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Indirect–Band-Gap Super-Radiant Laser in GaP Containing Isoelectronic Traps

R. E. Nahory, K. L. Shaklee, and R. F. Leheny
Bell Telephone Laboratories, Holmdel, New Jersey 07733

and

R. A. Logan
Bell Telephone Laboratories, Murray Hill, New Jersey 07974

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We report observations which can be interpreted as super-radiant laser emission in the indirect–band-gap semiconductor GaP. Optical gain, as high as 10^4 cm^{-1} at 2°K, has been measured over a range of temperatures from 2 to 300°K. The gain arises from a new process not important at low excitation intensities. It is demonstrated that isoelectronic traps are an important part of this new process.

We report observations which can be interpreted as stimulated emission in optically pumped GaP doped with either nitrogen or bismuth isoelectronic traps. For GaP(N) the stimulated emission occurs in the green over a wavelength range extending from ~5400 to ~5700 Å, and is observed at temperatures from 2°K to above room temperature. For a pump excitation intensity of 2×10^7 W/cm^2, the peak optical gain[1] associated with the stimulated emission exceeds 10^4 cm^{-1} at 2°K and is greater than 200 cm^{-1} at room temperature. For GaP(Bi) the optical gain is spectrally shifted, occurring in the yellow from 5550 Å to beyond 6000 Å. The gain, until now not measured in any indirect–band-gap semiconductor,[2,3] is quite large for both GaP(N) and GaP(Bi), and it is comparable in magnitude to that measured[1,4,5] in direct gap materials at these temperatures and excitation conditions. The measured gain spectra cannot be simply related to the oscillator strengths measured in this spectral region for low excitation intensities,[6-8] thus implying that the stimulated emission is due to a new process which does not contribute at low intensities. It is shown that isoelectronic traps (either N or Bi) are an important part of this new process.

The observations reported in this paper were made using an experimental technique in which emission spectra are studied as a function of the optical excitation length. Various modifications of this technique have been used extensively for optical-gain measurements in gases,[9] liquids,[10] and direct–band-gap semiconductors.[1,4,5] Shaklee and Leheny[1] have given the details of the experimental technique as applied to semiconductors, and no further description of the technique will be given here. In the present work, this technique is utilized to establish the presence of gain in the indirect-gap material GaP.

In Fig. 1, we show with dashed lines the GaP stimulated emission spectra for two different lengths of excitation. It is seen that doubling the excitation length leads to an increase in emission intensity of a factor of 10. These spectra were

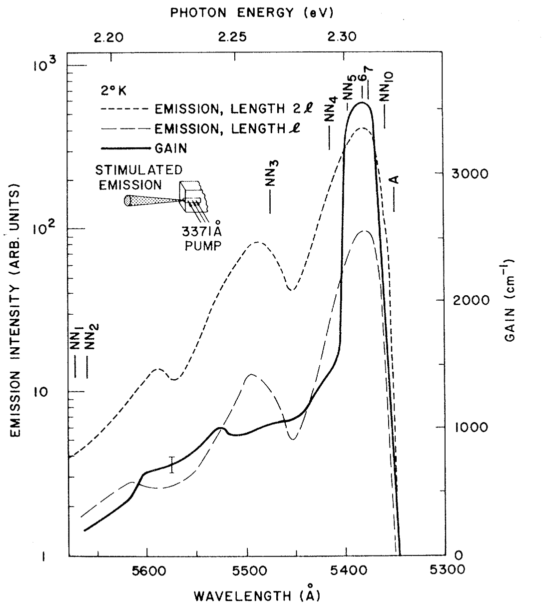

FIG. 1. Stimulated-emission spectra (dashed lines), measured as shown in the inset drawing, for two different lengths of optical excitation at the sample. Note that doubling the excitation length l gives an increase of a factor of 10 in intensity. The solid curve is the gain spectrum derived from the stimulated emission.

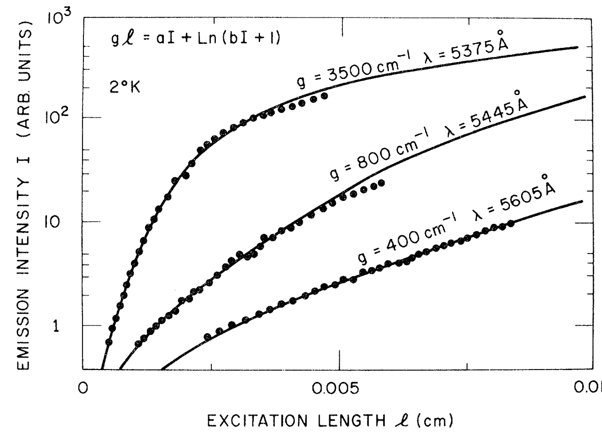

FIG. 2. Emission intensity at selected wavelengths plotted as a function of excitation length for a pump power density of 10^6 W/cm^2. The points were taken from continuous experimental data on an X-Y recorder. The solid curves are theoretical plots obtained from the equation shown, with fixed coefficients a and b and the values indicated for the small-signal gain g.

