

## **From Optically Pumped Molecular Lasers to Multi-Frequency Beam Splitters**

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### **Abstract**

In this paper, the two laser schemes of multi-frequency optically pumped molecular lasers are described. Molecular gas lasers based on optical pumping potentially produce separate laser beams with various different frequencies. Such lasers can serve as frequency splitter devices that will find a wide range of applications for example, in chemical sensors and communications.

**Keywords :** Lasers, Gas lasers, Optically pumped lasers, Optical frequency splitters

## 1. Introduction

Multi-frequency sources have gained increasing attention in recent years [1-5]. There are currently technologies that are capable to generate multi-frequency radiations in mid infrared (IR) region for example, optical parametric oscillators (OPOs), quantum cascade lasers (QCLs) and molecular cascade lasers (MCLs). These technologies will be potentially used as tools to serve variety of applications such as the interaction between several frequencies and various materials, chemical sensors, imaging and communications. However they have some stringent limitations. OPOs suffer for their cumbersome sizes and limited output powers. QCLs are promising sources to handle the multi-frequency criteria but they are still in their immature states. MCLs excited by the methods of gas dynamics, electronic-discharge, and chemical have been studied to pursue multi-frequency sources for high power operations [6]. However, the laser outputs obtained by such excitation techniques are inherently bulky. Furthermore, the compact versions of these types of MCLs are difficult to develop.

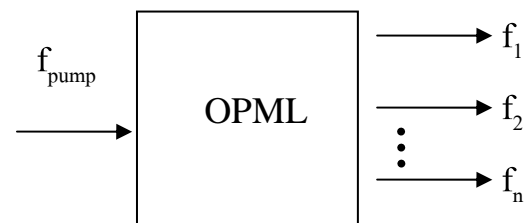
Gas lasers based on optical resonance pumping or also called optically pumped molecular gas lasers (OPMLs) have been proposed as promising candidates for MCLs [7-8]. Compared to other types of MCLs, optical pumping approach offers many advantageous properties. For example, a straightforward excitation can be employed by well developed lasers that would allow high laser efficiencies. Furthermore, it has been realized that the hollow waveguide technology can be applied to OPMLs [9-10]. Therefore, the technological advances in the hollow waveguide would bring a new class of compact multi-frequency sources.

This paper discusses the promising multi-frequency laser schemes based on OPMLs. The laser schemes presented here are restricted to mid IR transitions. The multi-frequency OPMLs will be a new class of multi-frequency sources. The

creation of multi-frequency OPMLs will impact to the advent of new gas phase media and development of pump source technologies. Furthermore, multi-frequency OPMLs will find range of applications that lead to sophisticated innovations in laser physics and laser spectroscopy.

## 2. Laser schemes

Molecular absorption and emission of OPMLs in mid IR region are generally due to the excitation of molecular vibrational motions in the electronic ground state [11]. OPMLs are considered as effective conversion systems that shift pump frequencies to desired output frequencies. The features of OPMLs can be extended to the multi-frequency laser systems. Fig. 1. shows an idea of the multi-frequency laser system that performs as a frequency splitter device. The laser system splits a pump frequency into several output frequencies by the principle of population inversion.



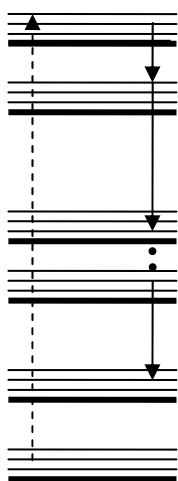
**Fig. 1.** Illustration of multi-frequency OPMLs. The pump frequency is denoted by  $f_{pump}$  and the laser frequencies are represented by  $f_1, f_2, \dots, f_n$ .

The multi-frequency OPMLs can be classified into two general groups according to the excitation processes. The direct OPMLs use a single molecule for pumping and lasing. The other type is the transfer OPMLs in which the energy transfer process involves to pump transitions. It should be mentioned that the definitions “direct” and “transfer” given by C. R. Jones [11] were adapted for multi-frequency OPMLs.

## 2.1 Direct OPMLs

The configuration of the multi-frequency direct OPMLs is shown in Fig. 2. As shown in Fig. 2., each vibrational state (thick horizontal lines) consists of rotational state levels (thin horizontal lines) and the dash arrow is the pump transition. In addition, the solid arrows denote the laser transitions.

Under the optical excitation of a vibrational-rotational state, population is transferred from the ground state to a terminal excited vibrational state. Laser transitions occur initially on the excited vibrational state and cascade to adjacent vibrational states.



**Fig. 2.** Schematic energy diagrams of multi-frequency operations of the direct OPMLs.

Laser transitions can exhibit different optical frequencies which depend on the type of laser molecules. A cascade hydrogen bromide (HBr) laser has generated laser transitions in the four micron region [12]. The same research group has extended their theoretical work to point out the conditions of the full cascade lasing of HBr lasers [13]. However, the laser frequencies obtained by diatomic molecules will be only few percent differences due to their natures of anharmonicity and favors in fundamental transition. The improvement can be achieved by using polyatomic molecules. Several normal

modes of vibration of polyatomic molecules offer multi-frequency transitions. Laser frequencies obtained by polyatomic molecules are governed by their dipole allowed transitions.

