PROPOSALS FOR PUMPING RADIATIONS

The use of nuclear transitions for generating stimulated emission in ultrashort wavelength region has been proposed nearly 28 years ago (Baldwin, 1962 and Vali and Vali, 1962). Here we will briefly discuss the proposals given for pumping the population to a suitable Mossbauer energy level. In practice, the pumping radiation must be intense (to provide an adequate concentration of active isomers), efficient (so that a negligible part of pumping radiations be expended in heating), and specific (so that the host may not be polluted).

Figs. 1, 2, and 3 show the possible approaches to the development of gamma-ray lasers based on nuclear transitions. In the first approach, shown in Fig. 1, the isomeric level itself is explored to be an upper laser level. The activation, separation, concentration and crystallisation of isomeric nuclei needs a time longer than $10^{-5}$ sec. But the Mossbauer effect is generally
Gamma-ray laser approach in a nuclide $^{Z}_{A}\text{Gr}$ taking isomer level as upper laser level. Wavy arrows show the gamma-ray lasing transition from isomeric level (I) either to lower laser level (L) or to ground state (G).
observed in the nuclear levels with lifetimes shorter than $10^{-5}$ sec. Moreover, for short-lived isomeric states, the high pumping density of hard x-radiations may overheat (or destroy) the Mossbauer and Borrmann effects of the solid host crystal. On the other hand, in case of long-lived isomeric states, the inhomogenous broadening becomes so prominent that it is difficult to achieve a gain. However, scientists (Balko, 1988 and Balko and Nicoll, 1988) have shown the possibility of reducing these inhomogeneous broadenings through either nuclear magnetic resonance or level mixing spectroscopy techniques (Coussement et al., 1988). Considerable efforts to observe some success in a few cases, viz. Ge, Zn, Ta, etc., are also going on.

Fig. 2 exhibits another approach for transferring the population to a nearby level that can lase either to ground level or to some other lower laser level. In this case a low pumping power is required. Such pumping may either be through thermal neutrons or through the x-ray photons. But the production of intense thermal neutrons causes a delay and other geometrical losses. On the other hand, the x-radiation pumping causes the electrostatic and magnetic inhomogenous effects that inevitably broaden the
Fig. 2  Gamma-ray lasing transitions from an intermediate transfer level (T).
Fig. 3 Two-step pumping approach. Arrow (1) shows the first slow step for storing the population in isomer level (I). Arrow (2) indicates the second fast pumping step. Double arrow represents a cascade to upper laser level (U).
Mossbauer line. Moreover, the non-resonant absorption of x-radiations may overheat the crystal to destroy the Mossbauer and Borrmann effects.
4.1 Two-Step Pumping

Fig. (3) shows a two-step approach to the problem. According to this scheme, the population is first transferred to the nearby transfer level which ultimately cascades to another lower energy level which can lase either to a lower laser level or to ground level. In Fig. (4) the block diagram for a two-step pumping mechanism is illustrated.

In this scheme the population is first stored in a long-lived isomeric level of an active nucleus having another short-lived level nearer (in energy) to this isomeric level. Such a storage level is implanted in the solid crystal, whose properties support the Mossbauer and Borrmann effects; then this medium is cooled. The first step can be performed with the help of slow excitation processes. The second step, to transfer the population from storage level to the intermediate transfer level, is to be performed within the lifetime of the transfer level. This fast step may be accomplished by transfer of resonant radiation through optical excitations (Baldwin, 1988, Cohen, 1988a, 1988b, Collins, 1988, 1987, 1986, Collins et al., 1989, 1982, Ho, 1988, and Winterberg, 1986) or
Fig. 4  Block diagram of two-step pumping mechanism.
through electronic excitations (Berger et al. 1988a, 1988b, Dyer et al., 1988, Rinker et al., 1988, 1987, and Solem and Biedenharn, 1988). These transfer processes will be discussed in detail in sections 4.2 and 4.3.

In two-step pumping schemes the existence of the transfer level can not be resolved unless some special spectroscopic techniques are involved. Haight and Baldwin (1986) have suggested the possibility of exploiting broadband radiations to excite such a level and then to confirm its existence. However, this method for searching such transfer levels would be useful only under some very restrictive conditions. If such a suitable gamma-ray laser candidate exists then the production and separation of the isomer nuclei from other nuclear products have to be tailored. At present, it can be done at least in some particular cases as will be discussed in section 4.5. The feasibility of this scheme also depends upon the rapid and efficient population transfer process.
4.2 Optical Pumping

Two-step upconversion processes for optically pumped nuclear reactions can be divided into coherent and incoherent categories.

