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Exciton-exciton and exciton-carrier scattering in GaSe†

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An investigation is presented of the photoluminescence at high excitation intensities, in monocrystals of GaSe which contain only low impurity concentrations. Four lines appear at high excitation, one of which corresponds to stimulated emission. The properties of these lines are described as functions of excitation intensity, temperature between 4.2 and 300 °K, and of excitation energy. Two lines, which at 4.2 °K lie 4 and 35 meV below the free direct exciton, are attributed to exciton-carrier scattering giving rise to spontaneous and stimulated emission, respectively. The two other lines are located 15 and 20 meV below the exciton. They correspond to two different exciton-exciton scattering processes. In the first, an exciton is scattered from the ground state $n = 1$ to the excited state $n = 2$, whereas the other recombines radiatively. In the second process, one of the excitons dissociates giving rise to an electron-hole pair in their respective bands, the other one emits a photon. The shifts of these lines with temperature and excitation intensity are interpreted by phenomenological theory.

I. INTRODUCTION

High concentrations of free carriers and excitons can be created in a crystal with intense laser or electron beams. At these high concentrations, the interactions between the particles become important and new processes of recombination occur. Several luminescence lines, which have been observed in different semiconductors, have been attributed to exciton complexes or to inelastic exciton-exciton or exciton-carrier scattering. A series of articles have recently been published reviewing this field.[1–4]

The semiconducting compound GaSe, which crystallizes in a layer structure, has a direct and an indirect gap lying very close to each other. In the past the exact energy difference between the two gaps has been subject to controversy, but recent measurements of photoluminescence[5,6] and of optical absorption,[7] show that the energy E_{DG} of the direct gap (Γ point) in the ϵ-γ polytype at 4.2 °K is

$$E_{DG} = 2.1275 \pm 1 \times 10^{-3}\ \text{eV},$$

and that the indirect gap (M point to Γ point) lies about 25 meV lower.

The selection rules state that direct dipolar interband transitions are allowed only for polarizations parallel to the crystal $\vec{c}$ axis, i.e., perpendicular to the plane of the layers. For polarizations perpendicular to the $\vec{c}$ axis the transitions are weakly allowed only on account of spin-orbit coupling. Of exciton levels associated to the direct gap, the singlet states are observed with polarizations parallel to the $\vec{c}$ axis, and the triplet states with polarizations perpendicular to the $\vec{c}$ axis.[8,9] A decomposition of the exciton ground states into series of closely spaced lines has been attributed to different stacking sequences of the layers. The ionization energy of the ground state of the most intense triplet is 19.5 ± 1 meV.[5]

The luminescence of GaSe at high excitation intensity has been investigated by various authors. Ugumori *et al.*[10,11] have described the properties of an emission line which at 80 °K lies 20–30 meV below the recombination line of the free-exciton ground state. Its intensity increases superlinearly with excitation and it has been attributed to exciton-exciton interaction. Similar experimental results obtained by Leite *et al.*[12] have been interpreted in terms of the recombination of an exciton-carrier complex. In a preceding paper,[13] we have described the behavior of an other radiative line, whose intensity increases superlinearly with excitation. At helium temperature this line is situated between 3 and 5 meV below the free-exciton line and has tentatively been attributed to exciton-electron interaction. Nahory *et al.*[14] have observed for the first time a stimulated emission in GaSe which, at 2 °K, lies at about 45 meV below the free-exciton line. From this observation they concluded that GaSe is a direct-gap semiconductor. The same stimulated emission has also been observed by Ugumori *et al.*[11] at 80 °K, and has been attributed by them to exciton-exciton collision combined with reabsorption. Recently Kuroda *et al.*[15] proposed two different explanations for the stimulated emission: recombination of a direct exciton associated either with the successive emission of two phonons or with the simultaneous emission of a pair of TO phonons.

