Exciton–exciton interaction in electroluminescence spectra of a GaSe crystal

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(Submitted 10 March 1994; resubmitted 8 September 1994)
Pis'ma Zh. Eksp. Teor. Fiz. **60**, No. 9, 622–624 (10 November 1994)

A stimulated emission due to an exciton–exciton interaction has been observed near the fundamental absorption edge of GaSe crystals at 77 K. © *1994 American Institute of Physics.*

According to Refs. 1–3, an exciton-exciton interaction shifts the emission line of a free exciton to the vicinity of the energy of a trapped exciton. The gain mechanisms responsible for the observed lines are thus linked with joint exciton processes.

In this letter we are reporting a low-temperature study of the electroluminescence spectra of GaSe for various strengths of the electric field (E) applied to the test sample in the observation configurations $\mathbf{q}\perp\mathbf{c}$ and $\mathbf{q}\parallel\mathbf{c}$, where $\mathbf{q}$ is the emission wave vector, and $\mathbf{c}$ is the optical axis of the crystal.

The experiments were carried out in GaSe samples grown by the Bridgman–Stockbarger method. The samples had a resistivity of 10^5–10^6 $\Omega\cdot$cm at 300 K and dimensions of $3\times5\times0.1$ mm. The samples were prepared by simple cleavage along a cleavage plane. Samples in which the ϵ polymorph was predominant among the various modification were selected on the basis of x-ray diffraction. Contacts were applied to the freshly cleaved surface. The electrode consisted of a eutectic In–Ga mixture. To ensure that contact effects at the electrode–crystal interface were small, we recorded current–voltage characteristics at 77 K in both the forward and reverse directions.

Figure 1 shows a series of electroluminescence spectra of GaSe at 77 K in the two observation configurations ($\mathbf{q}\perp\mathbf{c}$, $\mathbf{q}\parallel\mathbf{c}$) with increasing strength of the applied electric field. The insets show the relative intensity of emission line A versus the field E and the energy position of line A in larger scale. The energy positions of the electroluminescence lines observed are as follows: 595 nm for line A, 615 nm for line B, 667 nm for line C, and 688 nm for line D. One distinctive feature of the $\mathbf{q}\perp\mathbf{c}$ configuration is the presence of a strong resonant line, A, which is due to the insignificant level of reabsorption as the light passes through the test sample. Other distinguishing features of this configuration are the narrowness of peaks A and B and the absence of an energy shift of these peaks.

Analysis of the results reveals that two stimulated-emission bands, A and B, in the configurations $\mathbf{q}\perp\mathbf{c}$ and $\mathbf{q}\parallel\mathbf{c}$, are present near the fundamental absorption edge. These bands arise at various strengths of the applied electric field. In other words, two mecha-

