Quality of a Ferroelectric Material

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***Summary*—The paper describes a simple method of measuring the nonlinear properties of a ferroelectric material at microwave frequencies. A composition of commercial polycrystalline barium titanate, of stated Curie temperature of 20°C, and of polythene powder was investigated. The incremental permittivity of the specimen at both increasing and decreasing biasing field was observed at *X*, *C* and *S* band. The effective dielectric constant of the specimen, for the same biasing field, was lower for increasing field, contrary to the theoretical analysis. Diamond's model of polycrystalline ferroelectric materials provides a qualitative explanation for the observed anomalous behavior of the specimens. An experimental arrangement for the observation of the microwave equivalent of a low-frequency ferroelectric loop is suggested.**

THE PURPOSE of this paper is to describe a method of evaluating the quality of ferroelectric materials to be used primarily for parametric amplification. Not very much work has been done on the application of ferroelectric materials to microwave devices. This is due, perhaps, to the opinion expressed by Powles and Jackson [1] that the relaxation frequency of barium titanate, a widely investigated ferroelectric material, is below the microwave range. In recent years a number of workers [2]–[7] have measured the properties of different ferroelectric materials at high frequencies.

The use of ferroelectric materials for parametric amplification has been suggested by Cassedy [8] and others [9]. Two communications [10], [11] describe successful application of barium titanate for parametric amplification. It is appropriate, after the successful construction of ferroelectric parametric amplifiers, to discuss a method of finding the quality of a ferroelectric material. Let Fig. 1 be the equivalent circuit of a ferroelectric material where R is the resistance of the specimen assumed to be constant and C_0 is the zero-bias capacitance. When this is placed in a waveguide it looks, after a quarter-wave shift, like an admittance given by

$$Y = Y_0^2\left(R + \frac{1}{j\omega C_0}\right) \tag{1}$$

where Y_0 is the characteristic admittance of the waveguide. Let C_v be the capacity of the specimen at a bias bias of V volts/m. Then the

$$\frac{\text{change of susceptance}}{\text{conductance}}$$

$$\Delta B = \frac{\dfrac{1}{\omega C_v} - \dfrac{1}{\omega C_0}}{R} \tag{2}$$

$$= \frac{C_3}{C_0}\,\frac{1}{\omega C_v R} \tag{3}$$

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where C_3 is the maximum amplitude of the change in capacitance.

When Y is plotted on a Smith chart it will lie on a circle of constant conductance. It is possible to transform this circle to a unit conductance circle by the addition of lossless elements. The ratio of the

$$\frac{\text{change of susceptance}}{\text{conductance}}$$

for any two points at the plane of the specimen and for the corresponding two points at the unity conductance circle is the same. Thus, by matching the specimen to the waveguide through the use of lossless elements, the change in the susceptance can be measured for any applied dc bias. It can be shown that the change of susceptance on the unity conductance circle is

$$\Delta B = \sqrt{S} - \frac{1}{\sqrt{S}} \tag{4}$$

where S is the VSWR. By measuring the VSWR for a particular bias, the change of susceptance is directly obtained from (4). The advantage of the method is that a knowledge of the absolute values of the complex dielectric constant at different bias voltages is not necessary. A possible figure of merit of the specimen is

$$\frac{\text{maximum available change in susceptance}}{\text{conductance}}.$$

Fig. 2 shows [12] the experimental arrangement at X band. The position of the variable short circuit was initially adjusted to reduce the mismatch. Subsequently, complete matching was obtained by the tuning screw. Application of different magnitudes of dc bias changed the position of the voltage minimum and the VSWR.

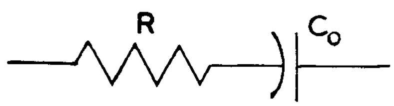


Fig. 1—Equivalent circuit of a ferroelectric material.

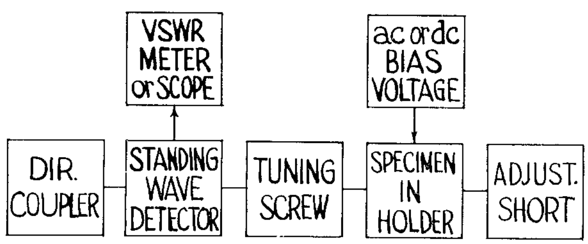


Fig. 2—Experimental arrangement for measuring the change of susceptance.

Fig. 3 shows a plot of the change of susceptance of a typical specimen. It is important to ascertain that, with zero bias, all the incident microwave power is absorbed by the specimen. Absorption of power by any circuit element will reduce the quality of the specimen. Similar measurements have been taken on variable capacitance diodes [13], [14] and recently on ferroelectrics [15]. Fig. 4 shows the specimen holder. Specimens were made up of a mixture of powdered barium titanate and polythene powder of different compositions. This was done to reduce the value of the dielectric constant. The specimens had a nominal diameter of 2 mm. For the application of a dc bias, a small thin copper wire was placed at the center of the specimen during its preparation. The specimen, referred to in Fig. 3, was composed of 90.3 per cent ceramic barium titanate and 9.7 per cent polythene powder. The ceramic material, prepared by Technical Ceramics Limited (England), had a stated Curie temperature of 20°C and a low-frequency dielectric constant of 6470 at the room temperature. Measurements taken at 3816 Mc in a cylindrical cavity for another specimen of similar composition gave a value of 36 for the dielectric constant and 0.028 for the loss tangent.

