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Spontaneous and optically amplified luminescence from exciton-exciton collisions in GaSe at liquid-He temperature

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We report on our investigations of the excitonic luminescence spectra of photoexcited GaSe crystals immersed in superfluid He. We measured the spontaneous and optically amplified luminescence due to exciton-electron and to direct-direct as well as to direct-indirect exciton-exciton scattering. The intensity of the exciton-exciton recombination lines is proportional to the squared intensity of the luminescence from simple excitonic recombination. We find good agreement, within experimental uncertainties, between our experimental results and theory. Gain due to exciton scattering processes is weak and difficult to observe in GaSe. From our measurements, we estimate net gain values of up to about 50 cm^{-1}.

I. INTRODUCTION

When investigating excitonic spectra of semiconductors one normally distinguishes three regimes. (i) At weak excitation intensities the excitons form a dilute and therefore ideal gas. (ii) At intermediate densities interactions between excitons become important. A broadening of the luminescence lines is observed, and new emissions connected with exciton-exciton (X-X) scattering appear. In such a scattering process, one exciton recombines optically while the other is raised into an excited state. The emitted photon has an energy which is roughly one excitonic Rydberg lower than that of the free-exciton ground state. Similar exciton-electron (X-e) and exciton-hole (X-h) processes are also possible. At low temperature and with adequate excitation, all these scattering processes can have optical gain. (iii) Finally, if the electron-hole (e-h) pair density is higher than the Mott value, i.e., at densities for which the screening of the Coulomb interaction is so strong that bound excitonic states do not exist any more, the carriers form an e-h plasma.

In this publication we present spontaneous and optically amplified photoluminescence spectra of GaSe measured at a temperature of 2 K and at intermediate carrier densities. We propose an explanation which involves different X scattering processes. In particular, in GaSe, the direct energy gap is very close to the indirect one. Therefore we observed scattering between direct and indirect excitons too.

Several years ago, luminescence from X scattering processes had been observed in different semiconductors such as CdS, CdSe, and ZnO.[1-3] More recently, the luminescence and gain spectra connected with these processes have also been calculated theoretically.[4,5] For GaSe, luminescence due to X scattering has also been observed by several authors.[6,7] For such experiments, it is important to make sure that light emitted only from the excited spot is detected. Otherwise, due to inhomogeneous excitation, emissions coming from different processes will be mixed. The recent development of experimental techniques[8,9] and a more detailed understanding of the Mott transition of the excitonic gas[10,11] permit us to distinguish the luminescence of X scattering from that of an e-h plasma. In particular, we believe that optical gain due to X scattering has not previously been observed in GaSe, because it is weak and in GaSe, a relatively strong optical absorption due to indirect transitions and crystal imperfections persists below the band gap. The strong emission, which at higher excitation intensities appears about two exciton Rydbergs below the line from simple direct excitonic recombination,[12,7,13] is, according to our present model, recombination luminescence from an e-h plasma.

II. THEORETICAL SUMMARY OF EXCITON-EXCITON AND EXCITON-CARRIER RECOMBINATION

In GaSe, the indirect exciton (at point M of the Brillouin zone) has an energy which is only 35 meV lower than the direct exciton at point Γ.[14] Therefore, in our luminescence experiments, both direct and indirect energy levels can be populated. In particular, scattering processes involving a direct and an indirect exciton are possible also. Thus, the X-X recombination processes to be considered are the following:

(a) $X^{\Gamma}(1,\vec{k}_1)+X^{\Gamma}(1,\vec{k}_2)\rightarrow X^{\Gamma}(n,\vec{k}_1,+\vec{k}_2)+\hbar\omega_n^{\Gamma}$,

(b) $X^{\Gamma}(1,\vec{k}_1)+X^{\Gamma}(1,\vec{k}_2)\rightarrow e^{\Gamma}h(\vec{k}_1+\vec{k}_2)+\hbar\omega_{e\text{-}h}^{\Gamma}$,

(c) $X^{\Gamma}(1,\vec{k}_1)+X^{M}(1,\vec{k}_2)\rightarrow X^{M}(n,\vec{k}_1+\vec{k}_2)+\hbar\omega_n^{M}$,

(d) $X^{\Gamma}(1,\vec{k}_1)+X^{M}(1,\vec{k}_2)\rightarrow e^{M}h(\vec{k}_1+\vec{k}_2)+\hbar\omega_{e\text{-}h}^{M}$.

