \Omega}{g}(e^{g l} - 1). \tag{1}$$

A plot of intensity as a function of length of the excited region (according to eq. (1)) is given in fig. 2 for different values of the gain parameter g. Note that the curves typically show two regions. For instance, for the curve marked $g = 10$, the intensity is increasing linearly with length for $l < 0.2$. In this regime, the exponential in eq. (1) can be expanded and we find that spontaneous emission dominates the output intensity. For $l > 0.3$, the exponential term dominates eq. (1). The out-

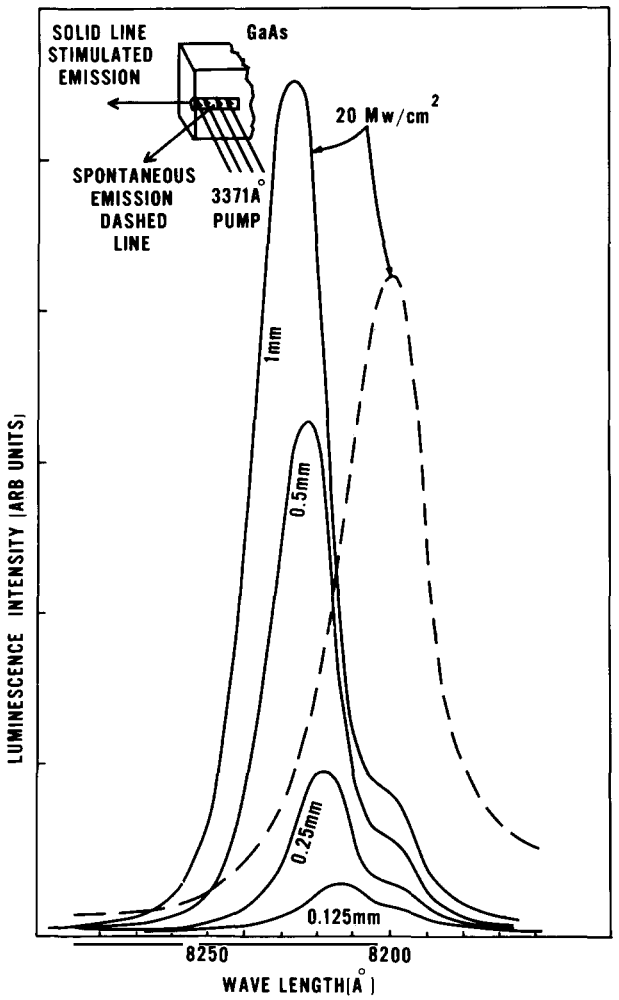

Fig. 3. Stimulated emission spectrum of GaAs at 2°K for several different excitation lengths. The spontaneous emission spectrum is shown as the dashed line. The excitation intensity for all spectra is 20 MW/cm^2.

put consists mainly of amplified spontaneous emission, and the slope of the straight line portion of the plot is the gain factor g. So we see that by measuring the amplified spontaneous emission intensity as a function of the length of the excited region, we can determine directly the optical gain factor g.

The above discussion has treated a highly excited semiconductor as a simple one-dimensional optical amplifier. This approach has been shown to work in gas laser [3, 4] and liquid-organic dye laser [11] systems. What we will show here is that the

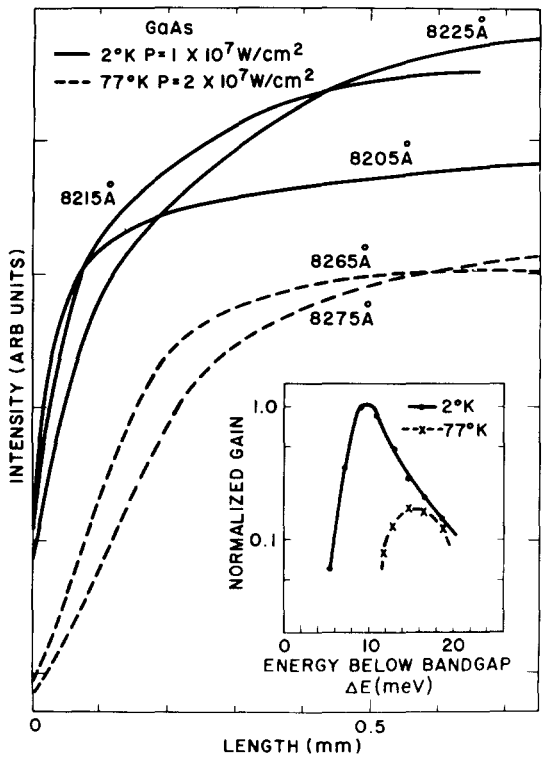

Fig. 4. Plots of the variation of the stimulated emission intensity with excitation length for 2°K and 77°K for several different wavelengths. The gain spectra obtained from a family of such plots are shown in the insert.

experimental observations on stimulated emission in semiconductors are, to a large degree, consistent with the assumption of a one dimensional amplifier. We will use these observations to gain insight into the physics of stimulated recombination processes.

The determination of gain described above is based on the dependence of the observed amplified luminescence on the length of the excited region. An example of this dependence is shown in fig. 3. Here we show the amplified luminescence spectrum of a lightly doped epitaxial layer of GaAs at 2°K for the different excitation lengths indicated for each curve. Note that the pump intensity is the same for all the spectra shown. The effects of a factor of 2 change in length are different for various portions of the spectrum. On the short wavelength side ($\approx$ 8210 Å) the luminescence intensity appears to vary linearly or sublinearly with increasing excitation length, while on the long wavelength side ($\approx$ 8230 Å) the luminescence intensity increases superlinearly with excitation length. Such a superlinear dependence

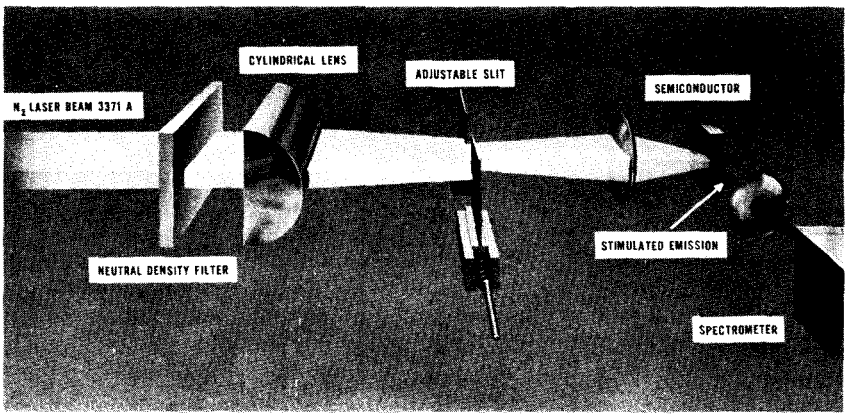

Fig. 5. Experimental arrangement for gain measurements.

is an indication of the existence of optical gain. This indication can be checked by plotting the luminescence intensity as a function of excitation length as suggested by eq. (1). Such a plot is shown for GaAs in fig. 4. Data is shown for both 2°K and 77°K. Note that exponential regions are observed for both temperatures. By measuring the slopes of these exponential regions, gain spectra can be obtained as shown in the inset. Saturation of the luminescence is observed at long lengths and will be discussed further later.

