CHAPTER - IV

SECOND HARMONIC GENERATION
AND
TANDEL EFFECT

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4.1 Introduction:

Number of materials in which ferroelectric behaviour has been studied, and the variety of phenomena were observed. Ferroelectrics show strong anomalies in dielectric constant and can be used to build special devices. The only practical application of ferroelectricity which was presented was on TANDEL. This device is based on the fact that a ferroelectric crystal under certain conditions is self stabilizing with respect to temperature. Second Harmonic Generation (SHG) has been previously studied in various ferroelectrics. In $BaTiO_3$ by R.C.Miller (1964), in KDP by Van Der Ziel (1964), in TGS by Smith (1964), Surorov V.S. and Sonin A.S.(1968). The dielectric nonlinearity of ferroelectric triglycine sulfate (TGS) was studied in a circuit of a ferroelectric frequency multiplier in the radio frequency range by A.M.Vincent (1951). Glanc et al (1963) have reported the use of triglycine sulfate as a thermoautostabilized nonlinear dielectric element.(TENDEL).

In the magnitude of the a.c. signal applied to the ferroelectric is increased, the dielectric heating raises the temperature of the material to near Curie temperature. In the neighbourhood of Curie temperature the losses begin to fall and as a result the material stabilizes its own temperature.

It stabilized at a crystal temperature near curie temperature where the nonlinearity usefully high; the signal magnitude can then be lowered below the critical value without loss of that stable state. The device can replace varactors as circuit elements in modulators, frequency multipliers and dielectric amplifiers. It may be used directly as a thermostat.

4.2 Tandel Effect:

Temperature autostabilization effect (Tandel) was studied by Glane et al (1963, 1964 a, 1964 b), Fousek (1965), Malek et al (1964). This effect can best be understood with reference to the Fig. (4.1), which represents the temperature dependence of the dissipation of a ferroelectric crystal switched with an alternating signal. The three curves marked W_2 (which show a peak around the Curie point T_C) represent the heat produced in the crystal by the alternating field, for three different values of the applied voltage. Part of this heat is due to normal dielectric lesses and partly due to hysteresis losses. The straight line marked W_1 represents the heat last by the crystal because of conduction and convection losses, consider the intersections 1, 2 and 3 of straight line with the middle one of the three curves W_2. These three intersections represent points at which the heat produced is equal to the heat lost, thus representing points of thermal equilibrium for the ferroelectric.

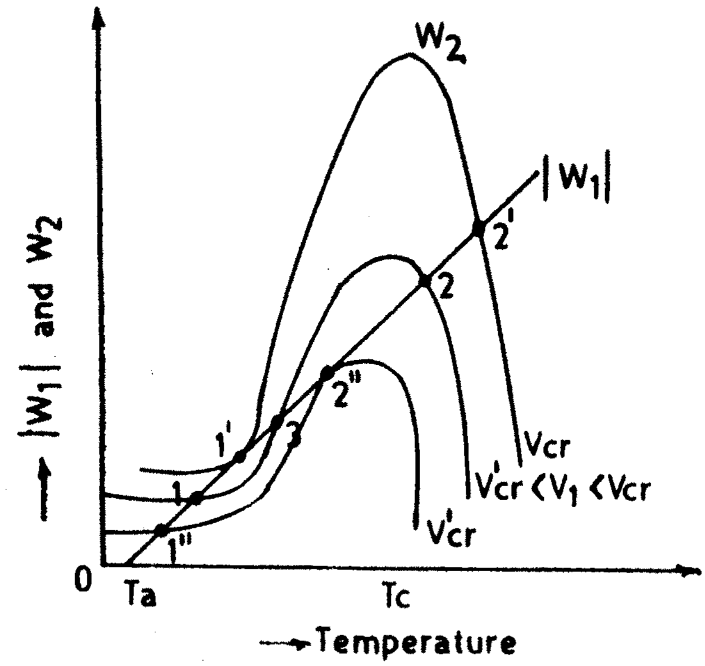

Fig. 4·1: The curves W_2 show the power dissipated in the crystal as a function of temperature, for three different values of the applied voltage. The line $|W_1|$ shows the power lost by the crystal to the surroundings. The curie point is Tc and room temperature Ta.

