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Optimum third harmonic generation efficiency in the far infrared in Si, GaAs and InP

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Abstract

We investigate by means of a Monte Carlo technique the nonlinear drift response of electrons to high power electromagnetic waves in Si, GaAs and InP. The first and third harmonic drift velocity amplitudes and phases are presented as functions of the pumping wave frequency in the range of 200 to 500 GHz. The third harmonic generation efficiency is found to reach a maximum at a pumping wave amplitude of 10Π25 kV/cm depending on the material and the lattice temperature. Cooling down to liquid nitrogen temperature results in an improvement of the efficiency by a factor of 2 to 10, depending on the material and the pumping wave amplitude. Cooled GaAs and InP are both an order of magnitude more efficient than Si at ambient temperature, for which to date the best measured performance has been reported.

Introduction

Powerful radiation sources are available in practically all regions of the electromagnetic spectrum, but are still missing in the submillimeter or far-infrared region in the wavelength range of 100 mm to 1mm, where they are required for high temperature plasma diagnostics in tokamaks. At the present time only free electron lasers are able to produce sizeable powers in this region. However, free electron lasers are large installations which are not readily available. Powerful radiation sources exist in the form of gyrotrons at larger wavelength. They require exceedingly powerful magnets when their operating frequency is increased and are currently limited to operation at about 500 GHz.

In this paper we investigate frequency conversion by means of third harmonic generation in solid state materials, in particular in Si, GaAs and InP. If a power conversion efficiency around 10% could be achieved, a powerful source in the range of 10 to 100 kW could be obtained from current day technology gyrotrons, by the conversion of their output radiation in an external frequency tripler.

A Monte Carlo simulation technique [1] is used to analyze the drift nonlinearity in technologically available materials. Comparison with experimental results in n-type Si has allowed us to demonstrate the usefulness of this method in explaining the 3^{rd} order nonlinearity in n-type Si for pumping intensities up to 2 MW/cm^2 at the fundamental frequency of 443 GHz [2]. However, Si is not the best possible choice with a power conversion efficiency not exceeding about 0.1%. GaAs or InP, especially when cooled down to liquid nitrogen temperatures, seem to be much more promising.

Theoretical considerations

In the range of powers well below the impact ionization threshold the main source of nonlinearity to be considered stems from the drift velocity of electrons affected by the non-parabolicity of the conduction band and by the energy-dependent scattering rate.

In order to achieve high powers, the harmonic radiation produced in a certain thickness of material is usually summed up. However, the result depends critically on the achievable phase-matching distance. Unfortunately the absorption of the fundamental beam in the case of drift nonlinearities is rather strong, so that usually only the first maximum of the Maker's oscillations [3] is observed. Therefore it is more appropriate to consider thin layers of materials only so as to maintain uniformity of the pumping field amplitude E_1 over the
layer thickness d. In this case the conversion efficiency is

$$\eta_3 = K_0 \left(\frac{2}{1+\sqrt{\varepsilon_L}} \right)^2 \left| \frac{(enV_3)d}{E_1} \right|^2 \tag{1}$$

where K_o is the material-independent constant, e_L is the lattice dielectric constant, n is the electron density, and V_3 is the 3rd harmonic drift velocity amplitude. The latter is obtained from the Monte Carlo simulation.

For the electron motion in n-type Si six ellipsoidal X-valleys are considered, taking into account the nonparabolicity. One type of acoustic phonons in the equipartition and elastic approximation was used to model intravalley scattering, whereas intervalley scattering is accounted for by six types of large-momentum phonons. The phonon parameters of Brunetti et al. [4] have been used and direct comparison of simulation results with experimental data of the third harmonic generation at room temperature has been very successful [2].

As far as the model is concerned in GaAs and InP, an improved efficiency is expected from the additional non-linearity due to the inelastic scattering of electrons on optic phonons and electron transfer between the G-, L- and X-valleys.

The band structure of InP is similar to GaAs with a somewhat higher energy separation of the valleys. The intervalley phonon energies and coupling constants are not much different from those in GaAs [5], but, all in all, we expect a competing 3^{rd} harmonic generation efficiency in InP.

We have selected a pumping field amplitude range of 10 to 60 kV/cm in order to avoid the exceedingly slow convergence of the Monte Carlo procedure at lower fields, and the increasing necessity to account for the full electron and phonon bands at higher fields.