Before discussing the details of stimulated emission in semiconductors, let us consider how the measurements have been made. The experimental arrangement is shown in fig. 5. Here we show pump radiation coming from the left. Typically the pump source is 3371 Å light from a pulsed (10^{-8} sec/pulse at 100 sec^{-1}) N_2 laser. The cross-section of the laser beam is rectangular with typical dimensions of 5 X 0.2 cm and the intensity distribution along the long dimension is uniform to within 2%. The laser beam passes through a variable attenuator (neutral density filter) and is focused to a line by a cylindrical lens. An adjustable slit assembly placed at the focal line of the cylindrical lens, is used to control the length of the exciting beam. The slit (and hence the laser light in the plane of the slit) is imaged on the sample by means of a spherical lens. The slit assembly has been constructed such that it has one fixed jaw and one movable jaw. The optics are arranged such that the image of the fixed jaw occurs at the edge of the sample, while the image of the movable slit moves away from the edge of the sample as the slit width is increased. This arrangement assures that the length of unexcited sample in the optical path does not change as the length is varied.

The magnification factor of the optics determines the size of the excited area of the sample relative to the size of the pump beam passing through the slit. Typically the magnification factor is 0.1, which results in an excited region with a width of less than 10 μ and a maximum length of about 1.5 mm. Under these conditions the maximum excitation intensity at the sample surface is $\approx 2 \times 10^7$ W/cm^2.

The amplified luminescence (stimulated emission) which passes out the edge of the sample, is focused on the entrance slit of a spectrometer. The output of the spectrometer is detected by a fast logarithmic amplifier averaged by a boxcar integrator, and digitally processed by a dedicated digital computer. The short light pulses and the wide dynamic range of light intensity used present stringent boundary conditions on the optical detection procedures. The photomultiplier should have a risetime short compared to the pulse length of 10^{-8} sec, should be linear over a wide dynamic range (at least four decades), and should have a large maximum anode current for ease of subsequent electronic signal processing. For this purpose we have chosen an Amperex tube with 14 stages and a typical operating gain of 10^7. The risetime is 2.5 nsec, and the peak anode current is greater than 100 ma. Both of these parameters are acceptable for most stimulated emission measurements. Detection and averaging of the photomultiplier current pose further problems. Since the stimulated emission intensity can range over several orders of magnitude for small changes in experimental parameters, it is necessary to convert the data to a form that can be handled and displayed (over at least four decades) by conventional electronics. For this purpose, it is convenient to logarithmically compress the data with a logarithmic amplifier as mentioned above [12]. The logarithmic amplifier provides an output voltage pulse proportional to the total energy in the measured light pulse (i.e., total charge per pulse from the photomultiplier). The voltage pulses are then detected and averaged by a boxcar integrator. The output of the boxcar integrator can be displayed on an X-Y recorder, or stored digitally on magnetic tape by a dedicated small computer.

The dedicated computer provides several useful services in this experiment. In addition to storing data and performing numerical analysis, it controls the important experimental parameters during data acquisition. These parameters are the spectrometer wavelength, the excitation length, and the averaging time at a given data point. These programmed functions are necessary due to the large amount of data required to characterize the gain spectra of a given material [13]. This will be demonstrated in the following sections.

There are three important independent experimental parameters that can be varied with the measurement technique, described above. Namely, length of excited region, wavelength of the detected amplified luminescence, and excitation intensity. Normally the order of importance is as listed above, and we are prompted to take data as shown in fig. 4. Here we have taken the excitation length as the independent variable, the wavelength as the experimental parameter, and have held the excitation intensity fixed. Alternatively, we can choose the wavelength as the variable, and either excitation intensity or excitation length as the parameter as shown in fig. 6A and B, respectively. Notice that these curves have several features in common: (1) the stimulated emission intensity increases more rapidly than linear with increase of either excitation intensity, or excitation length. (2) The maximum stimulated emission intensity occurs at longer wavelengths than the spontaneous luminescence. (3) The peak stimulated emission intensity shifts to longer wave-

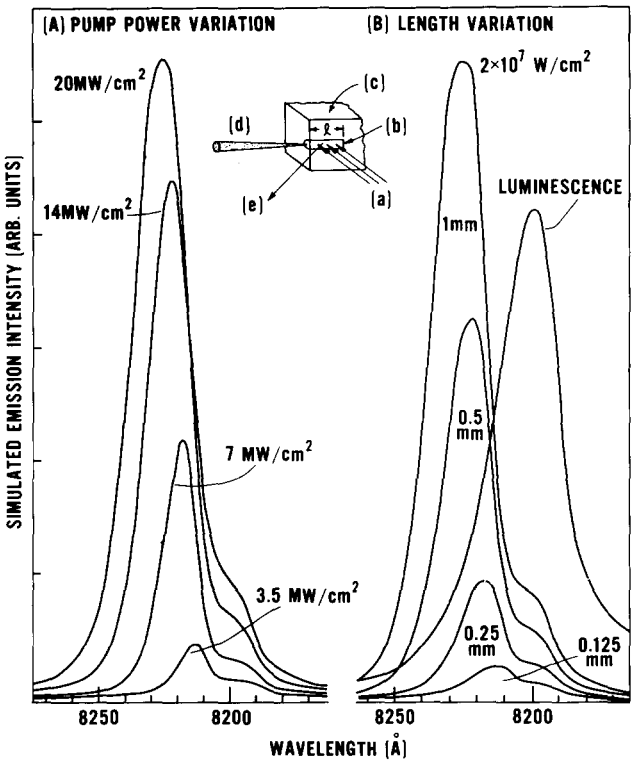

Fig. 6. Stimulated emission spectra of GaAs at 2°K. A) Spectra for an excitation length of 1 mm with various excitation intensities. B) Spectra for an excitation intensity of 20 MW/cm^2 with various excitation lengths. The spontaneous emission for this excitation level is also shown, labeled "luminescence".

lengths with an increase in either parameter. These observations have bearing on some of the observed properties of GaAs lasers [6] and on some of the mechanisms proposed for stimulated radiative recombination in GaAs, and will be discussed in more detail later.

3. Experimental results

A larger number of materials have been studied using the gain measurement technique. A list of these materials and a summary of the results are given in table 1. We have already shown data for GaAs, and will discuss this data in detail in a later section.

A length plot for another III-V semiconductor, GaN [8], is shown in fig. 7. This material is interesting in that the stimulated emission occurs in the near UV at

Table 1
Gain measurement results

	Material	Nominal gain (cm^{-1}) for 10^7 W/cm^2 and 2°K	Wavelength of peak gain Å =	Comments
III-V	GaAs	2,000	8200	pure material $n \sim 10^{15}$; $\mu \sim 10^5$
	GaP : N	10,000	5400	isoelectronic trap
	GaP : Bi	175	5554	induced stimulated emission
	GaN	10,000	3590	measured for 2.5×10^6 W/cm^2
II-VI	CdS	200	4905	Cd rich material
	CdSe	1,000	6840	Cd rich material
I-VII	CuCl	6,400	3920	Excitonic molecule
III-VI	GaSe	10,000	6000	ϵ polytype

3.46 eV, illustrating that the technique applies over a wide spectral range. Further, it is interesting to note the values of the observed gains as indicated on fig. 7. The maximum observed gain is greater than 10^4 cm^{-1} for an excitation intensity of about 2.5×10^6 W/cm^2, and the gain appears to vary linearly with excitation intensity. We would estimate at the maximum available excitation level of 10^7 W/cm^2, the gain would reach 10^5 cm^{-1}.

