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Far-Infrared Wave Generation by Frequency Tripling

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Abstract

In future fusion-oriented plasma devices α-particle diagnostics will be of great importance. It is generally recognized that collective Thomson scattering techniques show great potential in this field. For ion temperature measurements as well as for α-particle diagnostics a radiation source emitting in the submillimetre range is required. However, powerful radiation sources which meet the requirement of pulse durations in the millisecond range are not easily available at these wavelengths. A high frequency gyrotron, combined with an efficient frequency tripler could be an interesting alternative to the free-electron laser, the only source which has been considered up till now.

We report experiments on third harmonic generation of radiation in n-doped Si, using a source with a wavelength of 676μm. A power of up to 2 kW could be generated at 225μm with a conversion efficiency of 0.1% While the onset of saturation could be observed, the power limit was due to surface breakdown effects rather than saturation.

Introduction

The conversion of a sizable fraction of an electromagnetic wave to a wave at a harmonic frequency is by now well known and has been applied with great success in particular in the visible part of the spectrum, where very impressive conversion efficiencies of more than half of the initial power have been achieved. A range of experiments have also been reported in the mm and FIR regions. So the topic of this paper is not new, and indeed, at least a part of this presentation will be a review of the field, while a large fraction will, of course, be devoted to reporting our own experimental results. A novel aspect of our research, apart from specific results, is given by the motivation behind it: while we are also investigating frequency tripling in the FIR in its own right and with the hope to further the knowledge in this field, we have a very specific application in mind: collective Thomson scattering as a plasma diagnostic tool. This imposes certain constraints in our research and in particular makes "conversion efficiency" of primordial importance.

In the first part of this paper a short review of collective Thomson scattering and an outline of what we hope to achieve with it will be given. The proposed project requires the availability of a powerful radiation source in the FIR, which could probably only be supplied by means of a free electron laser at the present time. We have considered two possible ways to achieve this:

1) operation of a gyrotron at a higher harmonic and
2) passive frequency conversion of gyrotron radiation in solid state materials.

In actual fact a combination of the two will probably be required, since the ideal radiation source for collective Thomson scattering should have a wavelength around 250μm which cannot easily be achieved with a single step process.

A few remarks concerning work on frequency conversion in the FIR and on the operation of gyrotrons at harmonic frequencies will then be given. Finally our results on passive frequency conversion in solid state materials and molecular gases will be reported. We have also a project to investigate a gyrotron operating at the third harmonic, but work in this field is only just starting at our lab: so it will not be possible to report any results yet.

Collective Thomson scattering as plasma diagnostic tool

Thomson scattering has become the standard technique to measure the electron temperature in a tokamak. It can also be used for density measurements, determination of the toroidal current and to obtain information on the fluctuation levels in a plasma. The advantages of this method are mainly its precision and its good temporal and spatial resolution which allows one to follow the evolution of the parameters concerned. Although it is an active method, the resulting perturbation of the plasma is completely negligible.

The cross-section for Thomson scattering is in fact very small:

$$\sigma_T = 8\pi r_e^2 / 3 = 6.65 \bullet 10^{-29}\ m^2$$

Another way of illustrating this is to compare the power of the radiation collected in a typical scattering arrangement with the power of the radiation source. This is typically of the order of 10^{-12} to 10^{-15}. This small value is one of the main reasons why a Thomson scattering diagnostic is not an easy measurement and requires powerful radiation sources as well as sensitive detection systems.

It is considerably more difficult to determine with this same method the movement of ions, although a feasibility demonstration has been given at CRPP (1). On the TCA tokamak it has been possible to determine the ion temperature with reasonable precision, at least at higher plasma densities. The system comprised a molecular FIR gas laser optically pumped with a CO_2 laser of 500J energy in a single pulse. The scattered radiation was detected with a Schottky diode in a heterodyne arrangement. While the capabilities of the equipment were fully exploited, it was also realized, that insurmountable limits had been reached, mainly due to the fact that the pulse length of a high power, optically pumped molecular laser cannot be increased much beyond a few μs.