The adjacent sketch (see Fig. 5) shows the schematic diagram for the energetically excited levels of a typical nucleus of interest to the development of a gamma-ray laser. The arrow (1) illustrates the incoherent pumping of the storage level through the absorption of x-rays (from laser plasma or an exploding wire) that are resonant with the energy separation between the storage level and the next higher level of proper symmetry. The arrow (2) represents coherent pumping through the non-resonant absorption of a photon from the radiation field in order to create a virtual or dressed state of excitation. Ultimately the gamma-ray laser output results from the upper laser level populated by a cascade.
Fig. 5 Coherent (arrow 1) and incoherent (arrow 1') optical pumping schemes.
4.2.1 Coherent Pumping

In this technique the nuclear isomeric state is excited to a virtual (or dressed) state with high intensity optical pumping (the anti-Stokes upconversion of conventional laser radiation, see Fig. 5) (Collins et al., 1982). This is analogous to the dressing of atomic energy levels by a strong magnetic field (Collins and DePaola, 1985). The populated virtual level decays to the laser level leading to the stimulated emission. The stimulated emission cross-section for the composite process, i.e., transferring the population from storage level to virtual level and then to decay of the virtual level to the upper laser level, can be given by the Breit-Wigner value as:

$$\sigma_x = \frac{\lambda^4}{2\pi}$$

where \( \lambda \) is the wavelength of the stimulated emission. In order to diminish the direct transitions between the isomeric state to upper laser level, the natural width has to be preserved.

The possibility of the temperature rising in the pumped dressed layer can be avoided by diluting the active nuclei in a low Z host, e.g., \(^3\text{Be}^9\) (Collins, 1986). It has
also been computed (Collins et al., 1982 and Collins 1986) that for a critical intensity of $5.3 \times 10^5$ Watts per cm$^2$ and a typical $10^6$ Hertz spurious broadening the threshold fluence should be 167 milli-Joules per cm$^2$. This proposal describes a model with a laser medium of length 1.0 cm and a filament of diameter 0.04 cm and the requirement that each pump photon makes ten trips of the laser medium. For this, pumping threshold would be about 26 Joules in a pulse of duration 300 nano-sec and the output obtained will be 66 Joules at 10 KeV. Moreover, the laser threshold might be further reduced by manipulating the bulk ferromagnetic or ferroelectric properties of the material in which the nuclei are diluted. Presently, experiments are being performed for dressing a nuclear level in $^{57}$Fe (Sinor et al., 1989, Wagal et al., 1987, and Wagal and Collins, 1986) as a test case.

4.2.2 Incoherent Pumping

In incoherent pumping of nuclear material, the resonant excitation of energetic states of nuclei is done by the x-radiation from laser plasmas or exploding wires. Theoretical estimates show that a transfer level
decaying after 10 pico-sec through an El transition to the upper laser level with a 10 KeV photon will have $6.6 \times 10^{-5}$ eV bandwidth, assuming the complete resonance between the pump and transfer level (Collins et al., 1982). So the complete fluence from the optical to the upper laser level can be built up for a time equal to its lifetime (1 - 10 nano-sec).

For an x-ray laser plasma source (radius, R, to be about 30 micro-m and length, L, of about 0.1 cm), the total population of upper laser level can be given as

$$N_u = \frac{n \sigma_x N_o}{(2\pi R L)}$$

where $n$ is the number of x-ray photons produced in the source during the lifetime of upper laser level, $N_o$ is the concentration of transfer level and $\sigma_x$ is the Breit-Wigner cross-section for the x-ray wavelength. This model further suggests that a relative fraction of about $10^{-5}$ eV of the line energy would be used in El transitions. For an x-ray energy around 10 KeV, the threshold value is 10 Joules in the x-ray line and results in a threshold fluence of 131 Joules per cm$^2$ (Collins, 1986) for a 10 KeV output transition. However, this threshold can further be reduced by manipulating the properties of the material embedding the nuclei.
Ho and Pan (1988) have studied the optical excitation of a long-lived isomeric level to a short-lived Mossbauer isomeric level by means of a parametrisation approach. They conclude that the threshold laser intensity of about $2.0 \times 10^{17}$ watts/cm$^2$ may invert sufficient population leading to the coherent generation of gamma-rays.

