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MASTER

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"THE LASL PROGRAM IN NUCLEAR PUMPED LIQUID LASERS"*

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INTRODUCTION

Lasers based on the lanthanide rare earths in solution show potential for production of efficient, very high energy nuclear pumped laser output. Four of the rare earth ions which appear to be highly suited for nuclear pumped lasers are Eu^{+++}, Tb^{+++}, Nd^{+++}, and Pm^{+++}. In general, these systems must be excited by external flashlamps, however, one of the results of research on optical pumping of liquid lanthanide lasers is that in some cases the addition of uranyl ions (UO_2^{++}) can increase the pumping efficiency. Sixty fold enhancement of the output has been observed with a mixture of UO_2^{+++} and Nd^{+++} at 1.06 microns.[1] Direct nuclear excitation by fission fragments in large volumes of liquids, potentially self-critical reactors, appears to be quite feasible.[2]

HISTORY OF LIQUID BASED LANTHANIDE ION LASERS

Ions of the lanthanide rare earth sequence attracted interest as lasing species early in the development of lasers. These ions exhibit several features which lend them to use as laser materials. Many rare earth compounds have narrow line emissions at room temperatures and the fluorescent lifetime of the excited states can be quite long. The atomic structure of the rare earth ions, discussed in detail by Reisfeld and Jorgensen,[3] in unique in that partially filled 4f electron

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shells are optically active. These shells are shielded by outer electrons which help give rise to the well-defined energy levels and the resulting line emission. Transitions from most of the higher excited states to the ground state or low lying states are forbidden for electric dipole transitions which is the major radiation mechanism for the rare earth ions. The low probability of spontaneous emission results in long lifetimes for the upper states.

A long series of investigations was carried out during the 1960's in developing rare earth lasers in which liquid and solid state systems were studied. The major commercial success has been the neodymium glass and crystal lasers. Recent work has been reported on gaseous Nd^{+++} lasers.[4] The work in liquid based rare earth systems was successful in that lasing was demonstrated for many of the ions and in some cases at relatively high powers.

Deactivation of excited states by transfer of energy to OH bonds rule out the use of water based solutions for liquid lasers. The use of deuterated water increases the luminescent yields but not to the point of making practical systems. One successful approach was based on binding the lasing ion with an organic compound called a chelating agent.[5] The effect of the chelating agent is to produce an ion surrounded by a cage of organic molecules. The organic structure further isolates the ion from its environment resulting in extended fluorescent lifetimes of the upper excited states and enhanced definition of the energy levels.

The proper choice of chelating agent can result in enhanced optical pumping efficiency. The chelate molecule will, in general, be in a highly excited singlet state after absorbing a photon as shown in Figure 1. A radiationless transition then occurs to the triplet state of the molecule. By careful choice of chelating agent the molecular level will lie above the upper lasing level of the ion so that a second radiationless transfer will occur to the upper ion level.

This scheme has been employed with Eu^{+++}, and Tb^{+++} to produce a lasing at room temperatures.[6,7] The major problem found with these lasers was that the absorption bands, due to the chelates, were so intense that the thickness of material was limited to a fraction of a millimeter. The net result was that organic solution, optically pumped lasers could not be scaled to large sizes.

A second approach which proved to be more successful was the development of liquid lasers based on aprotic or hydrogen-free acids. Two aprotic solvents which were successfully used were $POCl_3$ and $SeOCl_2$. When these solvents are acidified with $SnCl_4$ it is possible to achieve ionic concentrations approaching $10^{21}/cm^3$. The effect of these acids is to produce a solvation shell around the ion which isolates the ion from interactions. The aprotic acids have no absorption bands in the visible spectrum. Hence, much larger volumes can be optically pumped than in the use of the organic solution lasers. The optical pumping in this case is mainly via the ionic absorption bands.

Laser and laser amplifiers have been built using aprotic acids demonstrated storage capabilities of 300J/ℓ and output powers on the order of 3Gw.[8] Buzhinski et al. found that by adding uranyl ion (UO_2^{+++}) to the laser solutions, gains of output could be obtained for optical pumping of Eu^{+++}, Sm^{+++} and Nd^{+++}.

RESEARCH AREAS

High power nuclear pumped lasers based on the lanthanide rare earths in solution have many excellent qualities:

a) Long lifetimes of the upper laser level (250-500 μsec) which implies low threshold and high energy storage capability.

b) High heat capacity relative to gases.

c) Since uranyl ions can be mixed with the lasing medium, uniform volume excitation is possible in large volumes.

d) In some cases, the uranyl ions provide increased laser output due to radiationless transfer from excited UO_2^{+++} to the lasing species.

e) High density of fissile material possible. The aprotic acid and organic solutions have a solubility limit of around 5×10^{20} ion/cm^3 if the liquid form is maintained. Above these concentrations increased polymerization leads to solid forms at room temperature.

