COMMENTS AND ADDENDA

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Stimulated Emission from Excited Semiconductors

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The potential inhomogeneities in actual excited semiconductors and the Fermi-statistical basis for the optical gain therein are recognized in the context of a composite-gain model. Spontaneous emission from a variety of physical processes arising in regions of differing excitation density is amplified by a smooth, broadband gain process, which is characterized by a width and peak position dependent on excitation density. A variety of poorly understood but generally observed characteristics of stimulated light emission from many semiconductor materials may be simply explained in terms of this model.

Recently, a great deal of effort has been devoted to understanding optical gain in excited semiconductor materials, yet the very nature of the gain process remains generally unsettled.[1] What might have been expected to be the "simplest" case, i.e., pure or lightly doped material at low temperature, appears indeed the most complex. More work has been directed to CdS[2] than to other materials in this context, and we may regard the characteristics of stimulated emission in that substance as prototypical of the problem we consider, since similar features are found in *all* I-VII, II-VI, III-V, II-IV-V_2, and I-III-VI_2 direct-gap compounds which have shown evidence of optical gain.[3]

The low-level luminescent spectra of pure or lightly doped semiconductors exhibit at suitably low temperatures (2–4 K) numerous narrow-line features due to various recombination processes typically involving excitons. As the excitation level is increased, stimulated emission is commonly observed at energies corresponding to one or several of these features; naturally enough, such excitonic processes have historically been identified as the source of the optical gain. In CdS, virtually *every* transition which can be observed in the low-to-moderate-level luminescent spectrum has been suggested to provide optical gain. Nevertheless, neither separately nor in conjunction can these processes—involving bound excitons; free excitons scattered by LO phonons, electrons, or other excitons; donor-acceptor pairs; even excitonic molecules—account for all the details of the observed stimulated emission in a simple way. The specific difficulties are as follows.

(i) Stimulated emission can be observed at energies which are incompatible with any of the processes identified in the low- or moderate-level spectra.

(ii) An essentially universal red shift of the emission with increase of excitation is observed. The possibility that hot-carrier effects or pump-dependent absorption might account for these observations has been proposed,[2] but there is as yet no independent evidence for such effects, and there is still the implausibility that several physically very different gain processes (as many as eight in CdS) should "turn on" always in wavelength-sequential fashion.[4]

A simple alternative model, as diagramed in Fig. 1, may resolve these problems in a logical and unified way. The excited semiconductor is represented as a distributed chain of noise-source elements and ideal optical amplifiers. The amplifier is assumed to have a relatively smooth and broad gain spectrum $A(\lambda)$, the *position* and *width*, as well as the peak amplitude, of which are pump-intensity dependent. The smooth spontaneous-emission noise associated with this gain process is represented as $N(\lambda)$, and all other spontaneous emission, particularly from sharp exciton features, is represented as $S(\lambda)$. Specifically, $A(\lambda)$ is identified with

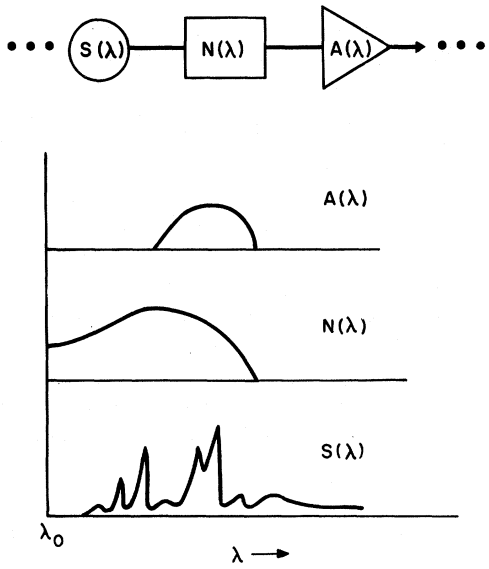

FIG. 1. Model of excited semiconductor with ideal optical amplifier of gain per unit length $A(\lambda)$, noise generator with power per unit length $N(\lambda)$ corresponding to spontaneous emission from physical process which gives rise to $A(\lambda)$, and noise generator $S(\lambda)$ corresponding to spontaneous emission from all other sources. Representations are sketched for A and N arising from band-to-band recombination including appropriate many-body interactions, and for S arising from exciton features, against wavelength, with λ_0 corresponding to the band-gap energy of the unexcited material.