The signal to noise ratio at the output of a heterodyne arrangement (which is the only detection method feasible in this wavelength region) can be expressed as (2)

$$S / N = (s / n) / (1 + s / n) \bullet \sqrt{(1 + \tau \Delta f)}$$

where s is the measured signal power, n the power of the noise due to whatever effect limits the performance of the system (usually either the noise temperature of the diode or plasma background radiation), τ is the pulse length or integration time and Δf the width of a frequency channel. For a small signal the signal-to-noise obviously increases linearly with source power, but this effect saturates for powers approaching the noise power. At this stage it can only be improved further by increasing the pulse duration and measurement integration time. Although the improvement is only proportional to the square root of τ, it must be realized that most plasma dynamic effects of interest in this context have time scales in the ms domain. If τ could be increased to 10 ms instead of the 1μs which is typical for a FIR laser, the signal to noise ratio could be improved 100-fold. Gyrotrons can indeed supply the required power levels and can - in principle -

be operated continuously. However, gyrotrons operate at mm wavelengths and up to now no sources of the required type exist in the FIR with the possible exception of the free electron laser FEL.

In fact, even if such sources existed, a system for collective Thomson scattering would probably not be developed for measuring the temperature of the majority ions because several other methods have meanwhile been developed to do this and some of these methods (charge-exchange spectroscopy, for example) have reached a degree of sophistication and a precision which can hardly be expected to be beaten by Thomson scattering (3). However, there are currently no proven methods to diagnose fast ions and α-particles which will be produced in future activated machines like ITER. Hence collective Thomson scattering is considered again with renewed interest. Fig. 1 shows that by judicious choice of the parameters, spectra can be produced where the features of electrons, majority ions and α-particles are nicely separated and information on either of these constituents can be obtained.

Possible radiation sources for collective Thomson scattering

The main source of noise at the frequencies considered is the emission of cyclotron radiation (ECE) by the plasma electrons. The spectrum of this radiation consists of a broadened peak at the fundamental frequency and several harmonics which are more or less resolved. Fig. 2 shows a typical emission spectrum. Beyond a particular maximum frequency the emitted power decreases rapidly. As a general guideline it can be assumed that beyond about $10\omega_0^{ce}$ the emission can be neglected. Thus, with respect to collective Thomson scattering, there are three possible frequency ranges:

1) One could choose a frequency which is smaller than the lower edge of the fundamental ECE emission. In ITER $\omega_0^{ce} \approx 120$ GHz and hence one could operate around 80 GHz using a gyrotron as source. In this frequency range refraction effects are important in a plasma and the beam trajectories of the incident and scattered beams would no longer be straight lines (4). Apart from this, one is close to resonances and even cutoff. The diagnostic would hence be limited to low plasma densities. Supra thermal electrons would absorb part of the radiation and since the radiation noise level is high, a powerful source would be required. It would typically have to supply peak powers of 1GW, a modest mean power of 1MW, short pulses (but > 10ns) and high repetition rate. Such sources are not currently available, but it is believed that they are within the range of current technology.

2) With a radiation source with frequency inside the emission band of the ECE one would have to operate at an emission minimum. Generally one would choose the frequency between the fundamental and the second harmonic, which for ITER would be around 180 GHz. The scattering angle would have to be around 90°, which is convenient, but care would have to be taken to choose the angle between the plane of the scattering vectors and the magnetic field direction with an offset of more than 10° from normal to avoid modulation of the spectrum by the magnetic field in a difficult to interpret way (5). This requires vertical access to the machine which is probably not possible on ITER. Measurements at high densities would still be difficult to achieve and the requirements with respect to power, pulse duration and repetition frequency would not be much less stringent than with the first option.
A system of this type has been installed on JET, but has not produced any results yet (6).

3) For the third high frequency option ($\omega > 1.2$ THz, $\lambda < 250$ μm) small scattering angles (~10°) in forward direction would be required. Radial ports available on ITER could be used. The spatial resolution of a few cm would be acceptable for most applications. The measurements of the fast ions would be largely insensitive to bulk plasma conditions.