We present in this paper an investigation on the photoluminescence of GaSe at high excitation intensity. The variations of the spectra as functions of excitation intensity, temperature between 4.2 and 300 °K, and excitation energy are described. The spectra contain four well defined lines, whose intensities increase superlinearly with the excitation intensity. At helium temperature the first line lies about 4 meV below the recombination line of the

free-exciton ground state. This line is attributed to exciton-electron and exciton-hole scattering processes. The second line is situated about 15 meV below the free exciton and results from inelastic exciton-exciton scattering, in which an exciton is scattered into excited state $n = 2$. The third line, which lies about 20 meV below the free exciton, results from a similar scattering process but in which one exciton is dissociated into a pair of electron-hole in their respective bands. The fourth line corresponds to stimulated emission. It is situated between 35 and 50 meV below the free exciton. We tentatively attribute this line to the same process giving rise to the first line, i.e., to the exciton-carrier scattering. The energy difference between the maxima of the spontaneous and the stimulated lines is explained by the self-absorption process resulting from high carrier and exciton densities.

The GaSe crystals on which the present investigation was carried out have low impurity concentrations (about 10^{17} cm^{-3}) and all exhibit at low excitation intensities the luminescence spectrum $A(h\nu)$ described in Ref. 5. This spectrum is characterized by strong free-exciton recombination lines and is free of impurity-induced emission which hides the lines of interest here.

II. EXPERIMENT AND RESULTS

A. Experiment

The crystals of GaSe were grown by the Bridgman technique. The measured samples issued from the ingots number XXII, VI, I, and II, whose properties have been described previously.[16] They were obtained by cleaving the ingots along the plane of the layers into slices of about 1-cm diameter and some tenths of a millimeter thick. The samples were put into a helium cryostat equipped with a temperature controller which allows any temperature between 2 and 300 °K to be maintained. The $\lambda = 0.532$-μm line of a frequency-doubled, pulsed, Nd:YAG laser was used as excitation. Moreover, a tunable dye cell served to determine the excitation spectra. Pulse duration was about 70 nsec at a repetition rate of 75 sec^{-1} and maximum peak power obtained with focalization of the beam was as high as several MW/cm^2. Lower excitation intensities were obtained by means of suitable calibrated grey filters. The luminescence spectra were recorded by means of a double spectrometer (resolution better than 1 Å), a photomultiplier, and a boxcar integrator.

B. Behavior with excitation intensity

Figure 1 shows a series of photoluminescence spectra of GaSe measured at four different excitation intensities J at 4.2 °K (a, b, c, d) and at 77 °K (e, f, g, h). For these measurements, the laser beam was focused on the center of a cleaved face of the sample into a spot of about $\frac{1}{10}$ mm in diameter and the luminescence was measured perpendicularly to this face, as it is shown in the insert of Fig. 1. At the lowest excitation intensity (1 KW/cm^2, Figs. 1a and e), the spectra are composed essentially of the recombination lines of the triplet states of the direct free exciton (FE). At 4.2 °K, two lines are visible about 3 and 5 meV below the FE line which correspond to the lines a_1 and a_2 described previously.[5] They are no longer visible at 77 °K. It is characteristic of crystals containing small impurity concentrations that they are weakly luminescent only at wavelengths longer than that of the FE line and at low excitation intensities.[16] When the excitation intensity is increased, new lines appear

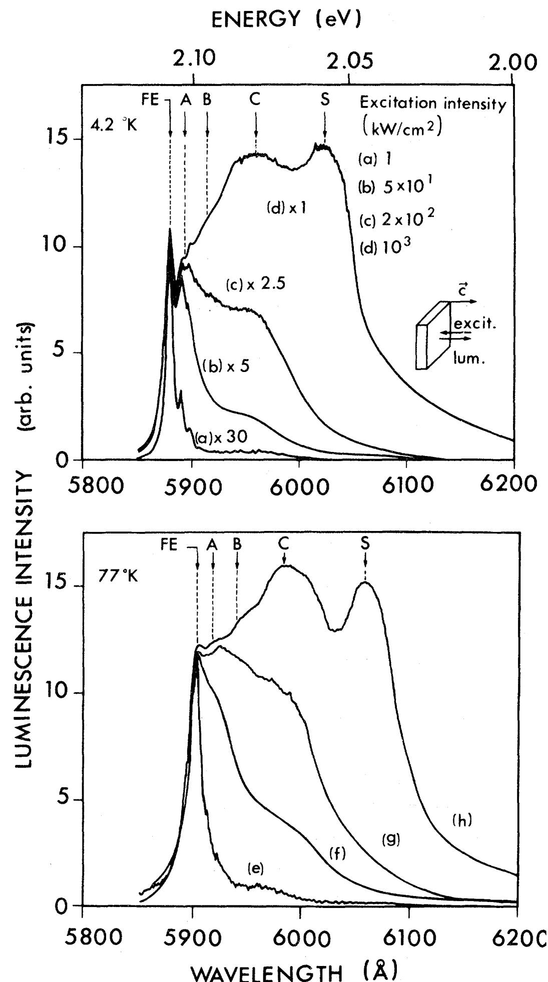

FIG. 1. Photoluminescence spectra of GaSe at different excitation intensities at 4.2 °K (a, b, c, d) and the corresponding spectra at 77 °K (e, f, g, h). The origin of the lines marked by arrows are discussed in the text. The values of the excitation intensities and the geometry of the experiment are given in the inset.

