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# THE DEVELOPMENT OF FAR-INFRARED LASERS FOR THOMSON-SCATTERING MEASUREMENTS ON TOKAMAK PLASMAS

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Centre de Recherches en Physique des Plasmas ECOLE POLYTECHNIQUE FEDERALE DE LAUSANNE The Development of Far-Infrared Lasers for
Thomson-Scattering Measurements on
Tokamak Plasmas

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#### Abstract

The article reviews the development of far-infrared (FIR) lasers suitable for making Thomson-scattering measurements on collective thermal electron-density fluctuations in a tokamak plasma, thereby determining its ion temperature. A brief summary of the scattering of e.m. radiation from a plasma is given. Thomson scattering in tokamaks using FIR lasers is discussed and the performance figures required of such lasers are derived. The development of these lasers up to the present is traced, with particular emphasis on the pulsed optically-pumped  $D_2O$  laser. The current performances of  $D_2O$  lasers are examined and various approaches to improving these to the requirements of a scattering experiment are discussed.

#### INTRODUCTION

Currently, about eight groups working in Europe and in the U.S.A are involved in the study of powerful pulsed far-infrared (FIR) or submillimetre lasers operating in the range 50-500 µm approximately. The goal is to develop a laser which enables cooperative scattering measurements to be made on thermal fluctuations in a tokamak plasma, thereby determining its ion temperature. Not surprisingly, the majority of these groups are attached to institutes in which research into tokamaks is performed.

At present, plasma ion temperature in tokamaks is routinely determined from the spectrum of charge-exchange neutrals. In the current generation of large tokamaks under construction, of minor diameters one metre or more, this technique will not be reliable for the plasma interior because the neutrals will have mean free paths shorter than the plasma radius. Spectroscopic analysis of impurity radiation in the ultraviolet will also be unsatisfactory in these plasmas because of their high purity and the relatively-large impurity radiation from their surfaces. FIR laser scattering is a technique which appears very attractive as an alternative to these, provided certain technical problems can be overcome.

In this article, we review the development of pulsed FIR lasers suitable for Thomson-scattering measurements on collective electron density fluctuations in tokamak plasmas. In the first section, the essentials of laser scattering are summarised and the performance figures required of the FIR lasers are estimated. The second section introduces the principles of pulsed optically-pumped FIR lasers. In the third and fourth sections we briefly trace the development of these lasers. The final section discusses how closely the performance of present-day lasers matches the requirements of a scattering measurement, and various approaches to meeting these are considered. The review includes up-to-date results presented at The Second International Conference on Infrared Physics, held at Zurich in March of this year.

## A Brief Review of Thomson Scattering

The principles of using Thomson scattering to measure various plasma parameters are now well established. For a comprehensive account of these, the reader is referred elsewhere 1-3.

Scattering of electromagnetic radiation from a plasma is a three-wave interaction, Figure 1. An incident e.m. wave, of frequency  $\nu_{0}$  and wavelength  $\lambda_{0}$ , interacts with electron density fluctuations, characterised by  $\nu_{f}$  and  $\lambda_{f}$ , to produce an e.m. wave, of frequency  $\nu_{s}$  and wavelength  $\lambda_{s}$ , at an angle  $\theta$  to the incident wave. Momentum conservation requires that  $\underline{k}_{s} = \underline{k}_{0} + \underline{k}_{f}$ , where  $|\underline{k}| = 2\pi/\lambda$ . Neglecting momentum transfer between photons and electrons,  $|\underline{k}_{s}| = |\underline{k}_{0}|$ . Hence  $|\underline{k}_{f}| = 4\pi$  sin  $(\theta/2)/\lambda_{0}$ . Only periodic fluctuations in a direction parallel to  $\underline{k}_{f}$  and of wavelength  $2\pi/|\underline{k}_{f}|$  will be detected. We may determine the scale length and propagation direction of the fluctuations that will be observed in any scattering experiment by a judicious choice of the collection angle  $\theta$  and the laser wavelength  $\lambda_{0}$ .

When  $\alpha=(k_f\lambda_D)^{-1}<<1$ , where  $\lambda_D$  is the Debye length,  $2\pi/|\underline{k}_f|<<\lambda_D$  and we only detect uncorrelated electron density fluctuations. The scattered radiation is Doppler broadened due to the electron thermal motion, Figure 2a. For a plasma in thermal equilibrium, the spectrum is a Gaussian of half-width  $2|\underline{k}_f|v_e/2\pi$ , where  $v_e$  is the electron thermal velocity, which is thus proportional to  $\sqrt{T}_e$ . When  $\alpha>>1$ ,  $2\pi/|\underline{k}_f|>>\lambda_D$  and we observe the correlated motions of the electrons, e.g. due to scattering off electron clouds Debye-shielding the ions and off plasma waves. If  $T_e^{-2\nu}T_i$ , the scattered-radiation spectrum is as shown in Figure 2b. The width of the central ion feature is proportional to  $\sqrt{T}_i$  for a thermal plasma. The two shoulders arise from scattering off ion-acoustic waves and the two detached features are due to electron plasma waves.

Two phenomena could cause problems in an attempt to measure  $T_i$  by Thomson scattering. The first arises because the presence of impurity ions in the

plasma can greatly distort the ion feature. Even concentrations as little as 10 ppm of highly-ionised Mo or W can be detected, the heavy ions manifesting themselves as an intense narrow spike at the centre of the feature due to the fundamental ions of the plasma. Lighter impurities of lower charge, such as 0, have a broader spectrum (due to their higher velocities), which is much less intense (because their Debye-shielding electron clouds contain fewer electrons and the spectrum is broader). Generally, the high-frequency part of the spectrum of the fundamental ions is not much changed by the presence of impurities at a concentration where the tokamak plasma might still be termed "clean". Under this condition, the half width of the ion feature will yield a reasonably-accurate value of T<sub>1</sub>. With adequate laser power and high spectral resolution it might even be possible to determine a value of the effective charge Z<sub>eff</sub> from the low frequency part of the ion feature, although this would be an ambitious project.

The second difficulty would arise if the tokamak plasma had a high level of microturbulence such that the radiation scattered off electron density fluctuations due to this was more intense than the radiation scattered off fluctuations due to ion thermal motion. In this event, the plasma ion temperature could not be determined from the spectrum of scattered radiation. However, the technique could now be used to obtain valuable information concerning the spectrum of turbulence in the plasma. In any case, provided  $\alpha$  is not much greater than unity, in a stable tokamak discharge it should be possible to select a direction for  $\underline{k}_f$  such that the level of the fluctuations due to microturbulence, in that part of the spectrum in which ion thermal motion manifests itself, is negligible.