It can be shown that 1 and 2 are points of stable and 3 of unstable equilibrium. Because of the steep decrease of the curves W_2 above T_C ; the temperature of the crystal (if this is in the stage 2) is extremely well stabilized against fluctuations of the ambient temperature T B which tend to shift the straight line parallel to itself. It should be noted, however, that the applied voltage may be so high or so low that the corresponding W_2 curve lies above the top curve or below the bottom curve of Fig.(4.1). In this case there is only one stable temperature at which the crystal can operate(2 or 1).

The Tandel effect can be used in several applications, when a high; temperature independent displacement current is needed. A large number of devices from electrometers to frequency multipliers and parametric amplifiers can be built by using the Tandel effect.

Glanc A et al (1963) had reported that the element has various applications e.g. in frequency multiplier, frequency mixer, frequency modulator, pulse position modulator, dielectric amplifier, electrometer, electromechanical trausducer, transducer sensitive to heat transfer etc. Tandel as a new electronic element offers possibilities for microminiaturization and assembling into micro modules and its shock resistance is outstanding. Other prominant features in contrast to the semiconductors are a high d.c. imput resistance insensitivity to power overloading, and a simple production technology.

4.3 Second Harmonic Generation:

Abe Z. et al (1971) had examined the possibility of using a second harmonic type modulator, made of ferroelectric material as an electrometer. It was established that if modulation is carried out at a temperature slightly above the Curie point utilizing old symmetry of the relationship between voltage and charge of a ferroelectric capacitor, then the delay in response to a large input signal (memory effect) is eliminated, both zero drift and noise are reduced and carrier amplitude is diminished largely in contrast to the results obtained at room temperature. In the experiment, solid solutions of $PbTiO_3$, $SrTiO_3$ and $CaTiO_3$, near the Curie point, were used with a carrier frequency of 50 KHz, reducing zero drift to the order of 10 μv/day and realising low noise as characterized by 6 μv p-p with 1 M ohm input resistance and in the bandwidth from d.c. to 1 Hz. This modulator is solid state, and its input resistance is about 10^{13} ohm. as determined by specific resistance owing to the absence of any vibrating part, the carrier frequency can be raised and the bandwidth is more than several kilohertz. Compared with the best vibrating capacitor electrometer, this modulator, having nonvibrating parts, has an advantage in bandwidth but a disadvantage in inpur resistance. There remain some practical problem such as the method of keeping the element temperature in the vicinity of the Curie point and initial zero drift.

Dolino G. et al (1969) had reported that during second harmonic generation of Ruby laser light in ferroelectrics, angular scattering by ferroelectric domains has been found in the plane perpendicular to the polar axis. The influence of an applied d.c. electric field on the generation and the scattering of second harmonic has been measured and it has been indicated that change in domain structure can be studied by this method.

4.4 Analysis of Second Harmonic Generation:

Matsuo and Terashima (1956) described the generation of second harmonic in ferroelectric by approximating the nonlinear relationship between P and E to a hyperbolic function without considering operating temperature. Abe et al(1971) estimated the efficiency of conversion of dc input to second harmonic by taking into consideration the temperature of the ferroelectric. According to Devanshire (1950,1954) the nonlinear relation between P and E of ferroelectrics in the vicinity of the Curie point is expressed as

$$E = 2 \lambda (T - T_C) P + 4 BP^3 \qquad - - - - \qquad (4.1).$$

Where E is the electric field, P is the induced polarization, T is the temperature of ferroelectric, T_c is the Curie point temperature and λ and B are material specific constants. If constant current having a pure sinusoidal wave with an angular frequency W and small d_c voltage are fed to a ferroelectric capacitor in superposition, the polarization of capacitor is given by

$$P = P_o + P_W \ \mathrm{Cos}\ t \qquad ---- \qquad (4.2)$$

Electric field across the capacitor E is obtained by putting equation (4.2) in equation (4.1)

$$\begin{aligned} &= \left(2\lambda\ (T-T_c)\ (P_o + P_W\ \mathrm{Cos} Wt) + 4B\ (P_o + P_W\ \mathrm{Cos}_2 Wt)^3 \right) \\ &= \left(2\lambda\ (T-T_c) + 4BP_o^2 + 6\ BP_W^2 \right) P_o \\ &\quad + \left(2\lambda\ (T-T_c) + 12BP_o^2 + 3P_W^2 \right) P_W\ \mathrm{Cos} wt \\ &\quad + 6BP_oP_W^2\ \mathrm{Cos}_2 wt + B\ P_W^2\ \mathrm{Cos}_3\ wt. \qquad ---- \qquad (4.3) \end{aligned}$$