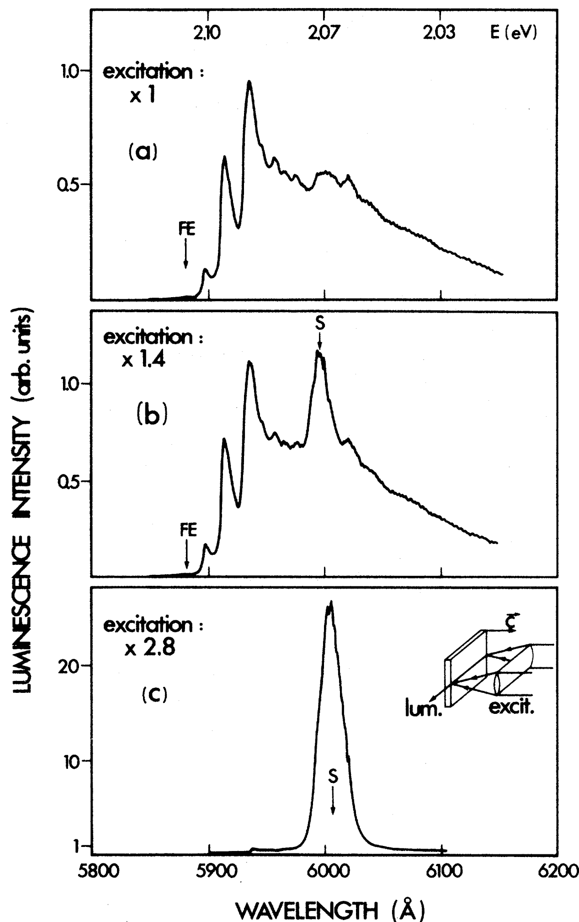

FIG. 2. Photoluminescence spectra of GaSe at 4.2 °K at excitation intensities in the ratios 1 : 1.4 : 2.8. The geometry of the experiment is shown in the inset.

in the low-energy tail of the spectra, and their intensities increase more than that of the FE line (Figs. 1b, c, d, f, g, and h). A line *A* which can easily be observed at 77 °K is situated about 4 meV below the FE line. At 4.2 °K it merges with the lines a_1 and a_2. Two other lines, marked *B* and *C*, lie about 15 and 20 meV, respectively, below the FE line. At high excitation intensity (10^3 KW/cm^2), somewhat below the threshold for crystal destruction, the lines *B* and *C* are more intense than the FE line. Moreover, a new line labeled *S* appears about 40 meV below the FE line (d and h). The intensity of the line *S* strongly depends on the excitation intensity and was shown by Nahory *et al.*[14] to correspond to a stimulated emission. In order to improve the amplification effect of the line *S*, the excitation beam was focused in such a way that the excited area of the crystal was rectangular 5 mm by 0.1 mm. The amplification of the light which propagates parallel to the large side of the excited rectangle is thus enhanced. This arrangement was used by Benoit à la Guillaume *et al.*[17] for measuring the gain of stimulated lines. The spectra shown in Fig. 2 were obtained with such an arrangement for three values of the excitation intensity in the ratios 1 : 1.4 : 2.8. The luminescence was detected in the direction of the largest amplification, as shown in the inset of Fig. 2(c). The spectrum (a), which was obtained with the smallest excitation intensity, is composed of a set of lines which has been attributed earlier to localized structural-defect states.[5] The difference between this spectrum (unpolarized) and that presented in Fig. 1(a) (polarization perpendicular to the $\vec{c}$ axis) is explained by reabsorption near the indirect gap and by the selection rules governing the interband transitions.[18] For the same reasons, the FE, *A*, *B*, and *C* lines cannot be observed in the spectra shown in Fig. 2. The spectrum in Fig. 2(b) was measured with an excitation intensity 1.4-times higher than that of the spectrum (a), immediately above the threshold for stimulated emission of the line *S*. If *J* is further increased by a factor of 2, only the line *S* persists, with an intensity increased by two orders of magnitude (Fig. 2c). It is noteworthy that impurities do not affect the stimulated emission: it is also observed in strongly doped crystals.