For the source parameters the following approximate rule could be used which links peak power P, repetition frequency F and pulse duration t:

$$P(\mathrm{MW})\,[F(\mathrm{kHz}) \bullet \tau(\mathrm{ns})]^{1/2} \geq 10^3 - 10^4$$

A possible combination would be P = 200 MW, F = 20 kHz and τ = 100 ns. A system comprising an optically pumped master oscillator and a free electron laser amplifier might constitute such a system. However, one should not forget that a FEL is an instrument of fair size and price tag.

The condition given above could also be satisfied with P = 30 kW, F = 100 Hz and τ = 1 ms. If it was not for the fact that we are talking about a frequency exceeding 1 THz, these conditions could be achieved quite easily with a gyrotron. For ITER the condition we specified for the high frequency option ($\omega > 10\omega_0^{ce}$) corresponds to a wavelength around 250μm. If the frequency of a 400 GHz gyrotron could be tripled with reasonable efficiency (10 to 20%) such a system would be an interesting solution.

A gyrotron operating at 300 GHz is in operation at the MIT in Boston (7). It produces beams exceeding half a MW power. At much higher frequencies, powerful magnets are required. Alternatively, the research on the operation of gyrotrons at higher harmonics of the fundamental frequency is quite advanced. We will not report on developments in this field but would just like to mention that a possible design for a Thomson scattering system could be based on a 500 kW gyrotron at 400 GHz, the radiation of which would then have to be converted with an efficiency of 10% to the required wavelength of 250μm. This passive frequency conversion is the main topic of this paper.

Frequency tripling in the far infrared

Efficient frequency converters have become commonplace in the visible and near infrared (8). High powers are required and hence these techniques have only been developed after lasers were available.

In the radio and microwave regions, nonlinear elements have been in use long before, where mixing techniques have mainly been used for detection purposes. However, these applications have practically always been restricted to low powers. For high power applications bulk materials are particularly attractive and hence our research program is devoted to the study of semiconductor bulk materials.

Since resistance to radiation damage mainly induced by surface breakdown effects becomes important, gaseous materials would be quite attractive too. If breakdown occurred in the gas and would alter its properties, it could easily be replaced. Near resonant multiphoton transitions could be used involving rotational energy levels and we have found that systems with appropriate selection rules could indeed be found for particular frequencies. However, transition probabilities depend on differences of population densities of the levels involved, and these differences in thermal equilibrium are all quite small. An additional tuned radiation source would have to be used to modify the population density of at least one of the levels involved. This makes it a rather complex system. Calculations showed, however, that sizable conversion efficiencies could be reached and with appropriate mixing-in of buffer gases the phase-matching problem could be solved as well. Due to its complexity this project has not been pursued further for the moment.

Apart from gases, doped semiconductors also show nonlinear properties. These are mainly caused by the motion of the free carriers, i.e. electrons in the conduction band for n-doped materials, under the influence of the electric field of the incident wave. First measurements of frequency-tripling in semiconductors were reported by Mayer et al (9). They used an optically pumped laser as source. This system did not operate in a single mode and hence showed large intensity fluctuations during the pulse. The power at the third harmonic is proportional to the cube of the instantaneous fundamental power, at least in the small signal regime. The power fluctuations at the fundamental frequency are thus considerably enhanced which makes the interpretation of the time integrated measurements rather difficult.

Power fluctuations of such systems are typically in the ns time domain and hence a detection system is required which is fast enough to resolve oscillations of such frequencies. Since speed and sensitivity of a detector are contradicting requirements, time resolved measurements are only possible by using powerful radiation sources. Such a system was available at our Institute from the feasibility studies of collective Thomson scattering on the TCA tokamak (1). Two Ge-hot electron detectors were used for the time-resolved frequency tripling experiments we are going to report. But before this we will briefly discuss the mechanisms responsible for third-harmonic generation in bulk semiconductors.