If we connect the TANDEL element having a capacitance C in series with a small resistance R, so that W C R 1, then the output voltage V_o across the resistor on applying an a.c. voltage V is given by

$$\begin{aligned} V_o &= W\ C\ R\ V \\ &= W\ C\ R\ E\ D \qquad ---- \qquad (4.4) \end{aligned}$$

Where d is the thickness of the crystal. If we assume that the resistance R is so small that the conditions of polarization are not influenced by it, the relation between electric field and polarization is still given by equation (4. 1). Then second harmonic voltage V_{2h} across the resistor can be obtained by replacing E in equation (4.4), by the term containing cos2wt in equation (4.3) and is given by

$$V_{2h} = 6\ W\ C\ R\ d\ B\ P_o\ P_w^2\ \mathrm{Cos}\ (2wt) \qquad - - - - \qquad (4.5)$$

It can be seen from equation (4.5) that the generated second harmonic is proportional to P_o (which is proportional to the applied d.c. bias) and to P_w^2 (proportional to the square of the applied a.c. voltage). The result of the equation (4.5) has been successfully we used only for a qualitative explanation of the experimental results.

4.5 Experimental:

In the present study, we have investigated the voltage response of the TANDEL elements, the solid solutions of $(Pb,Sr)TiO_3$ and $(Pb,Ca)TiO_3$ which were connected in series with a pure resistor of 10 ohms; Blockdiagram of the experimental set-up of Tandel is shown in Fig.(4.2). The pellets of above solid solutions having thickness of 1 mm were introduced in the crystal holder. Silver conducting paint was used with for electrodes (Silver paste with thin Al.foil).

