Laser action in a-Si:H

- A. M. Danishevskiĭ, V. Latinis, S. Yurshchenas, V. I. Gavryushin,
- M. M. Mezdrogina, and E. I. Terukov
- A. F. Ioffe Physicotechnical Institute, Russian Academy of Sciences, 194021, St. Petersburg

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Laser action between definite energy states in band tails has been observed in measurements of the photoluminescence spectra of films of amorphous hydrogenated silicon with intense picosecond excitation at $T \sim 80$ K. The particular energy at which the emission occurs is determined by a selective resonator whose preferred energies are associated with the film thickness.

The photoluminescence of amorphous hydrogenated silicon has been studied in detail with both cw and pulsed excitation. The usual spectrum of intrinsic photoluminescence has a broad band (0.22–0.25 eV) with a peak at about 1.33–1.35 eV (at 80 K). There is also a small band of extrinsic luminescence, again fairly broad, with a peak at 0.9 eV.

In the case of pulsed excitation² (by pulses with a length of several nanoseconds), the peak of the intrinsic luminescence band is initially near 1.4 eV; its short-wave tail reaches an energy of 1.6 eV at a substantial excitation level.

Luminescence spectra are usually measured on films deposited on substrate surfaces with a matte finish, in order to avoid possible interference effects. In all cases of

which we are aware, the intrinsic luminescence band has been broad and structureless.

In this letter we are reporting a study of the photoluminescence spectra of several undoped a-Si:H samples on a polished quartz substrate. The luminescence was excited by intense picosecond pulses. The samples were in a tetrode system in an rf glow discharge with decomposition of pure silane. The substrate temperature was 280 °C. The optical width of the energy gap in the test samples was 1.85–1.9 eV. The Fermi energy was 0.8–0.95 eV from the bottom of the conduction band. The concentration of intrinsic defects was less than 8×10^{15} cm⁻³. The results which we are reporting here were obtained on a sample with a minimal defect concentration $(N_d=3\times10^{15}~{\rm cm}^{-3})$, with the lowest energy parameter of the Urbach tail $(E_u=50~{\rm meV})$ in the absorption spectrum, and with an activation energy of 0.92 eV. Since the film with a thickness of 0.95 μ m has a region of UV photoconductivity, we conclude that the rate of surface recombination is low, so the film is not columnar.

The samples were excited by the second harmonic of an Nd:YAG laser ($h\nu=2.33$ eV) generating single picosecond pulses (35 ps) at a frequency of 2 Hz. The samples were mounted on the cold conductor of a nitrogen vacuum cryostat. The sample temperature was 80 K. The measurements were carried out in an arrangement of reflection from the film surface. The luminescence spectra were recorded with the help of an MDR-2 monochromator with an FÉU-83 photomultiplier. An OS-14 filter was positioned at the front slit to prevent detection of the scattered light of the exciting pulses. An OS-12 filter was placed at the lens which focused the recombination light onto the front slit of the monochromator. The electrical output signals from the photomultiplier were fed to a broad-band amplifier with a bandwidth of $(0.7-10)\times10^6$ Hz and then to a V9-5 boxcar converter. The digitized signals from this converter were fed through CAMAC units to a DVK-2 microcomputer. During the recording of the spectrum, each spectral point was found as an average over 30-40 pulses in a given interval of heights of the exciting pulses.

Figure 1 shows the luminescence spectra of sample M912 recorded under the conditions described above, for four values of the energy of the exciting pulses. All four spectra were measured with a delay of 80 ns after the beginning of the pulse.¹⁾

Spectrum 1 was recorded at a minimal intensity of the excitation pulses. This spectrum is considerably narrower than that of Ref. 2; the width of the line at 1.39 eV is less than 20 meV. Spectrum 2 was recorded at a higher intensity of the exciting light. This spectrum is considerably broader. Its peak and its short-wave part are at higher energies ($h\nu=1.405$ eV). The broadening of the main band is inhomogeneous. One might suggest that this broadening is the result of a superposition of two narrower bands, with energies of 1.395 and 1.41 eV. There are also several small peaks in the spectrum. Spectrum 3, recorded at an even higher excitation intensity, is even broader. Its main peak is at an energy of 1.415 eV. At an energy of 1.59 eV we find a fairly broad emission band. The peak of spectrum 4 is also at 1.415 eV. On its short-wave side there is a knee, and an intense narrow peak has appeared at 1.59 eV.