For frequencies which are comparable to or slightly higher than the scattering frequency of the carriers, the equation of motion of a single carrier can be written as

$$m^*(v)\, v + m^*(v)\, v / \tau(v) = qE$$

There are mainly two effects which give rise to nonlinearities: the velocity dependence of the effective mass $m^*(v)$ caused by the nonparabolicity of the conduction band and the scattering time $\tau(v)$ which also depends on the particle velocity. The nonlinear reaction of the carriers to an applied oscillating electric field leads to nonlinear polarization which can be described by a power expansion

$$P = \chi E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots$$

Due to the spatial symmetry of the system, even terms can be neglected and the lowest order nonlinear polarization $P(3\omega)$ leads to frequency tripling. If this term is introduced as source term into the Maxwell equations, the following relationship between the fundamental and third harmonic field components is obtained

$$E(3\omega) = 4\pi\chi^{(3)}\, C(d)\, E^3(\omega)$$

Hereby the factor C(d), which depends on sample thickness, describes phase matching and absorption. It can be shown that the imaginary part of the complex dielectric function $\varepsilon(\omega)$ is proportional to the conductivity and hence the doping level of the material. Hence higher doping does not only improve the nonlinear susceptibility, but enhances absorption in the same way and limits the maximum conversion efficiency. Further optimization can only be achieved with a careful selection of material parameters and a more profound theoretical treatment is required to be able to understand the mechanisms involved.

The basic mechanisms described above are not critically dependent on wavelength as long as one does not enter a regime where new effects have to be included. In particular for the silicon sample used, experiments in the range of fundamental frequencies between 100 and 400 GHz should yield directly comparable results. In our experiments we have used both a gyrotron at 140 GHz and an optically pumped far-infrared laser at 440 GHz. While with the former long-time effects can be investigated, the latter yields higher peak power and allowed us to study the onset of saturation. A much more detailed study could be done with the laser which was dedicated to these experiments, while access to a gyrotron has only been very limited up till now.

Experiments

The experimental set-up used is shown in Fig. 3. Our optically pumped far infrared laser has been described in the literature (10). The pump source comprises a hybrid TEA oscillator followed by a triple pass amplifier operated at 2 bar. Pulses of 1μs duration with energies up to 500J can be produced at the 9P20 line used in these experiments. Pyrex tubes of 20 cm diameter inside a pressure tight tank formed the FIR resonator which comprised an aluminium coated concave glass mirror and a flat Al plate with large circular hole as output coupler. The pump radiation was injected into the L-shaped resonator through a wire grid which acts as polarizing mirror for the FIR. The hole in the output coupler had been optimized for maximum output power, irrespective of the resulting stability properties of the resonator. While the output radiation showed a Gaussian-like spatial profile, it did probably contain several transverse modes and certainly covered a range of axial modes. The CH_3F gas in the tank at a pressure of 3.5 torr produced radiation at a wavelength of 676μm (11) in pulses of up to 120 mJ energy and 2 MW power. The beam was focused onto the wedge shaped sample by a Teflon lens of f=13.5 cm leading to a focal spot of 3 mm diameter. A wave-guide cut-off filter was used to reduce the fundamental frequency by 10 orders of magnitude while it transmitted 60% of the third harmonic power. Two Ge-hot-electron detectors were used for simultaneous time-resolved measurements at the fundamental and third harmonic frequencies, recorded on a Lecroy digital oscilloscope with sampling speed of 2.5 Gs/s.

Results and discussion

The two signals show series of peaks which are several ns long. The third harmonic peaks are much more pronounced as is to be expected from the nonlinear behavior and they are well correlated with the peaks at the fundamental frequency. In Fig 4 we show the heights of correlated peaks plotted against each other. The highest peaks correspond to a power of 2MW at the fundamental frequency and 2 kW at the third harmonic. The maximum conversion efficiency thus observed was 0.1%. Note that these numbers refer to directly measured power values and do not include any corrections due to the coupling of the radiation into and out of the sample.