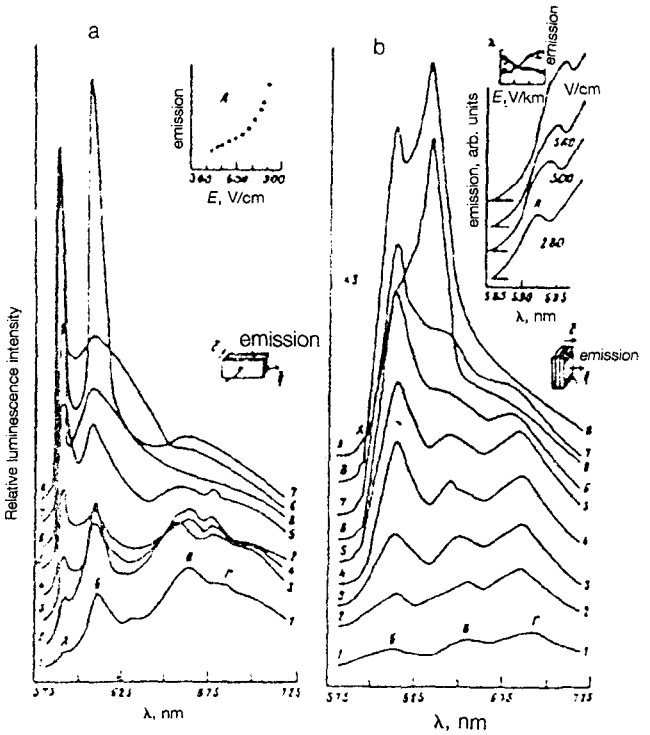

FIG. 1. Electroluminescence spectra of GaSe at 77 K in two observation configurations for various values of the field E. a: $\mathbf{q}\perp\mathbf{c}$. Line 1) 450 V/cm; 2) 530; 3) 550; 4) 660; 5) 720; 6) 760; 7) 820; 8) 840 V/cm. b: 1) 230 V/cm; 2) 280; 3) 340; 4) 390; 5) 450; 6) 500; 7) 560; 8) 670; 9) 780 V/cm. The insets show the relative intensity (for $\mathbf{q}\perp\mathbf{c}$ and $\mathbf{q}\|\mathbf{c}$) versus E and the energy position ($\mathbf{q}\|\mathbf{c}$) of exciton emission line A.

nisms are operating to create a population inversion in the GaSe crystals. These two mechanisms differ in threshold voltage.

For lines A and B, the superlinear E dependence rules out impurity emission, radiative recombination of a trapped exciton, radiative recombination of a free exciton, and a phonon repetition of a free-exciton line as explanations. The energy positions of lines A and B are not the same as those of the photoluminescence lines due to radiative recombination of free direct and indirect excitons. In addition, the behavior (curve 7) at $720 < E \leqslant 820$ V/cm in the configuration $\mathbf{q}\perp\mathbf{c}$ and that (curve 8) at $450 < E \leqslant 670$ V/cm in the configuration $\mathbf{q}\|\mathbf{c}$ are superlinear. Working from these results and the polarization and the directionality of the emission in lines A and B, we can suggest that we are observing a stimulated emission due to an exciton–exciton interaction.[4] Figure 2 shows the angular distribution of the emission at 820 V/cm in the configuration $\mathbf{q}\perp\mathbf{c}$. In contrast with bands A and B, lines C and D do not exhibit gain even at high values of the applied electric field. The short-wave shift with increasing E and the long-wave shift with the delay in measurement after the exciting pulse show that the most likely mechanism for the emission in bands C and D is a recombination of donor–acceptor pairs.[5]

We note in conclusion that the observation of a stimulated emission in layered compounds of this sort shows that these compounds hold promise as new laser active media.

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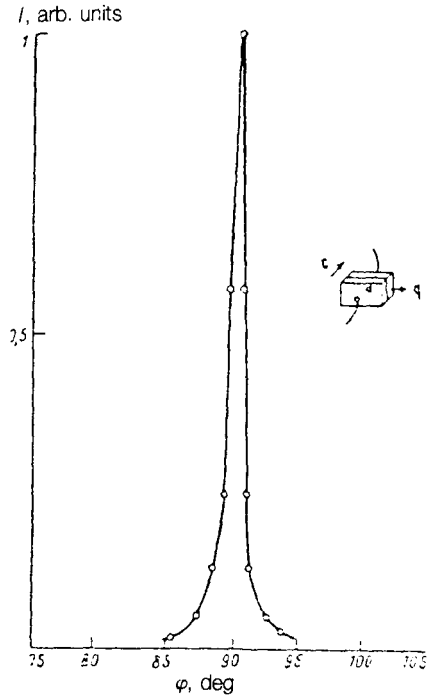

FIG. 2. Angular distribution of the emission at 820 V/cm in the configuration $\mathbf{q} \perp \mathbf{c}$. Here φ is the angle between the emission wave vector $\mathbf{q}$ and the plane of the monochromator slit.

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[4]T. Ugumori *et al.*, Solid State Commun. **12**, 389 (1973).
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Translated by D. Parsons