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Heat and density induced optical nonlinearities of Chinese tea extract

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This article is dedicated to Prof T Asakura

It has been reported that a Chinese tea dissolved in alcohol can produce very strong phase modulation. In this research work, we propose a self-align phase filter utilizing the phase modulation capability of Chinese tea. We have analyzed the spatial resolution characteristics in order to investigate the origin behind the optical nonlinearity of Chinese tea. A theoretical model has been presented based on the effects of thermal and density gradients. Our experimental results show clear evidence for the existence of density variations apart from the thermal variations to be the reason for the nonlinear effects observed in Chinise tea.

Keywords: Chinese tea, Optical nonlinearity, Phase modulation, Phase filter, Density gradients, Thermal gradients

1 Introduction

The nonlinear behavior of Chinese tea was first reported by Zhang *et al* [l] in 1989. They observed self-focusing, self-defocusing, self-trapping and self-phase modulation of a laser beam passing through a Chinese tea cell. Such observations have also been made by number of workers [2-5]. It has been suggested that the basic mechanisms behind the self-focusing phenomenon in tea may be laser-induced heating and formation of molecular aggregates in other words density variations. It has also been suggested that under high power pulsed laser illumination (< 70 psec), molecular reorientation plays a major role [3] and a practical application as a optical limiter has been proposed [6].

One of the purposes of our present study is to utilize this strong nonlinear property of Chinese tea in making a phase modulator controllable by light. Such a modulator finds application in optical metrology, image processing etc. Currently, liquid crystal cells are getting attention and they are commonly used due to their availability. Kadono *et al*. studied a common path phase shifting interferometer using a liquid crystal phase filter to realize high precision [7]. The main disadvantage of such a system is the difficulty involved in the alignment of the system. In this study, a tea cell has been employed instead of a liquid crystal cell. In order to realize a phase filter that has self-align capability and at the time faster response characteristics utilizing the optical nonlinear property of Chinese tea, it is necessary to understand the mechanism behind the origin of the nonlinearity of Chinese tea. Secondly, the active material responsible for such a function must be isolated. In this investigation, one of the possible materials present in green tea dissolved in alcohol chlorophyll has been considered.

Here, to study the origin, dynamic gratings have been formed inside tea and the spatial resolution has been analyzed. If the origin is different from thermal, the resolution limit will be greatly different. As chlorophyll is one of the materials present in tea when dissolved in alcohol, its nonlinear properties have been investigated.

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In section 2, the characteristics of an adaptive phase filter based on the self focusing properties of Chinese tea has been investigated. Section 3 discusses about the experimental and the theoretical investigation of the origin of nonlinearity. Finally, section 4 gives a conclusion and scope for further research.

2 Experimental

2.1 Tea as a spatial modulator

Here, we propose a self-align phase modulator using the self-focusing properties of Chinese tea. The main advantage of such a phase modulator is that it is optically addressable and the severe requirement on the exact alignment on the optical axis is greatly relaxed. In this section, the basic properties of the modulator are investigated with respect to its application in interferometry and image processing.

2.2 Preparation of tea plates

The Chinese tea leaves were first boiled in alcohol and then put through a centrifuge to remove solid debris. After filtering through an ultrafine filter (0.8 μm), the tea solution was evaporated to obtain tea extract. A known quantity of tea extract was dissolved in a known quantity of alcohol to get the required concentration. This solution was next sandwiched between two glass plates separated by a spacer element. Different kinds of plastic films with thickness ranging from 10 to 100 μm were used as spacer elements.

2.3 Demonstration of phase modulation ability

In order to investigate the phase modulation characteristics, a double diffraction optical system was employed. Figure 1 shows a schematic diagram of the optical imaging system employed. A He-Ne laser of wavelength l = 0.6328 μm was used as a light source. The laser light was expanded and collimated to illuminate an object by means of lenses L1 and L2. Lenses L3 and L4 of the same focal length f = 200mm form a double diffraction imaging system, and the object was imaged onto the observation plane. The Chinese tea plate was placed in the Fourier transform plane of the object, i.e., in the plane behind the lens at a distance f to examine the phase modulation effect of the Chinese tea. The images of the object were taken by a CCD camera placed in the observation plane. Total power of the incident light on the Chinese tea plate was controlled by the use of both a variable aperture arranged just in front of the object and a variable attenuator placed behind the laser. As an amplitude object, patterns of characters were recorded on photographic films. Phase type objects were also prepared by bleaching the amplitude objects.