TABLE I. Exciton-exciton recombination in GaSe (Ref. 14): emitted photons. For definitions of the symbols, see text. Note that the given numbers are mean values, and the energies of the excitonic states can vary up to ±1.5 meV (Ref. 15).

Process		Emitted photon energy (eV)	$E^{\Gamma}_{X(1)}-\hbar\omega^{\sigma}$ (meV)	Wavelength (Å)	
$X^{\Gamma}(1)+X^{\Gamma}(1)\rightarrow X^{\Gamma}(n)+\hbar\omega^{\Gamma}_{n}$	$\hbar\omega^{\Gamma}_{1}=E^{\Gamma}_{X(1)}$	2.110	0	λ_1	5875
	$\hbar\omega^{\Gamma}_{2}$	2.095	15	λ^{Γ}_{2}	5919
	$\hbar\omega^{\Gamma}_{3}$	2.092	18	λ^{Γ}_{3}	5927
	$\hbar\omega^{\Gamma}_{\infty}$	2.090	20	$\lambda^{\Gamma}_{\infty}$	5933
$X^{\Gamma}(1)+X^{M}(1)\rightarrow X^{M}(n)+\hbar\omega^{M}_{n}$	$\hbar\omega^{M}_{1}=E^{\Gamma}_{X}$	2.110	0	λ^{M}_{1}	5875
	$\hbar\omega^{M}_{2}$	2.088	22	λ^{M}_{2}	5940
	$\hbar\omega^{M}_{3}$	2.083	27	λ^{M}_{3}	5953
	$\hbar\omega^{M}_{\infty}$	2.080	30	λ^{M}_{∞}	5962

Here, $X^{\sigma}(n,\vec{k})$ represents a direct ($\sigma=\Gamma$) or an indirect ($\sigma=M$) exciton in a state with principal quantum number n and wave vector $\vec{k}$, $e^{\sigma}h(\vec{K})$ is a free-electron–hole pair with total momentum $\vec{K}$, and $\hbar\omega^{\sigma}$ represents the emitted photon. At low temperatures (i.e., neglecting kinetic energies) and assuming hydrogenlike excitons, we may write[3]

$$\hbar\omega^{\sigma}_{n}\simeq E^{\Gamma}_{X(1,0)}-E^{\sigma}_{R}(1-n^{-2}) , \tag{1}$$

and

$$\hbar\omega^{\sigma}_{e\text{-}h}\lesssim\hbar\omega^{\sigma}_{\infty} , \tag{2}$$

where $E^{\sigma}_{X(1,0)}$ is the energy of an exciton in its fundamental state and E^{σ}_{R} is the excitonic Rydberg. Numerical values for GaSe are reported in Table I. Following theoretical considerations,[5] the most probable X-X recombination is the process in which the second exciton is scattered from the $n=1$ band to the $n=2$ band.

As for the exciton-carrier recombination, X-e scattering is more probable than X-h scattering, because of the normally smaller electron mass.[5] For the same reason, scattering by an indirect electron is unlikely in GaSe. Thus, we are left with only one exciton-carrier scattering process,

$$\text{(e)}\quad X^{\Gamma}(1,\vec{k}_X)+e^{\Gamma}(\vec{k}_e)\rightarrow e^{\Gamma}(\vec{k}_X+\vec{k}_e)+\hbar\omega^{\Gamma}_{e} .$$

The spectral position of the peak of the emission connected with X^{Γ}-e^{Γ} collision in GaSe is given approximately by

$$\hbar\omega^{\Gamma}_{e}=E^{\Gamma}_{X(1,0)}-0.77k_BT_X-2W_0n^{\Gamma}_{X} , \tag{3}$$

where the second term is due to the thermal energy of electrons and excitons with temperature T_X,[7] and the third represents the self-energy of the excitons[5,16] [n^{Γ}_{X}: density of direct free excitons, $W_0=\frac{26}{3}\pi(a^{\Gamma}_{B})^3E^{\Gamma}_{R}$; a^{Γ}_{B}: exciton Bohr radius]. The self-energy of the electrons can be neglected if their density is low, i.e., at low temperatures as long as the density of the e-h pairs is much lower than the Mott density.