In a tokamak plasma, the combination of high temperatures (T $\sim$ 1-10 keV) and low densities (n $\sim$ 10<sup>13</sup>-10<sup>14</sup>cm-<sup>3</sup>) results in a relatively-large Debye length ( $\lambda_D^{\sim}0.1$  mm). Using an optical laser, the wavelength  $\lambda_o$  would be so short (<1  $\mu$ m) that in order to observe cooperative scattering ,i.e. to achieve a value of  $\alpha$  > 1, collection angles  $\theta$  of the order 0.1 would be needed. This would result in extremely-poor spatial resolution, since scattered

light would be collected from along almost the whole length of the incident light beam in the plasma. Even using a  $\rm CO_2$  laser, of wavelength 10  $\mu m$ , at most angles of a few degrees could be used. However, using FIR lasers of wavelength 50  $\mu m$ , or more, the collection angles needed are sufficient to permit adequate spatial resolution.

At present, the FIR laser most likely to be used for cooperative scattering measurements employs heavy-water vapour as active medium.  $D_2O$  exhibits strong lasing action on one of three wavelengths, viz 66, 114 and 385  $\mu m$ , when excited by the appropriate emission line of a pulsed  $CO_2$  laser. In principle, this permits the choice of three collection angles  $\Theta$  for a fixed value of the scattering parameter  $\alpha$ . Taking plasma parameters typical of those of a large tokamak -  $\bar{n}_e = 10^{14}$  cm-3,  $T_e(=T_1) = 2.5$  keV - for  $\alpha = 1.5$  and using the above wavelengths in increasing order we obtain the following angles:  $10.8^{\circ}$ ,  $18.7^{\circ}$  and  $66.7^{\circ}$ .

Currently, all FIR detectors that have adequate band-width to be useful in recording the ion feature have a high internal-noise level, termed noise equivalent power (NEP), when used in a video mode. The effective NEP may be considerably reduced if a heterodyne detection system³, in which the scattered signal is mixed with the output of a local oscillator of power  $P_{1o}$ , is adopted, since  $(NEP)_{het}^{\sim} (NEP)_{vid}^2/P_{1o}$ . At 66 and 114 µm a Ga:Ge photoconductive detector⁵ may be used while at 385 µm a GaAs Schottky diode6 or a Nb:Nb Josephson junction is suitable. In general, either the detectors themselves or the associated recording electronics are limited to band widths of a few GHz - this necessitates the choice of the differential scattering vector  $\underline{k}_f$  such that the Doppler broadening  $|\underline{k}_f|v_1/\pi \lesssim 1 \text{GHz}$ , where  $v_i$  is the ion thermal velocity. An optimal method of processing the detected signal is to record it using a transient digitiser and to perform a Fourier transform to yield the power spectrum of the plasma fluctuations8.

The scattered power  $P_s$  incident on the detector is related to the power  $P_s$ 

incident on the plasma by2

$$P_s = P_o n_e 1 d\Omega \sigma_T S(k)$$
,

where  $n_e$  is the plasma electron density, 1 is the interaction length over which scattered radiation is collected,  $d\Omega$  is the collection solid angle,  $\sigma_T = 6.94 \times 10^{-26} \text{ cm}^2$  is the scattering cross-section of an electron and  $S(k) \stackrel{\sim}{\sim} \alpha^4/(1+\alpha^2)(1+2\alpha^2)$  is the integrated dynamic form factor for the ions. The interaction length is  $1 \stackrel{\sim}{\sim} 2r/\sin\theta$ ; r being the radius of the focused beam incident on the plasma. The angle  $d\Omega$  is defined by the restriction, necessary for heterodyning, that the collection etendue (or light gathering power of the collection optics)  $<\lambda_0^2$ , i.e.  $d\Omega \stackrel{\sim}{\sim} 0.5 \lambda_0^2/\pi r^2$ . We obtain the minimum power  $P_0$  needed to obtain a signal-to-noise ratio of 1 by equating  $P_s$  with the post-detection noise power  $P_0 \stackrel{\sim}{\sim} (\text{NEP})_{\text{het}} \cdot (\text{N B}_{\text{if}}/2\tau)^{\frac{1}{2}}$ . In this formula N is the number of channels into which the signal is divided,  $\tau$  is the laser pulse duration (which needs to be at least 100 nsec to permit adequate accuracy in the determination of the power spectrum through Fourier transformation) and  $P_0$  is the intermediate frequency band-width.

In the case of the 66  $\mu$ m line of  $D_2O$ , for a focused spot size 2r = 0.8 cm, a bandwidth of 1 GHz, a pulse duration of 100 nsec, an (NEP) heta  $\frac{1}{2}$  3x10<sup>-18</sup> WHz<sup>-1</sup>; using 10 channels and with plasma parameters  $\bar{n}_e = 10^{14}$  cm<sup>-3</sup> and  $T_e$  (= $T_i$ ) = 2.5 keV, we obtain  $P_o = 350$  kW for a signal-to-noise ratio of unity. Turning to the 385  $\mu$ m line of  $D_2O$ , with conditions identical to those above, except for 2r = 2.0 cm, we obtain  $P_o = 150$  kW. These are minimum powers. In practice, to make measurements of  $T_i$  with reasonable accuracy we would need about ten times the incident power. Furthermore, taking into account losses in the collecting optics and inefficient coupling of the collected radiation into the detector (we have ignored these factors up to now) an incident power  $P_o \gtrsim 5$  MW would be desirable.Also,a pulse of longer duration,  $\lesssim 1$   $\mu$ sec, would be advantageous to obtaining a better statistical signal-to-noise ratio 8.

Finally, because of the band-width limitation of  $\sim 1 \mathrm{GHz}$ , the spectral purity of the FIR laser output must be high - the line width of the laser emission cannot exceed 50-75 MHz, or else it will be difficult to accurately determine the Doppler broadening of the radiation scattered by the plasma.

