3. Research on THz-wave applications using frequency-agile THz-wave source

3.1 Development of spectroscopic Stokes polarimeter by using tunable THz-wave source

(T. Notake, H. Minamide)

In THz frequency region, there are many functional group vibration modes of organic materials and vibrational circular dichroisms relating high order conformation of biological polymers such as proteins. Observation of such vibrational modes in THz frequency regions can serve new innovative knowledges which have never been obtained by conventional vibrational spectroscopy and circular dichroism measurements at from UV to IR regions.

We have been developing various coherent THz-wave sources, which possesses high peak power, narrow spectral bandwidth and ultra-wideband frequency tunability, by utilizing nonlinear optical effects. Since our THz-wave sources have high purity of polarization, they are suitable for application researches concerning polarization. Therefore, we have started to develop THz spectroscopic Stokes polarimeter covering frequency range of from 1 to 10 THz with DAST-DFG system. In particular, for THz-VCD spectroscopy, tiny absorption differences corresponding to right- and left- circular polarized incident THz-waves have to be detected. In order to do that, quarter waveplates which are capable of operating at wideband THz frequency region are required.

Quartz crystals have been predominantly used as wave plates in optical and low THz frequency regions below 3 THz. However, because quartz has poor transmittance at above 3 THz, we are developing special waveplates using artificial birefringence in silicon wafer. According to the effective medium theory, grating structure with periodical pitch less than wavelengths of the wave give rise to artificial birefringence with respect to TE- and TM-waves. By using numerical calculations, a number of grating structures, phase retardations and operating THz frequencies were iteratively investigated and several grating structures were actually fabricated on 300 µm thickness silicon wafers. Then, their frequency properties on phase retardations were experimental investigated by using DAST-DFG system. Lineary polarized THz-waves in horizontal direction from DAST-DFG were injected into the samples with 45 degree tilted grating angle with respect to THz-wave polarization. Rotation angles of wire grid polarizer installed in front of the detector were changed from -45 to 90 degrees with step of 45 degree and respective transmittance were measured. Fig.1 shows intensity ratios among them in a certain sample whose grating parameters are given in the figure. Value of 1 corresponds to just quarter waveplate behavior and it was obtained at around 4.8 THz in this case. This result is reasonable compared to design and broadband waveplates are currently designed. Fig. 2 shows an example of phase retardation in newly designed wideband quarter waveplate. Here, wideband operation is mainly realized by expanding volumetric ratio of high refractive index silicon part. After fabrication and experimental test of such wideband quarter waveplates, THz spectroscopic Stokes polarimeter will be constructed.
**Fig. 1** Performance of THz waveplate

**Fig. 2** Retardation of wideband THz quarter waveplate
3.2 Groundwork optical-damage analysis of organic nonlinear optical crystal for development of high-output terahertz-wave source (T. Matsukawa, K. Nawata, H. Minamide)

We have achieved coherent terahertz (THz) wave generation for organic and inorganic nonlinear optical (NLO) crystals by difference frequency generation. To build higher conversion efficiency THz-wave generation system, groundwork optical-damage analysis of NLO crystals in laser irradiation is required. However, the damage mechanism depended on the repetition frequency and pulse width has not been evaluated yet. In this study, we evaluate the dependence both a repetition frequency and a pulse width during optical damage using organic NLO crystal 4-dimethylamino-4'-methyl-4'-stilbazolium tosylate (DAST).

Platelet-shaped DAST single crystals grown by methanol solution. Laser beams with pulse width of 15 ns and 600 ps between 10 and 100 Hz were irradiated to the (001) plane of the DAST crystals. For monitoring the optical damage, the transmitted power through the DAST crystal was detected with a power meter during 90,000 pulses.

Figure 1 shows the optical damage-thresholds with various repetition frequencies of laser pulses (15 ns). The threshold of the DAST crystal was constantly to 40 Hz and drastically decreased over the repetition frequency of 50 Hz. The damage pattern was associated with melting on the crystal surface and burnt residue inside the crystal. In order to analyze the thermal effect by absorption of irradiated beam to the damage, we consider a thermal transient model with frequently irradiated laser beam. The absorbed thermal energy of irradiated laser \( E_T \) as damage threshold in the crystal is given by

\[
E_T = E_0 \frac{1}{e^{1/\tau_{rd}} - 1}
\]

where \( E_0 \) is thermal energy of a laser pulse coupled into the crystal, diffused with relaxation time \( \tau_r \) of DAST crystal, an additional energy with repetition frequency \( \omega \). In pulse duration above \( \tau_r \) during 10-40 Hz, DAST crystal was damaged by thermal energy of the one pulse. In pulse duration below \( \tau_r \) during 50-100 Hz, the short pulse duration may induce the thermal accumulation in the crystal rapidly. Figure 2 shows pulse width dependence of damage threshold. The input energy density at 10 and 50 Hz were decreased by \( \tau_{rd}^{1/2} \) dependence with decrease pulse width. In contrast, the damage threshold in 100 Hz was deviated from \( \tau_{rd}^{1/2} \) dependence. It means that DAST crystals might be not only damaged by thermal, but also by peak intensity. The peak intensity on 600-ps pulse was up to 13 times higher than that of 15-ns regime. The conversion efficiency for THz-wave generation using 600-ps laser will be expected to increase approximately 170 times higher than 15-ns laser.

