Extended Abstracts of the 16th (1984 International) Conference on Solid State Devices and Materials, Kobe, 1984, pp. 125-128

Invited

Laser Oscillations from the Internal Levels of an Impurity in a Semiconductor

P. B. Klein

U.S. Naval Research Laboratory

Washington, D.C. 20375, USA

A new class of laser, the semiconductor-impurity laser, is described and compared to the standard types of solid state laser systems. The single known example of this laser, InP:Fe (λ_L~3.53 μm) is described with emphasis on the processes involved in exciting stimulated emission and on the current limitations of this system. The possibility of extending this idea to other impurity/host combinations is also discussed.

In this paper a new type of solid state laser is described which could potentially make the laser transitions associated with transition metal and rare-earth ions in insulators available in semiconductor hosts. This type of laser system could offer some of the advantages of, e.g., the Nd:YAG and ruby lasers with the added features of electrical rather than optical excitation, and the obvious compatibility with existing semiconductor technology. This "semiconductor impurity laser" is analogous to the insulator-impurity lasers mentioned above in that the laser emission arises from transitions between the discrete, internal electronic levels of transition metal or rare earth impurities in the host material. But because of the semiconductor nature of the host, it is found that the host material plays a central role in the energy transfer process associated with the population of the upper laser level. This should be compared with optical excitation via the internal absorption by the impurity ion itself in insulator impurity lasers such as Nd:YAG. The semiconductor-impurity laser is also distinct from the familiar semiconductor laser in that the latter derives laser emission from the recombination of electrons and holes present in the junction of a semiconductor diode. Although impurities are, of course, involved in creating the p-n junction, the emission wavelength is tied closely to the semiconductor band gap, and is unrelated any internal impurity levels.

To date, laser oscillations near 3.5 μm from Fe^{2+} in InP, excited under pulsed optical excitation, is the only known example of a semiconductor impurity laser.[1] In this system the Fe center resides on a substitutional In site and is observed in two charge states, a neutral acceptor state $Fe^{3+}(3d^5)$ and a singly ionized acceptor state $Fe^{2+}(3d^6)$. Photoluminescence (PL) is observed from transitions within the lowest ^5D term of Fe^{2+}, which is split by the tetrahedral crystal field of the InP host into an excited $^5T_2$ multiplet and a ground ^5E multiplet, separated by ~0.35 eV. Further spin-orbit and spin-spin splittings result in a $^5T_2 \rightarrow {}^5E$ PL spectrum consisting of four sharp lines spaced ~14 cm^{-1} apart near 0.35 eV (~3.5 μm).

In order to develop other similar laser systems or to improve the InP:Fe system, it is important to develop an understanding of the processes that are responsible for populating the upper laser level. To this end, a study of the time dependence of the 3.5 μm Fe^{2+} PL signal was undertaken[2] in a series of Fe-doped samples ranging from n-type to semi-insulating (SI). For n-type and closely compensated samples it was found that after the end of the 8 nsec excitation pulse ($\lambda_L \cong 5800$ Å) the PL intensity continued to grow in time (with a characteristic growth time of ~200 nsec) long after the photoexcited e-h

pairs had recombined (≲1 nsec), reaching a peak at ~1 μsec, and then decaying exponentially (τ~11 μsec). In contrast to this, the PL signal in SI samples was observed to be weaker, and to decay immediately after the excitation pulse ended. This fact and subsequent data were interpreted in terms of the model depicted in Fig. 1. On the left side of Fig. 1a is shown a schematic representation of Fe levels in n-type material before optical excitation. All of the Fe has compensated shallow donors and is in the Fe^{2+} state. The net effect of an intense, above-bandgap optical pulse is to convert (via hole capture or direct photoionization of Fe^{2+}) some of the Fe^{2+} to neutral Fe^{3+} and an equal number of excess conduction band electrons. This non-equilibrium state is shown for times after the end of the exciting pulse on the right side of Fig. 1a. The system relaxes back to the equilibrium configuration via electron capture by the nonequilibrium Fe^{3+}, creating Fe^{2+} preferentially in the excited $^5T_2$ state (the upper laser level). This capture process can be relatively slow, and is associated with the 200 nsec characteristic growth of the PL signal after the exciting pulse.

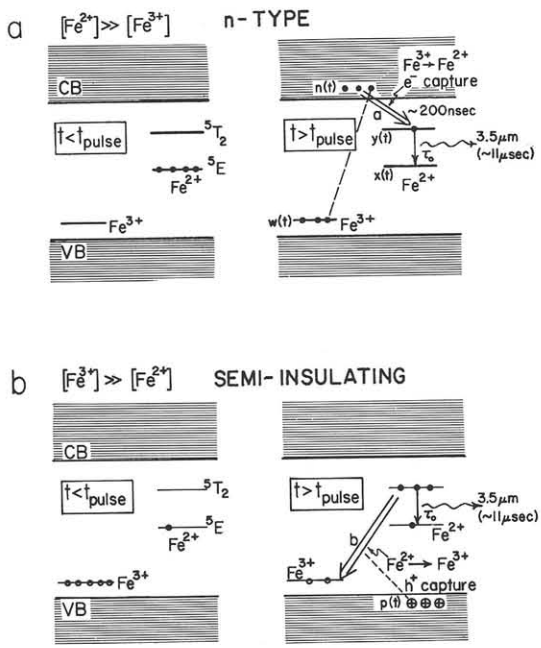

Fig. 1 Models for the kinetics of excitation and recombination at the Fe center in InP for a) n-type samples, and b) semi-insulating samples. The left side of the figures corresponds to times before the exciting pulse, and the right side corresponds to times after the end of the pulse and after photoexcited carrier recombination.

