Nano Quantum Dot Lasers: Advantages and Problems

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Abstract-Semiconductor quantum dots of different materials are novel materials developed in recent years and are attracting the researchers. Regarding semiconductor lasers, quantum dots are the next step in the development towards lower thresholds, better temperature stability and narrower emission line widths owing to their carrier confinement in all three dimensions. The emission wavelength of QDs depends on the dot size, and in the case of semiconductor nanocrystals, colour can be controlled precisely through simple chemistry. These current-generation lasers have high power output and low lasing thresholds, are stable over a wide range of temperatures, and are cheap and easy to produce. Still, there is scope for improvement as far as the performance of these lasers is concerned. This paper discusses the advantages and problems scientists are facing in the development of new type of Nano Quantum Dot lasers.

Keywords-Quantum dot laser, Surface defects, electron phonon interaction, Multiparticle Auger recombination

I. INTRODUCTION

Lasers made from bulk semiconductor materials have been used for last many decades. Semiconductor lasers play an important role in technologies from CD players to optical communications and from medical field to astrology. Laser performance has been improved with the introduction of quantum well lasers, in which charge carriers- ‘electrons and holes’ are confined to move in a plane or in a two dimensional quantum well. As compared with bulk semiconductors, the quantum well has a higher density of electronic states near the edges of the conduction and valence bands, and therefore a higher concentration of carriers can contribute to the band-edge emission. The higher density of electronic states is advantageous as it requires less intense “pumping” of energy into a quantum well laser to start lasing action. Additionally, quantum-well lasers show improved temperature stability and a narrower emission line. These current-generation lasers have high power output and low lasing thresholds, are stable over a wide range of temperatures, and are cheap and easy to produce. Still, there is scope for improvement as far as the performance of these lasers is concerned. This paper discusses the advantages and problems scientists are facing in the development of new type of Nano Quantum Dot lasers.

II.QUANTUM DOT LASERS

In recent years semiconductor quantum dots are developing as novel materials for many applications. At nano-scale cluster sizes containing only □ 100 to 10,000 atoms, semiconductors maintain their bulk crystal identity while exhibit strong size dependent variations in their electrical [1], optical [2], mechanical [3] and thermo-dynamical [4] properties. This is the consequence of increasing carrier (electron and hole) confinement with shrinking cluster sizes which causes an increase of the band gap and quantization of electronic levels.

In QDs, the charge carriers are confined in all three dimensions which results in discrete atomic-like energy spectrum of the electrons . In very small QDs, the spacing between these atomic-like states is greater than the available thermal energy, so thermal depopulation of the lowest electronic states is inhibited. It was therefore anticipated that a QD laser would have a temperature insensitive lasing threshold at an excitation level of only one electron-hole (e-h) pair per dot.

Lasing in QDs was first reported in 1991 by Vandysheve et al [5] and was practically achieved in an optically pumped device with CdSe nano particles of approximate size of 10 nm. The CdSe QDs were fabricated by high-temperature precipitation in molten glass. Lasing was also observed for QDs grown by epitaxial techniques in 1994 by Ledentsov et al [6]. The QD lasers showed an improved performance and exhibit a lower lasing threshold and enhanced temperature stability as compare with quantum-well lasers. These successes motivated the development of lasers based on Nano QDs of sizes less than 10 nanometers in diameter. For this size range, spacing between electronic levels can exceed hundreds of milli-electron-volts (meV), a much larger value than the room temperature energy scale of about 24 meV. Size-controlled spectral tunability over an energy range of 1 electron volt was expected. However, after a decade of research that provided some tantalizing hints of optical gain, NQDs failed to demonstrate lasing action. The main reasons for the failure of QD lasers were as follows-

1. *Surface defects*:- Material defects on the surface of the NQDs, which are a natural consequence of the large surface-to-volume ratio of the sub-10-nanometer particles. The defects lead to electronic states that lie within the material's energy gap. Electrons can relax into those states, whereupon they typically undergo either non radiative or radiative decay to the ground state. Thus the surface defects introduce carrier losses that inhibit the optical gain.
2. *Reduced electron-phonon interaction:-* Reduced efficiency of electron-phonon interactions that results from the discrete atomic-like energy structures, an effect that reduces the ability of carriers to enter into the band-edge states and hence reduces luminescence efficiencies.
3. *Multiparticle Auger recombination [7].*

III. MULTIPARTICLE AUGER RECOMBINATION AND OPTICAL GAIN

In order to produce optical gain, as in the case of other lasing media, QDs require a population inversion. The population inversion is the situation in which the number of electrons in a high-energy excited state is greater than that in the low-energy ground state. In small dots, the lowest "emitting" transition can be treated as a two-level system that contains two electrons in its ground state. To invert such a system, one has to transfer both electrons from the ground to the excited state. Thus the optical gain in QDs originate from nano particles that contain two e-h pairs (doubly excited nano particles).

Contradictory to the intrinsic decay of singly excited QDs which is due to the e-h recombination and the emission of a photon, the deactivation of two e-h pair states is dominated by non radiative Auger recombination as studied by Klimov et al. [8]. The group of Klimov came to the conclusion that Auger loss is intrinsically connected with the biexcitonic population inversion and can only be circumvented by using high NQD densities thus making the stimulated emission build up faster than the Auger decay, which is typically around 100 ps [8]. They were able to suggest a lower limit for the NQD volume fraction of about 0.2 %, which focused the research attention on solid lasing cavities incorporating close packed NQD films of high densities [9,10,11].