Note also that the growth of the luminescence bands as a function of the pump intensity is much faster than linear at relatively low intensities (spectra 1 and 2), while this growth slows down considerably (near the main band) at high pump intensities.

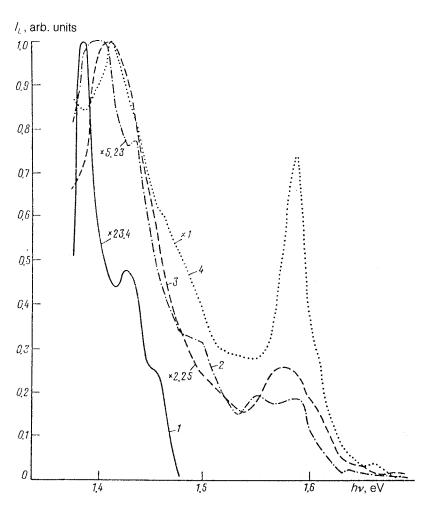


FIG. 1. Photoluminescence spectra of an a-Si:H sample (M912) with intense picosecond excitation at T = 80 K. 1—The average energy of the exciting pulses is 1.1 μ J; 2—2.25; 3—6.45; 4—17 μ J.

Estimates show that the excess density of carriers excited per pulse is greater than 10^{19} cm⁻³, even at the lowest pump intensity. The nature of the spectra shown here thus suggests that lasing is occurring near an energy of 1.39 eV in this film, even at the lowest excitation intensities. As the pump intensity is raised, the entire luminescence spectrum broadens toward higher photon energies. The band at 1.39 eV appears to reach saturation, but laser action arises on nearby transitions, and the band near 1.4 eV broadens and shifts in the short-wave direction. The reason for this behavior of the spectrum is that, as the generation rate increases in this range, states in the band tails become filled, and the time scales for the dissipation of this population are fairly long (microseconds). At sufficiently high intensities I, at which all the low-lying states are filled, laser action arises in the band at 1.59 eV.

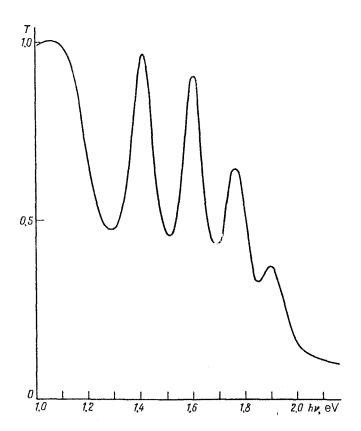


FIG. 2. Transmission spectrum of an a-Si:H sample (M912) at T = 300 K.

Why are the energies of 1.4 and 1.59 eV prominent in the luminescence spectrum? The answer to this question is provided by measurements of the transmission spectrum of the films. Figure 2 shows a T(hv) spectrum for sample M912. In its transmission region there are some interference peaks. Two of them, with energies of 1.405 and 1.59 eV in the transmission peak, fall in the range of the luminescence spectrum. Consequently, near these energies the film is a single-mode resonator with a very low loss. This is why these energies are selected in the superradiance spectrum.

The luminescence spectra recorded at a strobe delay of 180 ns are similar to those reported here. At an even greater delay (580 ns), however, the narrow bands nearly disappear from the spectrum; i.e., the population inversion has time to relax.

Translated by D. Parsons

¹⁾Because of the transient characteristic of the particular photomultiplier amplifier which we used, it was not possible to set the actual time at which the optical luminescence pulse was strobed. Nevertheless, the spectra recorded at various strobe delays were quite different.

¹R. A. Street, Adv. Phys. 30, 593 (1981).

²S. Kurita, M. Czaja, and S. Kinmond, Solid State Commun. 32, 879 (1979).