Results

We have performed calculations for the three materials at lattice temperatures of 300 K and 80 K and compared them with the room temperature data on silicon where experimental results on the 3^{rd} harmonic generation already exist [2]. The drift velocity amplitude and phase show a slight dispersion (Fig.1). The 3^{rd} harmonic generation efficiency, which is determined by the square of the 3^{rd} harmonic drift velocity amplitude ratio to the pumping wave field amplitude (Eq.1), can be improved by cooling the crystal down to liquid nitrogen temperatures (Fig.2). At T = 80 K GaAs shows the highest efficiency which is 20 times higher than in Si at ambient temperatures provided that the pumping field amplitude is sufficiently low, whereas InP is seen to be superior in the high-amplitude range (Fig.2).

It is obvious that at exceedingly low field amplitudes the drift response should become linear causing the 3^{rd} harmonic generation efficiency to approach zero, whereas at high field amplitudes the heating of electrons, randomizing their momentum, should reduce the 3^{rd} order nonlinear susceptibility again. Hence a maximum efficiency is expected in between which is indeed clearly observed (Fig.2). Optimum conversion efficiency in n-type Si at the fundamental frequency of 333 GHz is around 20 kV/cm. InP shows an efficiency maximum at pumping field amplitudes slightly above 20 kV/cm, whereas for GaAs it is around 10 kV/cm. The maximum shifts to slightly higher amplitudes with rising temperature.

The maximum efficiency at low field amplitudes means that the pumping wave intensity should be rather moderate. A high 3^{rd} harmonic output power seems to be accessible by an expansion of the pump beam in such a way that the optimum field amplitude is not exceeded in the material. This is not difficult to achieve and does not result in exceedingly large material diameters due to the quadratic dependence of power density on linear beam diameter.

From the 1^{st} harmonic drift velocity amplitude (Fig. 1) one can judge that the absorption of pump radiation in GaAs and InP is stronger than in Si. This is the result of the free motion of electrons before they reach a threshold energy for inelastic scattering on phonons. For the heat transfer the low-temperature operation of thin non-linear material layers is essential.

Conclusions.

As far as high pumping field amplitudes are concerned, silicon, as readily available and simple to use material performs practically as well as GaAs or InP. However, if one aims to optimize the output power and hence expands the pump beam in order to operate at the point of maximum efficiency, GaAs seems to be superior, in particular at the temperature of liquid nitrogen. On the other hand, InP performs better at higher pumping field amplitudes.

With the latter two materials an overall conversion efficiency in the percent region seems feasible. In order to achieve this, the harmonic radiation has to traverse the pumped layer of material several times. The layer would have to be made thin enough so that phase mismatch remains insignificant during one pass. If a stack of thin plates is used, provision has to be made to readjust the summed-up mismatch between fundamental and harmonic per transit with some suitable dispersive material between the plates. On the other hand it may be possible to achieve the desired conversion efficiency in one single plate, by multipassing the harmonic radiation, using a resonator type structure. Phase adjustment then boils down to selecting the appropriate distances of the two resonator mirrors and the converter plate in-between.

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References

[1] LEBWOHL P.A., *J. Appl. Phys.*, **44** (1979) 1744.

[2] URBAN M., SIEGRIST M.R., ASADAUSKAS L., RAGUOTIS R. AND BRAZIS R., *Appl. Phys. Lett.*, **69** (1996), 1776.

[3] TERHUNE R.W., MAKER P.D. and SAVAGE C.M., *Phys. Rev. Lett.*, **8** (1962) 404.

[4] BRUNETTI R., JACOBONI C., NAVA T., REGGIANI L., BOSMAN G. AND ZIJLSTRA R.J.J., *J. Appl. Phys..*, **52** (1981), 6713.

[5] BRENNAN K. AND HESS K., *Solid State Electronics*, **27** (1984), 347.

Figure captions

Fig. 1 Electron drift velocity amplitude (a) and phase (b) as a function of the fundamental frequency: 1st harmonic - dashed lines, 3rd harmonic - solid lines. Materials and temperatures are shown in the figure. E_1 = 20 kV/cm.