As far as the kinetics of the direct free-exciton population is concerned, under steady-state conditions we write

$$\frac{dn^{\Gamma}_{X}}{dt}=0=G-\alpha_r n^{\Gamma}_{X}-\alpha_D n^{\Gamma}_{X}-\beta_{\Gamma\Gamma}(n^{\Gamma}_{X})^2 -\beta_{\Gamma M}n^{\Gamma}_{X}n^{M}_{X}-\beta_{\Gamma e}n^{\Gamma}_{X}n^{\Gamma}_{e} , \tag{4}$$

where G is the direct free-exciton formation rate caused by optical excitation, α_r and α_D are the rate constants for simple radiative and nonradiative recombination of the excitons, and the three coefficients β are proportional to the probabilities for radiative X^{Γ}-X^{Γ}, X^{Γ}-X^{M}, and X^{Γ}-e^{Γ} scattering. Under steady-state conditions the intensity of the direct-direct exciton recombination lines should increase with the square of the density n^{Γ}_{X} of direct free excitons, as long as the three first terms on the right-hand side of (4) are the dominant ones. If one assumes that the ratio of indirect to direct e-h pair density is constant, then the direct-indirect exciton recombination becomes proportional to $(n^{\Gamma}_{X})^2$. Concerning X-e recombination, the emission intensity is expected to rise with $(n^{\Gamma}_{X})^{3/2}$ (Ref. 1). The gain coefficient g of X^{Γ}-X^{Γ} recombination can be estimated with[17]

$$g=\frac{\beta_{\Gamma\Gamma}(n^{\Gamma}_{X})^2c^2}{8\pi\nu^2\Delta\nu\epsilon} , \tag{5}$$

where ν is the photon frequency and $\Delta\nu$ the bandwidth of the corresponding spontaneous luminescence line, c is the velocity of light, and ϵ is the dielectric constant.

Finally, we suppose that within each valley excitons and free carriers thermalize to the same temperature T_X. Because of the short-lifetimes of the e-h pairs, this temperature (at intermediate densities) is usually about an order of magnitude higher than the lattice temperature (which is close to that of the He bath). This causes the emission bands of X-X and exciton-carrier processes to be rather wide [several meV (Ref. 4)]. Thus in good approximation we can use Boltzmann statistics in all our estimations.

III. EXPERIMENTAL TECHNIQUES

The crystals of GaSe investigated were grown either by the transport reaction or by the Bridgman method. In this case, the ingots were cleaved along the plane of the layers into about 0.1-mm-thick slices. The samples, immersed into superfluid helium, were optically excited by the emission at 532 nm of a pulsed and frequency-doubled yttrium-aluminum-garnet:neodium (YAG:Nd) laser. The light pulses had a duration of the order of 100 ns and a repetition rate of 1 kHz.

For the spontaneous luminescence spectra, only a small spot with a diameter of roughly 20 μm was illuminated on the sample. The luminescence was collected from the front face of the crystal and analyzed by a double spectrometer. The signal, detected by a photomultiplier, was recorded using boxcar techniques. For the excitation spectra (Fig. 3), a dye laser (Rhodamine 6G pumped by the YAG:Nd laser) was used as a variable monochromatic source, thus allowing resonant excitation of the excitonic levels. The dye-laser pulses had a duration of 50 ns and a repetition rate of 75 Hz.

For the amplified luminescence and optical-gain spectra (Figs. 4 and 5) the illuminated surface on the sample had the shape of narrow strip, one end of which coincided with the edge of the sample. Thus, the excited volume of the crystal had the shape of a rod. The luminescence, amplified on its way through the rod, was collected from the edge of the sample and was measured for different strip lengths (l).[18] Modulating l we directly recorded the gain spectra in function of the wavelength.[8,9] In our measurements l varied between 50 and 600 μm, which allows measurement of gain coefficients up to about 100 cm^{-1} since saturation effects become important for $gl \gtrsim 5$.[18,8] Further details on the experimental setup can be found in Ref. 9.