Expounding on point (e), one notes that a critical assembly can be built with a fissile density of 1×10^{19} atoms/cm^3 in 1×10^6 cm^3. For a flux of 1×10^{17} n/cm^2-sec, this implies an energy deposition into the liquid medium of 17kw/cm^3. A laser efficiency of 6% would imply a laser output power of 1kw/cm^3 of 1000 MW from the reactor. However, output energy would be limited to approximately 6-10 MJ in a 6-10 msec before thermal upset of the liquid media.

A program of experimental and theoretical investigating is planned at LASL to investigate nuclear pumping of liquid lanthanide ion lasers. A nuclear pumping cell, Figure 2, is currently being fabricated at LASL. This cell is specifically designed to be used in either an optical or nuclear pumping mode. The flashlamps will allow direct comparison to published results on both oscillators and amplifiers. In the design shown in Figure 2, the flashlamps pierce a neutron moderator made of transparent acrylic plastic. The moderator is covered with a layer of cadmium metal to prevent thermal neutrons from returning to the fast-pulsed bare critical reactor, Godiva IV. The Godiva IV reactor can typically produce a thermal neutron fluence of 3×10^{12} neutrons/cm^2 in a neutron moderated laser volume located 35 cm from the reactor in a pulse of 150 μsec half-width. If a uranyl ion density of 1×10^{19} atoms/cm^3 is maintained in the lasing volume, then under typically conditions, approximately 0.5 J/cm^3 can be deposited in the lasing volume with an average power density of 3.3 kw/cm^3. A 1% laser efficiency would have output powers of 50 kw/liter.

The liquid lasers represent a class of laser in which much additional research needs to be done. In addition, the details of nuclear energy deposition in liquid lasers are not fully understood. Several areas need to be explored before specific design questions can be approached. These areas include:

a) Energy channeling within liquid upon excitation by charged particles,

b) Overall nuclear pumping efficiency,

c) Radiation damage due to dissociation of solvent,

d) Optical properties of medium with nuclear excitation,

e) New reactor technology towards development of self-critical liquid reactor.

These research areas should receive immediate attention due to the high potential payoff that the liquid lanthanide ion lasers can provide.

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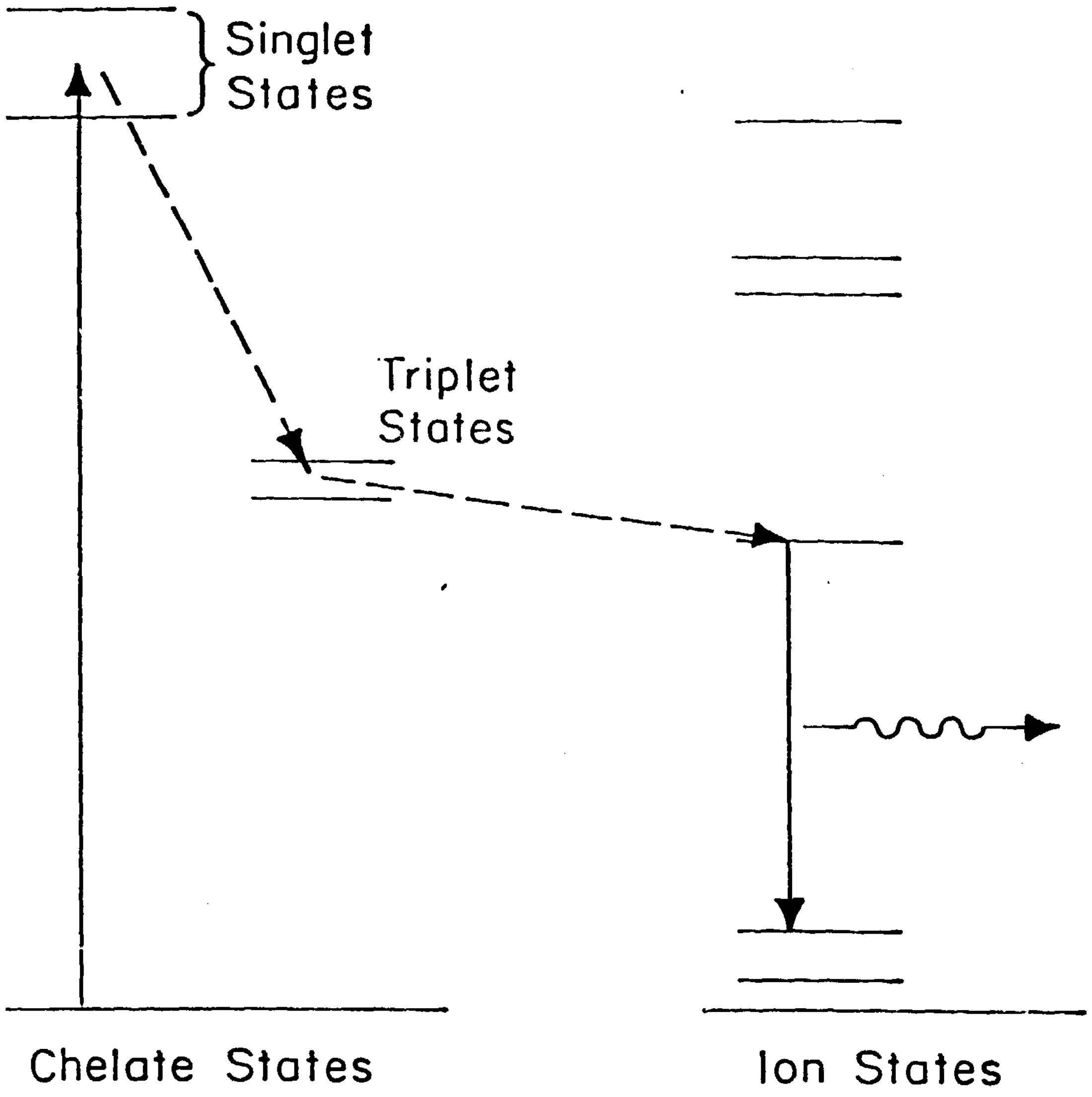