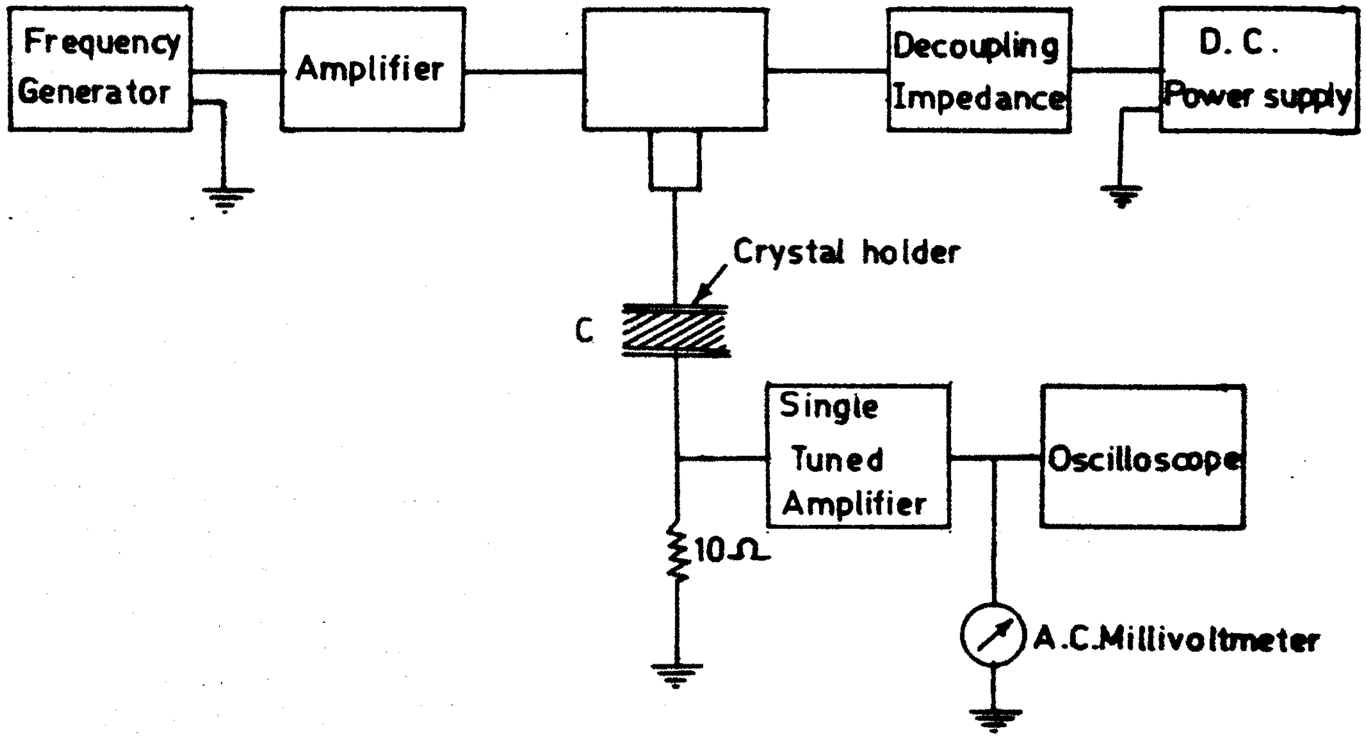

Fig. 4·2 : Experimental set up for Tandel.

A frequency of 10 KHz was used in the entire course of investigation. The single tuned amplifier was adjusted at 10K The frequency generator used was Philips GM 2308 type whose maximum output a.c. voltage was 90 V. Filter circuit, inductan in series and capacitance in shunts, was used as decoupling impedance. The A.C. millivoltmeter, (Philips - PP9001 X) was used for the response or measurement of voltage across the resistor. Experimental photograph of set-up for Tandel is w shown in Fig. (4.3).

4.6 (Lead, Strontium) titanate - TANDEL:

Preliminary investigation showed that the minimum peak voltage for autostabilization was 20 volt. Hence there amplitu levels 25, 30 and 35 volt peak were selected for the present studies. The ambient temperature was kept in the range 482 to 490°C with the use of temperature controller arrangement. The variation of the second harmonic voltage (measured by the A.C millivoltmeter) with d.c. bias for various peak voltages as mentioned above is shown in Fig. (4.4).

4.7 (Lead, Calcium) titanate - TANDEL:

Preliminary investigation showed that the minimum peak voltage for auto stabilization was 15 volt. Hence three amplitude levels 25, 30 and 35 volt peak were selected for the present studies. The ambient temperature was kept in the range 482 to 490°C with the use of temperature controll

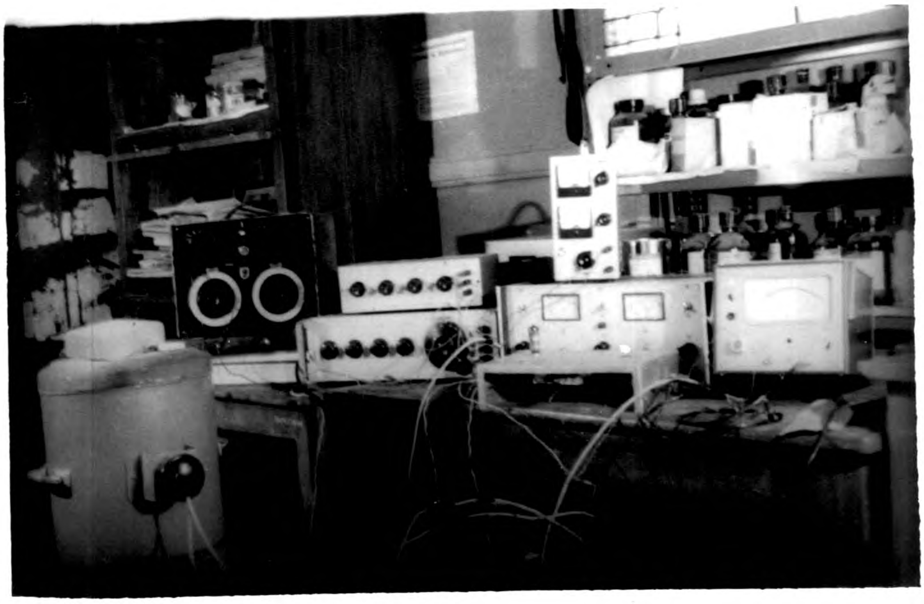

FIG. (4·3)