The intensity J absorbed by the crystals varied between 1 and 200 $kW\,cm^{-2}$. Assuming an absorption coefficient of 10^3 cm^{-1}, a dielectric constant of 8.4 (Ref. 19) and a carrier lifetime of 10^{-10} s,[20] electron-hole pair densities ranging from 3×10^{14} to 6×10^{16} cm^{-3} are created at the surface of the sample. The Mott transition of direct excitons is expected to occur for e-h pair densities of about 4×10^{16} cm^{-3}.[13] At the moment of their creation, the optically excited electrons will populate the conduction valley at the Γ point of the Brillouin zone, because the energy of 2.33 eV of the laser photons is not high enough to allow relaxation into an indirect valley.[21] The conduction valleys at M points can be populated through intervalley scattering of direct excitons[22]; this population can be quite important since the lifetime of indirect e-h pairs is usually significantly longer than that of direct ones.

In all our experiments a diaphragm was placed in the image plane of the collecting lens to stop the light leaving the sample at places other than the excited spot. Further, the gate of the boxcar had a width of 50 ns, which is comparable to the duration of the laser pulses. The diaphragm and boxcar gate allow the almost complete elimination of the perturbing extrinsic luminescence at energies lower than that of the free exciton, since such luminescence is often long lived and due to secondary excitation by intrinsic emissions. The use of a diaphragm also strongly reduces the risk of collecting luminescence from sample regions with completely different e-h pair densities.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Spontaneous luminescence

Figure 1 shows spontaneous photoluminescence spectra from a Bridgman sample of GaSe measured at a temperature of 2 K and at different excitation intensities J. The peak at 5875 Å, labeled λ_1^Γ, is due to simple recombination of the direct free exciton. It is well resolved for weak excitations and appears as a shoulder (on the high-energy side of the emissions due to exciton scattering) for J of the order of 100 $kW\,cm^{-2}$, i.e., for e-h pair densities close to the Mott value. Assuming that the luminescence at wavelengths shorter than λ_1^Γ is mainly due to the direct free-exciton emission, we fitted the high-energy tail of our spectra to $\exp[-\hbar(\omega-\omega_1^\Gamma)/k_B T_X]$. From this fit we got an estimate of the temperature of the carriers[23]: 15 K at the weakest excitations and 40 K for $J \simeq 100$ $kW\,cm^{-2}$. We note that for these temperatures and according to Ref. 4, the theoretical half-widths of the emission connected to the process $X(1)+X(1)\rightarrow X(1)+\hbar\omega_1$ are at most 20% smaller only than those of the measured line λ_1^Γ. Further, as seen from Fig. 2, the intensity of this line increases linearly with J.

Following Ref. 7, we attribute the peak around 5888 Å in Fig. 1 to X^Γ-e^Γ scattering. The increase of its intensity is proportional to $J^{3/2}$ (Fig. 2), and therefore proportional to $(n_X^\Gamma)^{3/2}$ which coincides with theoretical expectations.[1] Its spectral position, about 4 meV on the red side of the free-exciton line and almost independent on excitation intensity, however, does not match theoretical predictions. From Eq. (3), we calculate a shift which varies from 1.7 meV for an excitation intensity of 28 $kW\,cm^{-2}$ to 3.5 meV for $J=113$ $kW\,cm^{-2}$. It is also interesting to note that at our weakest excitation intensity of about 1 $kW\,cm^{-2}$, which corresponds to an e-h pair density of about 3×10^{14} cm^{-3}, we found an emission intensity of X-e scattering which is still comparable to that of simple excitonic recombination. This means that the population of free electrons is still quite high. Moreover, what we call "simple" excitonic recombination is normally a second-order process also (only excitons with wave vector $k=0$ can recombine); therefore, the corresponding luminescence line can be relatively weak.

At the lowest excitation intensities, a weak peak appears at about 5905 Å. This line can be attributed to the recombination of electrons from the extrinsic level D_1, as proposed in Ref. 22. As already reported earlier,[13] it saturates at excitation intensities of roughly 50 $kW\,cm^{-2}$.

As can be seen from Table I, we expect the emissions due to X-X scattering between about 5910 and 5970 Å. In the spontaneous luminescence spectra (Fig. 1) overlapping broad bands are indeed visible. We compared our spontaneous spectra with those calculated by Moriya and Kushida for GaAs.[4] For this, we first determined the carrier temperature from the high-energy tail of our spectra, as explained above. Then, using the results of the numerical calculations in Ref. 4, we got the theoretical half-widths and asymmetries of the luminescence lines of the two processes $X(1)+X(1)\rightarrow X(2)+\hbar\omega_2$ and $X(1)+X(1)\rightarrow e\text{-}h+\hbar\omega_{e\text{-}h}$. Finally, we decomposed our measured spectra into four lines of triangular shape: two for direct-direct and two for direct-indirect exciton scattering. The triangular lines have the theoretical widths and are centered on the wavelengths given in Table I; their intensities were adjusted in such a way that their sum matches the measured spectra. An example of such a decomposi-