The dynamic range of these results was 2 orders of magnitude of third harmonic power and was limited by the sensitivity of the detector on the low side and by surface breakdown of the sample on the high side. If breakdown could be suppressed, higher conversion efficiencies should still be possible since the slope in Fig 4 of about 1.5 at the highest powers suggests that a more than linear increase could still be maintained for a while. It is to be expected that not only the conversion efficiency, but also the third harmonic power reaches a maximum and cannot further be increased, although this limit could not be reached with our measurements.

Conversion efficiencies on the 0.1% level are obviously not good enough yet for the application we have in mind. Other, more promising materials have to be investigated and schemes have to be devised to couple the radiation into the sample in a more efficient way, using perhaps resonant enhancement techniques to increase the power inside the sample.

We are also investigating more complex techniques based on planar arrays of Schottky diodes with matched antennas. Conversion efficiencies of greater than 10% have been reported (12), albeit at power loadings which were considerably lower than what we have in mind.

In conclusion, we have investigated frequency tripling in the FIR regime with the aim of developing a powerful radiation source for plasma diagnostic purposes, using existing high power mm-wave devices in the form of gyrotrons as basic sources. While the required conversion efficiency has not been reached yet, the results are encouraging and allowed to identify the future direction of research.

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Figure captions

Fig 1: Typical spectrum obtained with collective Thomson scattering for plasma conditions representative of JET. Spectral regions exist where either the electron feature e, the majority ion feature i or the α-particle feature α is dominant.

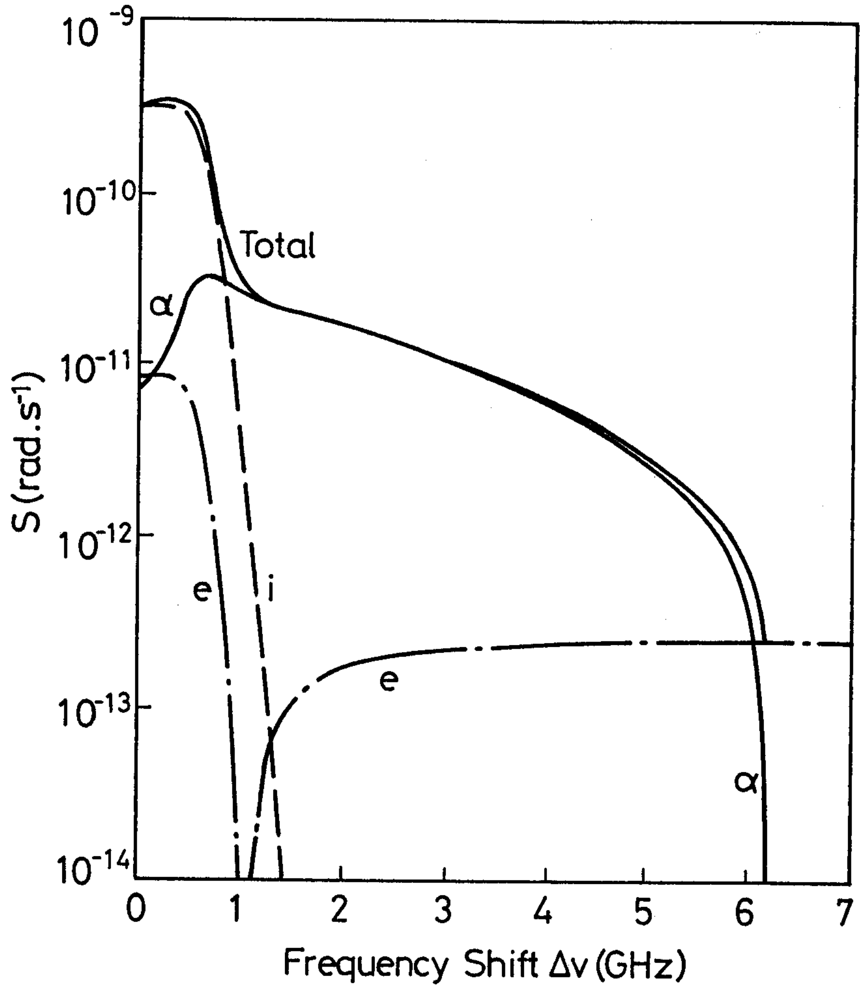
Fig 2: Typical spectrum of ECE radiation. To avoid interference from ECE, the source frequency for collective Thomson scattering could be chosen to be 1) smaller than the lower edge of the fundamental peak, 2) at the minimum between fundamental and first harmonic, or 3) higher than the highest significant harmonic ($\geq 10\ f_{ce}$)