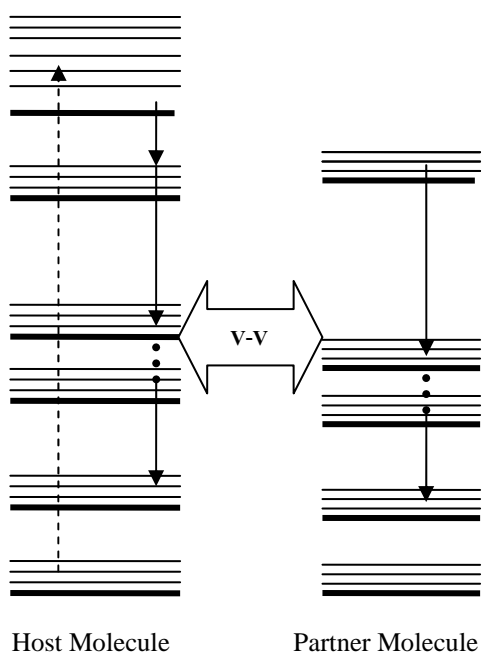
## 2.2 Transfer OPMLs

In mid IR region, the energy transfer lasers based on optical pumping offer possibilities of multi-frequency sources. In the past, there was considerable interest in the development of energy transfer lasers employed by optical pump schemes however all studies have attempted to obtain single frequency output. Continuous wave (CW) CO-CS<sub>2</sub>, CO-C<sub>2</sub>H<sub>2</sub>, CO-N<sub>2</sub>O, CO-OCS, and CO-CO<sub>2</sub> lasers based on vibrational-to-vibrational (V-V) energy transfers were reported [14]. In addition, CW optically pumped HF-HCN and HF-HCN lasers using V-V energy transfer processes were demonstrated [15]. These intensive studies ensure the capability of V-V energy transfer processes of these molecular gases to achieve population inversion. In the aspect of multi-frequency criteria, several vibrational levels associated with V-V exchange molecules can allow multi-frequency lasing.

The promising transfer OPMLs are required at least two laser species to enable V-V energy transfer process. Therefore the configuration of such the systems can be complicated. Fig. 3. shows an example of the simplified energy diagram of a multi-frequency transfer OPML. As shown in Fig. 3., the thick horizontal lines, the thin horizontal lines and the dash arrow are also represented to vibrational states, rotational states and the pump transition, respectively. In addition, the two-way arrow represents the V-V energy transfer process.

The host molecule serves as the absorbent for pump radiation and allows the rapid V-V energy transfer to the partner molecule. In this case, the pump laser excites

population from the vibrational ground state to the terminal pump state of the host molecule. Cascade lasing transitions can arise in the same behavior of the direct OPMLs. Furthermore, the V-V energy transfer between the excited population of the host molecule and population of the partner molecule in the ground state allows the excitation of the partner molecule. Thus the partner molecule is then capable to generate laser transitions which could also be the same manner of cascade lasers.



**Fig. 3.** Laser scheme of multi-frequency transfer OPMLs.

It should be mentioned that host molecules should be excellent population storages to be efficient transfer OPMLs. Furthermore, the excited levels of host molecules should be located above the upper laser level of partner molecules [16].

**3. Technical challenges**

Although OPMLs show promises to produce multi-frequency laser transitions, the invention of such the lasers will face several challenges. The search of suitable gas phase media to serve as the multi-frequency media is one of the concerns. To obtain a number of different frequencies, the

excitations of high overtones or combination bands of molecules are required. By the general nature of molecular gases, high overtones and combination bands suffer to weak dipole allowed transitions. Such the drawback would seriously enlarge the gas laser systems. As mentioned previously, however, the hollow waveguide technology has shown the promise of the gas laser based on population inversion [9-10]. The hollow waveguide provides long the interaction length between gas media and pump beams. The development of the hollow waveguide technology is on the route to pursue the next generation of gas lasers [17-19]. In addition, the development of the hollow waveguide for multi-frequency operations has been initiated in very recent years [20]. The fusion between such the development and gas lasers will guide to the creation of multi-frequency lasers based hollow waveguides in the future.

Furthermore, the choices of promising molecules also rely on the absorption lines of such molecules that should be matched with the currently well developed lasers [11]. In addition, the effective excitation can be gained by the developments of the pump sources in the aspects of precise narrowed frequency control and frequency stability.

The choices of molecules for the multi-frequency applications also relate to molecular relaxations such as V-V, vibrational-to-translational (V-T) and rotational-to-rotational (R-R) energy transfer processes which are the important mechanisms for molecular laser performances. The molecular kinetic processes affect to the available population for laser transitions. They also result to the bottleneck and local heat problems occurred in CW or long pulse operations [13]. In addition, the manipulation of frequency tuning can be a great challenge to accomplish by controlling the kinetic relaxation rates. Experimental investigations of involved kinetic relaxation rates will play critical roles for effectively utilizing OMPLs as desired multi-frequency generators.

#### 4. Conclusion

In recent years, the multi-frequency sources have gained remarkable momentum. Several endeavors both experimental and theoretical studies have been performed to achieve such the multi-frequency lasers. OPMLs can be multi-frequency sources in mid infrared region. They can also serve as frequency splitters for many potential applications (multi-frequency imaging, chemical sensing, communications, etc.). Multi-frequency sources based on OPMLs would combine excellent properties of gas phase media (the exceptional heat management, the high optical damage, the supreme beam quality, etc.) and commercial available lasers (solid state lasers, semiconductor lasers, etc.). In addition, they would easily handle kW or higher power operations that would be desirable for high power laser applications.

The hollow waveguide technology potentially supports the multi-frequency OPMLs. The marriage between the multi-frequency OPMLs and waveguide technology is a compelling alternative approach for the compact frequency splitter device.

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