It is appropriate to compare these gains observed in semiconductors with some common laser systems. A typical gas laser such as Ne–Ne may have a typical gain of 10^{-4} cm^{-1} [5], an argon ion laser may have a gain of 10^{-2} cm^{-1} [5], and dye lasers typically have gains of the order of 1–10 cm^{-1} [11]. The observations of gains in semiconductors that are several order of magnitude larger than in gas lasers is not so surprising if one considers that the density of excited states is much larger in a semiconductor than in a gas. We can compare the observed gain to typical absorption coefficients in semiconductors. It is known that absorption coefficients can easily reach 10^4–10^5 cm^{-1} for photon energies larger than the band gap energy [14]. Thus one might connect the large gain to band-to-band recombination of degenerate electron and hole gases in the highly excited semiconductors [15, 16]. Such a model cannot explain many of the observed properties of the stimulated radiative recombination in semiconductors, but it does provide some justification for the large values of the measured gains [17]. There has been much comment and speculation on the various processes which may give rise to gain in highly ex-

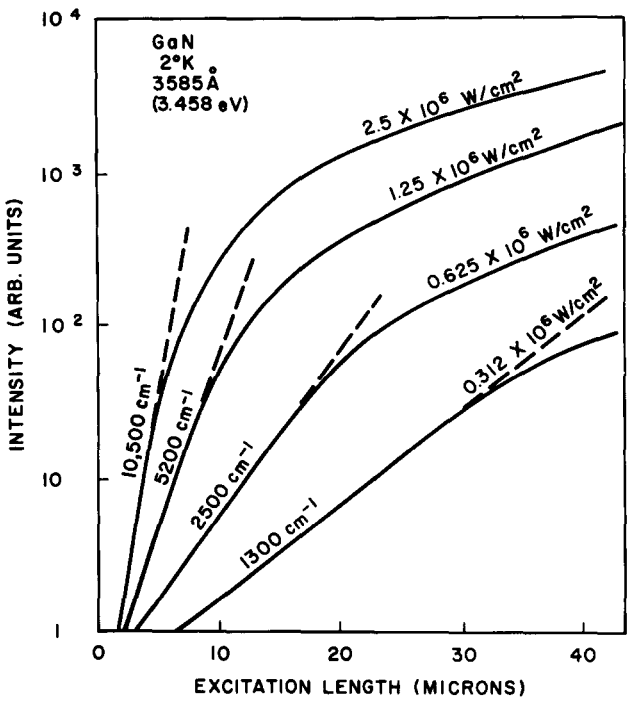

Fig. 7. Plots of the stimulated emission intensity of GaN at 2°K for several different excitation intensities.

cited semiconductors, but an adequate theory has not yet been presented. This problem has been discussed in detail by M. Pilkuhn in the preceding paper.

To illustrate that stimulated recombination need not be simply related to band-to-band recombination, we have investigated the recombination of excitonic molecules in CuCl. An excitonic molecule corresponds to an elementary excitation of the crystal that involves a bound state for two electrons and two holes analogous to the hydrogen molecule [18]. The excitonic molecule in CuCl has a binding energy of 44 meV and thus the luminescence associated with the radiative dissociation of this complex is shifted significantly below the energy gap in this material [19]. Thus CuCl is a unique material in which to study excitonic-molecule recombination radiation.

Since the excitonic molecule forms, in effect, the lowest energy excitation in the system, its decay is mainly radiative and it should provide a useful reservoir for stimulated emission [20]. The existence of stimulated emission from the excitonic molecule is suggested in fig. 8, where for a factor of 4 increase in excitation intensity (6.2×10^5 W/cm^2 to 2.5×10^6 W/cm^2) the intensity of the stimulated emission intensity due to excitonic molecule recombination has increased by a factor of

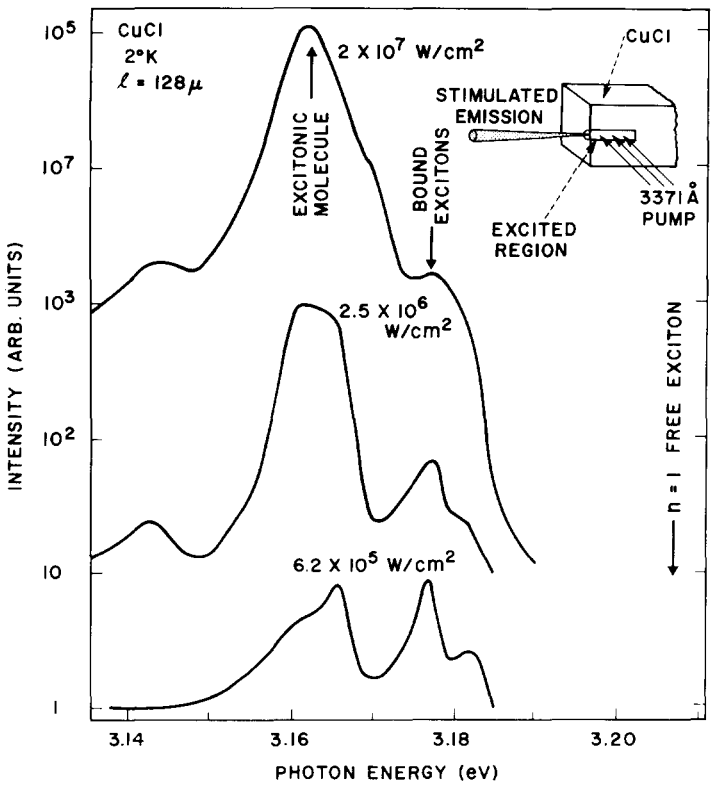

Fig. 8. Stimulated emission spectra of CuCl at 2°K for several different excitation intensities. The energies of the excitonic molecule and the $n = 1$ free exciton are shown by arrows.

100. A more conclusive test for the existence of stimulated emission or gain is to use the length variation technique described above [10]. The data obtained from such a measurement is shown for the excitonic molecule recombination in fig. 9 for several different excitation intensities. Note that well-defined exponential regions exist for all the various excitation intensities and values of the gain can be easily determined as shown. As in GaN [8], the gain is linear in excitation intensity, but the values of the gain are roughly a factor of 10 less than for GaN at the same excitation intensities. There are many possible reasons for this difference (matrix elements, surface recombination, etc.) and it is not pertinent to consider this feature in detail here. An analysis of this data in terms of a one-dimensional amplifier model will be given later in this paper. The important point to be made here is that there is gain associated with excitonic molecule recombination, and that the existence of this gain is readily and conclusively identified through the use of the length-variation gain-measurement technique [10].