Fig 3: Experimental arrangement for the generation of third harmonic radiation. The elements of the FIR laser are:

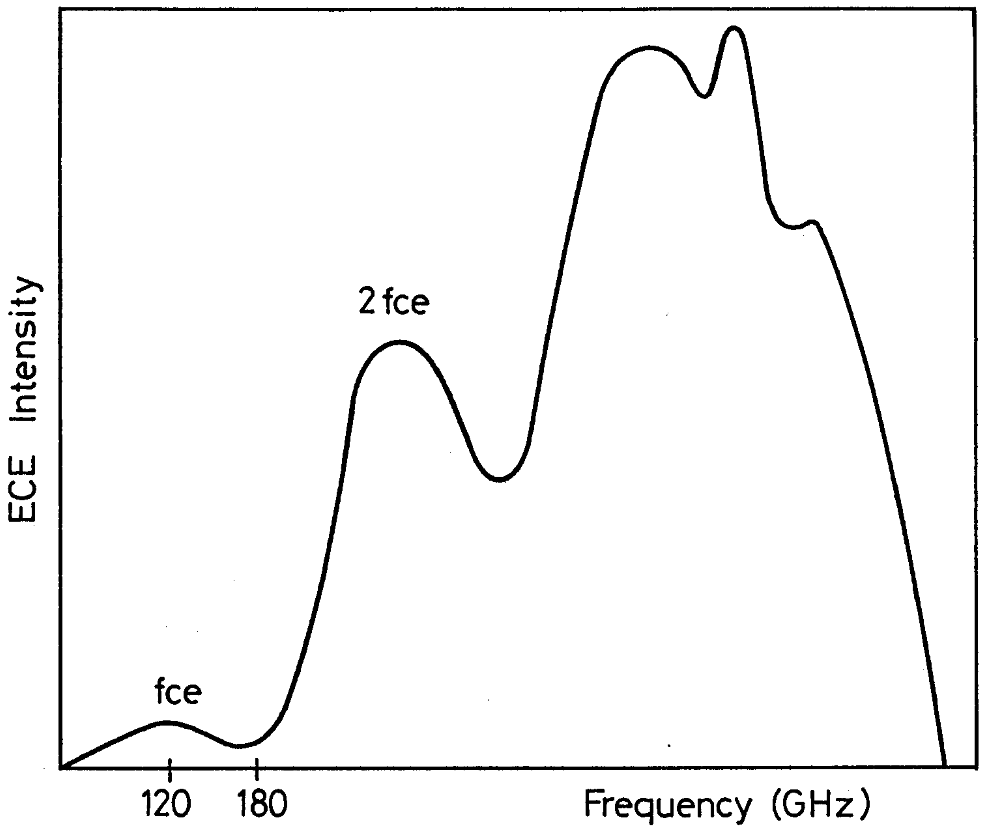
VV: vacuum vessel
PT: pyrex tube (20cm diameter)
W1: KCl window
W2: TPX window
WG: free standing wire grid
CM: concave aluminium coated glass mirror with RC=10m
OC: flat metal output coupler with central hole of 8 cm diameter
FM: focusing metal mirror
L: Teflon lens with f=13.5cm
BS: beam splitter (mylar)
S: wedged sample of n-doped Si
F: waveguide cut-off filter
D1,D2: identical Ge-hot-electron detectors
cavity length CM-OC: 4m