Figure 3 shows the variations of the intensities of the FE, *A*, *C*, and *S* lines vs the excitation intensity *J*. For the line *S*, these measurements were obtained in the geometry indicated in Fig. 2, for all the others in the geometry of Fig. 1. At values of *J* smaller than 2×10^2 kW/cm^2, the intensities of the FE, *A*, and *C* lines vary as $(J)^\alpha$. The values of α corresponding to the different lines are indicated in Fig. 3. At higher *J* the intensities of

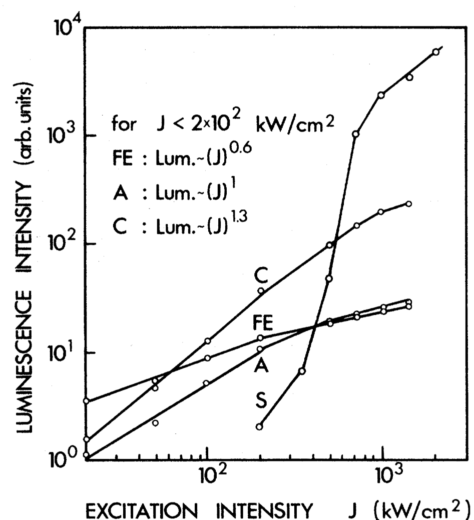

FIG. 3. Intensities of the FE, *A*, *C*, and *S* lines vs excitation intensity *J*. For $J < 2\times10^2$ kW/cm^2 the intensities of the different lines are proportional to $(J)^\alpha$, where α is indicated in the inset.

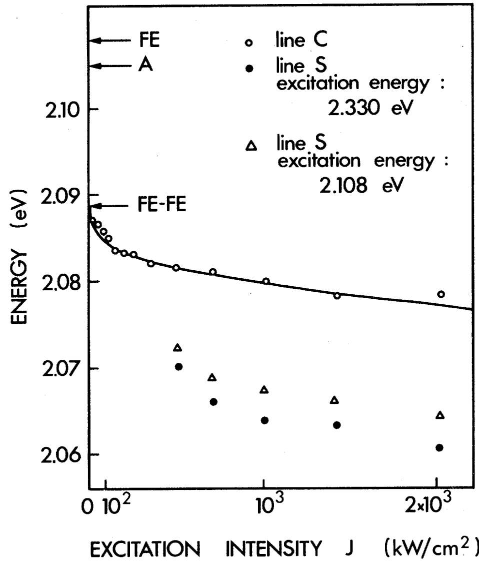

FIG. 4. Energy positions at 4.2 °K of the maxima of the lines C (open circles) and S (closed circles and triangles) vs excitation intensity J. The solid line corresponds to the theoretical considerations given in Sec. III B. The positions of the FE and A lines are indicated by arrows. The arrow marked FE–FE indicates the theoretical position at $J=0$ of the main line resulting from exciton–exciton scattering.

the FE, A, and C lines saturate and the line S increases very rapidly. The intensity of the line B which is not shown in Fig. 3 varies in the same way as that of the line C.

As shown in Fig. 4 the maxima of the lines C and S shift to lower energies as the excitation intensity increases. The experimental positions of the maxima of line C are indicated by circles, those of line S by black dots or by triangles depending on whether the value of the energy of the exciting photon was 2.330 or 2.108 eV. The continuous line represents the calculated shift of the line C as discussed in Sec. III B. The positions of the FE and A lines are indicated by arrows. The shift of these lines versus J is small and masked by the other lines. The theoretical position at $J=0$ of the main emission produced by inelastic exciton-exciton scattering (FE-FE) is also given in Fig. 4.