Radiative decay (~11 μsec) then occurs between the excited $^5T_2$ level and the ground ^5E states.

The situation is quite different in SI samples with $[Fe^{3+}]>>[Fe^{2+}]$, as depicted on the left side of Fig. 1b before the exciting pulse. As a result of the exciting laser pulse, some of the Fe^{3+} is converted to Fe^{2+} and an equal number of excess valence band holes via electron capture, so that the net effect of the excitation is to leave the nonequilibrium situation depicted on the right side of Fig. 1b. The system relaxes back to equilibrium via a hole capture process on Fe^{2+} that can compete with the radiative decay. There is also no mechanism for growth of the PL emission: the decay begins immediately after the end of the exciting pulse, as was observed in the experiment.

Upon increasing the excitation power, laser oscillations are observed, but only for n-type or closely compensated samples. This is shown in Fig. 2 as a plot of the time dependence of the 3.5 μm radiative emission for various excitation powers. At low power the PL time dependence exhibits growth after the exciting pulse followed by decay, and increases linearly with power. A very sharp laser threshold is reached (100-fold increase in intensity with a 4% increase in power), above which the intensity continues to increase nonlinearly with power. Several common characteristics of laser oscillations are reflected in the time-dependence of the radiative emission. The time delay before the onset of laser oscillations is observed to become shorter with increasing power, and the emission is dominated by a series of system-resolved spikes, presumably due to relaxation oscillation. The nonlinear increase in intensity is also found to be emitted in only one of the four spectral lines that characterize the Fe^{2+} radiative emission.

From the previous discussion the origin of the observed laser oscillations is relatively clear: the upper laser level ($^5T_2$) is populated at a much faster rate (~200 nsec) than it is emptied (~11 μsec), thus resulting in population inversion. It is noteworthy that the process that populates the $^5T_2$ excited state, electron capture by the Fe center, represents an energy transfer mechanism from the host to the impurity.

Thus the semiconductor host plays a central role in the excitation process, and in a sense may be viewed as a sensitizer for the Fe^{2+} lasing medium.

It can be seen from Fig. 2 that laser oscillations set in only after the exciting pulse has ended and the photoexcited electron-hole pairs have recombined. It is then that the system relaxes back to equilibrium, preferentially filling the $^5T_2$ excited state. For SI samples this is not the case, as decay of the $^5T_2$ population sets in immediately after the end of the exciting pulse. Population inversion in these samples must therefore be achieved <u>during</u> the exciting pulse. However, it is precisely during this time, when the photoexcited carrier density is high, that Auger effects[2,3] and competing hole capture processes, tend to de-excite the $^5T_2$ state nonradiatively. From these arguments it is understandable that laser oscillations are not seen in SI samples.

It is also important to note that the electron capture cross section associated with the process that populates the $^5T_2$ excited state is anomalously large for Fe in InP.[2] For example, the same process for GaAs:Fe is three to four orders of magnitude weaker (thus making population inversion impossible in this material). The large capture cross section in InP:Fe (and consequently the observed laser oscillations) has been attributed[2] to the involvement of a shallow level in the capture process. The important point to note here is that the achievement of laser oscillatons in a semiconductor impurity laser is very sensitive to the details of the energy transfer process between the semiconductor host and the impurity, and that this will be different for each different host/impurity combination.

There are several distinct limitations to the InP:Fe laser that will have to be overcome in order to develop this (or another host/impurity) system into a convenient device. First, the excitation density at threshold is very high, $\gtrsim 10^6$ W/ cm^2. Presumably this problem is related to the small hole capture cross section for Fe^{2+}. It is this process[2] that is responsible for the $Fe^{2+} \rightarrow Fe^{3+}$ conversion occurring during the exciting pulse that sets up the conditions (excess Fe^{3+} and an equal number of conduction band electrons) for population inversion after the pulse. As the capture cross section for holes is characteristic of the InP:Fe system, it would seem difficult to increase, except by possibly increasing temperature, without changing the host/impurity combination. The temperature is the second major problem: stimulated emission has only been observed for temperatures below 12 K. This effect is very probably due to the rapid broadening of the natural linewidth of the emission line with increasing temperatures. The

