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GENERATION OF cw FIR RADIATION USING 2- AND 3-PHOTON INTERACTIONS

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Abstract.— Recent experiments aimed at generation of FIR continuous radiation with limited tunability utilizing 2-photon and 3-photon transitions are described.

Coherent cw radiation sources in the far infrared, the wavelength region between 20 and 1000 μm took a step forward in the early 70 s. After being limited to a few laser transitions in HCN and H_2O for a number of years, the discovery of the technique of optical pumping of molecules /1/ in 1970 gave rise to an activity, which up to now has provided some 2000 laser wavelength in the FIR /2/. (Fig. 1) While the efforts were first directed to the generation of new wavelengths and appropriate resonator design /21/, interest arose subsequently in the physics of these systems. Early work attempted to describe these lasers on a basis of rate equation models /3/ motivated to understand and possibly remedy the low efficiency of these systems. This work recognized the role of collision processes /4/. However in 1975 it was suggested /5/ that since one is dealing with coherent excitation, a rate equation treatment is inadequate. Work began then to treat the lasers semiclassically i.e. treating the molecules as quantum systems and the radiations as classical fields. (Fig. 1) The theoretical treatment was based on earlier spectroscopic work on coherently driven 3-level systems /6/ using the density matrix formalism and generally applying a generalized rotating wave approximation. These calculations predicted coherence effects like anisotropic gain and line splitting due to AC-Stark effect. These two effects could be experimentally observed in /7/ /8/.

A quantitative comparison between calculated and measured laser properties such as gain saturation as functions of the relevant parameters gas pressure, field strengths, and field frequencies yielded satisfactory agreement for CH_3OH lasers /9/ /10/ and in particular of

NH_3 lasers /11/ for which the molecular parameters such as transition dipole moments, level energies, partition function, line broadening parameters which enter the calculations are known best.

It appeared therefore that the theoretical treatment could be trusted to properly describe 3 level coherently pumped lasers and that it could be used to predict the feasibility of new systems.

1. Since the theoretical treatment contains no limitations as to which degree the fields have to be resonant with the molecular transitions, it was possible to investigate theoretically the process of stimulated Raman scattering. Calculations showed that the two-photon process of stimulated Raman scattering (SRS) (Fig. 2) which had already been used for FIR generation using pulsed MW power pump sources /12/ could be made strong enough, even when using cw pump sources 5 orders of magnitude weaker, to provide Stimulated Raman Scattering gain adequate for the realisation of cw Raman lasers. A SRS FIR cw laser was experimentally demonstrated ^{*)} /13/ and after this the rules of the game were known so that additional systems could be made to work /14/. Gain measurements of the FIR Raman laser /11/ finally confirmed the adequacy of the 3-level theory also for Raman lasers.

These Raman systems show some interesting features compared to resonantly pumped systems

- a) Their emission line frequency can be tuned by the pump frequency and
- b) The FIR emission line width is approximately equal to the IR Doppler width and thus by an order of magnitude larger than for resonantly pumped systems. (Both as a consequence of the absorption-emission process (Fig. 2) being two-photon resonant)
- c) The SRS lasers are insensitive to molecular collisions since no population inversion in the usual sense is required.

The feature b) has been utilized technically already providing FIR laser frequencies 100 MHz apart for beat frequency modulated plasma interferometer, for Tohamak diagnostics /15/

^{*)} This constitutes the first demonstration of a cw SRS gas laser. Shortly after the publication of /13/ a SRS cw gas laser was realised by A. Brillet /20/ using optical pumping of electronic transitions in Li_2 dimers.

Feature C allows operation of these systems at higher pressure and consequently higher emitter density, limited only by the collision broadening of the nearby resonant absorbing FIR transition.

The gain saturation behaviour of SRS lasers differs markedly from resonantly pumped systems since gain saturation is only reached by two-photon saturation. Thus the gain saturation intensities are typically 1 - 2 orders of magnitude higher - possibly making SRS lasers more efficient converters from pump to FIR radiation.