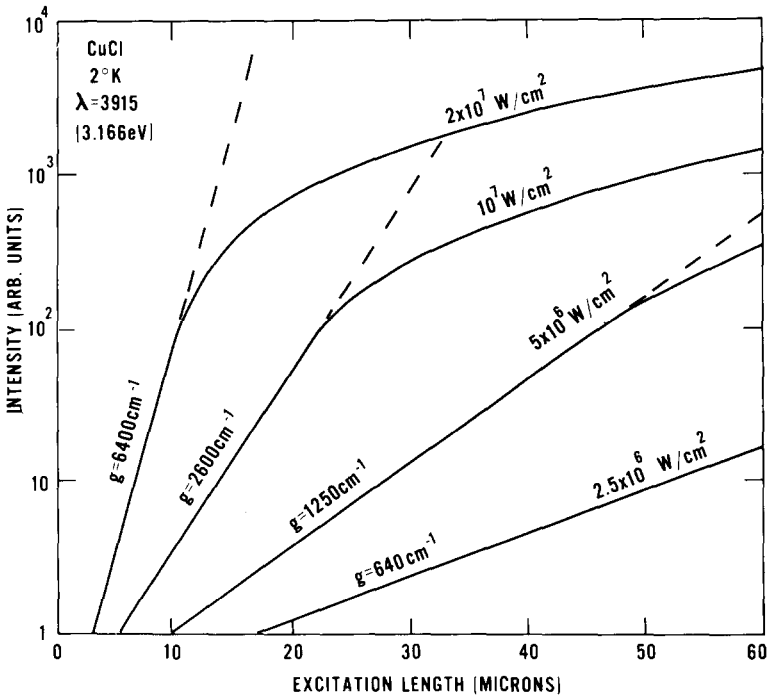

Fig. 9. Plots of the stimulated emission intensity of CuCl at 2°K for several different excitation intensities. The measured gain for each intensity is indicated.

All the materials we have described thus far have direct band gaps. It is of interest to ask if it is possible to observe optical gain in indirect gap semiconductors. This question has significance in that intrinsic radiative recombination in an indirect gap material is a second order process involving a momentum conserving phonon, and is therefore generally weak. Optical gain associated with the radiative recombination of free electrons and holes (or free excitons) in an indirect gap semiconductor has not been identified. Certainly such gain should exist, but it is quite likely that it is small and therefore difficult to observe [21].

It is possible to break the momentum selection rule in the indirect gap material GaP by the use of isoelectronic impurities such as N and Bi [22, 23]. Since these impurities are isoelectronic with phosphorous, the impurity potential has only a short range, and the wave function of any impurity found in this local potential should have a large wave vector spread [22]. For the case of excitons bound to N impurities in GaP, it has been shown that the bound exciton has significant magnitude of the wave function for all values of the crystal momentum between the X point and the Γ point in the Brillouin zone [24]. It is well known that the relaxation of the momentum selection rules by N impurities in GaP gives rise to more efficient luminescence than is observed in undoped GaP [22].

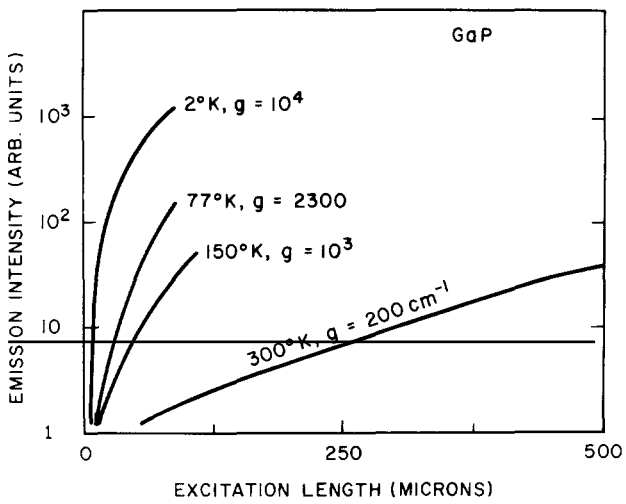

Fig. 10. Plots of the stimulated emission intensity as a function of excitation length for GaP doped with nitrogen. The plots are made at the peak of the stimulated emission spectra for several different temperatures. The values of the measured gains are indicated. The excitation intensity is 20 MW/cm^2.

We have thus investigated the possibility that this could give rise to stimulated recombination in GaP [9]. In fig. 10, we demonstrate stimulated radiative recombination, in nitrogen-doped GaP. We show plots of the stimulated emission intensity of GaP as a function of the length of the excited region of the sample (the sample was solution grown and contained approximately 10^{17} N/cm^3). Measurements were taken at the peak of the stimulated emission spectrum ($\approx$ 5400 Å at 2°K) for several temperatures between 2°K and 300°K. The excitation intensity was 10^7 W/cm^2. Note that at 300°K, the plot shows a well defined exponential behavior and a gain of 200 cm^{-1} is measured. As the temperature is lowered, the measured gain increases markedly, and at 2°K the gain is $\sim 10^4$ cm^{-1}.

Similar results are shown for Bi doped GaP in fig. 11. Here measurements were made on the "A" line of the Bi bound exciton for two different temperatures. Representative data points are indicated and the solid lines represent a theoretical fit to be described in detail in a later section. Note again the well defined exponential regions corresponding to gains of 175 cm^{-1} at 2°K and 2000 cm^{-1} at 23°K. In contrast to the N assisted recombination, the gain on the Bi "A" line increases with increasing temperature up to about 30°K. This can be understood in terms of thermal activation of the Bi "A" level [25].

The important observation [9] concerning stimulated emission in GaP is that the isoelectronic impurities N and Bi can contribute to a relaxation of the momentum selection rules and allow the observation of optical gain. The role of these isoelectronic impurities is confirmed by observing that the stimulated and sponta-

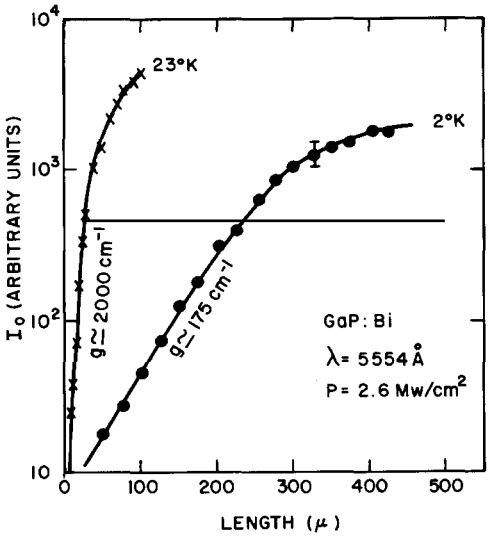

Fig. 11. Plots of the stimulated emission intensity as a function of excitation length for GaP doped with bismuth. Data is shown for sample temperatures of 2°K and 23°K. The solid lines represent fits to the data as described in the text.

neous emission associated with N occurs in a spectral region near 2.3 eV [22] while the stimulated and spontaneous emission associated with Bi occurs in a spectral region near 2.2 eV [23].

The details of the processes giving rise to stimulated emission in indirect gap materials doped with isoelectronic impurities are not clear, but the evidence supporting large-gain stimulated emission is quite extensive [26]. Stimulated emission associated with the isoelectronic complex Zn–O has also been reported [27]. A detailed discussion of the experimental observations in indirect gap materials will be given elsewhere.

We have presented data on stimulated emission and gain measurement for four materials covering a wide range of physical properties and have summarized the results for a wide range of materials in table 1. The characteristics of the stimulated emission in these materials have a good deal in common. The stimulated emission occurs below the band gap, and the measured gains (10^2–10^4 cm^{-1}) are large compared with the gain observed in gas and liquid laser systems. In order to identify further the stimulated recombination processes from the experimental data, it is necessary to extract, from the experimental data, those parameters that can be compared with the predictions of theoretical models.

In the following sections, we will develop our understanding of the gain measurement technique by treating the excited region of the sample as a one-dimensional optical amplifier. Then we will develop some methods for analyzing the data in terms of this simple model, and treat in detail the case of GaAs.