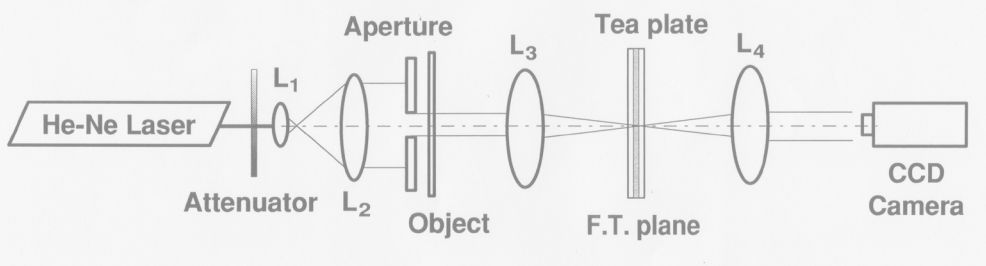

Fig 1. Optical system to demonstrate the phase modulation capability of tea

Figure 2 shows the observed images for an amplitude object of character pattern as the incident power on the tea plate is changed by means of the aperture in front of the object. Figure 2(a) is the normal image of the object observed at an incident power Pi = 0.39mW, there was no change in the image below this power. As the diameter of the aperture is increased, however, the contrast of the image becomes weak, and the image almost disappears at Pi = l.5mW as shown in Fig 2(b). For further increase of the incident power, the image appears again and the maximum contrast is attained at Pi = 2.8mW as shown in Fig 2(c). However, a noticeable feature is that the image appears with the reversed contrast compared with that in Fig 2(a), i.e., the

bright part in Fig 2(a) becomes dark and the dark part in Fig 2(a) becomes bright. By increasing the incident power furthermore, the image disappears again and appears with the normal contrast as shown in Fig 2(d) at the incident power Pi = 5.0 mW.

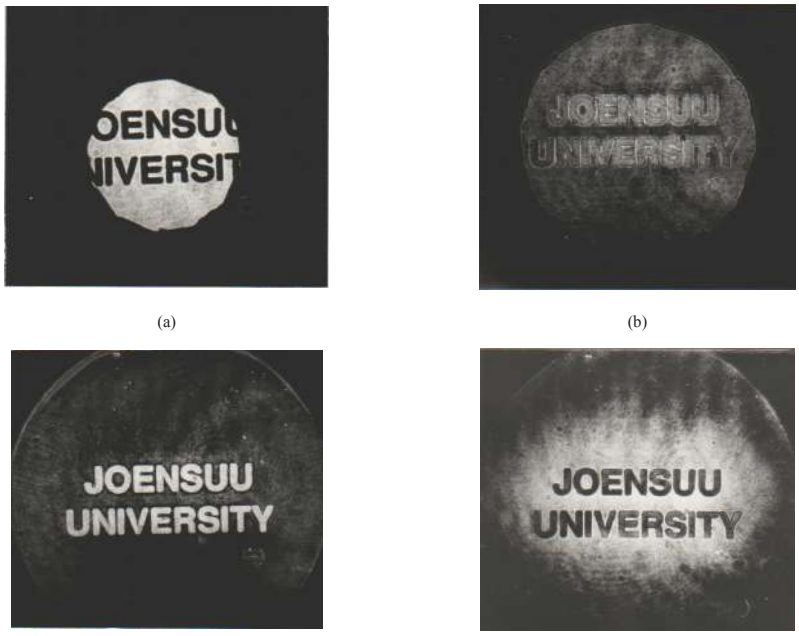

Fig 2. Observed images for incident powers of Pi (a) 0.39mW, (b) l.5mW, (c) 2.8mW and (d) 5 mW

The interpretation of this interesting phenomenon can be possible in the same manner as of Zernike's phase contrast method. In Zernike's method, a phase retarder of $\pi/2$ is placed in the Fourier transform plane. Therefore, only the phase of the specular component of the object is changed and this results in the conversion between the phase and the amplitude distributions of the image. In a similar way, regarding the fact that the object used in the experiment is a weakly diffusing object and hence yields a very strong specular component in comparison with the diffused component, it can be considered that only the strong specular component induces refractive index changes in the tea plate kept at the Fourier transform plane so that a phase retarder is formed on the optical axis. The amount of phase modulation due to induced refractive index changes can be controlled by the total incident power. Therefore, the phase difference between the specular and the diffused components can be considered to be $\pi/2$, 2π and $3\ \pi/2$ rad for Fig 2(b), (c) and (d), respectively.