## Pulsed Optically-Pumped FIR Lasers

The basic principles of optically-pumped FIR lasers are now generally well understood 9-11. In Figure 3, we illustrate the processes leading to the achievement of a population inversion and FIR laser emission, for the simple case of a polar symmetric top molecule such as CH3F. A chance nearfrequency coincidence between the emission line of the pump laser, generally emitting in the infrared, and a certain vibrational absorption line (transition  $v = 0, J \rightarrow v = 1, J$ ) of each of an ensemble of the polar molecules results in the selective population of a rotational level (v = 1.J) in an excited vibrational state. In general, the coincidence must be within a few hundred MHz. A population inversion is achieved between adjacent rotational levels (v = 1, J with respect to v = 1, J-1). Because of the high permanent dipole moment of each molecule the active medium has a high gain and laser emission occurs as a result of purely-rotational transitions ( $v = 1, J \rightarrow v = 1, J-1$ ). Frequently, there is additional laser emission due to accompanying cascade transitions ( $v = 1, J-1 \rightarrow v = 1, J-2$ ) and refilling transitions ( $v = 0,J+1 \rightarrow v = 0,J$ ). For frequency offsets between pump and absorption line of more than a few hundred MHz, under certain conditions of molecular concentration and pump intensity the emission process is of the stimulated-Raman type.

In principle, optical pumping of an active medium using a laser source is undesirable because of the inherent low efficiency in pumping one laser with another. However, in the case of pulsed FIR lasers there is generally no broad-band pumping mechanism that will selectively channel energy into

the desired upper state because the rotational levels are so closely spaced. Thus, we usually find that excitation methods are restricted to selective laser pumping which, in fact, works well for the generation of coherent FIR radiation. The most-widely used pump source is the pulsed  ${\rm CO}_2$  TEA laser, which is one of the most highly-developed lasers and is capable of generating high intensities at relatively-good efficiencies. Furthermore, with over 150 emission lines between approximately 9.1 and 11.3  $\mu$ m, it is possible to match the output frequency of the laser to vibrational modes in a wide variety of molecules. This is demonstrated by the existence of well over 1000 FIR laser lines between about 40  $\mu$ m and 2  $\mu$ m<sup>12,13</sup>.

Intense FIR pulse generation cannot be achieved using conventional Q-switching techniques because, due to molecular collisions, the short lifetime of the upper rotational levels makes significant energy strorage impossible. This lifetime is typically 10 nsec at 1 torr pressure; that of the excited vibrational state is at least four orders of magnitude greater. Under the conditions of saturating pump and FIR intensities, the power extraction from a FIR laser is proportional to the rate of pump absorption from the ground-state rotational level. This process is limited by the collisional refilling of the absorbing ground state and by the emptying of the upper rotational laser states, both of which occur at the rate  $v_r \simeq 10^8~{\rm sec}^{-1}~{\rm torr}^{-1}$ . If  $n_m$  is the molecular density and f is the fraction of molecules in the ground state at equilibrium, then the maximum pumping rate will be  $v_p = {\rm fn}_n v_r$ . The maximum FIR power that can be extracted is  $P_{\rm FIR} = 0.5~{\rm h}v_{\rm FIR}v_p$  (Wcm-3).

There is an optimum working pressure, governed by the pump duration and by molecular dynamics, above which the FIR output decreases due to a "bottleneck" effect. Molecules are pumped at a maximum rate  $\nu_p$  until the limiting excited vibrational-state population 0.5  $n_m$  is reached. The population of the ground and excited vibrational states are now equal, the

rotational levels achieve thermal equilibrium and FIR laser emission ends. The time scale for this to occur is  $_{\rm T} \simeq 1/2~{\rm f}_{\rm V}{}_{\rm r}$ . In symmetric top molecules, we can take f to be about 1% at best, which means for a pump pulse of order 100 nsec we must maintain the working pressure below about 5 torr or the collision rate becomes too high for efficient FIR power extraction. Taking a wavelength of 0.5 mm, and at a molecular density set by the 5 torr limit, the maximum FIR power that can be generated is  ${\rm P}_{\rm FIR} \simeq 150~{\rm kW/litre}$ . It is clear that in order to reach power levels of greater than 5 MW, systems of large volume will be needed.

#### Early Work on Pulsed FIR Lasers

FIR laser emission, as a result of optical pumping, was first observed in  $CH_3F$  <sup>14</sup> Early studies of this medium were performed in mirrorless cells pumped at 9.55  $\mu m$  by the 9P(20) line of a TEA  $CO_2$  laser <sup>15-19</sup>. These consisted, in general, of a tube a few metres long and of diameter  $\sim 20$  cm, with a window at one end transparent to the pump radiation (NaCl or KCl) and with a window transparent to the FIR radiation (PTFE or polyethylene) at the other. Laser emission occurred through the amplification of spontaneous emission by the high-gain medium in a single pass through the tube. This process is called superradiance or amplified spontaneous emission (ASE).

Good power conversion efficiencies were obtained and FIR pulses exceeding 1MW were achieved at 496  $\mu m^{18}$ , for 200 MW of pump power. Linewidths were typically 500-700 MHz FWHM¹7, with the pulse temporal shape being highly irreproducible. The optimum operating pressure of CH₃F was 2-4 torr; at higher pressures depopulation of the upper laser level by molecular collisions became important. The linewidth was much broader than could be accounted for by pressure broadening, the rate for this being  ${\sim}40$  MHz torr¹. This discrepancy was explained by the fact that in the CH₃F molecule the energy levels designated by  ${\sim}$  and J values, and depicted in a

simplified manner in Figure 3, are repeated many times for different values of the quantum number  $K^{9-10}$ . J is the quantum number representing the total angular momentum of the molecule and K represents the component of this momentum along the molecular axis. For several different values of K, the energy differences between the  $\nu = 0$ , J and the  $\nu = 1$ , J levels are almost the same. The  $CO_2$  laser, with an emission line-width of  $\sim 1.5$  GHz, simultaneously pumped the molecules to the  $\nu = 1$ , J rotational level for as many as 5-7 different K-values. This resulted in FIR laser emission on as many rotational lines<sup>17</sup>.

In an attempt to achieve the high intensities, the narrow line-widths and the pulse reproducibility necessary for Thomson-scattering measurements various oscillator configurations were studied  $^{17}-^{21}$ . However, the combination of the very high gain of the medium and its low saturation intensity to the pump makes the extraction of intense spectrally-narrow pulses difficult. Each oscillator produced an output, of line-width 30-150 MHz, on one or more longitudinal or transverse cavity modes, with peak powers of greater than  $^{10}$  kW. In particular, the use of longitudinal-mode-selecting elements in the cavity, such as using a mesh output coupler comprising a Fabry-Perot etalon  $^{17}$ ,  $^{21}$  or incorporating a Fox-Smith interferometer  $^{22}$ , ensured single-longitudinal-mode output. The use of stops eliminated high-order transverse modes.