the optical gain arising from direct recombination of electrons and holes from states reflecting the effects of many-body Coulomb interaction, so that $A(\lambda)$ depends explicitly on excitation density and temperature. As shown elsewhere,[5] this process gives rise to gain, the magnitude, width, position, and shift with excitation level of which are at least qualitatively in agreement with observations[1-3] in CdS and also ZnO, CdSe, GaAs, InP, CuCl, etc.

The "output" of the model in Fig. 1 is the total spontaneous emission $S(\lambda)$ plus $N(\lambda)$ amplified by the wide-band gain $A(\lambda)$. Even though the integrated power in S may be small compared to the integrated power in N, high-level peaks in S will still contribute importantly to spontaneous emission and significantly affect the output, particularly since the band-to-band gain mechanism is expected to saturate quasihomogeneously. Thus, even though the gain is itself spectrally smooth, sharp features at the wavelengths associated with exciton transitions may appear in the output. This will account for the common inference of a sharply peaked gain distribution when the stimulated-emission spectrum is observed as a function of pump intensity. However, this problem may be avoided if actual optical gain is measured rather than light output.

Recently, experiments have been reported in which the variation of stimulated emission as a function of the *length* of the excited region was investigated,[6] permitting direct inference of the gain characteristic. The results appear more complex than for similar experiments previously performed on gas- and dye-laser media.[7,8] In the latter, the spontaneous-emission and gain distributions are to an excellent approximation proportional functions and scale together linearly with excitation, as is to be expected for an assembly of independent or distinguishable Boltzmann-distributed two-level oscillators. Optical gain in a semiconductor differs in that it arises from inversion in a system described by Fermi statistics. The gain and spontaneous-emission distributions from the band-to-band transition are not proportional to each other; nor, in general, is the value of either at any specific wavelength linearly related to excitation level. Another consequence of the Fermi statistics is that saturation of the gain is neither homogeneous nor inhomogeneous in the usual sense; under the influence of a saturating electromagnetic field, the gain is reduced for higher energy more than for lower energy within the gain bandwidth. We note that the peculiar saturation phenomenon predicted for this gain mechanism—shorter wavelengths showing initially higher gain but saturating more readily—has in fact been observed in a variety of materials.[9] We suggest it is a simple consequence of the Fermi statistics for conditions of moderate degeneracy.[10]

In any real experiment, the excitation will be more or less nonuniform, owing, for instance, to exponential pump absorption, material inhomogeneity, or even possibly electron-hole drop condensation.[11] It is reasonable that the optical gain will first appear at "threshold" in the regions of *highest* excitation density. There is reason to believe that excitons do not exist at the densities apparently characteristic of onset of stimulated emission,[5] but they will still contribute to spontaneous emission arising from the less densely excited regions. In the context of electron-hole condensation, for instance, N and A in Fig. 1 would come from electron-hole drops and S from the surrounding excitonic gas. The model would thus be useful to describe optical gain in an electron-hole drop system, but certainly does not imply or require that condensation take place.

In spite of the apparent wealth of published experimental results, there are no measurements of the actual optical-gain spectra under conditions where the excitation density and temperature of excited carriers are sufficiently well defined to permit verification of this model. It is not even apparent that thermal equilibrium within the excitation gas exists when semiconductors are excited with pulsed sources of high intensity and brief duration, e.g., the popular 337-nm N_2 laser. Thus, quanti-

tative verification of this model by comparison with published data is unfortunately not now possible. It is also true that the treatment of Ref. 5 cannot be expected to provide quantitatively significant details of the shape of optical-gain functions for real semiconductors with anisotropic and/or nonparabolic bands. Improvement of the theory via more detailed numerical calculations is now underway,[12] but what is most needed are gain measurements as in Ref. 6 under conditions of demonstrably uniform and well-characterized, preferably steady-state, excitation.