2.4 Measurement of phase modulation characteristics

Next, the phase modulation characteristics were measured as a function of the absorbed power by using an interferometer. The same optical system as in Fig l was used for the interferometric measurement except that a phase grating was placed at the object plane. Only the 0^{th} and $+1^{st}$ order spectra of the diffracted light from the grating were selected at the Fourier transform plane by a slit type mask placed just in front of the tea cell. The interference fringes are formed as a superposition of these two diffracted lights at the observation plane. Depending on the power absorbed by the tea cell, the relative phase retardation felt by the 0^{th} beam on passing through the cell will introduce a shifting of the interference fringes. The corresponding phase retardation is given by

$$\Delta\theta = \Delta\theta_0(P_0) - \Delta\theta_1(P_1) \tag{1}$$

where P_0 and P_1 represent the incident powers of the 0^{th} and the 1^{st} diffracted orders, respectively. $\Delta\theta_0$ and $\Delta\theta_1$ are the phase modulations due to induced refractive index changes, respectively, for the 0^{th} and $+1^{st}$ order diffracted lights. The power ratio between those two diffracted lights entering the plate was 1/50 . Therefore, the phase modulation due to the weaker incident light, i.e., $+1^{st}$ order diffracted light, was negligibly small. Since the interferometer forms a common path arrangement, the measurements were performed with quite high stability. An attenuator was inserted for the 0^{th} order diffracted light just behind the tea plate in order to increase the visibility of the interference fringes.

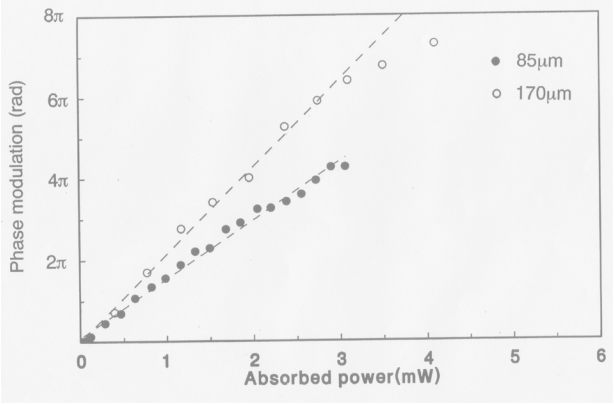

Fig 3 Dependence of phase modulation on the absorbed power.

The phase modulations obtained for the tea cells are shown in Fig 3 as a function of the absorbed power of 0^{th} order diffraction light. In Fig 3, the filled circles and the open circles represent the values obtained for the tea plates of thickness 85 μm and 170 μm, respectively, and dashed lines are drawn to fit those values. It can be seen that the phase modulation increases linearly with the absorbed power and a phase modulation of 2π is attained at an absorbed power of nearly 1.5mW and 1mW for the above samples.

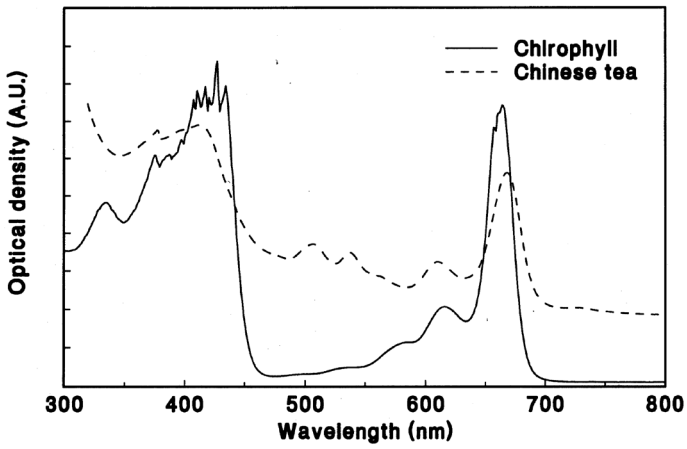

Fig 4. Absorption spectra of Chinese tea and chlorophyll.

Figure 4 shows the phase modulation as a function of absorbed power for two different wave lengths of 633 nm and 514 nm and tea plates of thickness 85 μm. It can be seen that the phase modulation obtained with red light (633nm) is slightly stronger than that for green light (514nm) under the condition of absorbed

power being the same. If the origin of the phase modulation effect is from thermal, then, in the actual case, the phase modulation with green light is expected to be 20% larger because of the shorter wavelength for the green light compared to the red light. However, the experimental results show the phase modulation to be larger for red light. This kind of dependence of phase modulation on the incident wavelength suggests the existence of some kind of resonant mechanism. Moreover, it has been found that the absorption spectrum of tea as shown in Fig 4 has a sharp peak in the red region 670 nm. Figure 5 shows the absorption spectra of chlorophyll and green tea.