Fig.2 3rd harmonic efficiency compared to the maximum efficiency in n-type Si at T = 300 K as a function of the pumping wave field amplitude. The fundamental frequency is 333 GHz. Materials and temperatures are shown in the figure.

Fig. 1 a

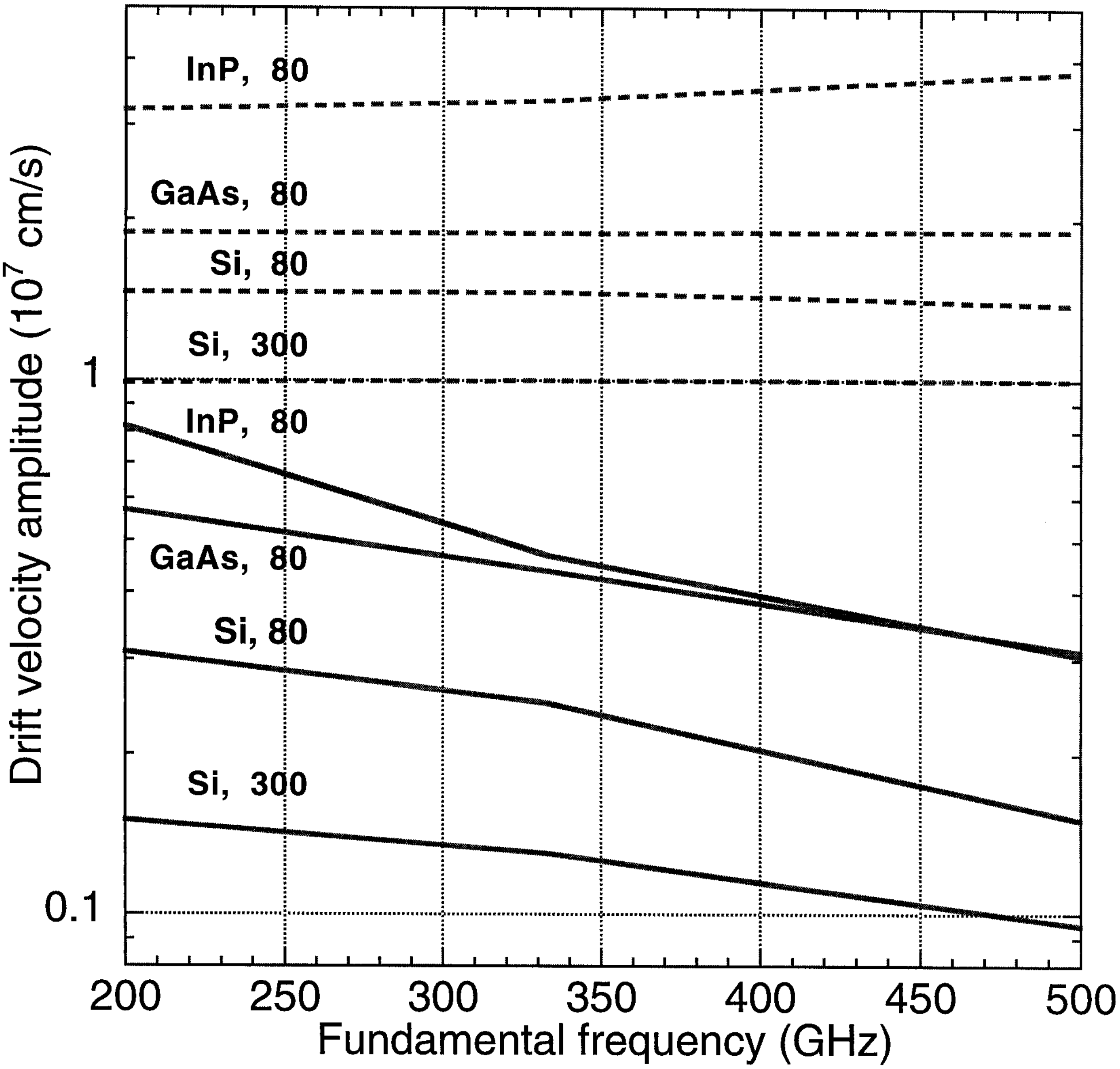