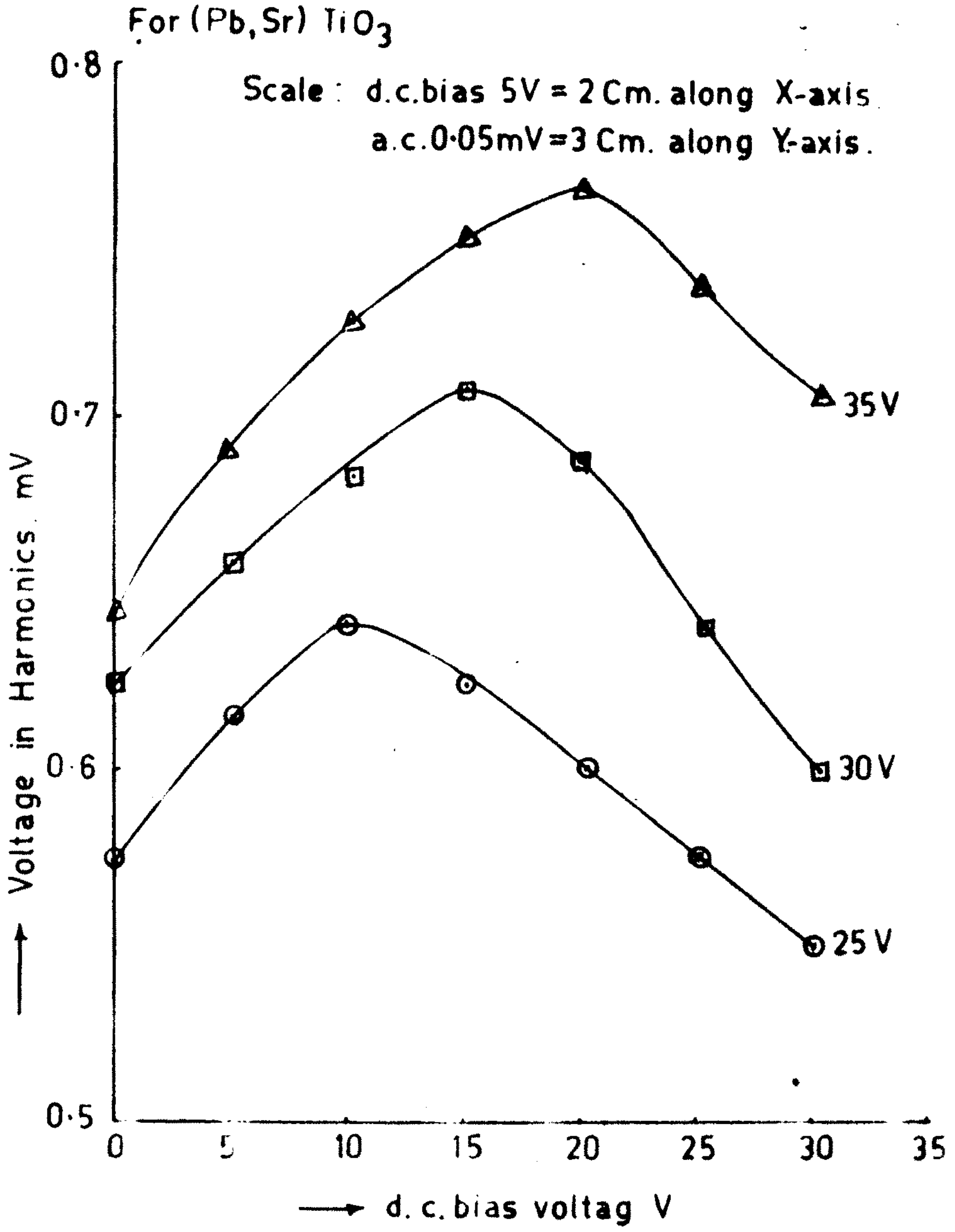

Fig. 4·4 : The variation of second harmonic voltage with d.c. bias for different a.c. voltages (volts peak) △35V, ⊡30V, ⊙25V.