The output powers of these narrow-line oscillations were further improved by the addition of an amplifier  $^{23-27}$ . Outputs of up to 600 kW were achieved, and it was generally believed that 1MW of spectrally-pure FIR radiation could be generated using from 100-500 MW of  $\mathrm{CO}_2$  laser power. However, a careful study of the FIR emission revealed that the spectra comprised a narrow central feature superposed on a broad low-level pedestal of ASE generated in the amplifier. This was inacceptable since in a scattering measurement the power scattered is typically  $10^{-14}$  of that incident, and parasitic reflections of the latter would completely mask the radiation scattered by the

plasma, which would be of comparable line-width. Alternative schemes, such as injecting the single-mode output of a c.w.<sup>28</sup> or pulsed<sup>22</sup> CH<sub>3</sub>F oscillator into a superradiant assembly, thereby forcing emission in a narrow frequency band, were not noticeably better.

It was not feasible to reach the desired power levels, in a narrow line, by scaling up the volume of the oscillator (and not using an amplifier) since its size is limited by those parameters of the optical cavity which govern the output of a narrow line-width fundamental mode. Also, the use of a pump source of narrow line-width was not considered worthwhile, since there was evidence that several of the closely-spaced K sublevels in the CH<sub>3</sub>F molecule could still be simultaneously excited through the phenomenon of off-resonance pumping<sup>29</sup>. Consequently, because of this and the fact that D<sub>2</sub>O promised to be several times more efficient, CH<sub>3</sub>F was steadily replaced by D<sub>2</sub>O as an active medium.

Initially, the use of  $D_2O$  seemed an easier way of reaching the desired performance levels. Firstly, the medium has three strong lasing transitions, viz. 66, 114 and 385  $\mu$ m, each pumped by a different  $CO_2$  laser line, permitting flexibility in the choice of scattering geometry for a given set of plasma parameters. Secondly, for each pump line  $D_2O$  has only one upper lasing level because it is an asymmetric top molecule, discounting the cascade and refilling transitions  $^{3O}$ , which are sufficiently different in wavelength from the principal transition as to present no serious problems in a scattering measurement. Thus, the prospects of obtaining narrow-band emission appeared good. Finally, any scattered signal would be further away in frequency from the radiation due to cyclotron emission which, it was believed, would cause problems in high-field tokamaks.

The earlier studies of  $D_2O$  were performed in superradiant assemblies at operating pressures of several torr<sup>30-33</sup>. Using a  $CO_2$  laser pump delivering pulses of power about 150 MW, superradiant emissions of average power 11, 6 and 2MW were reported for the transitions at 66, 114 and 385  $\mu$ m (together with the accompanying cascade and refill transitions),

respectively  $^{30}$ ,  $^{33}$ . However, as in the case of CH<sub>3</sub>F, the pulses were irreproducible and also of broad line-width, up to 370 MHz, which was broader than could be accounted for by pressure broadening.

The first attempt at producing a high-power narrow-line-width output from  $D_2O$  used an oscillator-amplifier combination<sup>34</sup>. A very short oscillator, of length 36 cm, was used so that the separation of the longitudinal modes in the cavity - c/2L  $\simeq$  417 MHz, where c is the velocity of light and L the mirror separation - was greater than the gain width of the lasing transition at 385  $\mu$ m. Output on a single longitudinal mode, of width  $\sim$ 20 MHz, was achieved and this was increased by use of an amplifier of length up to 8 metres. Almost 200 kW of power were produced in a line-width of  $\sim$ 70 MHz. However, there was a broad low-level background of superradiance as well.

Various other configurations have been investigated  $^{26}$ . The single-mode output from a short  $D_2O$  oscillator operating on 385  $\mu m$  was injected into the cavity of a second oscillator of length 155 cm which, without the injected signal, operated on as many as five longitudinal modes simultaneously. The length of the slave oscillator was adjusted so that one of its modes coincided in frequency with the output of the master oscillator. In this way, the injected signal caused this mode to dominate, although the other modes were not completely eliminated. Better results were obtained when only the short cavity (of length 36 cm) was used, pumped using all the available  $CO_2$  laser power. However, at intensities exceeding about 1 MW/cm<sup>2</sup> the output of the short oscillator itself became multimode.

One of the more promising techniques to obtain a single longitudinal mode was found to be the use of a Fox-Smith interferometer as mode selector<sup>26</sup>. This is illustrated in Figure 4. There are, in effect, two coupled resonator cavities. The main cavity, of length 170 cm, is formed between the mesh output coupler and the solid mirror with reflecting surface parallel to the mesh. The auxiliary cavity is formed between the two solid mirrors of the Fox-Smith interferometer via the mesh beamsplitter, which is set at 45° to the axis of the laser. With

suitable adjustment of the length of both cavities, only one of the longitudinal modes in the main cavity is of the correct frequency to also form a standing wave in the auxiliary cavity. Consequently, this mode grows at the expense of the others. The quartz plate mounted at  $45^{\circ}$  is used to couple the  $\text{CO}_2$  laser radiation into the FIR laser cavity by reststrahlen reflection; the plate is transparent to the FIR emission. At 385  $\mu\text{m}$ , the results  $^{26}$  showed that the output consisted of predominantly a single mode of width  $\frac{1}{6}$  30 MHz FWHM. Output pulses of power 70 kW were obtained for 250 MW of pump power.

#### Further Developments of FIR Lasers

In 1977 there was a significant advance in the understanding of the lasing mechanism in  $\rm D_2O$  with the demonstration that the 66  $\mu m$  line is produced by stimulated Raman emission  $^{35}$ . The difference between this type of emission and laser emission may be understood by reference to Figure 5. We depict the three rotational levels involved in FIR emission by optically-pumped molecules. Level 1 is in the ground vibrational state, level 2 is in an excited vibrational state and level 3 is adjacent to 2. A laser process consists of two separate steps involving single-photon transitions at the pump and FIR wavelengths. In the first step, the molecule absorbs a pump photon, of wavelength corresponding to the energy difference between levels 1 and 2, and is excited from level 1 to 2. Subsequently, in the second step the excited molecule undergoes a FIR laser transition by stimulated emission from level 2 to 3, emitting a FIR photon corresponding to the difference in energy between these levels. The two steps are uncorrelated in time.