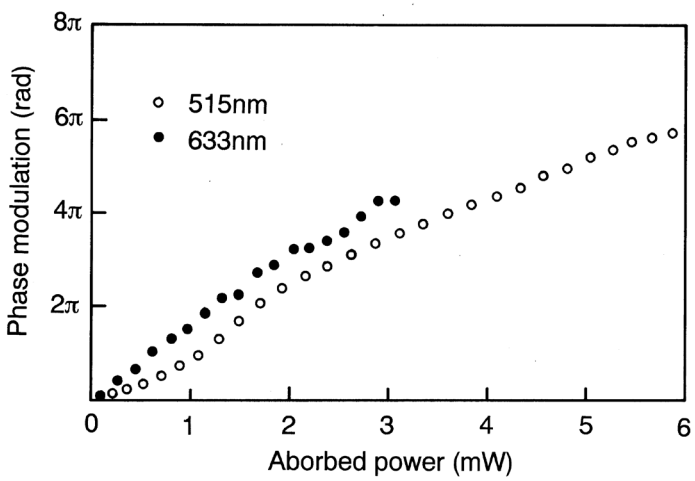

Fig 5. Dependence of phase modulation for different illuminating wavelengths

2.5 Measurement of response time

Next, response time of the tea plate was examined with the experimental system shown in Fig 1. A point detector of photodiode was placed at a certain point of the character pattern, and an electric shutter was arranged behind the laser. The incident power was set to yield the reversed image as shown in Fig 2(c), and the time to reach 90% of the maximum of the intensity after opening of the shutter was measured by the photodiode as the response time. The results are tabulated in Table 1. The rough tendency of the response can be seen from the Table 1 that faster responses are attained for narrower gap widths of the tea plate, and the minimum response time is 7.8 msec for the sample No. 1. It is remarkable that the response times are comparable or even faster than those of the nematic liquid crystal phase modulators for its considering the large phase modulation that could be obtained with tea cell. On the other hand, response times of nematic liquid crystals are usually in the order of tens of hundreds of milliseconds.

Table 1. Sample no. of the Chinese tea cells with their gap widths, transmittance and the corresponding response times to yield a reverse image.

Sample No.	Gap width (μm)	Transmittance (%)	Response time (msec)
1	40	56	7.8
2	85	42	9
3	85	48	9
4	85	79	13
5	170	70	30
7	255	57	42

3 Origin of Non-linearity

In order to investigate the origin behind the large phase modulation capabilities of Chinese tea, a dynamic grating was formed by two strong pump beams in the tea cell and simultaneously read by a probe beam. By investigating the spatial frequency characteristics of such a system, the maximum resolvable spatial frequency can be deduced. By means of a comparison with a theory based on thermal effects [8,9], it may be possible to arrive at some information regarding the origin of the phenomenon.

3.1 Theory

In the theoretical model used for investigation, it was assumed that the tea molecules first absorb light and then convert the absorbed energy to heat. This heat released is conducted to the surrounding by the alcohol which results in a temperature gradient. This in turn produces a refractive index gradient resulting in phase retardation of the beam passing through the cell. The change in temperature can be calculated as a solution of the heat conductivity equation given by,

$$c\rho \frac{\partial \Delta T(x,t)}{\partial t} = k \frac{\partial^2 \Delta T(x,t)}{\partial x^2} + q(x), \tag{2}$$

$$\Delta T\,(x,0) = 0 \tag{3}$$

where $\Delta T(x,t)$ is the temperature change, c is specific heat coefficient, ρ is density notation used in the model, k is thermal conductivity and $q(x)$ is the energy absorbed per unit length.

When the source function takes the form of a cosinusoidal intensity distribution given by,

$$q(x) = I_{abs}[1 + \cos(2\pi v_g x)], \tag{4}$$

with

$$I_{abs} = \frac{\boldsymbol{P}_{inc}[1-\exp(-\alpha L)]}{\pi w^2 L} \tag{5}$$

where $\boldsymbol{P}_{inc}$ is the incident power, α is the absorption coefficient w is the beam radius, L is thickness of the tea cell and v_g is the spatial frequency of the grating.