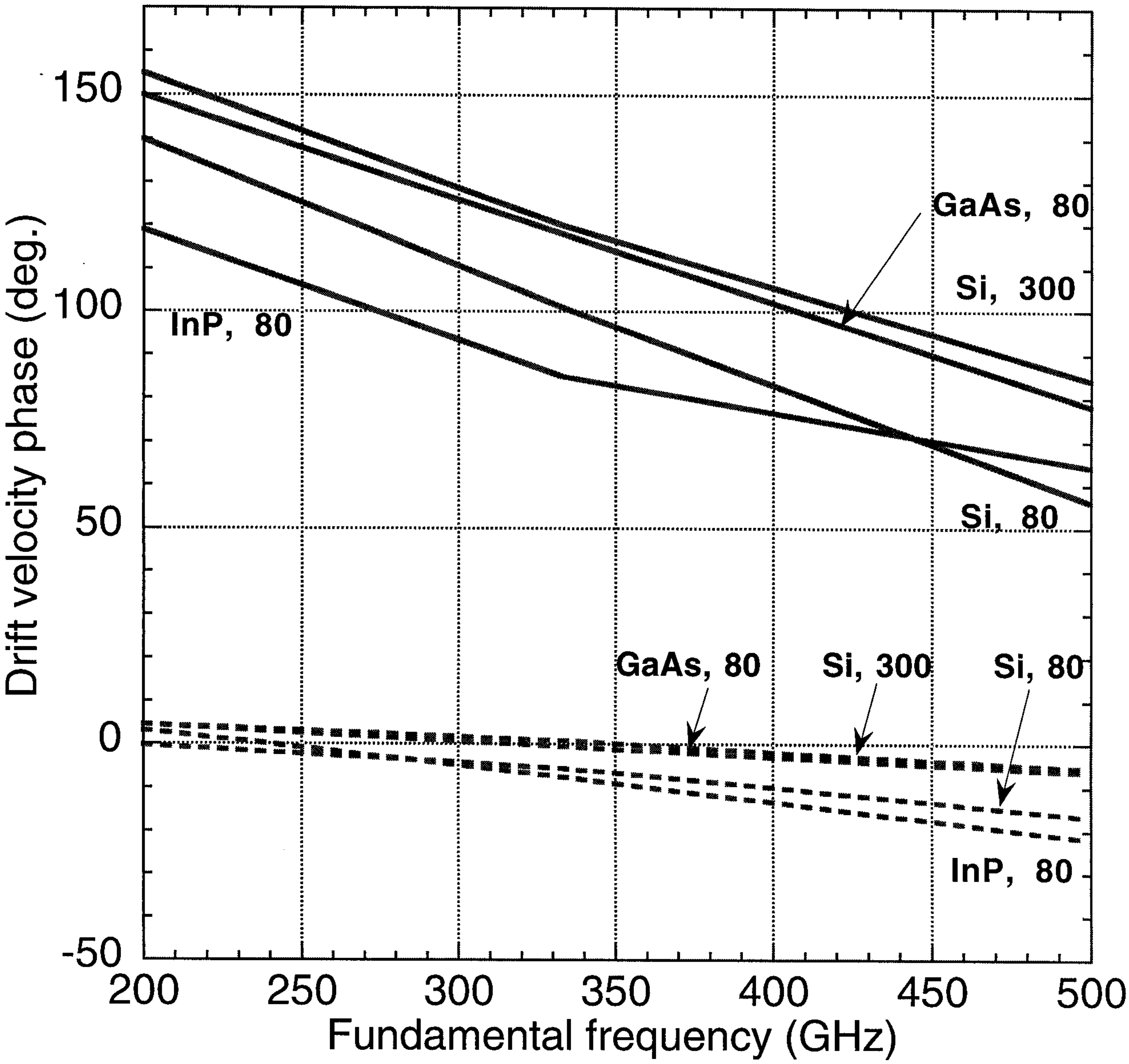
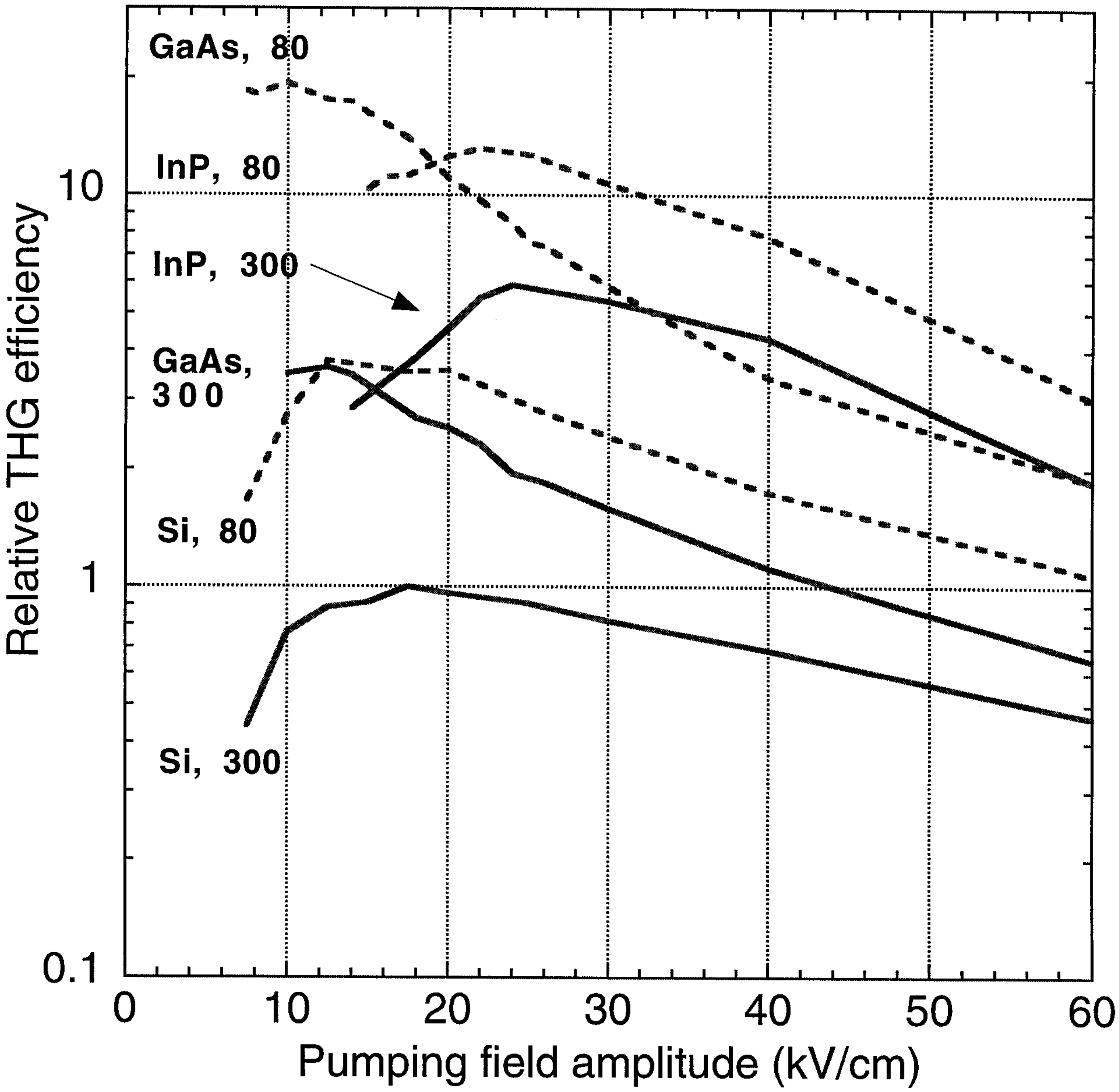

InP, 80
GaAs, 80
Si, 80
Si, 300
InP, 80
GaAs, 80
Si, 80
Si, 300
Drift velocity amplitude (10^7 cm/s)
1
0.1
200
250
300
350
400
450
500
Fundamental frequency (GHz)

Fig. 1 b



150
100
50
0
-50
200
250
300
350
400
450
500
Drift velocity phase (deg.)
Fundamental frequency (GHz)
GaAs, 80
Si, 300
InP, 80
Si, 80
GaAs, 80
Si, 300
Si, 80
InP, 80

Fig. 2


GaAs, 80
InP, 80
InP, 300
GaAs,
3 0 0
Si, 80
Si, 300
Relative THG efficiency
10
1
0.1
0
10
20
30
40
50
60
Pumping field amplitude (kV/cm)