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Lasing in a $ZnS_{0.12}Se_{0.88}$/ZnSe multilayer structure with photopumping

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Photopumped lasing in a $ZnS_{0.12}Se_{0.88}$/ZnSe multilayer structure up to 180 K is reported for the first time. The films were grown by metalorganic vapor phase epitaxy on (001) GaAs. The purpose of using the multilayer structure is to prevent the diffusion of the photoexcited carriers to have population inversion sufficient for lasing. The possibility of lasing at the higher temperature is briefly discussed.

ZnSe or ZnS_xSe_{1-x} alloys have been extensively studied to realize blue-light emitting diodes and laser diodes. For this purpose, the control of the electrical property, especially *p*-type, is necessary for current injection and the situation is being improved by trials with lithium[1-3] or nitrogen[4,5] dopings in the films grown on (001) GaAs substrates. As for the optical property to obtain lasing, electron beam pumping[6,7] and photopumping[8,9] are being tried at present. In the former excitation method, a high density of electron-hole pairs is easily excited deep into the semiconductors with the incident energetic electron beam. On the other hand, the photopumping is localized at the semiconductor surface and is more difficult to obtain lasing. But it is, in a sense, a convenient test of the optical property for laser action. Up to now, lasing in the blue region with photopumping remains at low temperature below 10 K.[8,9]

Although ZnSe films are nearly lattice matched to GaAs substrates, the presence of misfit dislocations above the critical thickness of 1500 Å was formerly identified by the increase of the minimum backscattering yield in the Rutherford backscattering[10] and by the interface diffusion enhanced along the misfit dislocations with thermal annealing observed electrically[11] and optically.[12] The influence of the misfit dislocations on the photoluminescence (PL) efficiency was studied by the comparison of the near-band-edge blue PL intensities in the undoped ZnSe films and those in the $ZnS_{0.06}Se_{0.94}$ films lattice matched to GaAs substrates.[13] The former ones gave at least one order of magnitude smaller PL levels than the latter ones, indicating the increase of nonradiative recombinations due to the misfit dislocations present in the ZnSe films. Therefore, photopumping in the lattice-matched $ZnS_{0.06}Se_{0.94}$ films with the thicknesses of 1–1.5 μm was tried, but lasing action was not obtained even at the low temperature near 10 K. The main factor to this failure was identified to be the diffusion of the photoexcited carriers from the $ZnS_{0.06}Se_{0.94}$ film into the GaAs substrate, which was identified by the abrupt increase of the GaAs PL level in the higher excitation range.[13]

In this letter, photopumped lasing up to 180 K is reported in $ZnS_{0.12}Se_{0.88}$/ZnSe periodic multilayers grown on GaAs substrates. The main factor for the improved lasing property is attributed to the suppression of the carrier diffusion with the heterobarriers. The possibility of lasing at the higher temperature is discussed following a simplified model developed to study the measured lasing properties.

ZnS_xSe_{1-x} films were grown by metalorganic vapor phase epitaxy at atmospheric pressure using diethylzinc (DEZn), diethylselenide (DESe), and diethylsulfide (DES). The films were grown at 500 °C with the fixed DESe/DEZn ratio of 6 and the DEZn flow rate of 2.5 μmol/min. The S composition was controlled by the flow rate of DES. The layer structure used for the optical pumping is shown in Fig. 1. After the 0.36 μm $ZnS_{0.06}Se_{0.94}$ buffer layer was lattice matched to the GaAs substrate, 12 periods of 300 Å $ZnS_{0.12}Se_{0.88}$/300 Å ZnSe layers were grown. The film thicknesses are the values expected from the respective growth rate. The $ZnS_{0.12}Se_{0.88}$ and ZnSe have 0.54% lattice mismatch and the critical thickness for causing misfit dislocations is about 620 Å,[14] which is much larger than the layer thicknesses used in the present experiment. In this structure, the free-standing in-plane lattice constant of the multilayer structure is very close to that of GaAs (or $ZnS_{0.06}Se_{0.94}$) and the lattice mismatch is just 4×10^{-3}%. The wafer was cleaved into a bar with the cavity length of 1.5 mm. The sample was excited with a pulsed N_2 laser (3371 Å) on the surface using a cylindrical lens.