Figure 1 - Generalized Energy Flow in Liquid Chelate Lasers

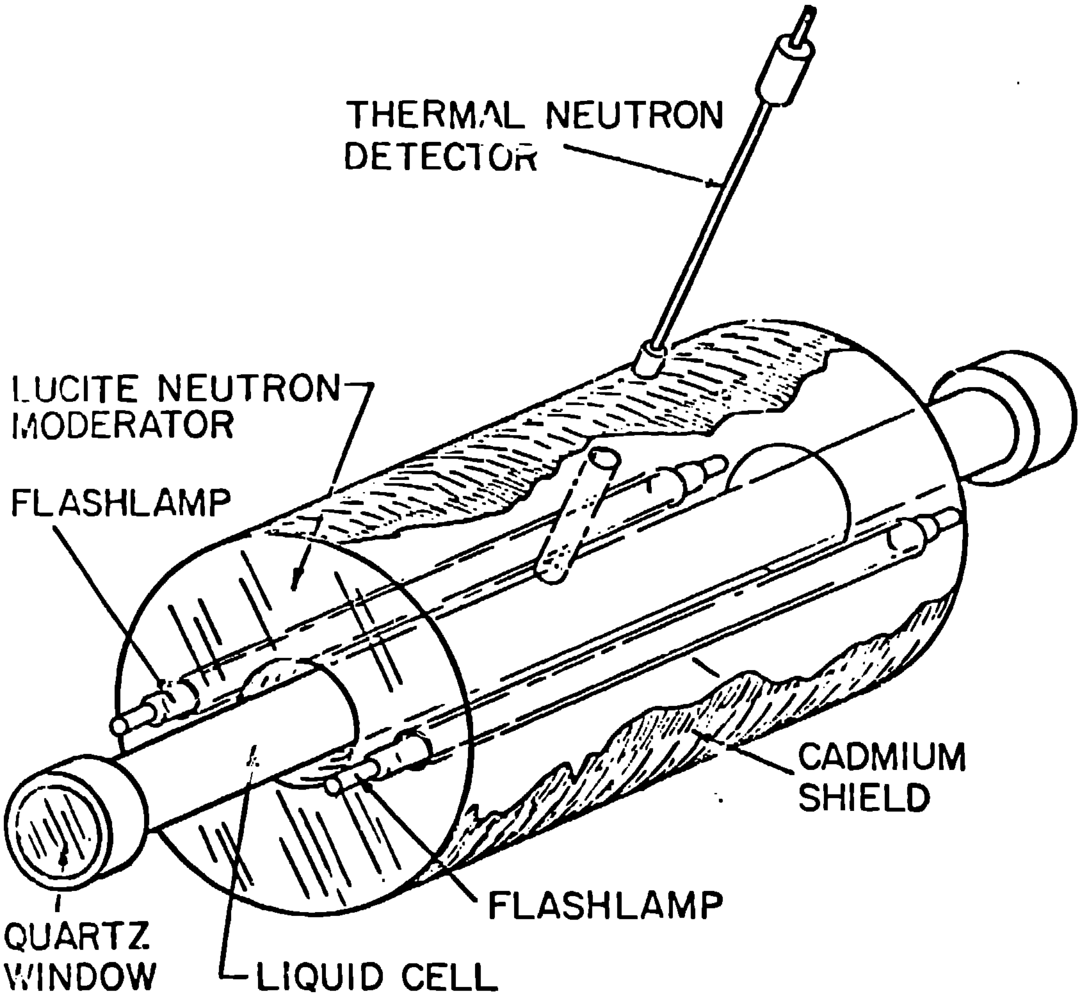

Figure 2 - Nuclear Pumped Liquid Cell