A unique solution satisfying the boundary conditions can be obtained as,

$$\Delta T(x,t) = \int_0^\infty Q(v) \frac{1-\exp(-4\pi^2 v^2 kt)}{4\pi^2 v^2 k} \exp(-i2\pi vx) dv \tag{6}$$

where $Q(v)$ is the Fourier transform of source function, v is the spatial frequency and k is the thermal diffusivity. Here, as it can be seen, the temperature change is obtained as a convolution of the point spread of heat function and the source function.

For a source function given by Eqs (4) and (5), the temperature change is given by,

$$\Delta T(x,t) = I_{abs}\left[\frac{t}{c\rho} + \frac{1}{2} \cdot \frac{1-\exp(-4\pi^2 v_g^2 \kappa t)}{4\pi^2 v_g^2 k} \cos(2\pi v_g x)\right] \tag{7}$$

and refractive index change is

$$\Delta n(x,t) = n_0 - \frac{dn}{dT} \Delta T(x,t) \tag{8}$$

where n is the refractive index of the medium and dn/dT is the thermo-optic coefficient. Thermally induced phase change is

$$\phi(x,t) = \frac{2\pi}{\lambda} \Delta n(x,t) L \tag{9}$$

with λ being the wavelength of the probe beam. Finally, the diffraction efficiency for the m^{th} order can be obtained as

$$\eta_m = J_m^2(\beta), \tag{10}$$

where

$$\beta = \frac{2\pi}{\lambda} L\frac{dn}{dT} \cdot \frac{I_{abs}}{2} \cdot \frac{1-\exp(-4\pi^2 v_g^2 \kappa t)}{4\pi^2 v_g^2 k} \tag{11}$$

Therefore, under the steady-state condition and β being small and m takes a value of 1, then

$$\eta_1 = J_1^2(\beta) = \beta^2/4 = 1/v^4, \tag{12}$$

The response time (τ_t), i.e., the time taken for η to reach 90% of its maximum value is given by

$$\tau_r = 1/v^2\kappa \tag{13}$$

Based on the theory developed, the diffraction efficiency should vary as a function of the inverse of the fourth power of spatial frequency and response time should vary as the inverse of the second power of the spatial frequency.

3.2 Experiments

Next, experiments have been performed to investigate the origin of the Chinese tea. A dynamic grating was formed inside a tea cell and the diffraction efficiency and the response and decay times of the grating were measured. A schematic diagram of the experimental system is shown in Fig 6. The light from Ar^+ laser after passing through a Michelson type interferometer interfere to produce interference fringes at the tea plate plane. This cosinusoidal intensity distribution produces a corresponding refractive index variation and hence a phase grating. Upon illuminating this grating by light from a low powered He-Ne laser, the diffracted light can be observed on a screen placed behind the sample cell. Figure 7 shows the diffraction patterns obtained under low and high powers of the pump beam with a sample cell of thickness 35 μm and made of green tea. Here, the diffraction efficiency (η) of the phase grating formed inside the tea cell is defined as the ratio of the power in the first order of the diffracted beam to the total transmitted power of the probe beam. The diffraction efficiency was measured for various cells made of tea, chlorophyll, ink and various dyes with values of thickness 35μm and 80μm. As mentioned earlier, one of the possible molecules present in tea is chlorophyll as it is soluble in alcohol and also the absorption and the fluorescence spectra of tea and chlorophyll are very much similar as compared in Fig 5.

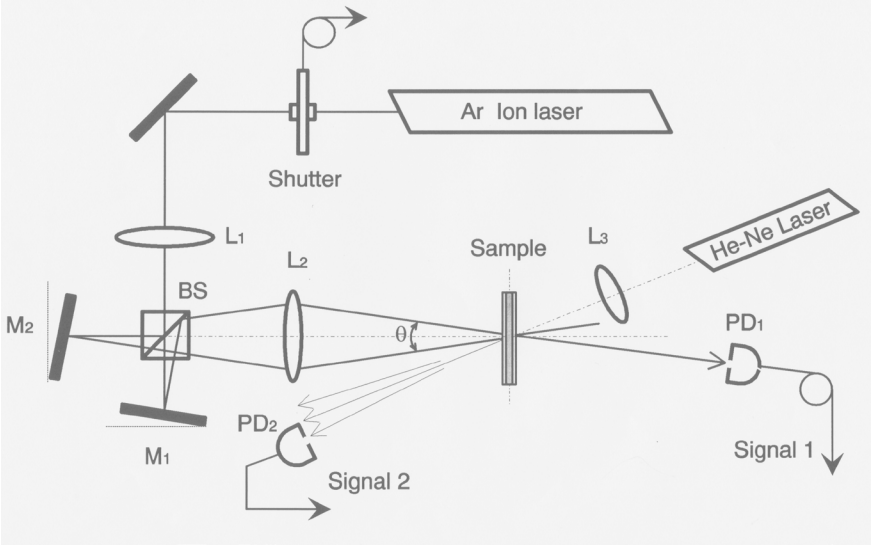

Fig 6. Experimental system to measure the response time.