obtained using nitrogen-laser uv excitation on as-grown faces of lightly doped, bulk, vapor-phase grown GaP. The nitrogen concentration was approximately 5×10^{16} cm^{-3}, but similar spectra are obtained from a wide range (up to 10^{19} cm^{-3}) of nitrogen concentrations and different material sources (vapor phase, liquid phase epitaxy). Figure 2 shows the stimulated-emission intensity at three fixed wavelengths as a function of continuous length variation. It has been shown[1] that in regions of unsaturated gain, the stimulated-emission intensity can be represented by

$$I=[\exp(gl)-1]/b, \tag{1}$$

where g is the small-signal gain, l is the excitation length, and b is a constant depending on matrix elements and geometry. According to Eq. (1), when $gl \gtrsim 2$, I will vary exponentially with excitation length. It can be seen that the three traces in Fig. 2 initially have such an exponential variation (a straight line on the semilog plot), which demonstrates that in GaP under our experimental conditions, g is positive, which in turn shows the presence of stimulated emission and optical gain.

The departure from exponential behavior at longer lengths and for higher intensities is due to saturation effects. Such effects can be included[11] in the calculation of length dependence by taking account of the dependence of electron population on emission intensity; then

$$gl=aI+\ln(bI+1), \tag{2}$$

where a is another constant depending on matrix elements. Equation (2) reduces to Eq. (1) when I is small such that aI is negligible compared with the logarithm. The solid lines in Fig. 2 represent a fit to the data using Eq. (2) and fixed values of a and b. (These values of a and b thus obtained have meaning in terms of matrix elements but only after further analysis, which will be discussed elsewhere.) The agreement is quite good; hence from such analyses the magnitude of the optical gain can be obtained. The maximum gain in Fig. 2 is 3500 cm^{-1} at 5375 Å with 10^6 W/cm^2 pump intensity. At the maximum available pump intensity of 2×10^7 W/cm^2 gains in excess of 10^4 cm^{-1} are observed.

Equation (2) can be applied to the stimulated emission spectra in Fig. 1 to obtain the spectral distribution of the gain, as shown by the solid curve in Fig. 1. The gain spectrum shows two distinct features, a large peak near 2.30 eV and a long-wavelength tail extending below 2.20 eV. The onset of the large gain peak occurs just below the energy of the nitrogen A line[6] (2.3171 eV) in lightly doped materials. This peak is pushed to lower energies as the N concentration is increased up to $\sim10^{19}$ cm^{-3}.

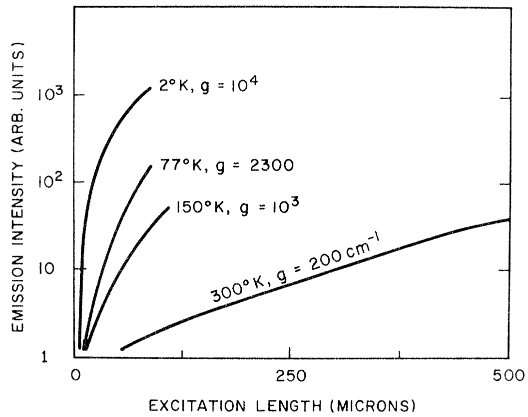

FIG. 3. Measured emission intensity versus excitation length for various temperatures. The curves are all superlinear and give the magnitudes of the gain shown using Eqs. (1) or (2) in the text.

With increasing temperature the gain was found to decrease in magnitude, but the gain was still present even at room temperature. For example, Fig. 3 gives the emission intensity versus excitation lengths for selected wavelengths at temperatures between 2 and 300°K. The data were taken for each temperature near the maximum gain in the spectrum. It is seen that the unsaturated gain varies in magnitude from $g \sim 10^4$ cm^{-1} at 2°K to $g \sim 200$ cm^{-1} at 300°K for a pump intensity of 10^7 W/cm^2. At room temperature the emission intensity varies exponentially with length, according to Eq. (2), over a large range before saturation takes place at lengths close to $\frac{1}{2}$ mm. This is, of course, to be expected since at such low values of gain saturation cannot be achieved so readily as with the high gains measured at low temperatures. (The crucial parameter in attaining saturation is gl rather than g alone.)