The observed spectra below and above the lasing threshold are shown in Fig. 2. Below the threshold, broad spontaneous emission was observed. Above the threshold, sharp stimulated emission peak superposed on the spontaneous emission at the longer wavelength appeared. This is because the gain peak appears in the longer wavelength of the spontaneous emission in the band-to-band transitions.[15] The interval of the longitudinal modes is about 0.23 Å in the present structure and the measured half width of the stimulated emission (about 7 Å) is limited by the measurement resolution. The lasing peak shifted to the longer wavelength with the increase of the temperature as shown in Fig. 3. The shift is linear to temperature up to the measured 180 K and is 0.94 Å/K. This value is smaller than the temperature dependence of the energy gap, 1.5 Å/K, which may be due to the increase

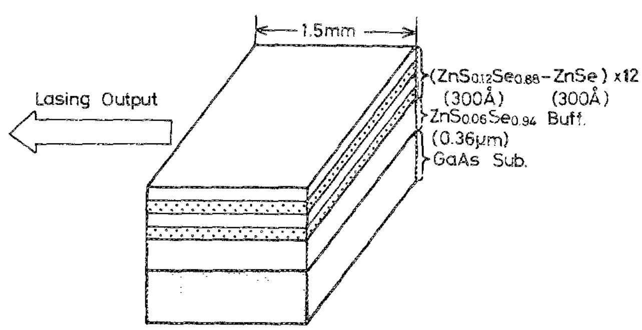

FIG. 1. Schematic layer structure for photopumped lasing.

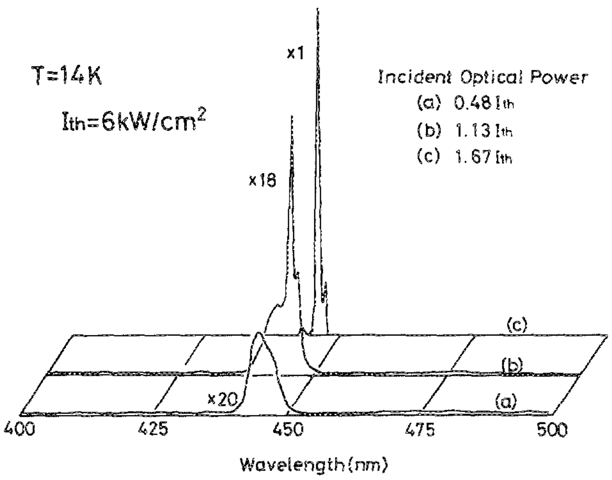

FIG. 2. Observed typical spectra below and above the lasing threshold.

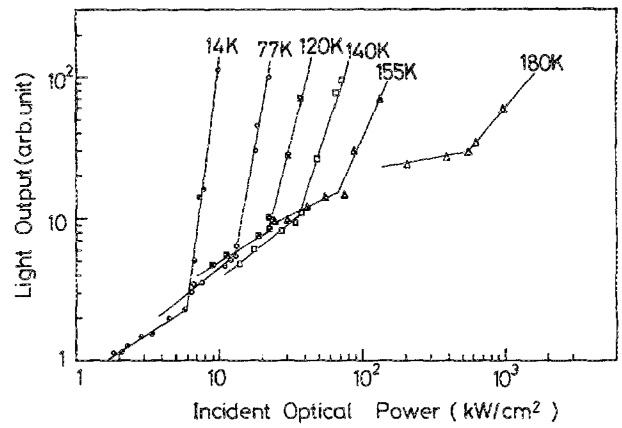

FIG. 4. Light output vs incident optical excitation power dependence measured at the different temperatures. Lasing up to 180 K was observed.

of the carrier concentrations necessary for lasing with the increase of the temperature and the resultant gain peak shift to the higher energy.

The light output versus incident excitation optical power characteristics are shown in Fig. 4. The characteristics show clear threshold properties inherent in lasing actions. Lasing up to 180 K is demonstrated in Fig. 4. The temperature dependence of the threshold optical power is shown in Fig. 5. The T_o value defined as $I_{th}(T_1) = I_{th}(T_2) \times \exp[(T_1 - T_2)/T_0]$ was about 74 K below 140 K and the threshold increased drastically above 150 K.

The measured temperature dependence of the threshold optical power was studied with a simplified model assuming the semiinfinite periods of 300 Å $ZnS_{0.12}Se_{0.88}$/300 Å ZnSe layers. The absorption coefficients of ZnSe at the photon energy of 3.68 eV of the excitation N_2 laser, which is about 0.8 eV above the ZnSe energy gap, will be very large and the photoexcitation will be restricted to the region very near to the surface, on the order of 1000 Å. Therefore, the photoexcitation of the electron-hole pairs was assumed to be restricted to the first ZnSe layer nearest to the surface to simplify the problem. The barrier height at the $ZnS_{0.12}Se_{0.88}$/ZnSe heterointerface was estimated in Ref. 16 taking the strain effects into account, and the band offsets in the conduction band and in the valence band were predicted to be $\Delta E_c \sim 0$ and $\Delta E_v \sim 100$ meV, respectively. Therefore, the electrons can diffuse easily, while the holes experience barrier heights. This may result in the different depth carrier distributions between the electrons and holes, but the resultant electric field will work to reduce the difference of the carrier distributions. The detailed description of these phenomena is rather complicated, and it was tentatively assumed that the electrons redistribute in depth to satisfy the neutral condition and that the carrier distribution is limited by the hole dynamics.

