# PHOTOLUMINESCENCE IN HIGHLY EXCITED GaSe

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ABSTRACT

The photoluminescence of GaSe at high excitation intensities is reported. Four lines, whose intensity increases superlinearly with excitation, are resolved and their behaviour is investigated as function of excitation intensity, excitation frequency and temperature. According to their spectral position and their behaviour, two lines are attributed to different exciton-exciton scattering processes while the remaining two which give rise to spontaneous and stimulated emission, respectively, are tentatively attributed to exciton-carrier scattering.

The luminescence spectra of GaSe at high excitation intensity have been studied in the past by various authors[1-7] who have proposed contradictory interpretations.

An emission line which at 80° K lies 20 - 30 meV below the free-exciton recombination line and whose intensity increases superlinearly with excitation, has been observed by Ugumori *et al.*[1] and has been attributed to exciton-exciton scattering. The same line has also been observed by Leite *et al.*[2] who have interpreted it as originating from exciton-carrier scattering. A new line which increases superlinearly with excitation has been reported by Mercier and Voitchovsky.[3] At 4.2° K this line lies 4 meV below the free exciton line and has been attributed to exciton-electron scattering. Even more confusing are the various reports of a stimulated emission line which at 2° K lies approximately 45 meV below the free-exciton line. Nahory *et al.*[4] first reported this line and concluded that GaSe is a direct-gap semiconductor in contradiction with other experimental data.[8] Stimulated emission in GaSe has also been observed by Ugumori *et al.*[5] and has been attributed to exciton-exciton scattering. Recently, Kuroda *et al.*[6] have interpreted the stimulated emission as the recombination of the direct

free exciton which is assisted by two-phonon emission.

In the present paper we briefly present the results of a detailed investigation of the photoluminescence of GaSe under high excitation intensity. A more extended version of the present work is given elsewhere.[7)]

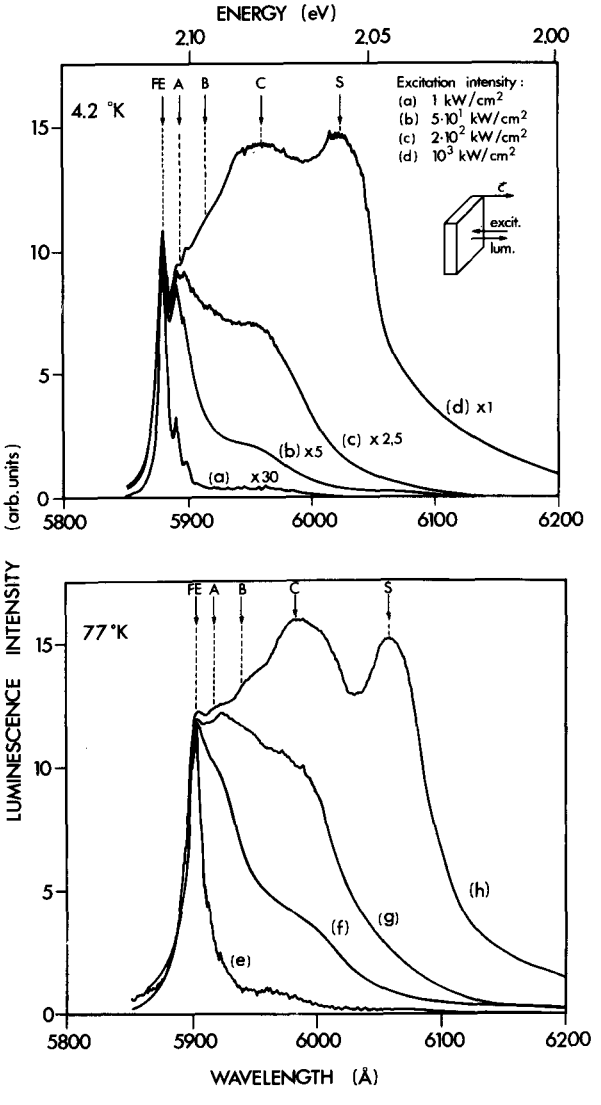


Fig.1. Photoluminescence spectra of GaSe at 4.2 K (upper portion) and 77 K (lower portion) for different excitation intensities. The geometry of the experiments is indicated.