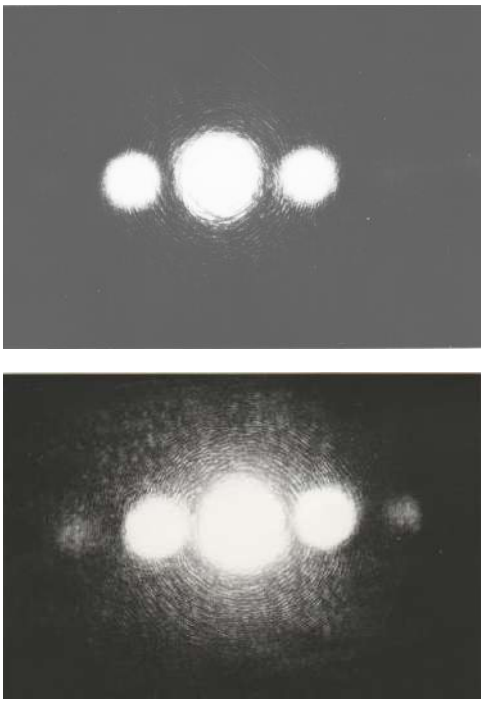

Fig 7. Diffraction patterns due to the read beam of He-Ne laser for low(top) and high (bottom) power of the pump beam.

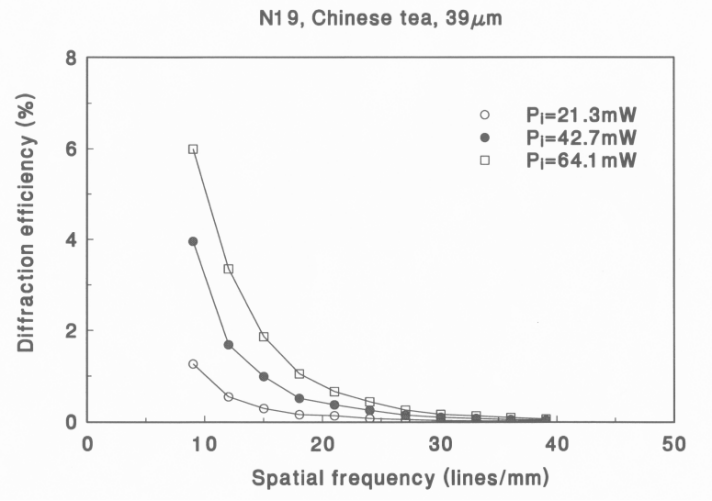

Fig 8. Variation of diffraction efficiency as a function of spatial frequency of the grating for a tea cell of thickness 39μm.

Figure 8 shows a typical example of the variation of the diffraction efficiency as a function of the spatial frequency of the grating for different values of the absorbed power for a tea cell of thickness 39μm. In the experimental system the spatial frequency of the grating can be simply varied by changing the tilt of one of the mirrors in the Michelson type interferometer. It can be seen that for all the cases of absorbed powers, the diffraction efficiency decreases rapidly with increasing spatial frequency. The maximum diffraction

efficiency obtained is around 20% with a cell of 55μm thickness. Figure 9 shows a comparison of the results obtained from tea, chlorophyll and other materials. It should be noted here that in all these measurements the amount of absorbed power is made the same. It can be seen from the figure that the results are almost the same for chlorophyll and tea while for Olong tea (a slightly different type of Chinese tea which is of brown colour) due to a larger value of the thickness, the absolute value of diffraction efficiency is comparatively large. However, as it can be seen, for the same power, the diffraction efficiency due to ink is very much low. This leaves behind a question regarding the origin of the phenomenon. In case, the origin is thermal and hence due to alcohol, then under the same absorbed power, the magnitude of the diffraction efficiency could be expected to be almost the same. Hence the theoretical model based on heat conduction presented in section 3.1 would give results different from that of experiment. Figure 10 shows the experimental variation of the diffraction efficiency as a function of the spatial frequency in a log-log scale and the slope is –3.2 which is quite different from the theoretical value of –4. The experimental slope varies between –3 to –5 and hence showing a large discrepancy.