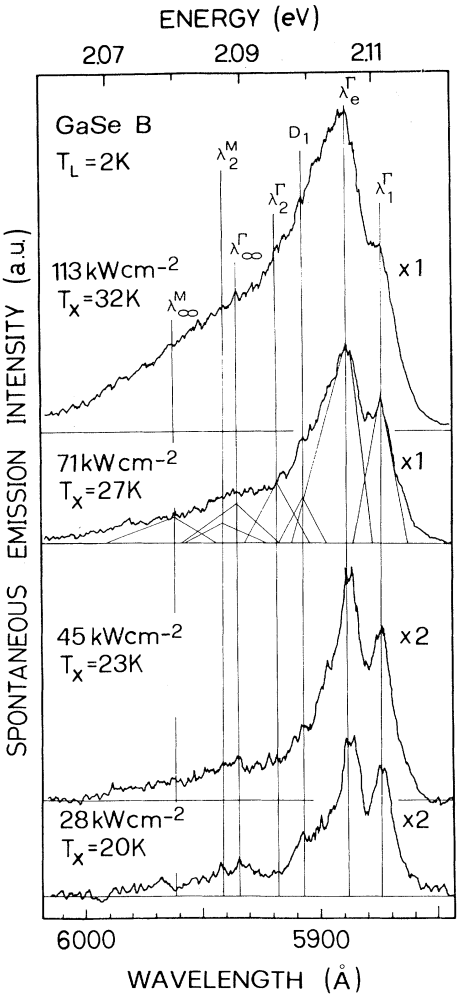

FIG. 1. Series of spontaneous photoluminescence spectra from a Bridgman sample of GaSe measured at a bath temperature of 2 K and for the indicated excitation intensities. Light was collected from the front surface of the crystals. Vertical lines mark the wavelengths, given in Table I, of the different emissions due to the exciton-scattering processes discussed in the text. T_X is the estimated free-exciton temperature. Factor indicated on the right of each spectrum gives the relative sensitivity which was used to record the spectra. Spectral resolution is 2 Å.

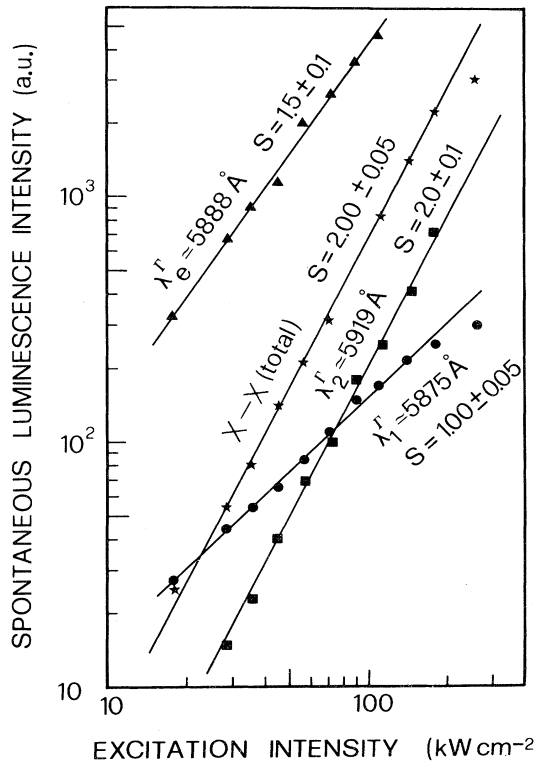

FIG. 2. Logarithmic plots of the integrated intensities I of the spontaneous luminescence due to direct free-exciton recombination (λ_1^Γ), exciton-electron recombination (λ_e^Γ), direct-direct exciton-exciton recombination (λ_2^Γ), and all exciton-exciton recombination processes together (X-X total) versus excitation intensity J. Symbols are explained in the text. Continuous lines are least square fits of the experimental intensities (dots) to a simple power law $I \sim J^s$. Exponents found are indicated.