The Raman effect is the inelastic scattering of photons by a quantised system  $^{36}$ . If the molecules in the system can undergo a transition of frequency  $\nu_{\rm t}$  the non-linear interaction of the incident radiation, of frequency  $\nu_{\rm i}$ , with the system, the polarisability of which is modulated at frequency  $\nu_{\rm t}$ , generates scattered radiation at frequencies  $\nu_{\rm R} = \nu_{\rm i} + \nu_{\rm t}$  (the Stokes

and anti-Stokes lines). These components have random phases since the vibrations of molecules are uncorrelated. Essentially, the Raman effect results in isotropic emission, of intensity  ${}^{\circ}10^{-6}$  of that incident. In the case of laser irradiation, as the incident intensity rises so that part of the Stokes radiation which is coherent with it plays an increasingly-important role. The interaction of the laser and Raman fields in the medium generates a frequency component  $v_i$ - $v_R$ , i.e.  $v_t$ . This component forces the molecules into resonant vibration, the motion modulates the laser field resulting in Raman scattering of greater intensity which, in turn, leads to more vigorous molecular vibration. For sufficient laser intensity, the process runs away and stimulated Raman scattering results in the generation of coherent radiation at high conversion efficiencies. Returning to Figure 5b, stimulated Raman emission is evidently a two-photon process; a photon is absorbed from the pump field and simultaneously a photon is emitted into the Raman field (at the FIR wavelength), the molecule being excited from level 1 to 3. A population inversion between levels 2 and 3 is not required - unlike the laser process - indeed level 2 itself need not exist. However, the process is resonantly enhanced by the proximity of level 2. If the pump frequency is off resonance by  $\Delta \nu$  with the frequency interval representing the energy between levels 1 and 2, then the FIR emission will also differ by  $\Delta v$  from the frequency corresponding to the energy difference between levels 2 and 3. In principle, a Raman process can be twice as efficient as a laser process in generating FIR radiation. In the former case, one FIR photon is emitted for each pump photon absorbed whilst in the latter case once 50% of the molecules - each of which was excited by absorbing a pump photon - have emitted a FIR photon a population inversion no longer exists and lasing ceases. Also, the Raman process relaxes the condition that a close coincidence, within a few hundred MHz, is needed between the centres of the pump and absoption transitions.

Spectroscopic data had shown that the line centre of the 9P (32) pump line was separated by  $\sim 1.4$  GHz from that of the absorbing vibrational transition

leading to FIR emission at 66  $\mu$ m and at 50  $\mu$ m (the latter is due to a refill transition). Careful measurements showed that the emission lines were detuned from their respective transitions also by about 1.4 GHz, when a CO<sub>2</sub> laser confined to working on a single longitudinal mode on line centre was used as pump<sup>35</sup>. Subsequent work<sup>37</sup> showed that emission at 385  $\mu$ m, where the frequency difference between the line centres of the 9R (22) pump line and the absorption transition is about 320 MHz, was also due to a stimulated Raman process. The broad line-width due to ASE in D<sub>2</sub>O lasers could now be explained in terms of the broad line-width ( $^{\sim}$ 1.5 GHz) of the CO<sub>2</sub> laser pump - each frequency increment of the pump beam, off resonance with the absorbing transition, giving rise to FIR emission detuned by the same amount. It thus became apparent that a narrow-line-width pump was necessary to achieve FIR emission of narrow line-width.

Another potential means of getting rid of unwanted ASE had been found in experiments on a D<sub>2</sub>O oscillator working at 66 µm<sup>38</sup>. The cavity was a rectangular waveguide of length 240 cm and cross-section 3 x 4 cm. It was pumped in a zig-zag manner by a  ${
m CO}_2$  laser beam which made an angle of about  $15^{\circ}$ with the FIR oscillator axis. For a pump pulse of energy about 6 Joules, over 40 mJ of FIR output were produced at a pressure of 2 torr. The emission spectrum had a half width of  $\sim 350$  MHz and was modulated at intervals of 56 MHz, which corresponded to the longitudinal mode separation of the cavity. Superradiant emission along the optic axis of the FIR laser was found to be very weak,  $\sim 10^{-3}$  of the normal oscillator emission. However, along the line of the zig-zag pumping beam an ASE output of the same strength as the laser output was detected. It had a spectral width of ~500 MHz. Subsequent work<sup>39,40</sup> showed that the spatial beam quality was poor and it was concluded that zig-zag pumping was not a satisfactory way of exciting an oscillator, but that the technique had potential as a means of reducing the level of ASE produced in an amplifier.

### Present Status of, and Future Trends in, FIR Lasers

To date, the highest power obtained in a narrow line-width output has been achieved at 385  $\mu\text{m}^{41}$ . Using a narrow-band pump working on a single mode, about 800 kW of FIR power has been generated in a 4-metre long oscillator, which did not have any mode-selecting element associated with it. The line width of the FIR emission was found to be about 50 MHz FWHM, compared with about 200 MHz when a broad-band  $\mathrm{CO}_2$  laser was used, and comprised three adjacent FIR cavity modes with no evidence of a broad superradiant background. Also, when the frequency of the CO2 laser was changed slightly, by tuning the oscillator to an adjacent cavity mode, the FIR emission changed by the same frequency, confirming that the transition emitted by a stimulated Raman process. Furthermore, the efficiency of the FIR laser was found to increase by almost a factor of two when narrow-band pumping was used instead of broad band. The largest output was found when the 9R (22) emission of the  $CO_2$  laser was on line centre, i.e. when detuned about 320 MHz from the line centre of the absorbing transition. When the  ${
m CO}_2$  laser output was tuned to be in coincidence with the transition the laser process was the dominant emission mechanism; for non-coincidence the Raman process, which is more efficient, became important. The FIR output was achieved using 25 J of CO2 laser energy in an 80 nsec pulse.

In recent work on the 66  $\mu$ m emission of D<sub>2</sub>O <sup>42</sup> single longitudinal and transverse mode output have been achieved using a short FIR oscillator, of length 45 cm, the length of which was stabilised using a frame of invar steel. Transverse-mode selection was obtained by restricting the diameter of the CO<sub>2</sub> pump beam to about 1.5 cm.

It is evident that at present there is sufficient accumulated knowledge on the behaviour of  $\rm D_2O$  lasers to be able to design a system that meets the requirements, previously outlined, of a scattering experiment. The most promising approach seems to be the simultaneous use of a single-mode-output  $\rm CO_2$  laser to pump a FIR oscillator. The latter could be achieved either by

the use of a short resonator cavity or by the use of a Fox-Smith interferometer. The second approach is the more attractive since an oscillator of sufficient length could be used that its output would be adequate to saturate a subsequent amplifying section, thus extracting the energy in the desired mode and suppressing the ASE. Zig-zag pumping of the amplifier could be used as a means of obtaining additional ASE suppression.