The GaSe crystals used in the present experiments have low impurity concentration (about $10^{17}$ $cm^{-3}$) and therefore the luminescence spectra at low excitation intensity consist only of the direct free exciton emission, as is shown in Fig.1. As the excitation intensity increases, four new lines (labelled A, B, C, and S in Fig.1) appear in the low-energy tail of the spectra. At low temperature, these lines lie about 4, 15, 20, and 40 meV below the free exciton line.

The intensity of these lines varies superlinearly with excitation intensity as is shown in Fig. 2. In particular, the intensity of line S is strongly dependent on the excitation intensity as one expects in the case of stimulated emission. Using the intensity of the free-exciton recombination line as a measure of the exciton density D, the data given in Fig. 2 show that the intensity at line A is proportional

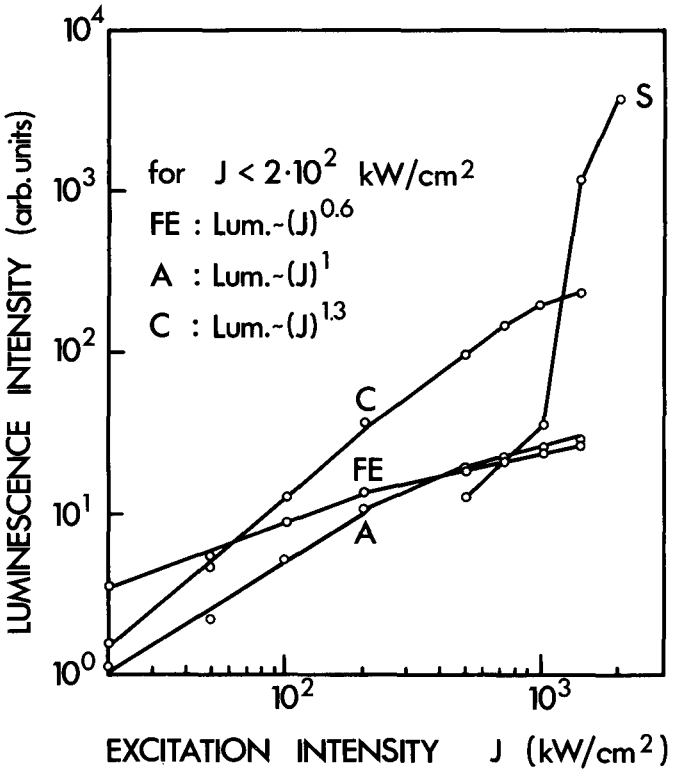


Fig.2. Intensity of the photoluminescence lines of GaSe as function of excitation intensity. The intensity variation of line B is similar to that of line C. The power laws, which are indicated, are valid at low excitation ($J < 2 \times 10^2$ kW/cm$^2$).

to $D^{1.6}$ while that of lines B and C is proportional to $D^2$. With increasing excitation intensity, the luminescence lines shift to lower frequencies with the exception of line A which approximately keeps its energy position. These results are given in Fig. 3 which shows that,