tion is shown in Fig. 1. With the exception of the low-energy tail, our crude construction reproduces quite well the measured spectrum. The integrated intensity of each of the four emissions increases exactly with the square of the intensity of the simple excitonic recombination line (for example, as shown in Fig. 2 for the line λ_2^Γ). For the λ^Γ emissions which are connected with direct-direct X scattering, this behavior is in excellent agreement with theory. The fact that direct-indirect exciton recombination also follows the square law indicates that the population ratio of the direct and indirect levels is constant and does not depend on the carrier temperature which varies by more than a factor of 2 in our experiments. This indicates that the intervalley thermalization time might be quite long. In Ge under [111] strain, e.g., at a lattice temperature of a few kelvins, an intervalley scattering time as long as 1 μs has been observed.[24] This is much longer than the duration of 100 ns of our laser pulses, i.e., intervalley thermal equilibrium cannot be reached. In any case, we have to note the precise proportionality to J^2 of the integrated intensity of the low-energy part of the spontaneous luminescence [measured starting from λ_2^Γ=5119 Å, labeled X-X(total) in Fig. 2].

The linear increase of the simple free-exciton luminescence intensity with J tells us that up to densities close to the Mott value, the first three terms on the right-hand side in Eq. (4) are the most important ones. Further, the spontaneous luminescence lines due to the different radiative recombination mechanisms are of the same order of magnitude, therefore nonradiative recombination must be much more frequent than radiative recombination.

Figure 3 shows the excitation spectra of the spontaneous emission line at λ_2^M=5940 Å for three different excitation intensities. We observe that the intensity of the spontaneous luminescence has a maximum when the energy of the exciting photons equals the energy of the levels of the direct free exciton. As before, these maximum intensities are proportional to J^2. The same behavior was

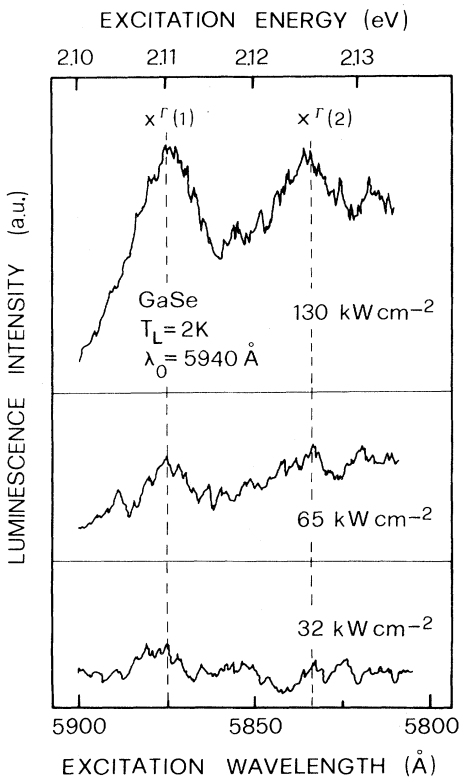

FIG. 3. Excitation spectra of the spontaneous luminescence at $\lambda_0 = \lambda_2^M = 5940$ Å from a Bridgman sample of GaSe at a bath temperature of 2 K and for the three indicated excitation intensities. Dashed lines give the energies of the two lowest excitonic levels. Light was collected from the front surface of the crystal. Spectral resolution is 5 Å.