C. Behavior with temperature

All the lines shift to lower energies when the temperature is increased. Figure 5 shows the positions of the maxima of the FE, A, C, and S lines at different temperatures between 4.2 and 300 °K. It can be seen from this figure that linear shifts $\Delta(h\nu) = -nkT$ occur at temperatures higher than 90 °K. The values of the coefficient n for the different lines are shown. The shift of the FE line is essentially the same as that of the direct gap, and has been attributed to hole–optical-phonon interaction.[19] Moreover, the stimulation threshold of the line S increases as the temperature increases. Above 120 °K, the crystal is destroyed by the excitation light, before the threshold is reached.

III. DISCUSSION

A. Line A

At 4.2 °K the maximum of the line A lies about 4 meV below the FE line. This line appears up to temperatures as high as 300 °K (Figs. 1 and 5). It follows from Fig. 3 that for excitation intensity $J < 2\times 10^2$ kW/cm^2 the intensity of line A increases like $(J)^1$, whereas that of the free-exciton line increases like $(J)^{0.6}$. As a consequence, the intensity of A varies as the power 1.6 of that of the FE line. This variation corresponds approximately to the theoretically expected behavior of the line resulting from exciton-carrier scattering.[17]

From the relations of energy and momentum conservation, Bille[1] has computed the shift versus temperature of the maximum of the spontaneous line due to exciton-carrier scattering. Assuming that the temperature of the excitons is identical to that of the carriers, and that the thermal distributions of excitons and carriers are of the Boltzmann type, Bille showed that the maximum $h\nu_m$ of this line can be deduced from the relation

$$K_0(\beta CX)/K_1(\beta CX) = A/C \ , \qquad (1)$$

where K_0 and K_1 are modified Bessel functions and where the other symbols are defined as follows:

$$\beta = 1/kT \ ,$$

$$X = E_G - E_{FE} - h\nu_m \ ,$$

$$C = [(m/M)(1 + (M-m)^2/4mM)]^{1/2} \ ,$$

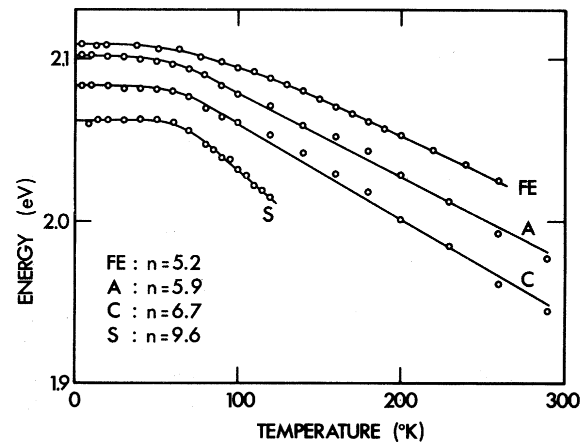

FIG. 5. The maxima of the FE, A, C, and S lines vs temperature. For $T > 90$ °K, the lines shift as $-nkT$. The values of n for the different lines are indicated.

A = $(M-m)/2M$,

M = total mass of the exciton,

m = mass of the carrier,

k = Boltzmann constant,

E_G = direct-gap energy,

E_{FE} = ionization energy of the direct exciton at $\vec{K}=0$.

The values of the density-of-state masses in GaSe deduced from electrical measurements are[20]:

$(m_\perp^2 m_\parallel)^{1/3} = 0.5\ m_0$ for the holes

$= 0.2\ m_0$ for the electrons at the direct minimum (Γ point)

$= 0.73\ m_0$ for the electrons at the indirect minimum (M point) ,

where m_0 represents the free electron mass. The value of the total mass of the direct exciton is thus $M = 0.7 m_0$. From these values one readily deduces for scattering of excitons by Γ-point electrons

$$A = 0.36 \text{ and } C = 0.64 ,$$

whence, with the relation (1),

$$\beta X = (1/kT)(E_G - E_{FE} - h\nu_m) = 0.77 ,$$

and

$$h\nu_m = E_G - E_{FE} - 0.77\ kT . \qquad (2)$$

Similarly, for an exciton-hole scattering one finds

$$A = 0.14 ,\ C = 0.86 \text{ and } \beta X = 0.064 ,$$

which in this case gives

$$h\nu_m = E_G - E_{FE} - 0.064\ kT . \qquad (3)$$

Because the exciton mass is smaller than the mass of the electron at the M point, the line corresponding to a scattering between these particles is expected to shift towards high energies as the temperature increases. Moreover, the probability of such a scattering is smaller because of the $\vec{k}$ dependence of the Coulomb interaction. From these arguments we conclude that the scattering of excitons by M-point electrons cannot explain the line A.