In the periodic potential wells in the valence band of $ZnS_{0.12}Se_{0.88}$/ZnSe multilayers, the diffusion effect in the thin 300 Å ZnSe layer can be neglected and the hole depth distribution is limited by the thermoionic emission over the $ZnS_{0.12}Se_{0.88}$/ZnSe heterobarrier in the measured low-temperature range. The tunneling effect through the 300 Å $ZnS_{0.12}Se_{0.88}$ barrier was neglected. Then the balance equation among the electron-hole recombinations within the layer and the thermoionic emission and injection from the neighboring layers can be obtained in each layer except for the first layer nearest the surface, where the photoexcitation of electron-hole pairs is assumed. Considering that the car-

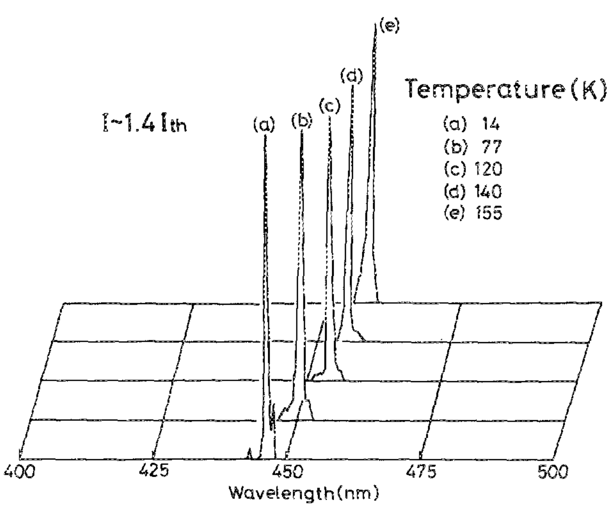

FIG. 3. Temperature dependence of the lasing spectra. Wavelength shift is linear to the temperature up to the measured 180 K and is 0.94 Å/K.

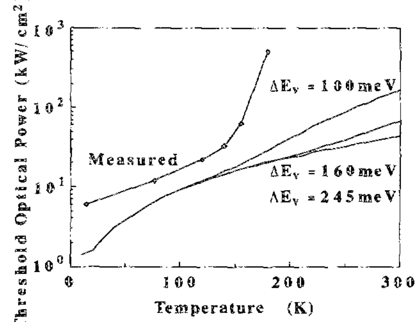

FIG. 5. Temperature dependence of the lasing threshold optical power. The measured threshold increased abruptly above 140 K. The theoretical curves were calculated with a model which took into account the thermionic emission. The calculated curve with the valence-band offset of $\Delta E_v = 100$ meV corresponds with the measurement.

rier concentration in the infinite depth is negligible, the following simple relation is derived under the Boltzmann approximation:

$$\Phi = J_0 \exp[(E_v - E_{fv1})/kT](\alpha - 2 + \sqrt{\alpha^2 - 4})/2, \quad (1)$$

where Φ is the incident photon flux, J_0 is the thermionic emission term given by $J_0 = [4\pi m_v^*(kT)^2/h^3] \times \exp(-\Delta E_v/kT)$, and α is defined as $\alpha = N_v W/(J_0\tau) + 2$, where W is the ZnSe layer thickness and a value of 300 Å was used. The threshold condition was simply assumed to follow the Bernard–Duraffourg relation $F_c - F_v = E_g + \delta E > E_g$ in the first ZnSe layer. Then the Boltzmann factor in (1) is given by $\exp[(E_v - E_{fv1})/kT] = (N_c/N_v)^{1/2} \exp(\delta E/2kT)$, where N_c and N_v are the effective density of states in the conduction and valence bands, respectively. The extra Fermi level separation above the energy gap δE and the recombination lifetime τ were tentatively assumed to be 3 meV and 0.3 ns, respectively, in the calculations. The mass values for ZnSe used for the conduction and valence bands are 0.17 and 0.60, respectively.

In Fig. 5 the calculated results for the three values of the valence-band offset are shown. The corresponding S composition in the ZnS_xSe_{1-x}/ZnSe structure is 0.12, 0.2, and 0.3 for the ΔE_v values of 100, 160, and 245 meV, respectively. Therefore, the curve for $\Delta E_v = 100$ meV corresponds to the measurements and the temperature of 140 K above which the extra increase of the threshold power is seen in reasonable agreement with the measurements. Figure 5 also indicates that the $ZnS_{0.3}Se_{0.7}$/ZnSe structure is sufficient to suppress the excess increase of the threshold up to room temperature and the study is under way.

In conclusion, lasing in the blue-light region was realized up to 180 K with the photopump of the $ZnS_{0.12}Se_{0.88}$/ZnSe periodic multilayer. Diffusion of the photoexcited electron-hole pairs was effectively suppressed in the multilayer structure, and the main factor which limits the maximum lasing temperature was shown to be the thermionic emission over the heterobarrier at the $ZnS_{0.12}Se_{0.88}$/ZnSe interface.

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[15] For example, see H. C. Casey and M. B. Panish, *Heterostructure Lasers* (Academic, New York, 1978), Chap. 3.
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