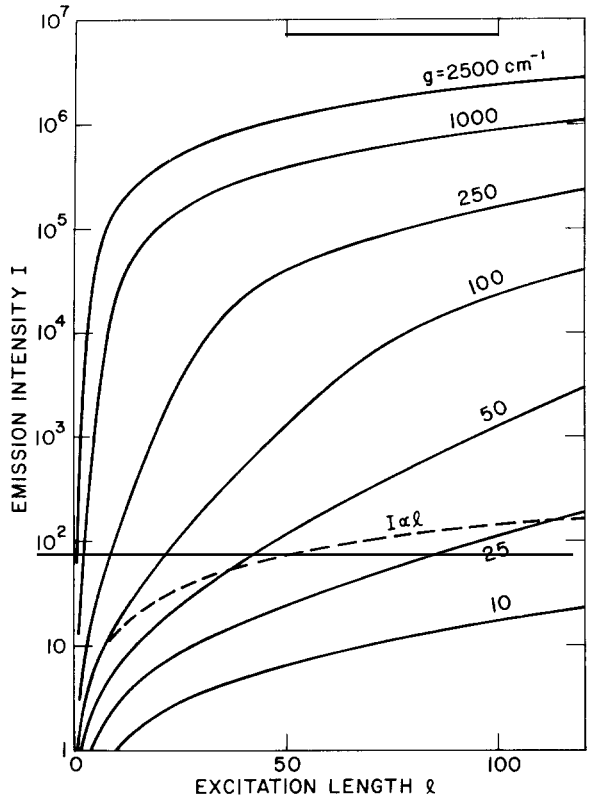

Fig. 12. Plots of expected variation of stimulated emission intensity with excitation length when saturation is included in the one-dimensional optical amplifier model. The different curves correspond to different values of the gain as indicated. The dashed curve, representing a linear relationship $I \propto l$, is given for reference. The length scale is in units of 10^{-4} cm.

4. One dimensional optical amplifier

We return now to the question of saturation of the stimulated emission at long excitation lengths as observed in much of the data presented above. This saturation is to be expected since the maximum output intensity of the amplifier must ultimately be limited by the intensity of the pump source exciting the amplifier. When the amplifier is completely saturated, the maximum output intensity will vary linearly with excitation length. This can be included in eq. (1) by writing it in the following form [28]:

$$gl = \alpha I + \log_e (\beta I + 1). \tag{2}$$

For $\alpha I \ll 1$ the logarithm term dominates the rigth-hand side and eq. (2) is equivalent to eq. (1) with $\beta = g/J_s\Omega$, hence the intensity varies with length as shown in fig. 2. When I becomes large such that $\alpha I \gg 1$, the linear term dominates the right-hand side of eq. (2) and the system is in saturation. Thus α determines how the system saturates and is defined to be the saturation parameter. Fig. 12 shows plots of eq. (2) for different values of the gain parameters g.

Let us examine in detail the curve marked $g = 100$. Note the three rather distinct regions in this plot. For $l < 5$ the intensity varies linearly with excitation length. As noted above, the output intensity is dominated by spontaneous emission in this region. The region of unsaturated gain is seen for $10 < l < 60$. For $l > 60$, the stimulated emission begins to saturate, and finally becomes linear in excitation length. For large gains the system saturates at short lengths as illustrated by the curve marked $g = 2500\ \text{cm}^{-1}$. The variation with length indicated in fig. 12 is in qualitative agreement with the data shown in the previous figures, and it appears that saturation effects do play an important role in the stimulated emission processes in semiconductors.

The applicability of eq. (2) to the experimental data can be checked by a fitting procedure. A least squares fit of data on CdS at 80°K to eq. (2) has been reported

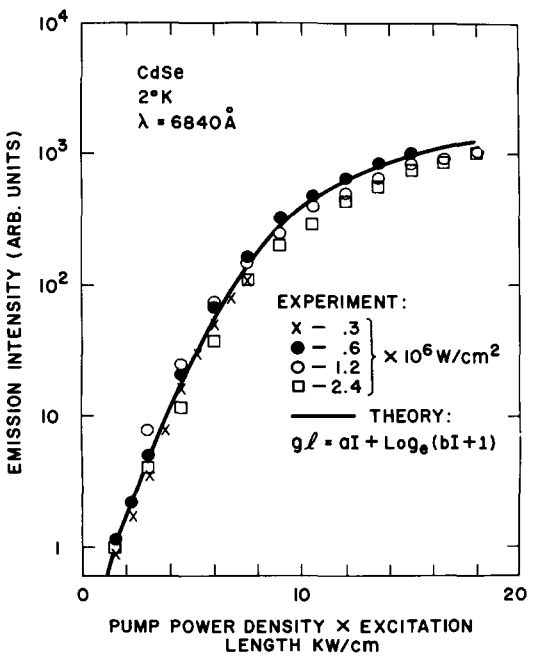

Fig. 13. Composite plot of stimulated emission intensity as a function of excitation length for CdSe at 2°K. The abscissa is plotted as the product of the excitation intensity and excitation length. The solid line is a theoretical fit to the one-dimensional optical amplifier model with saturation included.

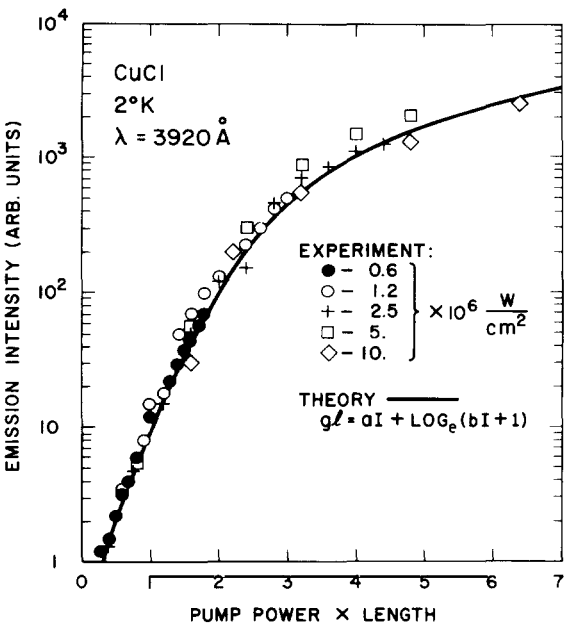

Fig. 14. Composite plot of stimulated emission intensity as a function of excitation length for CuCl at 2° K. The abscissa is plotted as the product of the excitation intensity and excitation length. The solid line is a theoretical fit to the one-dimensional optical amplifier model with saturation included.

previously. Through the use of the fitting procedure spectra of the parameters α, β, and g were obtained.