The incoherent optical pumping of isomeric nuclei requires a threshold energy fluence of about 100 - 300 Joules/cm$^2$ to be deposited in the gamma-ray laser material (Collins et al., 1982 and Collins, 1987). This fluence might produce a pressure of about $10^5$ Newton/cm$^2$ and temperature of about $10^3$ K. The difficulty may be resolved by using the electronic heat conduction under high pressure as proposed by Winterberg (1986). He has suggested that the gamma-ray laser active material be kept either in a static high press or in a high explosive pressurizing geometry (see Fig. 6). Provided the outer medium is of good heat conductivity and at low temperature, the gamma-ray laser material can be made to lose sufficient heat within the pumping time.
Fig. 6    Rapid heat removal under high pressure. Symbols
M represents the metallic tamp under high
pressure and at low temperature, P represents
externally applied pressure, and C represents
the conical optical laser beam.
Recently, Baldwin (1988) has suggested that a superlattice geometry in which the active material is doped in a single crystal host in a regular array (to support the Borrmann effect) may lead to recover the Mossbauer effect before the destruction of the crystal.

We have proposed an alternative technique to preserve the Mossbauer and Borrmann effects within a single crystal during the second step of fast pumping (Gupta and Husain, unpublished). We have explored the transfer of population from isomeric level to a nearby transfer level through successive excitation of atoms by some high intensity x-radiation pulses of much shorter widths (of few pico-sec). The defects in the crystal structure may be removed within a fraction of a nano-sec by non-thermal laser annealing techniques (Cullis, 1985 and van Vechtan et al., 1979).

Cohen (1988a and 1988b) has shown the possibility of generating coherent gamma-rays through nuclear recoil transitions embedded in a gas. The Doppler-recoil breadth of nuclear levels can be suppressed by obtaining a macro-sample of nuclei at rest with respect to each other. Such samples may be realized either by the atomic cooling through lasers (to velocity zero of all atoms) or through
accelerating the sample to a good fraction of the velocity of light. But this idea requires major technological developments in atomic cooling mechanisms or in particle accelerators.
4.3 Nuclear Excitation by Electron Transitions

An alternate to direct photon absorption for exciting the nuclear isomeric storage level to the short-lived intermediate level is the possibility to induce the transfer by first exciting the atomic electrons with high brightness ultraviolet lasers and then by exciting nuclei by exchange of virtual photons in the near field of electrons (Biedenharn et al., 1986a, 1986b, Berger et al., 1988a, 1988b, Noid et al., 1987, Reiss, 1986, Rinker et al., 1988, 1987, Rinker, 1986, Solem, 1986, and Solem and Biedenharn, 1988). The free electron system, in the vicinity of a strong laser field, possessing large multipole moments and broad resonances, forms a mixed state with the isomeric nuclear system. The non-linear response of the electron cloud can supply energy, angular momentum, and parity to the isomeric level. This can excite it to a short-lived intermediate level which ultimately cascades to the upper laser level. Goldanskii and Namiot (1981) observed the excitation of ground state uranium nuclei to 73 eV $^{235}$U in hot plasmas as a result of capture of continuum electrons to outer shell in ionized atoms. This has also been evidenced by the
experiments of Saito et al. (1980) on $^{237}$Np and Otozai et al. (1973) on $^{189}$Os. Before extending this idea to a real gamma-ray laser candidate, scientists at Los Alamos National Laboratory, U.S.A., (Bounds et al., 1987 and Dyer et al., 1988) have been investigating this mechanism for excitation of the $^{235}$U nucleus via an ultraviolet laser. Their preliminary experiments (Dyer et al., 1988) with laser intensities of about $10^{15}$ Watts/cm$^2$ have demonstrated an encouraging probability of about $10^{-4}$ of exciting a $^{235}$U atom. It is expected that intensities of about $10^{17}$ Watts/cm$^2$ will be required to excite a nucleus with significant probability.
4.4 Steps for the Search of the Appropriate Gamma-Ray Laser Medium