An oscillator configuration which appears attractive, and which merits detailed investigation, is that of the unstable resonator43,44. This combines large-volume energy extraction with excellent discrimination against high-order transverse modes, even for oscillators with diameter-to-length ratios as high as 1:5. By using a resonator of length about 100 cm, and by pumping it with a CO2 laser of narrow line-width, single-mode output in the FIR could be achieved. At 66  $\mu\text{m}$ , KCl has a very useful property in being highly reflective - through the phenomenon of reststrahlen reflection and it is also extremely transparent to CO2 laser radiation. In Figure 6, we illustrate an unstable resonator which makes use of this effect44. The CO2 pump radiation enters through the KCl plate and also passes through the KCl output coupler, thereby uniformly pumping the active volume. To the 66 µm radiation the KCl output coupler is highly reflective; the oscillator output is turned through 90° by the KCl plate through reststrahlen reflection. In the near field the output pattern is annular but in the far field it is an Airy pattern, with no hole on axis. An inverted Cassegrainian telescope could be used to shorten the distance over which this occurs.

We may now estimate the amount of  ${\rm CO_2}$  laser power needed to achieve the 5 MW level of FIR power. The best results to date - 800 kW at 385  $\mu m$  in a pulse of duration  ${\sim}100$  nsec and of line width  ${\sim}50$  MHz  ${}^{41}-$  have required about 25 J of  ${\rm CO_2}$  laser energy on the 9R (22) line centre in a narrow bandwidth. To achieve 5 MW, extrapolating linearly, a system capable of delivering about 150 J on the 9R (22) line in  ${\sim}100$  nsec would be required. This represents a large and complex  ${\rm CO_2}$  laser chain; typically it might comprise an oscillator of active volume 3 x 3 x 50 cm, a preamplifier of active volume 5 x 5 x 100 cm, through which the beam makes 3 passes, an amplifier

of active volume 7 x 7 x 100 cm and a final amplifier of active volume  $12 \times 12 \times 100$  cm. The first three modules would operate at a pressure of 1 atmosphere while the final amplifier would operate at 3 atmospheres, to increase the stored energy.

Most of the cost of a FIR scattering diagnostic is invested in the pump laser. This has its advantages; since dozens of molecules that will lase in the FIR when pumped by a  $CO_2$  laser are known - and doubtless there are many that remain unknown - if a better active medium than  $D_2O$  is found it would be a relatively-simple matter to tune the output of the pump laser to the appropriate absorption wavelength.

In the case of the 66  $\mu$ m emission of D<sub>2</sub>O, the Manley-Rowe limit implies, all other factors being equal, that this line is more efficient than that at 385  $\mu$ m by a factor of  $\sim$ 6.8. In practice, factors of up to three have been observed in superradiance experiments<sup>33</sup>. Thus, it would seem that about 60 J of CO<sub>2</sub> laser energy in  $\approx$  100 nsec would be needed on the 9P(32) line to achieve 5 MW of FIR. However, this line has a lower gain than the 9R(22) line; furthermore the line centre of the D<sub>2</sub>O absorption transition is about 1.4 GHz away from the line centre of the 9P(32) line. The CO<sub>2</sub> laser emission thus has to be off tuned by about 1 GHz to bring the two lines into reasonably-close proximity. Consequently, the output off line-centre on the 9P(32) line is much less than if the CO<sub>2</sub> laser was run on line centre on the 9R(22) line. Typically, the same size laser chain would be needed to achieve 60 J on the 9P(32) line as would be needed to achieve 150 J on the 9R(22) line.

As to the question of whether 66 or 385  $\mu m$  is better for performing a scattering measurement on a tokamak plasma, it is not an easy matter to decide between them. At 385  $\mu m$ , the detectors have good bandwidths, Schottky diodes can be operated at room temperatures and the collection angles for the scattered radiation are large; many tens of degrees. However, the

detectors have very small areas so that efficient coupling of the scattered radiation into them is a problem, they are very delicate and cyclotron radiation from the plasma will probably set the limit on the achievable NEP. At 66  $\mu$ m, cyclotron radiation is much reduced, the photoconductive detectors have large areas and they are relatively robust. On the other hand, the detectors have band widths which are only adequate, they operate at liquid He temperatures and the collection angles, 0, needed are somewhat narrow,  $\lesssim 10^{\circ}$ . The latter drawback to using 66  $\mu$ m could be overcome, whilst maintaining all the advantages associated with this wavelength, by developing a D<sub>2</sub>O laser emitting on the 114  $\mu$ m line (or the 94  $\mu$ m line, which is a branch transition) pumped by the 9R(12) CO<sub>2</sub> laser line.

In fact, it is highly desirable to have the capability of using several different FIR wavelengths, since this permits greater flexibility and, for example, would enable a number of values of the scattering parameter  $\alpha$  to be obtained for the same scattering geometry. Given sufficient FIR laser power and adequate spectral resolution at the detector to determine the feature due to scattering off impurity ions in the plasma, the effective charge  $Z_{\mbox{eff}}$  of the plasma may be determined from the ion spectra obtained at two values of  $\alpha$ .

In conclusion, our understanding of FIR lasers and their performance have progressed considerably since the pioneering work of the early 70's. Present—day knowledge is sufficient to design and construct a D<sub>2</sub>O laser capable of making cooperative scattering measurements on a tokamak plasma. Certainly, within a year we should look forward to the first successful FIR scattering experiment being performed on such a plasma.