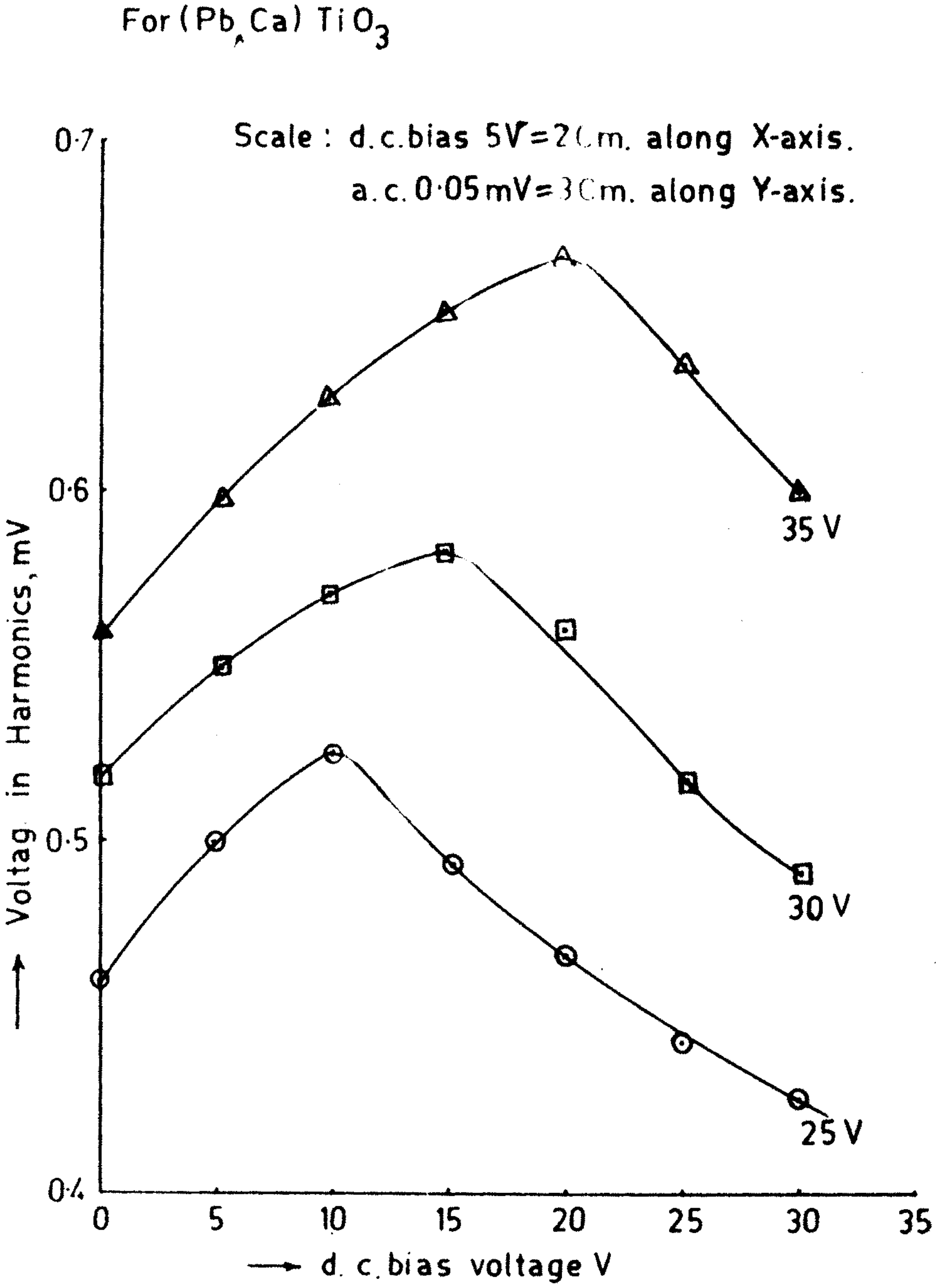

Fig.4·5: The variation of second harmonic voltage with d.c.bias for different a.c. voltages (volts peak) △35 V, ⊡30V, ⊙25V.

arrangement. The variation of the second harmonic voltage with d.c. bias for various peak voltages as mentioned above is shown in Fig.(4.5).

The list of experiments conducted on (Pb,Sr)TiO_3 and (Pb,Ca)TiO_3 TANDELS is given in Table (4.1) in order to get a comprehensive view of various TANDEL experiments.