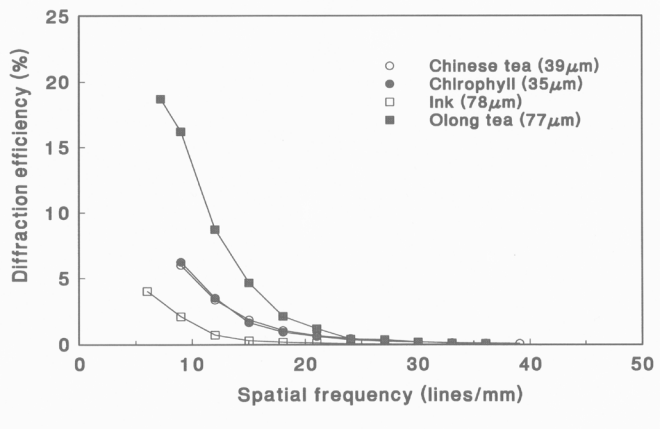

Fig 9. Comparison of diffraction efficiencies of various materials.

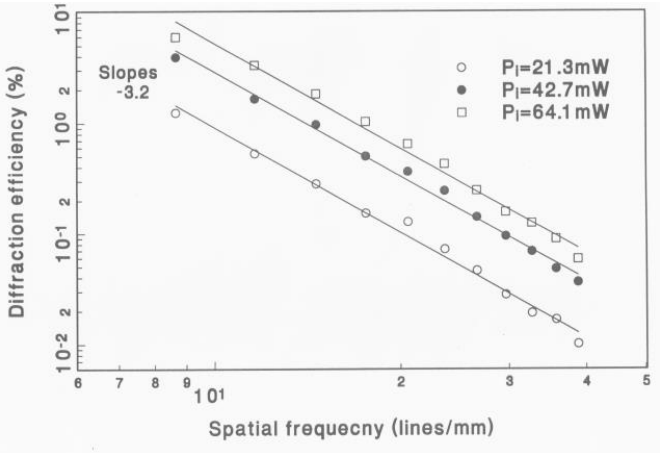

Fig 10. Log-log plot of diffraction efficiency versus spatial frequency for the same sample as of Fig 8.

Next, regarding the origin, we have got strong evidence supporting the existence of density gradients in a tea cell on illuminating with strong pump beams. This is due to the fact that the diffraction pattern

remains faintly for a few seconds (10-15 sec depending upon the duration of illumination) even after shutting off the pump beam. It should be pointed out that our green tea liquids are of high concentration in comparison to that of earlier reports [1-6] and the cells used are relatively very thin. This can be one of the strong reasons for observation of density changes apart from the heat-induced changes of refractive index. Now, due to the obvious presence of density effects, our theory needs modifications to take into account this density changes which will be discussed in the following section.

3.3 Discussion

There exist several possibilities which contradict the simple theory discussed in section 3.1. One is the thickness effect which may become strong when the cells are thin. So taking into account of this, and modifying the theory, Eq (6) will become

$$\Delta T(x,t) = L\int_0^\infty Q(v)\,\frac{1-\exp(-4\pi^2[v^2+(\pi/L)^2]\kappa t)}{4\pi^2[v^2+(\pi/L)^2]\kappa}\,\exp(-i2\pi vx)dv \tag{14}$$

After performing asymptotic approximations on L, it has been found that the diffraction efficiency should vary as a polynomial of degree 4, however, it has been found that the results are very much different from the expected.

We have considered the possibility of density variations arising from temperature variations. For such a case, it becomes necessary to solve a set of coupled equations given by

$$\frac{\partial\Delta T}{\partial t}-\frac{\kappa_T}{C}\left(\frac{\partial\mu}{\partial\rho}\right)\frac{\partial\Delta\rho}{\partial t}=\kappa_{th}^2\frac{\partial^2\Delta T}{\partial x^2}+q(x) \tag{15}$$

$$\frac{\partial\Delta\rho}{\partial t}=\kappa_m^2\frac{\partial^2\Delta T}{\partial x^2}+(\kappa_T/T_0)^2\,\Delta T \tag{16}$$

where $\Delta\rho$ is the change in concentration κ_T and κ_m are the thermal and mass diffusion constants, κ_{th} is the Sorel constant, $(\partial\mu/\partial\rho)$ is the chemical potential. By solving the set of coupled equations, the change of temperature can be obtained as,

$$\Delta T(x,t)=\int_0^\infty \frac{Q(v)}{C}\cdot\frac{1-\exp(\gamma_m\kappa_m+4\pi^2v^2\kappa_m)t]}{\gamma_m+4\pi^2v^2(\gamma_m\kappa_m+\kappa_m)}\,\exp(-i2\pi vx)dv \tag{17}$$

where

$$\gamma_m=\frac{\kappa_T^2}{T_0C}\left(\frac{\partial\mu}{\partial\rho}\right) \tag{18}$$