The properties of the measured gain are at present not readily understood. The gain gives rise to broad stimulated-emission spectra, Fig. 1, completely different in shape from the photoluminescence spectra measured at low excitation intensities.[7] Further, the observed gain spectrum cannot be simply related to measured absorption spectra since gains as high as 10^4 cm^{-1} are measured in a spectral region where the low-level absorption coefficient[7] is less than 1 cm^{-1}. Apparently, the oscillator strength available for stimulated emission far exceeds that measured in this spectral region at low excitation intensities, indicating that the optical gain arises from a new process which is not important at low intensities.[12]

The mechanism responsible for the stimulated emission is influenced by the presence of nitrogen isoelectronic traps. Note that the gain spectrum (Fig. 1) spans the wavelength range between the nitrogen *A* line and the longest-wavelength NN-pair line.[6] In a pure sample of GaP with negligible nitrogen, no emission at all was seen.[13] Zn-doped samples containing no nitrogen showed red luminescence with no optical gain. Moreover, as discussed below, one might expect isoelectronic traps to be important ingredients for obtaining stimulated emission in indirect-band-gap semiconductors. To test this idea we have made further measurements using GaP doped with Bi, a known isoelectronic hole trap.[8] In these samples stimulated emission *is* observed in the yellow. The measured gain extends from 5550 Å to beyond 6000 Å, i.e., between the known Bi *A* line and the longest-wavelength Bi emission.[8] This result reinforces the above conclusions concerning nitrogen and demonstrates the importance of isoelectronic traps in the observed stimulated emission processes.

We expect that a given center can provide the requirements necessary for stimulated emission, even in an indirect material, if it satisfies certain criteria. It must possess a large free-carrier capture cross section and it must also exhibit efficient radiative decay. Isoelectronic traps appear to satisfy these criteria. These centers are characterized by short-range potentials which give rise to large capture cross sections for free carriers.[7] Further, the resulting bound-electron wave functions extend throughout the Brillouin zone,[7] making possible essentially direct transitions with large[14] radiative efficiencies. Thus nitrogen or bismuth traps in GaP could satisfy the above criteria for stimulated emission. It is clear, however, from the properties of the measured gain spectrum, that the actual mechanism involved is more complicated than the known bound exciton recombination[6] which occurs at low intensities.

In summary, we have presented data which demonstrate the presence of stimulated emission in the indirect-band-gap semiconductor GaP. The measured gain, which is very high and spectrally broad, apparently arises from a new process which does not contribute at low excitation intensities. It is demonstrated, utilizing nitrogen and bismuth impurities, that isoelectronic traps play an important though little understood role in this new process.

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[12]This observation of large gain, g_ν, in a spectral region where the observed low-intensity absorption, $\alpha_\nu^{\rm low}$, is small, i.e., $g_\nu > |\alpha_\nu^{\rm low}|$, is not unique in semiconductor physics to GaP. Indeed, stimulated emission and laser action with $g_\nu > |\alpha_\nu^{\rm low}|$ occur in low-absorption regions well below the band gap in many semiconductors. GaAs {Ref. 4 and N. G. Basov, O. V. Bogdankevich, V. A. Gancharov, B. M. Lavrushin, and V. Uy. Sudzilovskii, Dokl. Akad. Nauk SSSR 168, 1283 (1966) [Sov. Phys. Dokl. 11, 522 (1966)]} is an excellent example where $g_\nu > |\alpha_\nu^{\rm low}|$. In addition to inferences drawn in the text, the observation $g_\nu > |\alpha_\nu^{\rm low}|$ implies rapid depopulation of the terminal state for the radiative transition; i.e., this transition does not correspond to a simple two-level system.

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Parametric Excitation of Plasma Instabilities in Semiconductors

J. I. Gersten and N. Tzoar*

Department of Physics, City College of the City University of New York, New York, New York 10031

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The parametric excitation of density waves in semiconducting plasmas is considered. A new nonlinear mechanism for the direct conversion of photons into plasmons is presented, resulting from the nonparabolic momentum-energy relations for the single electron. Applications to InSb yield threshold intensities typically in the range 10^4–10^6 W/cm^2.

The parametric excitation of density waves in gaseous and solid-state plasmas has been of considerable interest recently.[1] In this Letter we discuss a new nonlinear mechanism for the direct conversion of photons into plasmons which, to the best of our knowledge, has not been considered before. It results from the nonparabolic momentum-energy relation for the single electrons in semiconductors and becomes important in the case of an intense field, e.g., laser radiation. Next we show that for InSb under typical conditions, modest radiation intensities can give rise to growth rates of about 10% per period. Thus, a strong instability can be generated with current state-of-the-art laser fields. The large-amplitude plasma waves obtained in the instability can be monitored in light-scattering experiments where the strength of the anti-Stokes line is proportional to the induced plasma-wave amplitude. The efficient conversion of radiant energy into plasmons provides us with a useful tool for studying large-amplitude plasma oscillations as well as such processes as stimulated Raman scattering.