Other important conclusion of the study by Klimoc et al [8] is that in the e-h recombination energy is not released as a photon but is transferred to a third particle (an electron or a hole) that is re-excited to a higher energy state. Auger recombination has a relatively low efficiency in bulk semiconductors because of restrictions imposed by energy and momentum conservation. But linear or translational momentum conservation is a consequence of the translation symmetry of bulk crystals and this symmetry is broken in QDs .Therefore, translational momentum conservation does not apply to QDs, so the probability of Auger effects is greatly enhanced. Since Auger recombination and optical gain develop from the same initial state (that is two e-h pairs in a dot), the Auger decay is unavoidable in the regime of optical amplification and will always impose an intrinsic limit on optical gain lifetimes. For example, in CdSe NQDs, Auger recombination leads to the deactivation of doubly excited nanoparticles on time scales from approximately 400 picoseconds to approximately 10 picoseconds, depending on the dot size (the smaller the dot, the faster the recombination). These time scales are significantly shorter than the time of the radiative decay (approximately 20 ns to 30 ns) which obviously should hinder the development of lasing.

IV. NQD SOLIDS: A NEW TYPE OF LASING MEDIUM

Role of Auger recombination was realized at the end of 1999, after that detailed studies of multiparticle dynamics in CdSe NQDs by Klimov et al [8] has been started. Soon after it was found that how to overcome this problem. Studies showed that optical gain depends on the effect of stimulated emission, the rate of which can be enhanced by simply increasing the concentration of NQDs in the sample. It was estimated that the stimulated emission rate would exceed the Auger decay rate in a medium with NQD filling factors of 0.2 to 1 percent [12] . In close-packed NQD films (also known as NQD solids) such densities are readily achieved. For example NQDs capped with trioctylphosphine oxide (TOPO) will self-assemble into a thin film that can have filling factors as high as 20 percent, which is much above the estimated critical loading required for the development of stimulated emission.

For the first time, Klimov et al [12] in 2000 demonstrated optical gain by using close-packed, matrix-free films of CdSe NQDs. In the experiments, the NQD samples were optically excited by the output of an amplified titanium: sapphire pump laser. At pump intensities of 8 milliwatts, the development of a sharp, amplified spontaneous emission (ASE) peak situated on the low-energy side of the spontaneous emission band is observed. The value of this peak depends on the pump-laser intensity. Ii was also observed that the frequency of the ASE peak changed with the size of the dot. Due to strong quantum confinement effect , the peak in the smallest dots was blue-shifted with respect to that in bulk CdSe by more than 0.5 electron volt. These experiments are also performed at cryogenic temperatures in order to slow down the NQD degradation that results when the sample heats up. Mikhailovsky et al.[13] in 2002, demonstrated optical gain in NQDs at room temperature.

One of the most advantageous feature of the NQD lasers is that it shows enhanced temperature stability in lasing applications. The reason for this is that pump fluences (i.e. number of photons per pulse per centimeter squared) that are required to excite room temperature ASE in CdSe NQDs are not sufficient to produce light amplification in bulk CdSe samples. This is because of the fact that light amplification in bulk CdSe can be due to both low-threshold excitonic and high threshold e-h plasma mechanisms.

In order to achieve lasing action, the NQD gain medium must be combined with an optical cavity that provides efficient positive feedback [14,15] . Klimov et al. in 2001and Malko et al. in 2002 fabricated “a laser in a beaker” by incorporating NQD solids into a micro capillary tube. The cylindrical micro cavity can support two types of optical modes: planar waveguide-like modes that develop along the tube length and whispering gallery (WG) modes that develop (because of total internal reflection) around the inner circumference of the tube. The modes propagating along the tube can only achieve the ASE regime because no optical feedback is present. The WG modes can support a true lasing action (microring lasing). After several attempts , Klimov et al.(2001) and Malko et al. (2002) [14,15] were able to uniformly fill the interior of the tube with the NQDs and achieved the first occurrence of NQD lasing. Several types of cavities have since been utilized to demonstrate NQD lasing, including polystyrene microspheres (Klimov and Bawendi 2001) [14], and distributed-feedback resonators (Eisler et al. 2002) [16].

V. OUTLOOK

Scientists have only achieved lasing action by using a pump laser to create the population inversion in NQDs. An important conceptual challenge, however, is in the area of electrical injection pumping. Currently, developed lasing media consist of NQDs suspended in a non conducting matrix, and it is not possible to excite the dots electrically. One possible strategy to achieve electrical injection is by combining “soft” colloidal fabrication methods with traditional, epitaxial crystal growing techniques and incorporate dots into high-quality injection layers of wide gap semiconductors.

A possible technique which is compatible with colloidal dots is energetic neutral-atom-beam epitaxy. This method utilizes a beam of neutral atoms carrying significant kinetic energy of several electron volts. The beam energy is sufficient for the activation of non thermal surface chemical reactions, eliminating the need to heat the substrate in order to grow high quality films for NQD encapsulation. Because of Auger recombination, electrical pumping of NQD lasing devices would be significantly more difficult than pumping of simple “nonlasing” light emitters.

To overcome the problem of Auger recombination and to increase the efficiency of NQD lasers , there is a possible approach that is to completely eliminate Auger recombination from NQDs. It stems from the realization that the optical-gain requirement of two e-h pairs (the same initial state that allows Auger recombination to occur) is a consequence of the electron-spin degeneracy of the lowest emitting transition. Two electrons occupy the same ground state; therefore, both must be excited to achieve a population inversion. If the ground-state degeneracy could be broken (perhaps through interactions with magnetic impurities) the gain can, in principle be realized with a single e-h pair and Auger decay would no longer be a problem for either optically or electrically pumped NQDs.

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