In conclusion, two important differences exist between excited semiconductors and other optical-gain media: (a) Gain in an excited semiconductor arises from inversion in a *nonclassical* system obeying Fermi-Dirac statistics, and (b) significant spontaneous emission from unrelated processes may occur within the amplification bandwidth afforded by the gain-producing transition in the semiconductor. A simple model recognizing the implications of these peculiarities is consistent with the multitude of published experimental observations of stimulated emission in many semiconductors, can explain the puzzle of the always sequential appearance of stimulated emission peaks at the positions of numerous low-level excitonic transitions, and can provide a qualitative account for very generally observed wavelength shift and saturation phenomena for which no other simple explanation is now available.

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[1]J. L. Shay and W. D. Johnston, Jr., Phys. Rev. B **6**, 1605 (1972); N. Holonyak, Jr., D. R. Scifres, H. M. Macksey, and R. D. Dupuis, J. Appl. Phys. **43**, 2302 (1972); W. D. Johnston, Jr., J. Appl. Phys. **42**, 2731 (1971); D. Magde and H. Mahr, Phys. Rev. B **2**, 4098 (1970).

[2]A recent comprehensive description of stimulated emission in CdS has been given by K. Era and D. W. Langer [J. Appl. Phys. **42**, 1021 (1971)].

[3]Cf. Ref. 1; K. L. Shaklee, R. F. Leheny, and R. E. Nahory, Phys. Rev. Lett. **26**, 888 (1971); J. L. Shay, W. D. Johnston, Jr., E. Buehler, and J. H. Wernick, Phys. Rev. Lett. **27**, 711 (1971); J. L. Shay, B. Tell, and H. M. Kasper, Appl. Phys. Lett. **19**, 366 (1971); R. D. Burnham, N. Holonyak, Jr., D. L. Keune, and D. R. Scifres, Appl. Phys. Lett. **18**, 160 (1971); C. Benoit a la Guillaume, J. Debever, and F. Salvan, Phys. Rev. **177**, 567 (1969).

[4]This latter problem was specifically recognized in Ref. 2. Holonyak and co-workers have suggested that the solution required an electron-hole-lattice interactive approach (Holonyak *et al.*, as cited in Refs. 1 and 3, and references therein).

[5]W. D. Johnston, Jr., Phys. Rev. B **6**, 1455 (1972).

[6]K. L. Shaklee and R. F. Leheny, Appl. Phys. Lett. **18**, 475 (1971).

[7]W. T. Silfvast and J. S. Deech, Appl. Phys. Lett. **11**, 97 (1967).

[8]C. V. Shank, A. Dienes, and W. T. Silfvast, Appl. Phys. Lett. **17**, 307 (1970).

[9]Cf. Shaklee *et al.*, as cited in Refs. 1, 3, and 6.

[10]In detail, this follows from the dependence of the chemical potential and Fermi distribution function on average density at constant finite temperature. Gain is possible for photons of momentum $\vec{k}$ only if the electron and hole distribution functions satisfy $n_e(\vec{p}+\vec{k}/2) + n_h(\vec{p}-\vec{k}/2) \geq 1$ for some range of $\vec{p}$. The required light-carrier density depends on the ratio of electron and hole masses; the minimum at $m_e = m_h$ corresponds to a Fermi energy $\approx 1.02\,kT$, while for a material such as CdS or CdSe with $m_h \sim 5m_e$, the threshold Fermi energy for intrinsic optical gain is $\sim 2.5\,kT$.

[11]W. F. Brinkman, T. M. Rice, P. W. Anderson, and S. T. Chui, Phys. Rev. Lett. **28**, 961 (1972).

[12]W. F. Brinkman and P. A. Lee (private communication).