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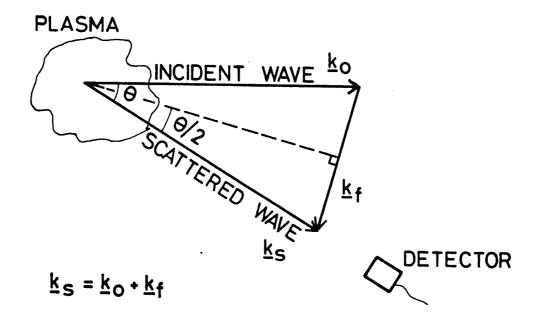
#### References

- 1. D.E. Evans and J. Katzenstein, Rep. Prog. Phys. 32, 207 (1969).
- 2. J. Sheffield, Plasma Scattering of Electromagnetic Radiation (Academic Press, New York, 1975).
- 3. D.E. Evans, Physica 82C, 27 (1976).
- 4. D.E. Evans and M.L. Yeoman, Phys. Rev. Lett. 33, 76 (1974).
- 5. G. Magyar, Fontenay-aux-Roses Report EUR-CEA-FC-815 (1976).
- 6. H.R. Fetterman, P.E. Tannenwald, B.J. Clifton, C.D. Parker,
  W.D. Fitzgerald and N.R. Erickson, Appl. Phys. Lett. 33, 151 (1978).
- 7. T.G. Blaney, D.J.E. Knight and E.K. Murray Lloyd, Opt. Commun. 25, 176 (1978).
- 8. H.C. Praddaude and P. Woskoboinikow, NASA Contractor Report 2974 (1978).
- 9. T.Y. Chang, IEEE Trans. Micro. Theory Tech. MTT-22, 983 (1974).
- 10. D.T. Hodges, Infrared Phys. 18, 375 (1978).
- 11. D.T. Hodges, Proceedings of the Second International Conference on Infrared Physics (Zurich 1979) p. 50.
- 12. M. Rosenbluh, R.J. Temkin and K.J. Button, Appl. Opt. 15, 2635 (1976).
- 13. J.J. Gallagher, M.D. Blue, B. Bean and S. Perkowitz, Infrared Phys. 17, 43 (1977).

- 14. T.Y. Chang and T.J. Bridges, Opt. Commun. 1, 423 (1970).
- 15. F. Brown, R. Horman, A. Palevsky and K.J. Button, Opt. Commun. 9, 28 (1973).
- T.A. DeTemple, T.K. Plant and P.D. Coleman, Appl. Phys. Lett. <u>22</u>, 644 (1973).
- 17. F. Brown, S. Kronheim and E. Silver, Appl. Phys. Lett. 25, 394 (1974).
- 18. D.E. Evans, L.E. Sharp, B.W. James and W.A. Peebles, Appl. Phys. Lett. 26, 630 (1975).
- 19. R.J. Temkin, D.R. Cohn, Z. Drozdowicz and F. Brown, Opt. Commun. <u>14</u>, 314 (1975).
- 20. L.E. Sharp, W.A. Peebles, B.W. James and D.E. Evans, Opt. Commun.  $\underline{14}$ , 215 (1975).
- 21. D.R. Cohn, T. Fuse, K.J. Button, B. Lax and Z. Drozdowicz, Appl. Phys. Lett. 27, 280 (1975).
- 22. D.E. Evans, L.E. Sharp, W.A. Peebles and G. Taylor, IEEE J. Quant. Electr. QE-13, 54 (1977).
- 23. Z. Drozdowicz, R.J. Temkin, K.J. Button and D.R. Cohn, Appl. Phys. Lett. <u>28</u>, 328 (1976).
- 24. F. Brown, P.D. Hislop and S.R. Kronheim, Appl. Phys. Lett. <u>28</u>, 654 (1976).
- 25. A. Semet and N.C. Luhmann, Jr., Appl. Phys. Lett. 28, 659 (1976).

- Z. Drozdowicz, P. Woskoboinikow, K. Isobe, D.R. Cohn, R.J. Temkin,
   K.J. Button and J. Waldman, IEEE J. Quant. Electr. QE-13, 413 (1977).
- 27. F. Brown, P.D. Hislop and J.O. Tarpinian, IEEE J. Quant. Electr. QE-13, 445 (1977).
- 28. T.K. Plant and T.A. DeTemple, J. Appl. Phys. 47, 3042 (1976).
- 29. H.R. Fetterman, H.R. Schlossberg and J. Waldman, Opt. Commun.  $\underline{6}$ , 156 (1972).
- 30. D.E. Evans, R.A. Guinee, D.A. Huckridge and G. Taylor, Opt. Commun. 22, 337 (1977).
- 31. T.K. Plant, L.A. Newman, E.J. Danielewicz, T.A. DeTemple and P.D. Coleman, IEEE Trans. Micro. Theory Tech. MTT-22, 988 (1974).
- 32. F. Keilmann, R.L. Sheffield, J.R.R. Leite, M.S. Feld and A. Javan, Appl. Phys. Lett. 26, 19 (1975).
- 33. D.E. Evans, L.E. Sharp, W.A. Peebles and G. Taylor, Opt. Commun. <u>18</u>, 479, (1976).
- 34. P. Woskoboinikow, Z. Drozdowicz, K. Isobe, D.R. Cohn and R.J. Temkin, Phys. Lett. 59A, 264 (1976).
- 35. S.J. Petuchowski, A.T. Rosenberger and T.A. DeTemple, IEEE J. Quant. Electr. QE-13, 476 (1977).
- 36. G.W. Chantry, Submillimetre Spectroscopy (Academic Press, New York, 1971).
- 37. J.D. Wiggins, Z. Drozdowicz and R.J. Temkin, IEEE J. Quant. Electr. QE-14, 23 (1978).

- 38. G. Dodel and G. Magyar, Appl. Phys. Lett. 32, 44 (1978).
- 39. G. Dodel, G. Magyar and D. Véron, Infrared Phys. 18, 529 (1978).
- 40. L. Allen, G. Dodel and G. Magyar, Opt. Commun. 28, 383 (1979).
- 41. P. Woskoboinikow, H.C. Praddaude, W.J. Mulligan, D.R. Cohn and B. Lax, J. Appl. Phys. <u>50</u>, 1125 (1979).
- 42. G. Dodel, Proceedings of the Second International Conference on Infrared Physics (Zurich 1979) p. 223.
- 43. D.P. Hutchinson and K.L. Vander Sluis, Conference Digest on the Second International Conference and Winter School on Submillimeter Waves and Their Applications (Puerto Rico 1976) p. 80.
- 44. M.R. Green, P.D. Morgan and M.R. Siegrist, J. Phys. E: Sci. Instrum. 11, 389 (1978).



$$\left|\underline{\mathbf{k}}_{\mathsf{f}}\right| = 2\left|\underline{\mathbf{k}}_{\mathsf{O}}\right| \sin(\theta/2) = \frac{4\pi}{\lambda_{\mathsf{O}}} \sin(\theta/2)$$

Figure 1 Vector diagram depicting the scattering of an e.m. wave by a plasma. The magnitude of the differential scattering vector  $\underline{k}_{\hat{f}}$  is defined.