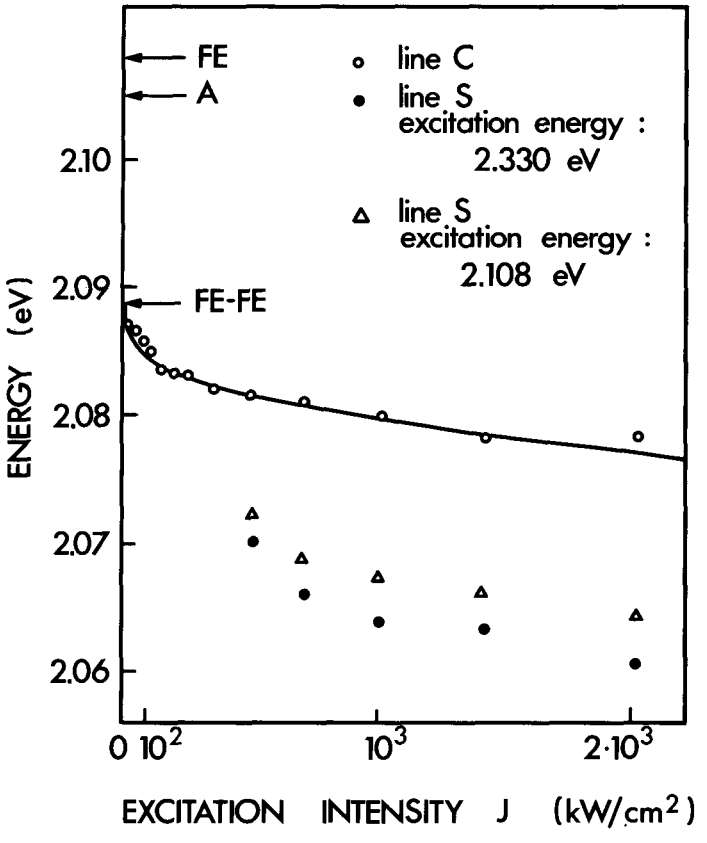


Fig.3. Spectral position at 4.2 K of the photoluminescence lines of GaSe as function of excitation intensity. The free-exciton (FE) and A lines do not shift with excitation intensity. Notice the strong dependence on intensity of the position of lines C and S. The arrow denoted FE - FE shows the theoretical prediction for the position of line C at J = 0.

for line S, the energy position is a function of the excitation frequency, too. The temperature dependence of the energy position of the four luminescence lines has also been studied and the results are shown in Fig. 4 for temperatures between 4.2 K and 300 K.

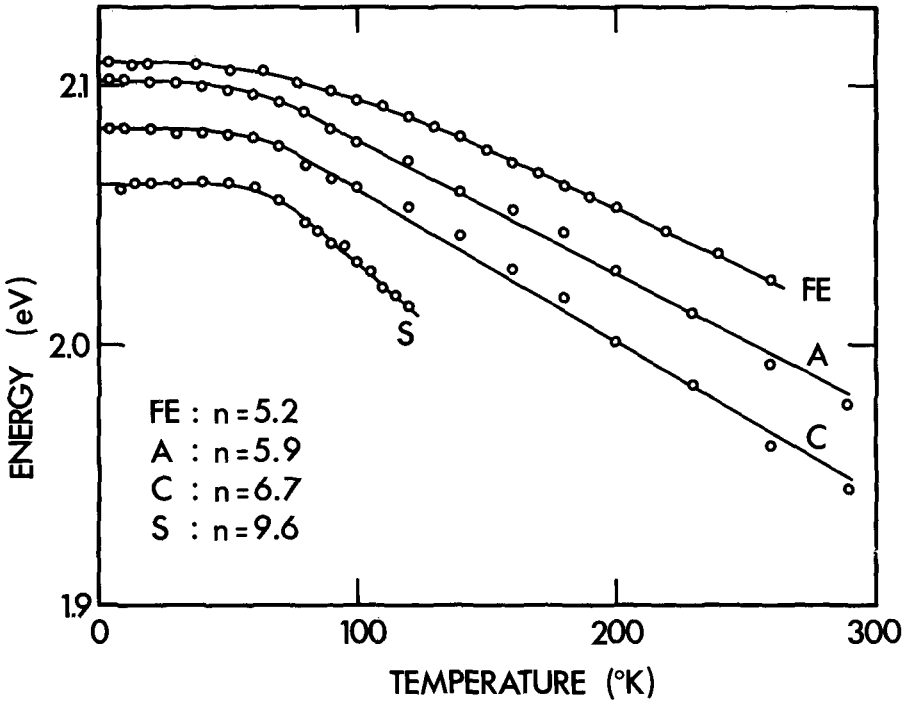


Fig.4. Spectral position of the photoluminescence lines of GaSe as function of temperature. The coefficients n of the linear shift (-nkT) of the various lines at higher temperatures are indicated.