As further evidence of the applicability of the one-dimensional model, we show data for CdSe, and CuCl. In fig. 13 we show a composite of data for CdSe taken at the peak of the stimulated emission spectrum at 2°K. The data were taken at four different pump intensities and then plotted on the same length scale by using the pump intensity as a scaling factor. Note that the experimental points all lie close to the single theoretical fit to eq. (2) given by the solid line. It is perhaps surprising that data taken with pump intensities ranging over an order of magnitude can be scaled and fit with one theoretical expression. However, if we assume that the gain is proportional to the pump intensity, i.e., $g = \gamma P$, then we can write

$$\gamma Pl = \alpha I + \log_e (\beta I + 1). \tag{3}$$

It is easily seen that the product of the pump intensity and excitation length (Pl) is a convenient variable for displaying data obtained for a wide range of pump intensities. A similar plot is shown for CuCl in fig. 14. Here the data was taken on the peak of the stimulated emission line associated with excitonic molecule recom-

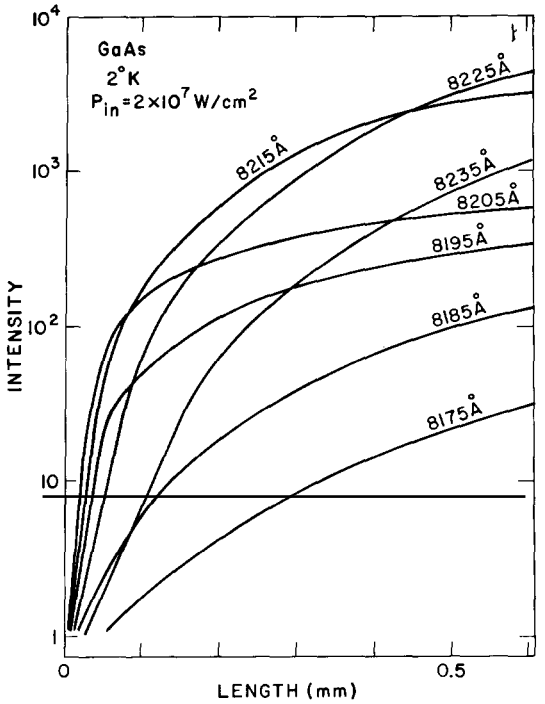

Fig. 15. Plots of the variation of the stimulated emission intensity as a function of excitation length for several wavelengths.

bination. Again the data scales with pump intensity and is fit by a single theoretical curve.

From these observations, one must conclude that the gain is linearly proportional to the pump intensity. Similar observations have been made in several other materials. However, in the case of CuCl this result is quite surprising since the spontaneous emission intensity and the saturated stimulated emission have been observed to vary quadratically with excitation intensity. A detailed discussion of this effect will be given elsewhere [17].

The agreement between the experimental observations in CuCl, CdSe, CdS and GaP and the one-dimensional model justify the use of this model as a starting point for understanding the physics of the various possible recombination processes. A more detailed discussion of eq. (3) starting from a simple four-level system has been given elsewhere [17]. For the ensuing discussion, we apply this one dimensional model to discuss effects observed in GaAs and consider the consequences of these effects on a usable device.

5. GaAs

Referring to figs. 3 and 4, we have illustrated how gain spectra can be obtained for a given material like GaAs. In addition, in fig. 6 there are two other important, and quite general, observations that can be made. First, the maximum intensity of the stimulated emission occurs at lower energy (longer wavelength) than the maximum intensity of the spontaneous luminescence. Second, the stimulated emission peak shifts to lower energies with increasing excitation intensity. These two effects are quite common to a rather large class of undoped or lightly doped semiconductors. There has been considerable discussion of the origin of this effect [27, 30], and in particular, for the case of GaAs, an electron-hole-lattice interaction [16] that results in a shift of the energy gap to lower energy [31], has been proposed as an explanation. However, using the measurement technique described earlier in this paper, we are able to obtain more detailed information on these effects [6], and thereby more closely limit their possible origins. Note in fig. 6 that as either excitation length or pump intensity is increased, the stimulated emission spectrum shifts to longer wavelengths. These observations are not consistent with a bandgap shift caused by excitation density. In the case of fig. 6B, the excitation intensity is held contant, while only the excitation length is increased. Thus, it is only possible for the equilibrium density of electrons and holes to decrease as the stimulated emission intensity saturates at long excitation lengths. Yet, we still observe a shift of the emission to longer wavelengths.

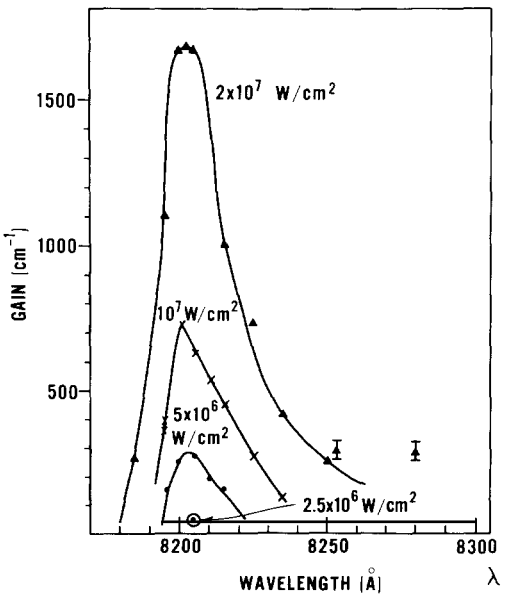

Fig. 16. Optical gain spectra of GaAs at 2°K for several different excitation intensities.

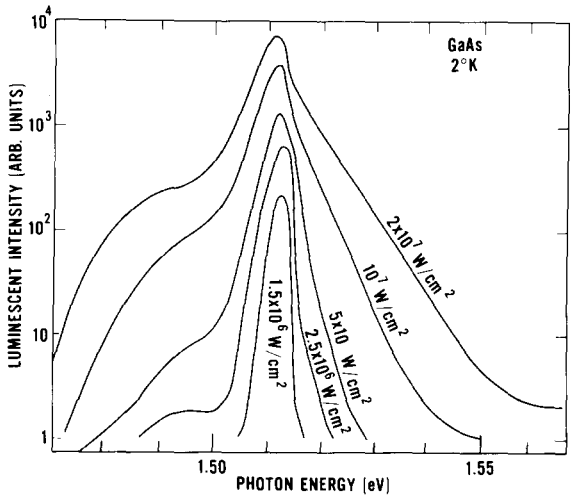

Fig. 17. Spontaneous luminescence spectra of GaAs at 2° K for several different excitation intensities.

We can gain more insight into this problem by studying the spectral dependence of saturation using the data shown in fig. 15. For $\lambda = 8205$ Å, the wavelength of the peak of the spontaneous luminescence shown in fig. 6B, the intensity rises initially with large gain but saturates at a relatively low stimulated emission intensity. At longer wavelengths (say 8225 Å) the initial gain is smaller than that at 8205 Å, but the saturated stimulated emission intensity is almost an order of magnitude larger.

It is clear now from the above that the saturation properties are distinct from the small-signal gain (g), so that it is necessary to consider separately the saturation. This can be done by measuring the saturation parameter α in eq. (2). The measurement technique described here provides just the means for accomplishing this. In fact, fitting eq. (2) to experimentally determined length dependence can provide both spectral and pump intensity information on the gain (g) and saturation (α) separately. In fig. 16, we show gain spectra, determined in this manner, for various pump powers. The peak in the gain spectrum occurs very near the peak in the spontaneous emission spectrum (see fig. 6B). However, the peak saturation intensity occurs at lower energies. Further, as shown in fig. 16, the peak of the gain spectrum does not shift appreciably when the excitation intensity is varied by almost a factor of 10.