From Fig. 5 one deduces that, for $T > 90\ ^\circ$K, the shift of line A with respect to the position of the free-exciton line is

$$\Delta h\nu(A) = -0.7kT .$$

This value is in excellent agreement with the computed value (2).

It should be noted that if the dispersion due to exciton-photon interaction is neglected, the energy difference between the exciton and the line arising from exciton-carrier scattering is expected to be zero at $T = 0$. Experimentally, at 4.2 °K a difference of 4 meV is found. This relatively large difference can be explained by the fact that at high concentrations the temperature of carriers and excitons differs markedly from that of the crystal. It follows from (2) that the temperature of the particles which corresponds to the position of line A at 4.2 °K is equal to 65 °K. With this particle temperature, the width of the free-exciton line should be 5–6 meV at 4.2 °K, which is in a good agreement with the experiment (see, e.g., Fig. 1). In conclusion, we may say that the dependence of line A on excitation intensity corresponds to that produced by exciton-carrier scattering. Moreover, the energy shift with temperature of line A suggests that the scattered carriers are the electrons at the Γ point.

B. Lines *B* and *C*

At 4.2 °K and with a low excitation intensity, lines B and C are situated about 15 and 20 meV below the FE line, respectively. These lines appear to temperatures as high as 300 °K. As seen in Fig. 3, the intensity of line C varies as $(J)^{1.3}$, i.e., approximately as the square of the exciton density and the same variation is observed for line B. This behavior corresponds to that expected for inelastic exciton-exciton scattering. Indeed, the probability of such a process varies as the square of the exciton density.[17]

(a) In order to explain the two lines, we consider two different processes of exciton-exciton scattering:

exciton $(n=1)$ + exciton $(n=1)$

→ photon + exciton $(n=2)$,

where $n=1$ and $n=2$ correspond to the ground state and the first excited state of the exciton, respectively. Energy conservation at $\vec{K}=0$ implies that

$$2(E_G - E_{FE}) = h\nu + E_G - \tfrac{1}{4}E_{FE} ,$$

where $h\nu$ is the energy of the emitted photon and where $\frac{1}{4}E_{FE}$ is the ionization energy of the first excited exciton state. Whence,

$$h\nu = E_G - E_{FE} - \tfrac{3}{4}E_{FE} .$$

$E_{FE} \simeq 20$ meV in GaSe, i.e., the line corresponding to this scattering process is expected to lie 15 meV below the FE line. Line B satisfies this expectation very closely.

(b) The second scattering process corresponds to the ionization of an exciton,

exciton $(n=1)$ + exciton $(n=1)$

→ photon + electron-hole pair.

In this case conservation of energy at $\vec{K}=0$ gives

$$2(E_G - E_{FE}) = h\nu + E_G ,$$

whence

$$h\nu = E_G - 2E_{FE} . \qquad (4)$$

The corresponding line lies 20 meV below the FE line and thus can be identified with line C.

In the above expressions for energy conservation the kinetic energies of excitons and free carriers have been neglected. At high excitations where high particle densities are present, this neglect is no longer justified. Lévy *et al.*[21] have evaluated the correction $\Delta(h\nu)$ to the energy (4) resulting from the gradual filling of elliptical bands:

$$\Delta(h\nu) = \left(\frac{m_0}{m_e}\frac{m_0}{m_h}\right)\frac{\hbar^2}{2m_0}\pi^2\left(\frac{3}{8\pi}\right)^{2/3}\rho^{2/3} ,$$

where m_0 is the mass of the free electron, m_e and m_h are the effective masses of the electron and the hole, respectively, and where ρ is the number of carriers per cm^3. With the effective mass values given by Ottaviani *et al.*[20] one obtains

$$\Delta(h\nu) = 9.1\times 10^{-15}\rho^{2/3} \text{ eV} . \qquad (5)$$

It follows from the recombination kinetic that in the stationary state and at high excitation the number of carriers is proportional to the number of excitons. Taking the number of excitons to vary approximately like $J^{1/2}$ over the whole range of excitation intensities covered by the experiment (see Fig. 3), one therefore obtains

$$\rho \sim J^{1/2} \text{ and } \Delta(h\nu) \sim J^{1/3} .$$

The solid curve shown in Fig. 4 corresponds to this J dependence. For $J=0$, $\Delta(h\nu)=0$ and the curve originates in the point given by (4) marked FE-FE