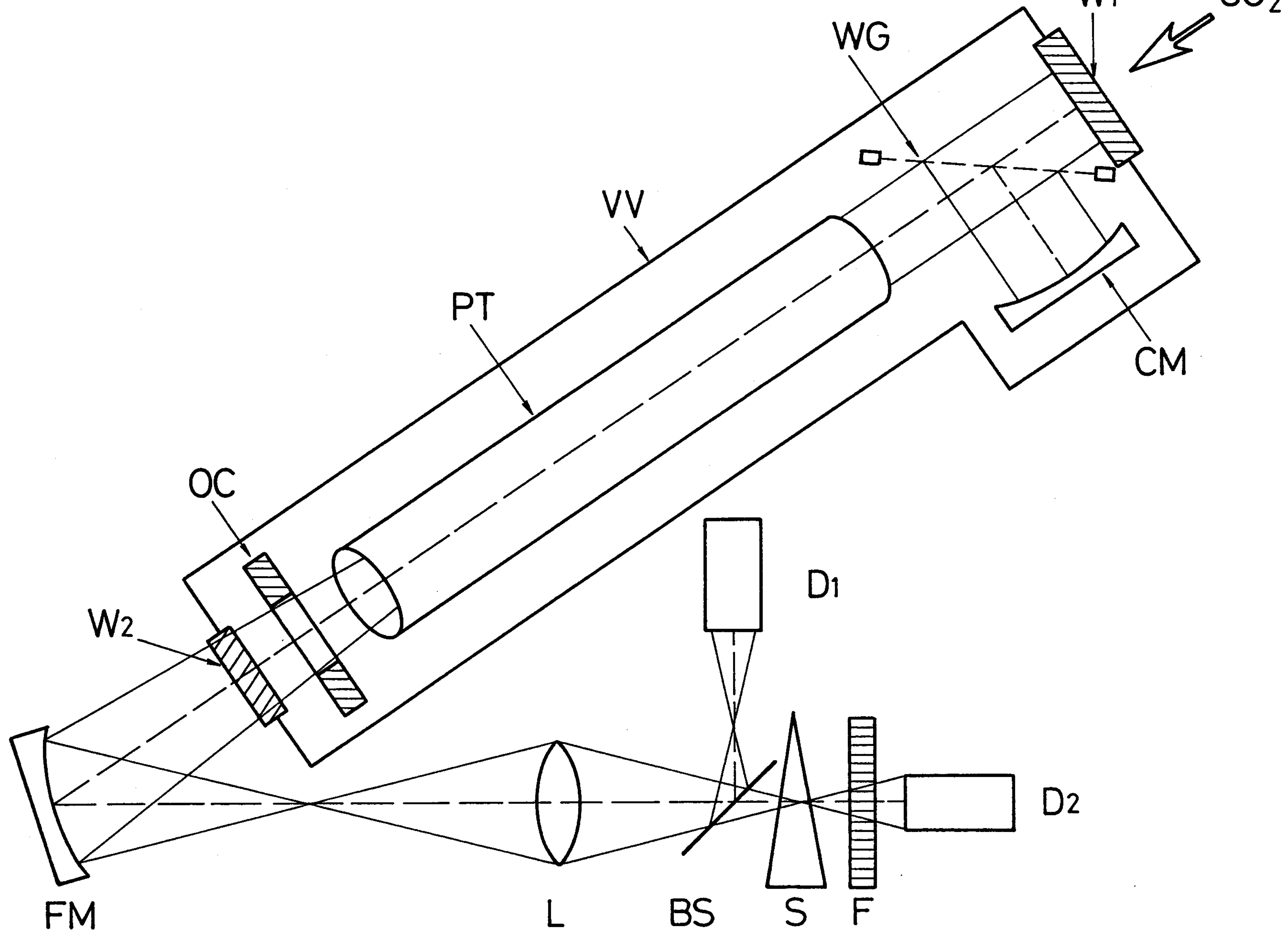
Fig 4: Third harmonic power vs. fundamental power of correlated peaks (several shots). The slope decreases from 2.5 to 1.5 over the range shown

Fig 1



10^-9
10^-10
10^-11
10^-12
10^-13
10^-14
S (rad.s^-1)
Total
α
e
i
e
α
0
1
2
3
4
5
6
7
Frequency Shift Δν (GHz)