Observations of the spontaneous, unamplified luminescence of GaAs (observed from the front surface) under various excitation conditions are shown in fig. 17. The peak of the spectrum does not shift as the excitation intensity is increased. Rather the peak appears to broaden and the high and low energy wings increase

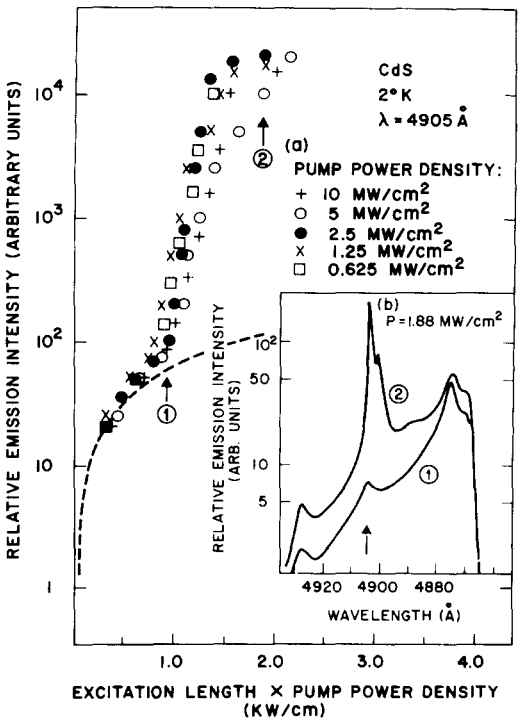

Fig. 18. Stimulated emission intensity as a function of excitation length for a CdS platelet cleaved to form a laser cavity. The abscissa is plotted as the product of excitation length and excitation intensity so that data taken for several different excitation intensities may be displayed on the same graph.

rapidly with increasing excitation intensity. The high energy wing is due to band-to-band recombination of the hot electron-hole gas [32]. The origin of the low energy wing is not clear, but it is this wing which gives rise to the large saturated stimulated emission (fig. 6). The maximum in the gain spectrum (fig. 16), however, occurs at nearly the same energy as the peak in the luminescence spectrum. A detailed understanding of these observed effects in high excited GaAs is not yet available.

6. Optical oscillators

In the preceding sections, we have demonstrated how we can obtain information on the characteristics of stimulated-emission in semiconductors by using the gain measurement technique. Now, let us consider how we might use this information to evaluate a potential semiconductor laser material.

In eq. (2), we described the one-dimensional optical amplifier in terms of a gain parameter g, a saturation parameter α, and a spontaneous emission parameter β. What relationship do these parameters have to the important characteristics of a laser oscillator, namely threshold and efficiency? The relationship between the gain parameter and laser threshold is quite well understood. It remains only to convince ourselves that the gain obtained from the length variation technique is indeed the same gain that is used to determine laser threshold. In the case of the one-dimensional optical amplifier, it is easily shown that a feedback of value R (Laser mirror reflectivity) yields a threshold gain g_0 given by

$$g_0 = \frac{1}{l} \log_e R. \tag{4}$$

To experimentally check the agreement between our measured gain and the threshold gain, measurements of gain are made first on a sample in which the optical cavity is definitely spoiled. This measurement then gives a value for the gain parameter at a given excitation intensity. Next, an optical cavity of mirror reflectivity R is fabricated from the sample, and the gain measurement is performed again. Now data is obtained as shown in fig. 18 for CdS at 2°K. The insert shows spectra taken at an excitation intensity of 1.88 MW/cm^2 for two different excitation lengths. For the shorter length (1) the peak at 4905 Å is barely visible. For a factor of 2 increase in excitation length (2) the intensity of this peak grows by nearly a factor of 100. In high resolution spectra, this peak is resolved, revealing well defined laser cavity modes. If we now follow the line at 4905 Å as a function of excitation length, we obtained the data shown in fig. 18. Here, as in figs. 13 and 14, we plot as the abscissa the product of excitation length and excitation intensity, but the excitation length is the quantity that is continuously varied and hence the gain remains fixed for a given curve. Note that the emission intensity shows a sharp break at about 1 kW/cm. It is at this break that the total gain (gain-length product) exceeds the cavity losses, i.e., laser threshold occurs at approximately 1 kW/cm. This threshold gain agrees with values obtained from the direct measurement of the gain discussed above.

The question of efficiency is related to the saturated output intensity. Saturation is seen to occur in fig. 18 at about 2 kW/cm. Since the output intensity will increase only linearly with additional excitation, efficiency should be determined by the ratio of the emission intensity to the excitation intensity at the onset of saturation. From the gain measurements, the efficiency can be determined from the saturation parameter α of eq. (3). Hence, from gain measurements, we can obtain information with which potential laser materials can be characterized and evaluated without the necessity of fabricating an actual laser oscillator.

7. Conclusions

We have described some experimental concepts and techniques which enable us to obtain a large amount of information about stimulated radiative recombination in semiconductors. Using a simple one dimensional model of an optical amplifier applied to gain measuring technique, the properties of a large number of materials have been studied. We have analyzed in detail, gain measurements on GaAs and have shown that the data acquired under the carefully controlled conditions of the gain measurement technique can be used to obtain information about the processes responsible for the optical gain. For the case of CdS, we have compared gain measurements with measurements on laser oscillators and have shown that the parameters obtained from the gain measurement can be used to predict the characteristics of the oscillator.

Acknowledgements

The authors wish to acknowledge helpful discussions with W.F. Brinkman and C.K.N. Patel, and the excellent technical assistance of R.W. Epworth.

References

[1] K.L. Shaklee and R.F. Leheny, Appl. Phys. Letters 18 (1971) 475.
[2] C. Benoit à la Guillaume, J.M. Debever and F. Salvan, Phys. Rev. 177 (1969) 567.
[3] W.T. Silfvast and J.S. Deech, Appl. Phys. Letters 11 (1967) 97.
[4] A.D. White, E.I. Gordon and J.D. Rigden, Appl. Phys. Letters 2 (1963) 91.
[5] B.A. Lengyel, *Lasers* (Wiley, New York, 1971).
[6] K.L. Shaklee, R.F. Leheny and R.E. Nahory, Appl. Phys. Letters 19 (1971) 302.
[7] R.E. Nahory, K.L. Shaklee, R.F. Leheny and J.C. DeWinter, Solid State Commun. 9 (1971) 1107.
[8] R. Dingle, K.L. Shaklee, R.F. Leheny and R.B. Zetterstrom, Appl. Phys. Letters 19 (1971) 5.
[9] R.E. Nahory, K.L. Shaklee, R.F. Leheny and R.F. Logan, Phys. Rev. Letters 27 (1971) 1647.
[10] K.L. Shaklee, R.F. Leheny and R.E. Nahory, Phys. Rev. Letters 26 (1971) 888.
[11] C.V. Shank, A. Dienes and W.T. Silfvast, Appl. Phys. Letters 17 (1970) 307.
[12] R.W. Epworth, unpublished.
[13] K.L. Shaklee, R.E. Nahory and R.F. Leheny, in *Proceedings of the 11th International Conference on the Physics of Semiconductors, Warsaw, 1972,* to be published.
[14] M. Cardona, *Semiconductors and Semimetals,* Vol. 3, Ed. R.K. Willardson and Albert C. Beer (Academic Press, New York, 1967).
[15] G. Lasher and F. Stern, Phys. Rev. 133 (1964) A553.
[16] J.A. Rossi, D.L. Kenne, N. Holonyak, Jr., P.D. Dapkus and R.D. Burnham, J. Appl. Phys. 41 (1970) 312.
[17] K.L. Shaklee, R.F. Leheny and R.E. Nahory, to be published.
[18] M.A. Lampert, Phys. Rev. Letters 1 (1968) 450.

