Biexcitons in the emission spectrum of uniaxially deformed silicon

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A new line corresponding to the radiative decay of a biexciton was observed in the emission spectrum of Si elastically deformed along the ⟨100⟩ axis at large excitation densities and at low temperatures. The binding energy of the biexciton is estimated at $\Delta \approx 1.3$ meV.

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The question of the experimental observation of biexcitons in the emission spectra of the indirect semiconductors Si and Ge remains open. The complications that arise here are due to the fact that the biexciton in these crystals is much less stable than the electron-hole liquid (EHL).[1] The biexciton gas therefore remains strongly dissociated even at saturated exciton-gas densities at which condensation into EHL sets in. Conditions for the observation of biexcitons become more favorable under uniaxial stresses. It is known that in the indirect semiconductors Si and Ge uniaxial strains lift the degeneracy in the bands, as a result of which the average kinetic energy per pair of particles in the EHL increases.[2,3] As a result, the binding energy of the EHL decreases, and the saturated-vapor density of the exciton gas increases appreciably. Estimates show that in silicon deformed along ⟨100⟩ the partial pressures of the exciton and biexciton gases become practically equalized along the exciton gas—EHL coexistence line.

We investigated the emission spectra of silicon strongly deformed along the ⟨100⟩ axis [so-called Si(1—2), where the first number denotes the multiplicity of degeneracy of the valence band and the second the number of electron valleys]. We used high purified samples with electrically active impurity concentration $N_a \lesssim 3\times10^{12}$ cm^{-3}. The samples were rectangular parallelepipeds with dimensions $1\times3\times7$ mm. The homogeneity of the strains along the largest linear dimension of the crystal was high enough and was monitored against the shape of the free-exciton emission line. The spectra were excited with a pulsed copper-vapor laser and with a cw GaAs laser and were registered by the procedure described in[4].

The kinetics of the emission spectra of Si(1—2) under pulsed excitation with an average e-h pair density $\bar{n}\sim3\times10^{17}$ cm^{-3} and at $T=1.8$ K is shown in Fig. 1. For comparison, Fig. 1 shows the spectrum of the undeformed Si. At the instant of the excitation pulse, the EHL radiation predominates (the L band in Fig. 1). The spectra show clearly the emission lines of the free excitons—FE—and there are no exciton lines bound on the impurity. From an analysis of the shape of the EHL spectrum, which will be reported separately, it follows that in Si(1—2) the carrier density in the EHL is $n_L=4.5\times10^{17}$ cm^{-3} and the binding energy is $\phi=2\pm0.2$ meV. These values agree well with calculation.[5] We note that in Si(2—6) we have $\phi=8$ meV.[1,4] As a result of the decrease of the bind-

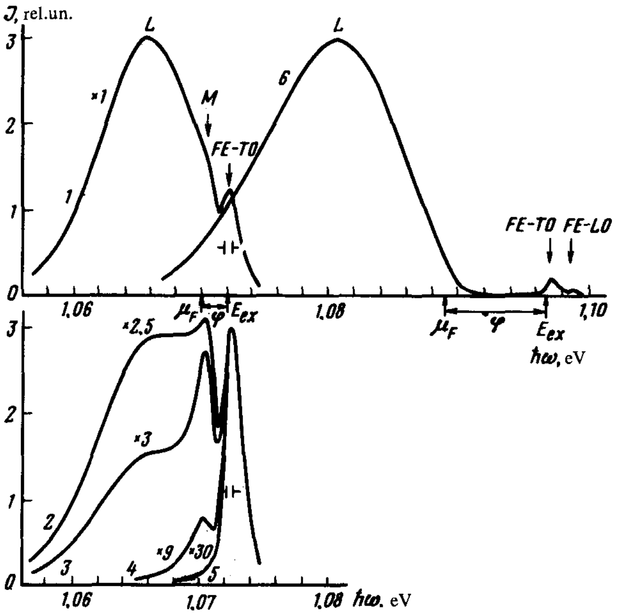

FIG. 1. Kinetics of the recombination radiation spectrum of Si(1—2) under pulsed excitation (~3.10^{17} cm^{-3}) and at $T = 1.8$ K. Spectra 1—5 correspond to respective delays τ_d (μsec) 0, 0.25, 0.35, 0.7, and 1.1 relative to the exciting pulse. Spectrum 6 is that of the undeformed silicon at the instant of the excitation pulse.

ing energy of the EHL in Si(1—2), the density n_{ex} of the saturated exciton gas increases by approximately one order of magnitude.

The kinetics of the Si(1—2) spectra show that in addition to the *L* and *FE* lines the spectra contain one more line, *M*, which appears at stresses $P \gtrsim 20$ kg/mm². The distance between the maxima of the *M* and *FE* lines is independent of the pressure and is equal to 2 meV. The shape of the long-wave wing of the *M* line remains unchanged after the damping of the EHL emission line.

The behavior of the *M* and *FE* lines when the temperature is varied is illustrated by Fig. 2. The spectra were measured under conditions when there was no EHL emission and the ratio of the intensities at the maxima I_M and I_{FE} was constant. This was accomplished by registering the spectra at different delays relative to the laser pulse. With increasing temperature, the spectra correspond to smaller delays, and consequently to larger values of n_{ex}. It is seen from Fig. 2 that it is the violet edge of the *M* line which is sensitive to temperature.