The essential feature of two-step pumping mechanism is to transfer the population from a storage isomeric nuclear level to a nearby short-lived level. This population can then enrich an upper laser level. At present a major problem is to identify the proper gamma-ray laser candidates from the different parts of the periodic table. Despite of some low-energy nuclear gamma-ray source compilations (Artna-Cohen, 1988, Arthur and George, 1988, Arthur et al., 1988, and Martin, 1986), no laser grade data-base exists yet and highly precise experiments need to be performed for the examination of nuclear properties. Scientists at Los Alamos National Laboratory (Arthur and George, 1988 and Strottman et al., 1986) have examined the appropriate experimental and theoretical data and have indicated the most fruitful mass regions. They have identified some pairs of the isomeric states (lifetimes > 5 sec) with short-lived levels within a specified excitation energy window of width 1 - 10 KeV, viz., Os, Te, La, Lu, Hf, etc. They have found that the odd-odd nuclei in rare-earth region would be a likely place to begin a search for appropriate gamma-ray laser
candidates and on the basis of their nuclear model they find $^{186}$Re as a viable candidate (Madland and Strottman, 1988, Strottman, 1988, and Strottman et al., 1986).

The Rochester-Stanford joint research programme (Gove et al., 1986) for the evaluation of gamma-ray laser candidates has developed a tandem Van de Graff accelerator for the study of high spin states in deformed nuclei on a recoil mass spectrometer. It is hoped that this joint programme will take the results from Los Alamos National Laboratory and evaluate the proper potential candidates.

At Lawrence Livermore National Laboratory scientists are setting an experiment to detect suitable shape isomers whose deformed nuclear shape, different from the shape of the ground state, exhibits an unusual stability in light actinide region (Bauer et al., 1988, Henry et al., 1988, NTIS Report No. DE86008153, 1986, and Weiss, 1988,). In such isomers the dominant transition mechanism between the two laser levels is gamma-ray decay.

At University of Texas at Dallas, a group working under the Strategic Defense Initiative Project (Collins, 1988, 1987, Collins et al., 1989, Davanloo et al. 1987,
and Wagal et al., 1987) has identified a slate of twenty-nine candidates being examined on an x-ray flash lamp delivering high peak powers of hard x-rays at high repetition rate of about 100 Hertz (Bowen et al., 1987, Collins et al., 1987, 1988a, and Davanloo et al., 1987, 1986). The ultimate success of these pump schemes involving flash x-rays will require investigation of the nuclear properties of those materials that are analogous to the kinetics of a conventional laser medium. In some selected cases, this examination can also be performed in a little time by means of using either the laser plasmas or large electron beam machines producing the synchrotron radiations, but the economics involved do not permit for a large number of examinations (Collins, 1986). In 1988 and 1989 the pumping of $^{180}$Ta isomer, which is not necessarily a gamma-ray laser candidate, has been successfully demonstrated (Collins et al., 1990, 1989, and 1988b and Collins, 1988). The results show a large increase (by two orders of magnitude) in cross-section for yielding the upper laser level of $^{180}$Ta over similar levels of other isomers, viz., $^{119}$Cd (Anderson et al., 1988), $^{115}$In (Collins et al., 1988a), and $^{77}$Se and $^{79}$Br (Anderson and Collins, 1987a, 1987b). Such a success with $^{180}$Ta isomer may lead to significant implications for future
experiments for searching a real material. Very recently, an experiment for the excitation of $^{123}$Te and $^{125}$Te isomers (Richmond et al., 1991 and Carroll et al., 1991) has also been reported.

If storage and transfer level possibilities (as discussed in sections 4.1 to 4.3) are found to be unfeasible then practical considerations at the current state-of-the-art require the nuclear lifetimes, offering the Mossbauer effect, to be about few seconds. Recent experiments (Hoy and Rezaie-Serej, 1990, Hoy et al., 1990, and 1989, Hoy and Taylor, 1988, Hoy, 1986, Rezaie-Serej and Hoy, 1989, and Taylor and Hoy, 1988) exhibit a small probability of finding the Mossbauer effect in 40 sec and 88 KeV first excited state of $^{109}$Ag. These experiments are based on gamma-ray self-absorption technique.
4.5 Isomer Separation

From the discussion given in the previous sections, it is clear that for developing a nuclear gamma-ray laser based upon nuclear transitions one has to invert the ground state population to the isomeric level and to separate such levels from the rest of the other nuclear reaction products -- to get the isomeric enrichment. In literature, there are some laser techniques for isomer separation, analogous to laser isotope separation. These are:

(1) Resonance ionization -- in which the ionization step is performed by laser beams. Dyer et al. (1985) have demonstrated the isomerically selective photo-ionization of $^{197}\text{Hg}$ nuclei.