ECE Intensity
2 fce
fce
120
180
Frequency (GHz)



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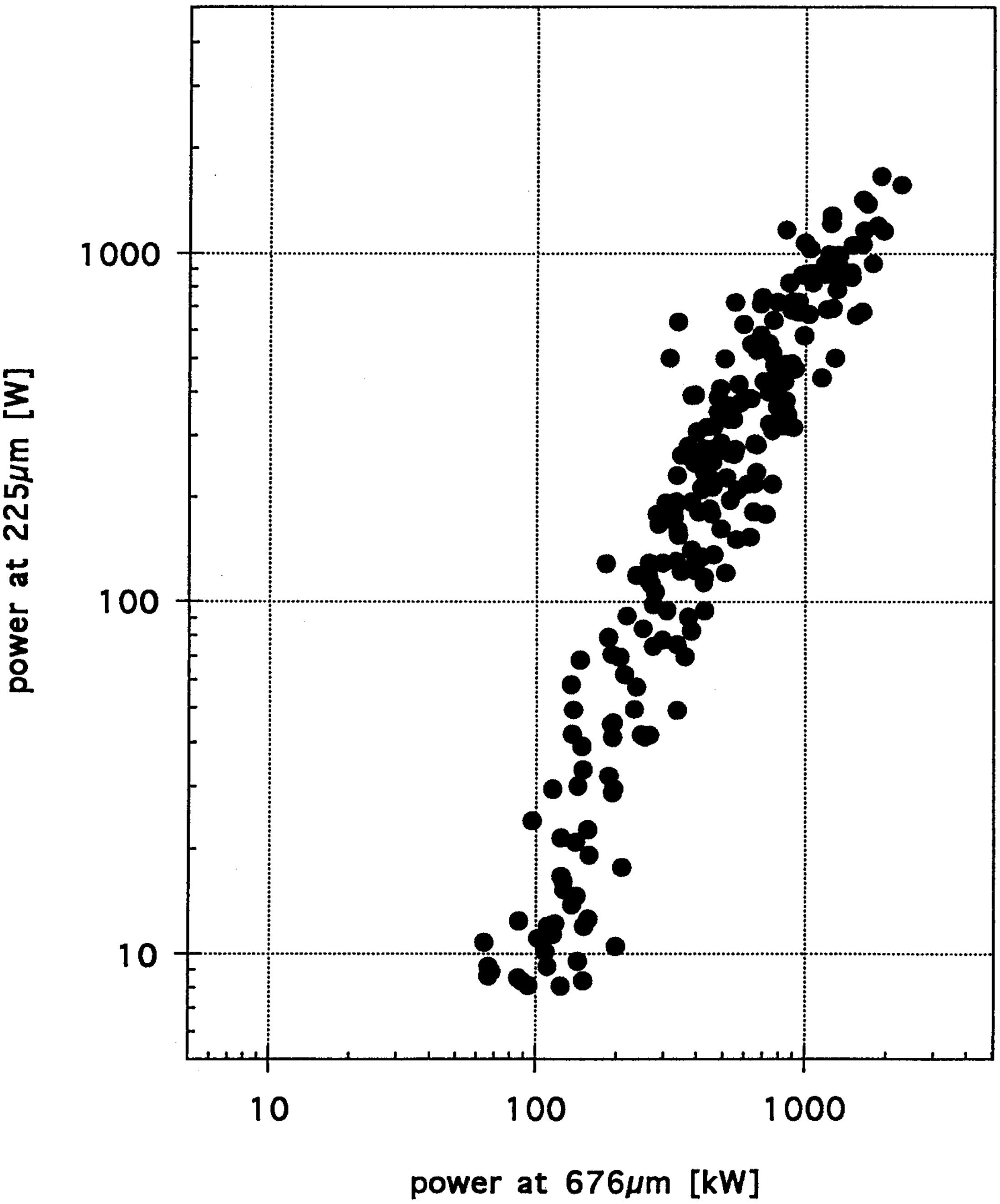
1000
100
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power at 225μm [W]
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1000
power at 676μm [kW]