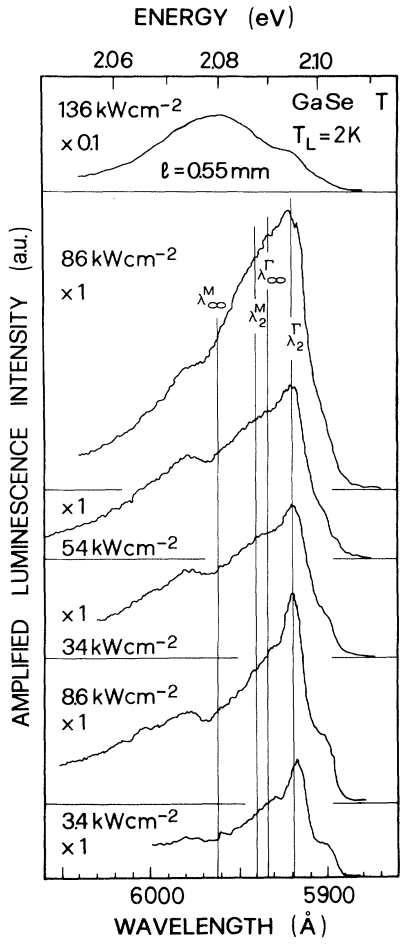

FIG. 4. Series of amplified luminescence spectra from a transport-grown sample of GaSe measured at a bath temperature of 2 K. Excitation intensity varies between 3.4 and 137 kW cm^{-2}, as indicated. Special "gain geometry" used for this experiment is discussed in the text. $l = 0.55$ mm is the length of the illuminated strip on the crystal. Factor indicated on the left on each spectrum gives the relative sensitivity which was used to record the spectra. The vertical lines denote the wavelengths, given in Table I, for different exciton scattering processes discussed in the text. Spectral resolution is 5 Å.

found in the excitation spectra of the three lines λ_2^Γ, λ_∞^Γ and λ_∞^M. The results of these measurements confirm our interpretation in terms of *X-X* scattering.

B. Optically amplified luminescence

The effects of *X-X* recombination processes can also be observed in the amplified luminescence and optical-gain spectra. The situation is somewhat difficult, though, because in the special experimental "gain geometry" extrinsic luminescence lines can be enhanced and, therefore, can cover intrinsic luminescence. While the spontaneous spectra—for high-quality crystals, of course—are always very similar to each other, the amplified spectra sometimes show narrow peaks whose positions vary from sample to sample. The previously discussed *X-X* recombination processes, however, show up as a broad band with weak structures only (as one would expect after having seen the spontaneous spectra of Fig. 1).

In Fig. 4, a series of such amplified spectra is shown. The peak at about 5920 Å coincides with the emission from the process $X^\Gamma(1) + X^\Gamma(1) \rightarrow X^\Gamma(2) + \hbar\omega_2^\Gamma$. On the low-energy side, a wide shoulder due to the other *X-X* scattering processes follows and a long tail extends far into the red (the little peak at ~ 5980 Å is probably extrinsic). We note that the emissions due to simple excitonic recombination ($\lambda_1^\Gamma = 5875$ Å) and to *X-e* scattering ($\lambda_e^\Gamma = 5888$ Å) are completely absent in the amplified spectra, because the absorption coefficient at the energy of the direct free exciton and in its vicinity is too strong. The first line on the high-energy side of our amplified spectra is due to the extrinsic level D_1. As in the spontaneous spectra, this line saturates with increasing J. At higher *e-h* pair densities, the structures on the spectra become weaker, and at 137 kW cm^{-2} we already observe a wide smooth band, which is shifted to the red and which we attribute to the emission of an *e-h* plasma. Thus, the transition from the excitonic gas to the *e-h* plasma occurs at lower laser intensities in the "gain geometry" than in the "spontaneous geometry," probably because the lumines-

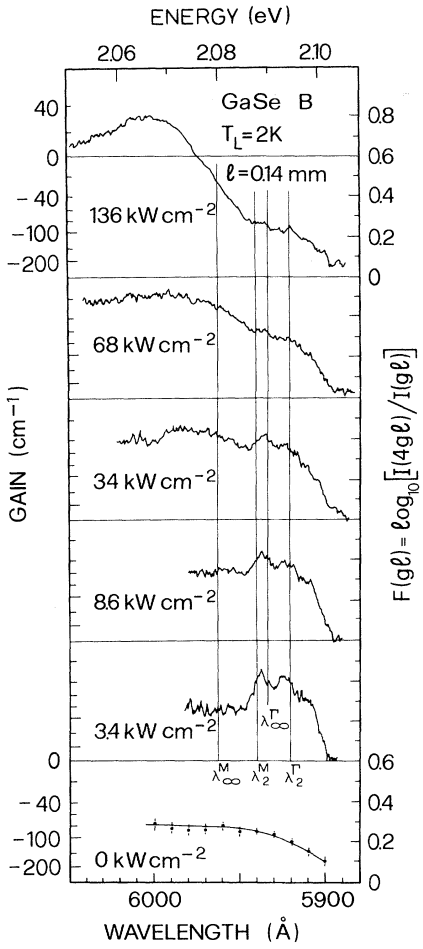

FIG. 5. Series of optical gain spectra of a GaSe single crystal grown by the Bridgman method, measured at a bath temperature of 2 K. Excitation intensity varies from 3.4 to 136 kW cm^{-2}, as indicated. Geometry of the experiment is discussed in the text. Vertical scale, on the right, is linear for the function $F(g\,l)$ (Ref. 9). $I(g\,l)$ is the amplified luminescence intensity, g the gain coefficient and l the length of the illuminated strip. On the left, the (nonlinear) scale permits one to directly read the gain coefficient. Vertical lines mark the wavelengths given in Table I for the different exciton scattering processes discussed in the text. At the bottom, the spectrum of the apparent absorption coefficient is shown (see text), it is an average of measurements on several samples. Spectral resolution is 5 Å.