(2) Optical piston -- in which the separation is done by exciting the isomeric atoms from the rest of the mixture kept in a capillary cell (Werij et al., 1984). Dyer has suggested this technique as a model for atoms like Sodium (Dyer et al., 1985).

(3) Atomic beam methods -- in which the isomeric atoms in a beam are first optically pumped into particular magnetic substates with a circularly polarized laser. They can then be filtered by means of a magnetic-
moment analyzer (Baldwin et al., 1981). Alkhazov et al. (1984) have shown the resonance ionization of $^{142m}_{\text{Eu}}$ by three laser beams.

(4) Photochemistry -- in which the excited atoms possessing the isomeric nuclei are chemically separated.

In radiochemical methods (Baldwin, 1982) the enrichment of isomeric nuclei, being produced in nuclear reactions, is achieved through Szilard-Chalmer’s process, in which recoil energy of the nucleus is greater than the chemical binding of the nucleus in a compound.

Despite of all these techniques, the optimum choice of any isomer enrichment process will depend on the properties of the actual graser material.
4.6 Nuclear Superradiance

Traditionally, a laser consists of an active medium pumped in some manner to produce an inverted population between two levels, and kept between mirrors to form a resonant cavity. But because of the non-availability of good mirrors for gamma-ray region, a gamma-ray laser based on nuclear transitions would be a single shot, high gain system comprising a long crystal without mirrors (or without a resonant cavity). Furthermore, the large internal conversion coefficient in Mossbauer transitions rapidly depopulates the upper laser level, hence build-up time for the inversion density is relatively short for ordinary stimulated emission. Therefore, the preferred process is superradiance (Baldwin, 1986a, 1986b, Baldwin and Feld, 1986a, 1986b, Hartmann et al., 1988, and Kudenko and Kuzmin, 1985). It requires a short build-up time for inversion density and leads to the coherent deexcitation of upper laser level. In this process the nuclei are coupled together by their common radiation field. This collection of indistinguishable nuclei is treated as a collective wave function. For a two-atom system the superradiant state is:

\[ \frac{1}{2}(|a+\rangle + |a-\rangle) \otimes (|b+\rangle + |b-\rangle) \]
where $|a+\rangle$, $|a-\rangle$ and $|b+\rangle$, $|b-\rangle$ are the upper and lower laser levels of the two atoms labeled a and b respectively (Eberly and Milonni, 1990). Such a superradiant state decays cooperatively leading to the intensity emitted by N nuclei being proportional to $N^2$ and pulse width to $N^{-1}$. Such a state can be formed in a single crystal of low absorption coefficient for the outgoing radiations. Baldwin and Feld (1986a) have given model superradiance calculations for $^{119}$Sn (diluted in diamond) and for $^{133}$Ba (diluted in borazon). The output performances of their results are given in table 1.
<table>
<thead>
<tr>
<th>Assumed and calculated parameters</th>
<th>Nuclide</th>
<th>119\text{Sn}</th>
<th>133\text{Ba}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Length of SR sample* (cm)</td>
<td></td>
<td>1.0</td>
<td>1.0</td>
</tr>
<tr>
<td>Diameter of SR sample (cm)</td>
<td></td>
<td>8.1 \times 10^{-5}</td>
<td>1.13 \times 10^{-4}</td>
</tr>
<tr>
<td>Active nuclei in upper laser level</td>
<td></td>
<td>7.8 \times 10^{12}</td>
<td>5.0 \times 10^{13}</td>
</tr>
<tr>
<td>Peak Power (Mega-Watts)</td>
<td></td>
<td>4.90</td>
<td>35.9</td>
</tr>
<tr>
<td>Pulse width (nano-sec)</td>
<td></td>
<td>1.64</td>
<td>0.67</td>
</tr>
<tr>
<td>Power enhancement</td>
<td></td>
<td>5.0 \times 10^9</td>
<td>6.74 \times 10^9</td>
</tr>
</tbody>
</table>

*SR sample: material sample containing superradiant nuclei*
4.7 Miscellaneous Ideas

In addition to the proposals discussed above, other ideas based upon giant resonances and nuclear isomers (Collins, 1986), nuclear single exciton states (Hannon and Trammell, 1990), an anomalous Mossbauer spectrum (Yukalov, 1989), and novel nuclear levels (Photonics Spectra, 1990) have also been mentioned in the literature but no detailed work has been reported yet.
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