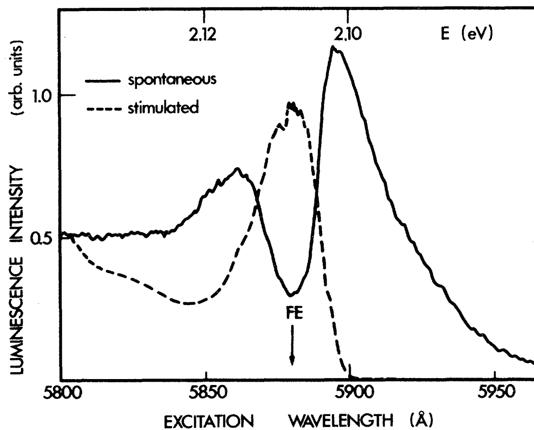

FIG. 6. Intensities at 4.2 °K and at $\lambda = 6000$ Å of the stimulated (dashed curve) and spontaneous (solid curve) luminescence as functions of the wavelength of the excitation light. The arrow marks the position of the free-exciton (FE) line.

in Fig. 4. At 2×10^3 kW/cm^2 it corresponds to a carrier density which according to (5) is $\rho = 1.3 \times 10^{18}$ cm^{-3}.

The shift of the maximum of line C as a function of temperature is shown in Fig. 5. Below 90 °K, this shift is small, above this temperature, it increases linearly with temperature. Because of the large number of particles involved in the exciton-exciton scattering process, it is difficult to evaluate theoretically the shape of the luminescence line and the shift of its maximum with temperature.

C. Line S

Line S is visible in Fig. 1 in the spectra measured with the highest excitation intensity. It follows from Figs. 2 and 3 that the behavior of this line is that expected for a stimulated emission. At 4.2 °K the maximum of line S is situated between 35 and 50 meV below that of the FE line, depending on the excitation intensity and on the energy of the excitation light (Fig. 4). Figure 6 shows the excitation spectrum (dashed curve) of the stimulated line observed in the arrangement indicated in Fig. 2(c). Also drawn in Fig. 6 is the excitation spectrum (continuous curve) of the spontaneous line which appears at the same energy as line S in Fig. 2(a). These excitation spectra have been measured by recording the intensities of stimulated and spontaneous luminescence emitted at $\lambda = 6000$ Å as functions of the excitation energy. The free-exciton line is marked in Fig. 6 by an arrow. As seen in this figure, the intensity of the stimulated light strongly increases when the excitation energy coincides with that of the free exciton, whereas the intensity of the spontaneous light decreases at this energy. The free exciton therefore plays an important role in the process which produces the stimulated emission, whereas the spontaneous emission at the same energy is not directly related to the free exciton.

It is noteworthy that the position of the maximum of the line S depends on the energy of the excitation light. When this energy coincides with that of the free exciton, the maximum of line S (triangles in Fig. 4) lies 3–4 meV higher than when the excitation energy is higher than the direct gap (black dots in Fig. 4).

Shaklee *et al.*[22] have measured that the gain for line S at 2 °K is about 10 000 cm^{-1} at an excitation intensity of 10^7 W/cm^2. Ugumori *et al.*[11] have proposed that the line is due to an exciton-exciton collision process, like line C, and that the stimulated emission is shifted with respect to the spontaneous emission because of reabsorption by the crystal states as was also observed in CdS.[23] It would appear that reabsorption cannot explain the present measurements. Thus we have shown in Fig. 7 the