cence traveling in the direction of the strip is quite intense.

Figure 5 shows a series of optical-gain spectra from a Bridgman sample. The structure, due to the different scattering processes, is rather weak and becomes smoother for increasing excitation intensity. In order to get the net coefficient we have to know the "apparent" optical-absorption coefficient for photons traveling through the excited rod and having energies below the band gap. Indirect transitions, absorption by impurities, but also scattering of light at crystal defects and at the crystal surface, contribute to this apparent absorption. For its estimation, we illuminated the crystals on a narrow strip with a dye laser, collected the laser light scattered into the direction of the strip from the edge of the sample, and used strip-length modulation in a way similar for a gain experiment. The result is given in Fig. 5; at the wavelengths where the luminescence due to exciton scattering occurs the coefficient of apparent absorption is of the order of 100 cm^{-1} which is quite strong. Thus, subtracting the bottom spectrum of Fig. 5 from the other ones, we find for the net optical gain due to X-X recombination in GaSe values up to 50 cm^{-1}.

Using this result and Eq. (5), we can estimate the rate coefficient $\beta_{\Gamma\Gamma}$ for X^Γ-X^Γ scattering. We know that under an excitation intensity of $\sim$70 kW cm^{-2}, the spontaneous luminescence emission connected to X^Γ-X^Γ scattering occurs at wavelengths around 5930 Å and has a width of $\Delta\lambda$=40 Å (Fig. 1). Assuming as before an absorption coefficient of 10^3 cm^{-1}, a dielectric constant of 8.4, and a total excitonic lifetime $(\alpha_r+\alpha_D)^{-1}\simeq(\alpha_D)^{-1}$ of $\sim 10^{-10}$ s, we get a density $n_X^\Gamma = n_{70}^\Gamma$ of $\sim 2\times 10^{16}$ cm^{-3}. With these numbers, Eq. (5) yields $\beta_{\Gamma\Gamma}\simeq 3\times 10^{-8}$ cm^3 s^{-1}. Furthermore we know that at this same excitonic density n_{70}^Γ, the spontaneous luminescence intensity due X^Γ-X^Γ scattering equals that of simple excitonic recombination, i.e., $\beta_{\Gamma\Gamma}(n_{70}^\Gamma)^2=\alpha_r n_{70}^\Gamma$. Using this, we get a radiative lifetime $(\alpha_r)^{-1}$ of the direct free excitons of roughly 2 ns, a number which has a reasonable order of magnitude.

V. CONCLUSIONS

We investigated spontaneous and amplified luminescence from excitons in GaSe at low temperature. We worked at intermediate e-h pair densities, i.e., at densities for which X-X scattering processes are important. The spectral positions of the luminescence lines as well as the dependence of their spontaneous intensity on excitation permitted us to assign them to different recombination mechanisms. The intensity of the spontaneous emission due to simple excitonic recombination increases linearly with excitation intensity J; the line due to X-e recombination rises as $J^{3/2}$, whereas the whole luminescence band connected with direct-direct and direct-indirect X-X scattering is proportional to J^2. It is possible to decompose this band into four lines, two for direct-direct and two for indirect-direct exciton collisions. For the linewidths, theoretical results[4] have been used. Excitation spectroscopy clearly confirms the connection between exciton population and intensity of these four lines. Gain due to X scattering processes is weak and difficult to observe in GaSe, because of the strong absorption which persists below the band gap. From our measurements we estimate net-gain values up to 50 cm^{-1}. At densities which are quite close to those predicted theoretically for the Mott transition of the excitonic gas,[10] the structures in the emission spectra become weaker, and with further increase of the e-h pair density, the luminescence spectrum has the typical shape of a plasma emission.

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