Table No.(4.1)

The Variation of Second Harmonic Voltage with d.c. bias

Substance	Threshold peak voltage for autostbilization V	Peak a.c. voltage V	Magnitude of second harmonic at zero bias. mV	Magnitude of d.c. bias for going out of autostabilization.
(Pb,Sr)TiO_3	20	25	0.57	12
		30	0.62	18
		35	0.64	22
(Pb,Ca)TiO_3	15	25	0.46	11
		30	0.52	17
		35	0.56	22.5

4.8 Results and Discussion:

The second harmonic voltage response of both the solid solutions is shown in Fig.(4.4) and Fig.(4.5) respectively.

If we observe these groups, we see that the generated second harmonics are linear with the applied a.c. voltage as predicated by the equation (4.5) for low biasing fields. Similar results were reported by Malek et al (1964) and Mansingh and Eswar Prasad (1977) and Chavan and Patil (1980). But at higher biasing voltages, there are deviations from linear behaviour and sharp decrease in the amplitude of second harmonic is observed. The region over which the second harmonic is linear with the applied d.c. bias is extended by increasing the heating a.c. voltages. This is due to the fact that at low biasing fields the P_W polarization is counteracted by d.c. bias field so that the second harmonic is generated. At high biasing fields the polarization is counteracted till the substance does not fall out of the state of autostabilization. As the a.c. voltage is increased, higher bias would be required to drive the substance out of the state of autostabilization. This has been explained from the impedance voltage hysteresis behaviour by Malek E. et al(1964).

From equation (4.5), it can be observed that generated second harmonce should be proportional to the square of the applied a.c. voltage. But in the present investigations and also in the work of Manshingh and Eswar Prasad (1977), Chavan & Patil (1980), such variations have not been observed.

The explanation for this comes from the basic TANDEL behaviour. It adjusts its impedance against variations of applied a.c. voltage. So that product CP_W^2 in equation (4.5) remains constant and this is consistent with the theory of TANDEL outlined by Dvorak et al (1964). It can be very easily seen from the graphs that when d.c. bias voltage is zero, there is still some second harmonic output. This is in conformity with the result of Abe et al (1971) and Mansingh and Eswar Prasad (1977) and Chavan and Patil (1980), but in contrast to those observed by Malek et al (1964), Mansingh and Eswar Prasad (1977) and Chavan & Patil (1980), have attributed this offset to the presure of defects, giving to an internal bias which in turn generates a second harmonic.

In order to confirm the role of the defects in crystals in giving rise to the internal bias, the experiments were conducted on annealed crystal plates. All these observations are consistant with each other in view of the explanation of the Tandel behaviour, as confirmed by Chavan and Patil (1980).

Our results establish that $(Pb,Sr)TiO_3$ and $(Pb,Ca)TiO_3$ can be used as TANDEL elements which provide the autostabilization state. This would make them interesting from the point of view of various applications.

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REFERENCES

Abe Z.,Koto Y and Furuhata Y. : (1971). Rev.Sci.Instum.42,805.

Chavan S.H. and Patil R.N. : (1980). Pramana,15,No.1,85-90.

Devonshire A.F. : (1950). Adv.Phys.8,10.

Dolino G., Lajzerowicz J.,and Vallade M. : (1969). Solid State Communications, 7, 1005.

Fousek J. : (1965). J.Apply.Phy.36,588.

Glano A.,Dvorak V., Janovec V., Rechzigel E.,and Janousek V. : (1963). Phys.Letters 7,106.

Glane A.;Malek Z. Mastner J.,Novak M. and Strojobova J. : (1964a). J.Appl.Phys.35,1870.

Glane A.,Malek Z. Mastner J.,Novak M. and Strajblova J. : (1964b). J.Appl.Phys.35,1870.

Malek Z.Mastner J. Hridicka J.,and Janta J. : (1964). Solid St.Electron.7,655.

Mansingh A., and Eswar Prasad S.R.J. : (1977). J.Phys.Chem.Solids, 38, 1207.

Matsuo M. and Terashima T. : (1956). Res.Nat.Conv.Inst., Elec.Commn.Eng.Japan, 191.

Miller R.C. : (1964) Phys.Rev.A134, 1313.

Smith.A.W. : (1964). Appl.Opt. 3, 147.

Suvorov V.S. : (1968). Soviet Phys.JETP., 27, 557.

Vincent A.M. : (1951 Dec.). Dielectric Amplifier Fundamentals, Electronics.

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