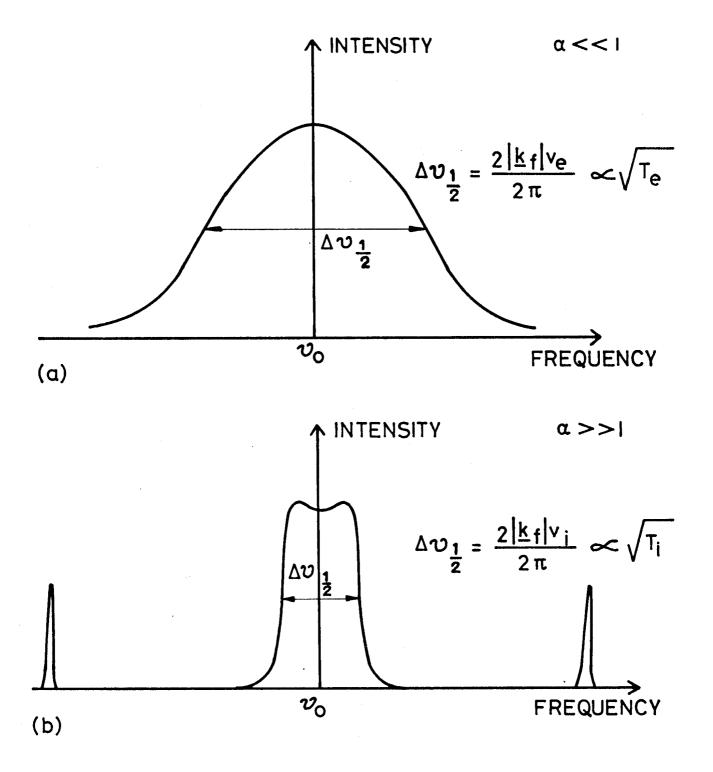


Figure 2 Intensity versus frequency profile of e.m. radiation scattered from a plasma in which  $T_e = T_i$ , for two ranges of the scattering parameter  $\alpha$ .

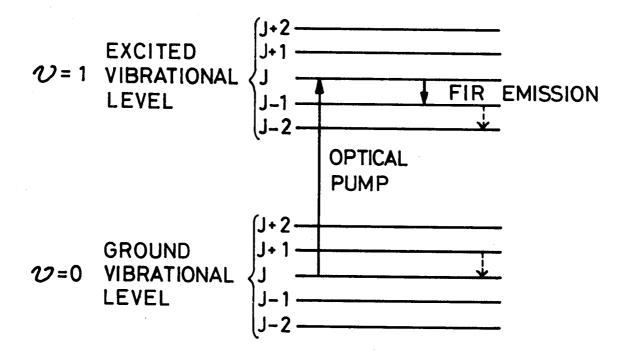
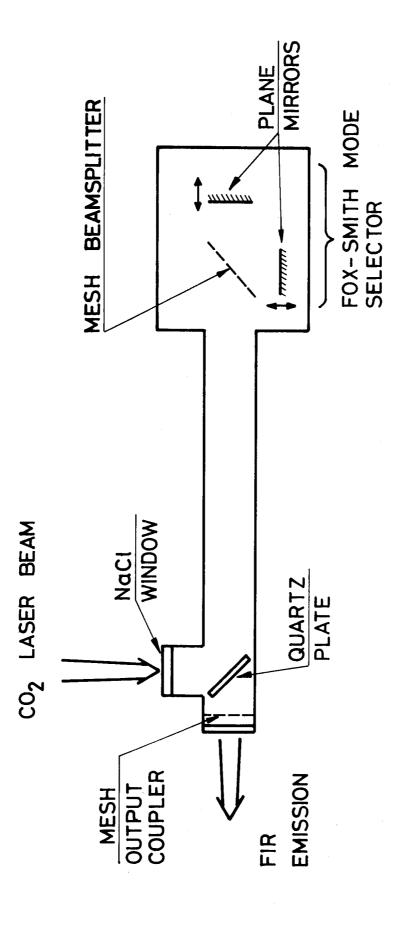


Figure 3 Energy level diagram of an optically-pumped far-infrared laser. The FIR transition occurs between two rotational levels of the upper vibrational state. Also shown are possible cascade and refilling transitions.



FIR oscillator, incorporating a Fox-Smith mode selector, for the 385  $\mu m$  line of  $D_2 \text{O.}$ Figure 4

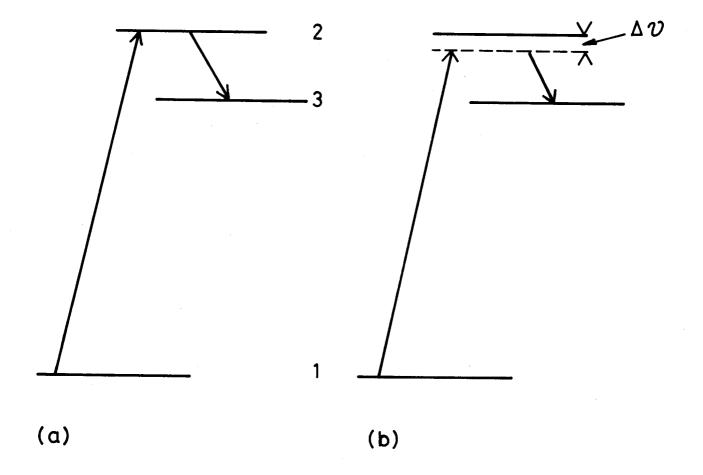


Figure 5 Energy level diagrams illustrating differences between
a) emission by a laser process and
b) emission by a stimulated Raman process.
For further details see text.

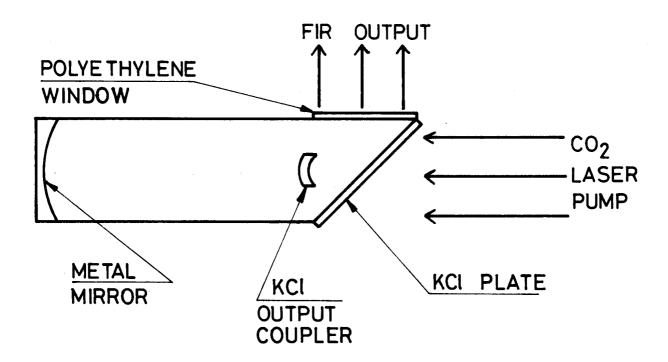


Figure 6 FIR oscillator for the 66  $\mu m$  line of D<sub>2</sub>O, incorporating an unstable resonator and utilising the effect of reststrahlen reflection in KCl.