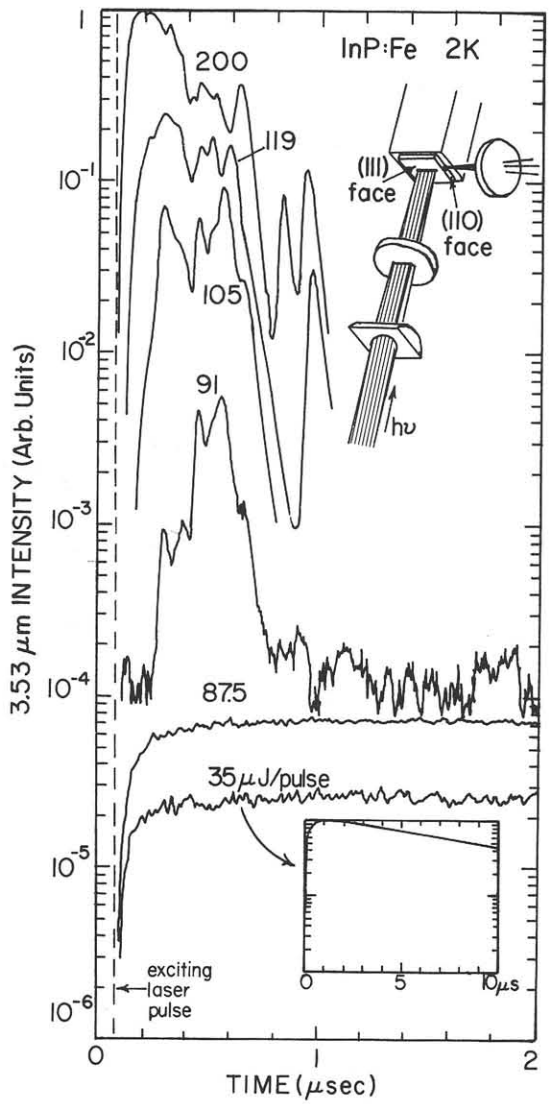

Fig. 2 Time dependence of the intensity of the 3.5 μm radiation for several values of the incident laser power above and below the threshold for laser oscillations.

linewidth figures directly into the threshold and gain of the laser system.[4] Temperature dependent CW PL measurements of InP:Fe exhibit[5] a rapid decrease in intensity and increase of the linewidth with increasing temperature, such that by 40K the sharp four-line PL spectrum has become a very broad, featureless signal. Unfortunately, this type of behavior is not unexpected for PL from other transition metal impurities as well, as the electron phonon coupling of the various 3d configurations is probably not all that different.

However, an excellent possibility for a higher temperature laser system appears to be rare-earth-doped semiconductors. Ennen et al.[6,7] have recently observed sharp-line PL spectra in InP, GaAs, GaP and Si implanted with Er and Yb. Because of the weaker coupling of the 4f levels to the lattice, as compared to the 3d levels of transition metals, the emission lines associated with internal transitions from rare earth impurities remain relatively narrow up to room temperature (increasing in width from 2 to 11 cm^{-1} between 4.2K and 300K). However, the process by which the rare earth impurities are excited is still unknown. Ennen et al.[6] have suggested the possibility of exciton migration, but this assignment is only tentative. Since the excitation processes are not understood, the conditions necessary for efficient pumping of the excited state are also not known. It is apparent, then, that further investigations of the excitation kinetics for the rare earth impurities are necessary if one hopes to observe laser oscillations from these materials.

Because they are able to be incorporated into Si, the rare earths also offer the advantages of Si-based device technology over the more difficult processing problems associated with III-V materials. In addition, the emission wavelength of Er (~1.54 μm) is very well matched to the high transparency range of SiO_2 optical fibers, while the 2.1 μm emission of Ho could prove compatible with halide glass optical fibers.

The transition from optical to electrical excitation in these systems may also prove to be challenging. Even for the case of InP:Fe, where the excitation processes are relatively well understood, results at 4.2K are not particularly encouraging. For several variations of Fe-doped p-n junctions, only weak Fe^{2+} electroluminescence has been observed.[8] The idea, of course, is to re-create the same conditions necessary for population inversion under optical excitation in an electrically excited device structure. This may prove to be difficult, particularly if Auger processes[3] tend to play a significant role. Further investigations in this area are clearly necessary, however, before any of these questions may be answered satisfactorily.

This work has been supported in part by the Office of Naval Research.

References

1. P.B. Klein, J.E. Furneaux and R.L. Henry, Appl. Phys. Lett. 42, 638 (1983).

2. P.B. Klein, J.E. Furneaux and R.L. Henry, Phys. Rev. B 29, 1947 (1984).

3. J.M. Langer, in Light Emitters and Detectors, ed. M. Herman (Pergamon, New York, 1983), p. 303.

4. A. Yariv, in Quantum Electronics (John Wiley and Sons, New York, 1975).

5. S.G. Bishop and S. Sundaram, unpublished.

6. H. Ennen, J. Schneider, G. Pomrenke and A. Axmann, Appl. Phys. Lett. 43, 943 (1983).

7. H. Ennen, U. Kaufmann, G. Pomrenke, J. Schneider, J. Windscheif and A. Axmann, J. Cryst. Growth 64, 165 (1983).

8. R. Magno and P.B. Klein, unpublished.