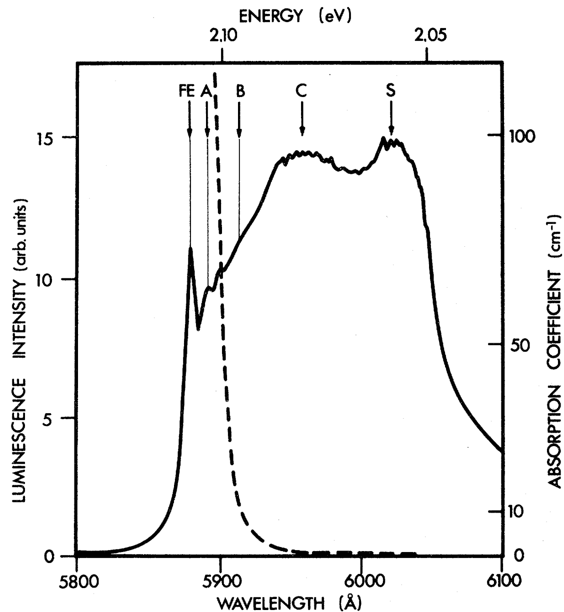

FIG. 7. Absorption coefficient of GaSe at 4.2 °K vs the wavelength (dashed curve). For comparison, the luminescence spectrum (solid curve) of the same crystal (as in Fig. 1 d) is also shown.

optical-absorption spectrum[7] (dashed line) of a pure single crystal of GaSe at 4.2 °K with light polarized parallel to $\vec{c}$ (allowed transitions). Also drawn in Fig. 7 is the emission spectrum (solid line) of the same crystal obtained in the geometry indicated in Fig. 1. It follows from this figure that the optical absorption spectrum near the lines *C* and *S* is nearly flat and that the corresponding absorption coefficients are too low to give rise to strong reabsorption. Moreover, the stimulated emission was observed in crystals containing up to 10^{20} cm^{-3} of impurities which produce a considerable increase of the optical absorption in the pertinent range. In spite of this, no significant displacement of line *S* was observed. In the absence of reabsorption, line *S* and line *C* would have to coincide[17,23] if they were both produced by exciton-exciton scattering as proposed by Ugumori *et al.*

Line *S* has also been observed by Kuroda *et al.*,[15] who describe it as a two-phonon replica of the free exciton. However, this interpretation cannot account for the large shift of line *S* towards lower energies which is observed when the excitation intensity or the temperature is increased.

We here propose that line *S* results from the same process as line *A*, i.e., from inelastic exciton-carrier scattering. At high excitation intensities, the gain curve corresponding to this process shifts towards lower energies with respect to spontaneous curve because the inverse process gives rise to considerable self-absorption. This inverse process corresponds to the absorption of a photon and the simultaneous creation of an exciton accompanied by the transition of a carrier from a higher to a lower state in the band. The probability of this self-absorption process is of the same order as that of the direct process[17] and may thus account for the energy difference between the maxima of line *S* and line *A*. In support of this interpretation we first of all note that the gain of the exciton-carrier scattering process is expected to be higher than that of the exciton-exciton scattering process.[17] Second the large shift of the stimulated line towards lower energies with increasing excitation (Fig. 4) and temperature (Fig. 5) can be understood by the dependence of the self-absorption on the density and temperature of excitons and carriers.[17] Finally, the self-absorption is enhanced by an increase of temperature, so that the gain diminishes with temperature: At $T > 120$ °K, the stimulated emission can no longer be observed.

The stimulated emission was investigated with excitation energies larger than the direct gap as well as with excitation energies close to the free exciton. In the first case, high densities of free carriers at Γ (direct gap) are created. In the second case, because of the resonance between the free exciton and the indirect conduction band,[6] holes are still created at Γ but only states near the indirect gap *M* are available for electrons. While exciton-hole and exciton-electron scattering contribute to stimulated emission in the first case, only exciton-hole scattering is probable in the second case. This might well explain the fact that the stimulated line appears at slightly different energies for the two types of excitation (Fig. 4).

IV. CONCLUSION

At low temperatures and with excitation intensities of about 1 kW/cm^2, the photoluminescence spectra of high quality GaSe crystals contain only lines corresponding to the free-exciton recombination. GaSe thus represents a material which is particularly well suited to study the interactions between excitons and carriers which appear at high excitation intensities and give rise to new radiative transitions. We have presented the properties of four luminescent lines *A*, *B*, *C*, and *S*, which result from such interactions. Line *A* is attributed to the spontaneous emission arising from exciton-carrier scattering. Lines *B* and *C* result from exciton-exciton scattering. In particular, line *B* corresponds to the scattering of an exciton from the state $n = 1$ to the state $n = 2$ with the radiative recombination of the other exciton. The emission of line *C* is associated with simultaneous dissociation of an exciton giving rise to a pair electron hole.

Finally, line S, which corresponds to a stimulated emission, can be attributed to exciton-carrier scattering. The energy difference between line A and line S is due to self-absorption.

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