The *M* line cannot be connected with recombination of *e-h* pairs or excitons belonging to the split bands, since the quantity $\hbar\omega_{FE} - \hbar\omega_M$ does not depend on the pressure. One should exclude from consideration recombination processes in which impurities take part, since the emission line of the excitons bound on neutral impurities (*BE*) is 2 meV farther towards the red than the *M* line. In

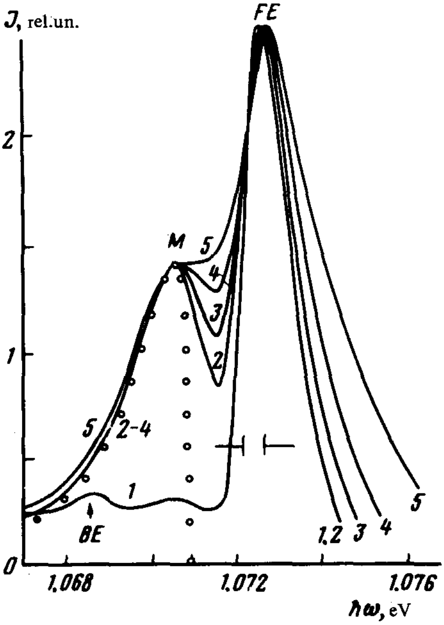

FIG. 2. Temperature dependence of the emission spectra of the excitons (*FE*) and of the biexcitons (*M*) under pulsed pumping ($\sim 3\times10^{17}$ cm^{-3}). Spectra 2—4 were recorded at the following temperatures and time delays relative to the excitation pulse: 2—2 K, 0.3 μsec; 3—4.2 K, 0.25 μsec; 4—7 K, 0.17 μsec; 5—10 K, 0.1 μsec. Curve 1 pertains to stationary pumping ($\sim 10^{15}$ cm^{-3}).

Fig. 2, spectrum 1 corresponds to a stationary pump at $n \sim 10^{15}$ cm^{-3}. At such relatively low excitation densities I_{BE} has a slower-than-linear dependence on I_{FE} and is practically saturated, whereas the *M* line is barely started and increases in faster than linear fashion with further increase of the excitation.

In our opinion, the most probable recombination mechanism responsible for the appearance of the *M* line in the Si(1—2) spectrum is radiative decay of a biexciton with production of an exciton and a *TO* phonon. The emission spectrum of a biexciton and an indirect semiconductor is given by

$$I(\hbar\omega) \sim \iint d\mathbf{K}\,d\mathbf{k}\,|D|^2 \exp\left(-\frac{h^2\mathbf{K}^2}{4mk_0T}\right)\ \delta(E_M(\mathbf{K}) - E_{ex}(\mathbf{k}) - \hbar\Omega^{TO} - \hbar\omega), \tag{1}$$

where E_M and E_{ex} are the ground-state energies of the biexciton and exciton, **K** and **k** are the corresponding quasimomenta, and D is the matrix element of the dipole moment, for the calculation of which it is necessary to know the wave function of the biexciton. Since the wave function of the biexciton has a characteristic dimension on the order of the exciton radius (a_{ex}), it is natural to expect $D(\mathbf{K},\mathbf{k})$ to be significant at $|\frac{1}{2}\mathbf{K}-\mathbf{k}| < a_{ex}^{-1}$. Then the characteristic exciton recoil energy, which determines the line width, is $\hbar^2/2ma_{ex}^2$. The emission line width Γ of the biexciton is therefore on the order of $\Gamma \sim (\mu/m)R_{ex}$, where μ and m are the reduced and translational masses of the exciton, while R_{ex} is the exciton Rydberg. Thus, the width of the biexciton emission band in Si should be approximately 3 meV, corresponding to the observed width of the *M* line. Next, according to (1), the long-wave wing of the biexciton line is independent of T at $k_0T < (\mu/m)R_{ex}$, whereas the violet edge should broaden in accordance with the distribution of the biexcitons in the band, a fact which likewise agrees with experiment.

If we use for $D(\mathbf{K},\mathbf{k})$ the model-dependent expression[6]

$$D(\mathbf{K},\mathbf{k}) \sim [(\tfrac{1}{2}\mathbf{K}-\mathbf{k})^2 + a_{biex}^{-2}]^{-2} \qquad (2)$$

then the calculation for $I(\hbar\omega)$ at $T=0$ K describes well the long-wave wing of the experimental contour of the M line (points in Fig. 2). At this approximation for the line shape we obtain for the biexciton binding energy in Si a value $\Delta \approx 1.3$ meV or $\Delta/R_{ex} \approx 0.09$. Calculation[7] yields $\Delta = 0.4$ meV and $\Delta/R_{ex} \approx 0.03$.

We note finally that we have investigated also the recombination spectra of Si crystals deformed along the axes ⟨111⟩ and ⟨110⟩ [Si(1—6) and Si(1—4), respectively]. We were unable to observe the biexciton emission line at these deformations, but the spectra did reveal a noticable increase of the radiation background between the EHL and exciton bands in comparison with the undeformed crystal.

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