The data presented above on the variation of the luminescence spectra as function of excitation intensity, excitation frequency and temperature, can be used to understand the mechanisms which are responsible for the various recombination processes.

The intensity of line A, which lies 4 meV below the free exciton line is proportional to the 1.6 power of the exciton density. This is approximately the expected behaviour for a line originating from exciton-carrier scattering.[9] The energy position as function of temperature of a line originating from exciton-carrier scattering depends on the effective-mass of the carrier, as shown by Bille.[10] The effective-mass values obtained by Ottaviani *et al.*[11] together with the experimental data shown in Fig. 4 suggest that the carriers are the electrons at the

center of the Brillouin zone.

Lines B and C (15 and 20 meV below the free-exciton line) have an intensity which varies with the square of the exciton density. They are therefore interpreted as being due to inelastic exciton-exciton scattering processes in which one exciton recombines radiatively while the other is excited to the n = 2 state (line B) or is ionized into an electron-hole pair (line C). This interpretation is consistent with the observed value of the binding energy of the free exciton (19.8 ± 0.1 meV).[12]

Finally, line S (which lies about 40 meV below the free exciton) corresponds to stimulated emission as is evident from the data of Fig. 2. In order to understand the nature of this emission band, we give in Fig. 5 the excitation spectrum corresponding to the luminescence at λ =

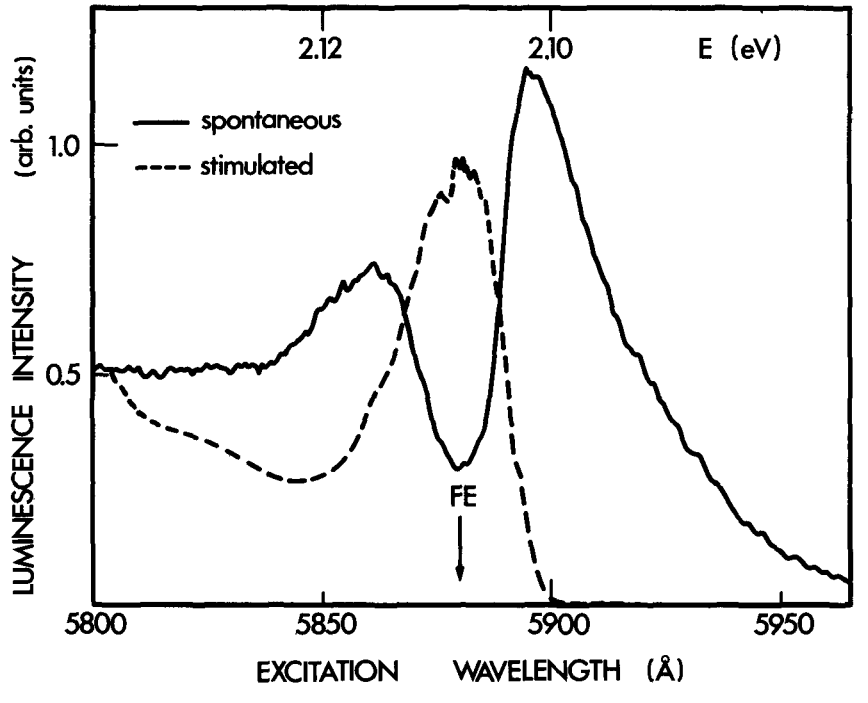


Fig.5. Excitation spectra for the spontaneous (solid line) and stimulated (broken line) luminescence at λ = 6000 Å and at 4.2 K. The spectral position of the free exciton is indicated by the vertical arrow thus showing the relevance of the free exciton in the stimulated luminescence.

6000 Å and at 4.2 K. From these data, it is obvious that the free exciton plays an important role in the recombination process which gives rise to this luminescence. This process, however, remains still unexplained. In particular, it is not clear at present why the stimulated line is at much lower energy than the free-exciton line and why its energy position is so strongly dependent on excitation intensity and on excitation frequency.

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