2. Optical pumping of molecules by IR lasers relies on close coincidences of lines of suitable IR pump lasers with absorption lines of polar molecules. Although a large number of such coincidences was found using a variety of molecules frequency-tuneability of the pump radiation source is of course desirable. E.g. no pump coincidence exists between the FIR laser molecule with the best properties $^{14}\text{NH}_3$ and the regular CO_2 -laser. As there seems to be little prospect for widely tuneable IR cw lasers with powers of several tens of watts, another application of two-photon interactions appeared to be promising. A two-photon transition involving an IR photon (fixed frequency) and a microwave (MW) photon (tuneable frequency) can be utilized as the pump source, if it can be made strong enough (Fig. 3). Estimates showed that cw 2-photon pumping of NH_3 laser transitions should be possible using pump intensities of $10 \text{ W/cm}^2 \text{ IR}$ and $1 \text{ W/cm}^2 \text{ MW}$.

An experiment was undertaken which successfully demonstrated the first cw two-photon pumped laser /16/. The key element in this experiment is the FIR laser resonator which was chosen to be of the circular metallic waveguide type (Fig. 4). This resonator can act at the same time as an overmoded FIR waveguide resonator of low loss and as a MW resonator to resonantly enhance and confine to a small volume the MW field. Coupling of the MW to the resonator could be done without substantially increasing the FIR resonator losses using $\lambda/2$ -slit coupling through the waveguide side wall. With this system two-photon pumping in cw mode was demonstrated of a number of NH_3 FIR laser transitions.

/14/

Apart from demonstrating two-photon pumping of cw laser transitions for the first time, these experiments yield lasers which are electrically modulatable in amplitude and in frequency because the laser process involves a MW-field, which is easily electrically controlled.

3. A combination of the SRS laser technique with the two-photon pumping technique suggests itself. This would be a two-photon pumped Raman Laser (also known as Hyper-Raman Laser) operating by a three-photon process Fig. 5). A two-photon pumped laser would be converted into a Hyper-Raman Laser simply by detuning the MW-frequency from the two-photon pump resonance.

Of course the Hyper-Raman gain will be smaller compared to the gain of a 2-photon pumped laser by roughly the same factor by which SRS gain is smaller than gain of a resonantly single photon-pumped system. This gain decrease will have to be compensated for by an increase in pump field strengths.

Evidently the laser emission of a Hyper-Raman laser would be tuneable in frequency (by tuning the MW-frequency) due to the fact that the process is 3-photon resonant. Realization of tuneable FIR cw laser is of course a challenging goal. In order to know the pump power requirements for a cw Hyper-Raman laser we have extended the semi-classical treatment of 3-level systems interacting with 2 coherent fields to a 4 level system interacting with 3 coherent fields.^{*)}

The calculations show a number of interesting physical features of coherently driven 4-level systems such as AC-Stark Splitting of the Raman emission line, 2-photon-AC-Stark Splittings and -shifts. Pump fields of 100 W/cm intensity will be required for a few percent/m Hyper-Raman gain, 1 %/m being the FIR laser threshold gain of our experimental system.⁺⁺⁾

^{*)} It should be mentioned that we had to restrict the calculations to the case in which all field polarizations are parallel since for orthogonal polarizations interference can occur between the transitions between the various magnetic sublevels, giving rise to AC-Hanle effects induced by one-photon /19/ and two-photon level shifts and splittings. Thus orthogonal polarizations would require the treatment of a many-level system and this exceeds reasonable computer times.

⁺⁺⁾ Interesting experiments could be done to observe multiphoton coherent transients in 4-level systems by pulsing the MW field.

There are other schemes which could be used to generate tuneable FIR cw laser radiation in 4-level systems e.g. by resonant single-photon pumping and inducing two-photon transitions involving the FIR radiation and a microwave (Fig. 6). Tuning the MW would then tune the frequency of the FIR radiation. An experiment which we performed on a 4-level system in HCOOH /17/ yielded very limited FIR tuneability due to the low gain in HCOOH which results from the large partition function of this asymmetric molecule /18/. A proper molecule for this purpose; simple symmetry for low partition function and accessible microwave transitions in the excited vibrational state has yet to be found.

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Fig. 1 Resonantly pumped FIR lase

Fig. 3 Two-photon pumped laser

Fig. 2 Stimulated Raman scattering laser

Fig. 4 Waveguide resonator used for two-photon cw pumping of FIR laser transitions

Fig. 5 Hyper-Raman-scattering laser

Fig. 6 Energy level scheme for generation of tuneable FIR radiation using a FIR-MW two-photon transition.