In this case also, the temperature change given by Eq (17) takes the same form as Eq (6) and hence will lead to same kind of results. Next, when the chemical potential arising from a mixture of large mass differences is negligibly small, there will be only thermal gradients and hence Eq (l8) will reduce to Eq (6).

4 Conclusion

In this paper, based on the phase modulating capability of Chinese tea, we proposed a self-align phase filter. The phase modulation was found to vary linearly with the absorbed power and a modulation of 2p can be obtained with an absorbed power of 1.5mW for a cell of thickness 85 μm and a transmittance of 48%. It was also found that the response time is of the order of a few milliseconds and this is relatively faster compared to nematic liquid crystal, a probable candidate for its use as a phase modulator.

From an experimental investigation of the spatial resolution characteristics of tea, it was found that the origin of nonlinearity comes from a combination of different mechanisms including thermal and density changes. Some clear evidence was obtained for the existence of density gradients or in other words

for the formation of aggregates. The resolution limit was found to be around 10μm. Though, our comparison of results with that of chlorophyll shows some agreement in the magnitude of the induced changes, there exists differences due to the presence of different varieties of tea in green tea itself. Each of these tea types is different on the type of processing they undergo and we do not know what is the reason behind such large differences in the optical nonlinearities of the different tea types [10,11]. Finally, it is worth to mention that our experiments on phase modulation with a tea cell in an electric field of 5×10^6V/cm failed to produce any change in the phase modulation.

Acknowledgement

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From 2008 to 2011, he worked as Research Director at Center for Environmental Science in Saitama and moved back to Department of Environmental Science and Technology, Graduate School of Science and Engineering, Saitama University in 2011. His research interest includes statistics of laser speckle field and its application in metrology, interferometry, speckle interferometry, optical coherence tomography etc. Recent research interest also includes applications of the optical techniques on the environmental sensing. High precision interferometer based on the statistics of a fully developed speckle was developed in his lab. and applied to investigate ultra-short term growth dynamics of plant in a time scale of second under the environmental stresses aiming to develop a novel environmental sensing and assessment techniques.

Uma Maheswari Rajagopalan

Uma Maheswari Rajagopalan is from India and now a permanent resident of Japan. From 2018 April onwards, she is working as a Professor in SIT research laboratories, Shibaura Institute of Technology. After receiving her Master's in Physics from the American College, Madurai in 1985 and Master's in Applied Optics from the Indian Institute of Technology, New Delhi, India in 1987, she got her Ph D in Engineering from Hokkaido University in 1991.

She also holds a master's degree in Education with specialization in Applied Linguistics that she obtained from Open University, the United Kingdom in 2014. She worked as a postdoctoral visiting researcher from 1992 to 1993 in the Department of Physics, Joensuu University (currently called as University of Eastern Finland).

From 1993 to 1998, she worked as a postdoctoral researcher in 'Ohtsu Photon control project' of Kanagawa Academy of Science and Technology, Kawasaki and worked on the development of ultra high-resolution optical microscope. From 1998 to 2005, She was a research scientist in Brain Science Institute (BSI), RIKEN, Japan and a research consultant from 2005 to 2013 in the same institute. During her time in BSI, she proposed and implemented Optical Coherence Tomography for live imaging of brain functions and succeeded for the first time in the world. From 2012 to 2017, she worked as Science and Technology adjunct lecturer at the national University of Electro-communications, Tokyo. From April 2014 to till March 2018, she worked as an assistant professor in the Department of Food and Nutrition studies at Toyo University. Her research interests include application of optical technology to biology and cognitive neuroscience apart from genre-based pedagogical approaches to teaching subjects of science and technology to non-native students. She is currently doing research on the application of laser speckle based measurement technologies to the determination of shelf life of fruits, environmental stress on plant growth, probe microscope for studying plant wall dynamics and wearable brain imaging technologies in attention and memory studies.