Fig. 1 The damage threshold depended on repetition frequency. The data were normalized such that 1 is equal to 10 J/cm². (○): experimental data, (solid line): fitting using eq. 1

Fig. 2 Pulse width dependence of damage threshold: (●) 10 Hz, (◆) 50 Hz, (▲) 100 Hz.
3.4 The non-thermal effects of external THz wave irradiation on living cells (S. Hayashi, N. Yaekashiwa, K. Kawase)

- **Introduction**

  The effects of external electromagnetic fields on living cells are still not well understood. Strong electrically polar character of biological constituents makes possible longitudinal oscillations generating electric field. Some protein structures are excited by energy supply from metabolic sources. H. Fröhlich formulated hypothesis of coherent vibrations in biological systems as their fundamental biophysical property. Some vibration modes are excited far from thermal equilibrium but the majority of modes in the system remain close to thermal equilibrium. Coherent excitation of some polar vibration modes can generate endogenous electromagnetic field with dominant electrical component.

  The endogenous electromagnetic field seems to be important for biophysical mechanism of organization. External electromagnetic field can change some properties of living matter. There is no uncertainty concerning the effects of the electromagnetic fields producing thermal effects. The main question of the effects of electromagnetic fields on biological systems concerns the exposure to the fields not producing thermal effects. The understanding of the mechanisms of non-thermal effects is far from comprehensive.

- **Terahertz vibration of a biomembrane ~ Fröhlich model ~**

  Molecular evolution has led to the development of a number of properties that cannot be predicted from theoretical considerations alone, but require intimate collaboration with experiment. Theory can and must provide, however, the general concepts in terms of which experimental evidence should be discussed; activities in particular cannot be discussed in terms of structure alone, except in the realm of linear response, which is unlikely to hold in the case of biological activity.

  High electric fields in membranes (10^5 V/cm or more), abundance of fixed and mobile ions (yielding 10^5 V/cm at a distance of 10^-6 cm from a unit charge), suggests strong electric polarization in many molecules. Such polarization will deform the molecule and lead to a nonlinear response invariably connected with a decrease in electrostatic energy.

  When applied to a finite system like a polymer, the result of calculation implies the existence of a metastable state with high dipole moment. In a sufficiently strong external field, the excited state will be depressed below the ground state, but even smaller fields will lead to increased thermal excitation of the polar state. The results are based on a coupling of polar with elastic modes of the system, and they result in its deformation when the metastable state is excited. In actual cases such deformations might imply larger conformational changes which cannot be treated in such general fashion. One further general consequence does arise, however, for the change in dipole moment upon excitation will frequently lead to rearrangements of counter ions and hence to further stabilization of the metastable state. This state may then be expected to have a very long lifetime.

  The high dipole moment of the metastable state implies in general its excitation to be coupled with excitation of homogeneous electric vibrations, i.e., giant dipole oscillations. It has been demonstrated theoretically on a simple model that random supply of energy to various polar modes coupled nonlinearly can lead to a strong (coherent) excitation of a single mode provided the energy supply $S$ exceeds a critical $S_0$, $S > S_0$. When the thickness of membranes and acoustic velocity in the cell are 5 nm and 3000 m/s respectively, the resonance frequency of around 300 GHz have been estimated.

  An essential feature of this model is its coupling with the rest of the material treated as a heat bath in thermal equilibrium at a temperature $T$ which it attempts to impose on the polar modes. When the energy supply to those modes exceeds the threshold, then the most favorable distribution is the one in which all modes but one are nearly in thermal equilibrium while the one mode becomes highly excited. Far-reaching biological consequences may be expected from such excitation.

![Fig. 1 Cell membrane.](image)
**Experimental setup and results**

We investigated the non-thermal effects of THz/MMW radiation on human dermal fibroblasts by measuring the reactance at an irradiating power of less than $1 \mu W/cm^2$. The activity levels of cells set in the 12 wells were measured using AC impedance using the HIOKI Chemical Impedance Meter 3532-80. The diameter of each well was 10 mm. The bottom of the well was made of an indium tin oxide (ITO) electrical pole. The irradiating source was a widely tunable uni travelling carrier photodiode (UTC-PD) that covers from 90 to 1000 GHz. The output power was less than 1 μW. Therefore, the irradiated power density on the cell was below 10 nW/cm² considering the absorption loss of millimeter wave in the ITO well. Fig. 2 shows the view of experimental setup.

An example of the measurements is shown in Fig. 3. The horizontal axis shows the elapsed time in minutes, and the vertical axis shows the reactance ratio, which is proportional to the area of cells adhering to the ITO glass at the bottom of the well. The UTC-PD placed just below the B1 well. During the irradiating period as indicated by red area, the frequency was tuned from 220 to 300 GHz. The activity (i.e. the reactance ratio) of B1 well represented by red solid line increased compared with other wells. However, we need further experiments as the reproducibility is still poor.

We also investigated the non-thermal effects of THz/MMW radiation on the same sample by the MTT assay. The MTT assay observes the mitochondrial dehydrogenase in the cells. Assays are performed by adding a small amount of reagent directly to culture wells, incubating for 3 hours and then recording the absorbance at 490 nm with a 96-well plate reader. The quantity of reduced product as measured by the absorbance is directly proportional to the cell proliferation and enzyme activity. We compared two groups; with or without THz/MMW radiation. As shown in Fig. 4, the absorbance increased with the cells which were radiated with MMW frequency of 100 - 300 GHz.

![Fig. 2 The view of experimental setup.](image)

![Fig. 3 Possible non-thermal effect of terahertz wave on human dermal fibroblast cells measured using a reactance meter.](image)

![Fig. 4 Results of MTT assay. Control contains only MEM medium.](image)