[19] S. Nikitine, J. Ringeissen, M. Certier, C. Wecker, S. Lewonczuk, J.C. Merle and C. Jung, in: *Proceedings of the Tenth International Conference on the Physics of Semiconductors, Cambridge, Mass., 1970,* Ed. S.P. Keller, J.C. Hensel and F. Stern (ONF–70081) CU. S. AEC Division of Technical Information, Springfield, Va., 1970) 196–J.B. Grun, S. Nikitine, A. Bivas and R. Levy, J. Lumin, 1,2 (1970) 241.
[20] H. Souma, T. Goto, T. Ohta and M. Ueta, J. Phys. Soc. Jap. 29 (1970) 697.
[21] P.T. Landsberg, Solid State Electron. 10 (1967) 513. A review of speculations on indirect bandgap lasers is contained in this paper.
[22] D.G. Thomas and J.J. Hopfield, Phys. Rev. 150 (1966) 680.
[23] D.G. Thomas, J. Phys. Soc. Jap., Suppl. 21 (1966) 265.
[24] P.J. Dean, J. Lumin. 1,2 (1970) 398.
[25] R.E. Nahory, K.L. Shaklee, R.F. Leheny and R.A. Logan, Bull. Am. Phys. Soc. 17 (1972) 233.
[26] N. Holoynak, Jr., D.R. Scifres, R.D. Burnham, M.G. Craford, W.O. Groves and A.H. Herzog, Appl. Phys. Letters 19 (1971) 254.
[27] N. Holoynak, Jr., D.R. Scifres, H.M. Macksey, R.D. Dupis, Y.S. Moroz, C.B. Duke, G.G. Kleinman and F.V. Williams, Phys. Rev. Letters 28 (1972) 230.
[28] This equation is obtained by coupling the Einstein Radiation Equation with the rate equations governing recombination. The departure from exponential behavior at long lengths (saturation) results from the dependence of electron population on emission intensity. Similar expressions for other systems have been obtained previously, see e.g. W.W. Rigrod, J. Appl. Phys. 34 (1963) 2602.
[29] N.G. Basov, O.V. Bogdankevitch, V.A. Gancharov, B.M. Lavrushin and V.Uy.Sudzilovskii, Sov. Phys. Dokl. 11 (1966) 522.
[30] W.D. Johnston, Jr., Phys. Rev. 6 (1972) 1455.
[31] The electron-hole-lattice interaction would give rise to a lattice dilation through screening, and hence to a decrease of the band gap.
[32] J. Shah and R.C.C. Leite, Phys. Rev. Letters 22 (1969) 1304.

Discussion

R.N. Bhargava: Would you care to comment on the work of Holonyak and coworkers on GaP where they claim that they have observed stimulated emission in GaP : N, and possibly in GaP : Zn, O, by observing the mode structure?

Reply: The observations of Holonyak and coworkers are consistent with our results. The mode structure can be understood in terms of saturation effects.

P.T. Landsberg: This is a comment on "band shrinkage" as it occurred in both Pilkuhn's and Shaklee's lectures. The notions of plasma effects, exchange effects and correlation effects as exemplified by "band shrinkage" are really all aspects of the Coulomb interaction many body effects. A convenient way of seeing this effect intuitively[1] (see also section 2.4 of our paper in this book) is to note that the conduction-band hole left by a radiative transition has its lifetime shortened, and hence its width broadened, by conduction band Auger transitions into this hole. This leads to a broadening of the individual conduction band levels in a manner which decreases to zero as the Fermi level of the (degenerate) conduction band is approached. This effect is marked only for a degenerate band. The "band shrinkage" effects could mostly be understood (at least qualitatively) in the past in terms of this model. Does one of the experiments described by Shaklee cast doubt on this interpretation? If so, I would say that as the

[1] Phys. Stat. Sol. 15 (1966) 623, where pumping to effect degeneracy in a pure radiating material to the low energy tails was advocated with a view to seeing these many-body effects.

effect just described by me is bound to be present, the experiment described introduces a new unknown effect which more than nullifies the broadening effect. It is extremely difficult to see what such an effect might be and why it has not been observed before.

Reply: It is quite possible that the excitation density is not large enough to initiate the band shrinkage effect. It seems clear that some other effect is shifting the photon energy below the band gap. There has been evidence of this in the literature for several years.

M.H. Pilkuhn:

(1) Can you give some comments about the physical mechanisms involved in the gain saturation which you observed? In particular, why does the high energy gain saturate faster?

(2) Since you report very low diffraction losses you might assume to have guiding. Can you comment on the refractive index changes with excitation intensity?

(3) What is the situation at high temperatures, e.g. in materials where excitonic contributions to the gain are unlikely?

Reply:

(1) Saturation occurs when the lifetime of the level in question becomes short compared to the time to fill this level. I have no good ideas as to the actual nature of the saturation mechanism. Actually several types of saturation have been observed, and these must be accounted for in any theory attempting to explain the stimulated emission in semiconductors.

(2) Guiding is certainly a possibility since there must be an index change corresponding to the large optical gain. However, since the gain appears to be linear in pump power, there is little experimental justification for invoking guiding.

(3) In GaAs we have made measurements over a wide temperature range. The structure observed at low temperatures is completely gone by 150°K and some new process appears at much lower energies. It seems, however, that this effect is easily explained in terms of the temperature dependence of the Urbach absorption tail rather than exciton dissociation.

O. Svelto: Since the intensity of your N_2-laser is varying in time, the gain will vary in time and so will the emitted light intensity. Since the relationship between emitted light intensity and pump intensity is not a linear one, it seems to me that one should take into account the time behavior of the pump light. Have you done it?

Reply: By appropriate alignment of the N_2 laser, the output pulse can be made to have a rather flat top of about 5 nsec width. In this case the time variation of the pump does not seem too important. We have not corrected for the remaining time variation in the results presented.

E. Mooser:

(1) Any localized state in GaSe such as free and bound excitons does of course contain the states of the whole conduction band including the indirect region. The indirect recombination mechanism which you mentioned for GaP should therefore be valid also for GaSe. It should be noted that if the lowest gap is indirect the localized states are resonant (F. Bassani, Il Nuovo Cimento, 1970) and this may lead to an enhancement of the indirect recombination.

(2) Since direct and indirect gaps in GaS are separated by 0.5 eV, measurements on this compound should allow to attribute the radiative recombination to direct and indirect gap in an unequivocal fashion.

Reply:

(1) We see no evidence of such a resonance. Further, the saturation behavior of GaSe is more characteristic of direct gap than indirect gap materials.

(2) Measurements have been made on the GaSe–GaS alloy system. The stimulated emission can be seen only for S concentrations of less than about 20%. This indicates that there is very little gain associated with the indirect transition.

D. Langer: The gain values which you measure are obviously some average values, due to the boxcar techniques which you use. It may be a question how representative these averages are. We measured the luminescence in CdS under equal excitation conditions (N_2 laser, 10 n sec

pulse width), but we used sampling techniques for a time resolution of about 1 n sec. We found that at higher excitation levels the luminescence narrows and at highest excitation levels double peaks occur with individual half-widths of about 4 n sec.

Reply: Such instabilities are certainly possible. We have not observed such pulsing.