Ferroelectric materials exhibit hysteresis at low frequencies. A number of properties [16], including the incremental permittivity of these materials as a function of the applied voltage, can be found out from the hysteresis loop. It was of interest to find out whether hysteresis could be observed at microwave frequencies. No information is available, to the best knowledge of the author, in the literature regarding the observation of hysteresis loop at microwave frequencies. Presence of hysteresis will result in different values of incremental permittivity for increasing and decreasing values of bias voltage of the same magnitude. Using the experimental arrangement of Fig. 2, the specimen was biased by 50-cps voltage. The output of the standing-wave detector and a fraction of the bias voltage were fed to the Y amplifier and X plate of a dc oscilloscope. This resulted in the formation of butterfly loops (Fig. 5) which are plots of the microwave field reflected from the specimen at each position of the low-frequency hysteresis loop. Matching could be done for any particular value of the biasing voltage. For Fig. 5 a perfect match was obtained at the maximum (right-hand side) value of the biasing voltage. Fig. 5, (a) and (b), shows photographs of the butterfly loops with the standing-wave detector at the position of voltage minimum and maximum, respectively. The zero level is indicated by the second beam on top. The hysteresis loop of the ferroelectric specimen is traced out at 50 cps. The microwave system is measuring the incremental dielectric constant at each point of this low-frequency hysteresis loop. By proper calibration the VSWR, and hence the change in susceptance for any value of the biasing voltage, either increasing or decreasing, can be found out from Fig. 5.

Often it is of interest to know the properties of these materials when they are placed inside a resonant cavity or when they are used as dielectric resonators. To investigate the first case CW microwave power was fed to a cavity loaded with a ferroelectric specimen. Bias of 50 cps was applied to the specimen. The detected output of the cavity and a portion of the bias voltage were fed to the Y amplifier and X plate of an oscilloscope. This resulted in butterfly loops, a selection of which is shown in Fig. 6. The resonant frequency, and as such the output of the cavity, was changing due to the ac bias applied to the specimen. Whenever the resonant frequency of the loaded cavity was equal to the klystron frequency, a maximum output was obtained. When the klystron frequency was at f_1 and f_3 [Fig. 6, (a) and (c)] two loops were obtained because twice in each cycle of the biasing field the resonant frequency of the loaded cavity and the klystron frequency were the same. When the two frequencies were equal four times in each cycle of the biasing field, four loops [Fig. 6(b)] were obtained. To get a better picture let us study Fig. 7. The effect of the application of varying dc field is to change the polarization of the specimen around the hysteresis loop. When a small ac signal is applied to the specimen biased to a point M, a small subsidiary loop is traversed. The microwave signal is expected to measure the incremental dielectric constant given by the slope of the subsidiary loop and is equal to $\Delta P/\Delta E$ where ΔP is the small change in the polarization due to a small change in the electric field ΔE. For very small ac signals and for high permittivity materials the incremental dielectric constant is equal to the slope of the hysteresis loop at the operating point.

The slope at each point, plotted in Fig. 8(a), of a hysteresis loop gives the incremental dielectric constant at the corresponding biasing field. The resonant frequency of a cavity, loaded with a specimen having the above characteristics, will vary as shown in Fig. 8(b). On reversing the direction of the biasing field, a decreased value of the incremental dielectric constant is obtained and, consequently, the resonant frequency of the cavity will follow the ascending portion of the curve.

Matthias and von Hippel [17] measured the dielectric constant of a single crystal barium titanate with a small low-frequency signal. They applied a dc bias to the specimen and varied it cyclically. Fig. 8(a) resembles their experimental results. Fig. 9 gives the results [12] of microwave measurements of the resonant frequency of a cavity loaded with a specimen subjected to an applied dc voltage which was varied over a cycle. Heating effect is partially responsible for the irregular shape of the curve. The theoretical variation [Fig. 8(b)] of the resonant frequency of the cavity is opposite to the experimental variation (Fig. 9).

A number of observations were made both in the cavity as well as in the waveguide (Fig. 2) at X, C and S band. All these observations showed consistently that the effective dielectric constant of a specimen, for the same biasing field, is lower for increasing field. Diamond [18], while experimenting with perovskite-like polycrystalline substances such as $BaTiO_3$, and mixed compositions of the type $Ba_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$, etc., found that "if the variation in permittivity with electric field is held to be in some way associated with

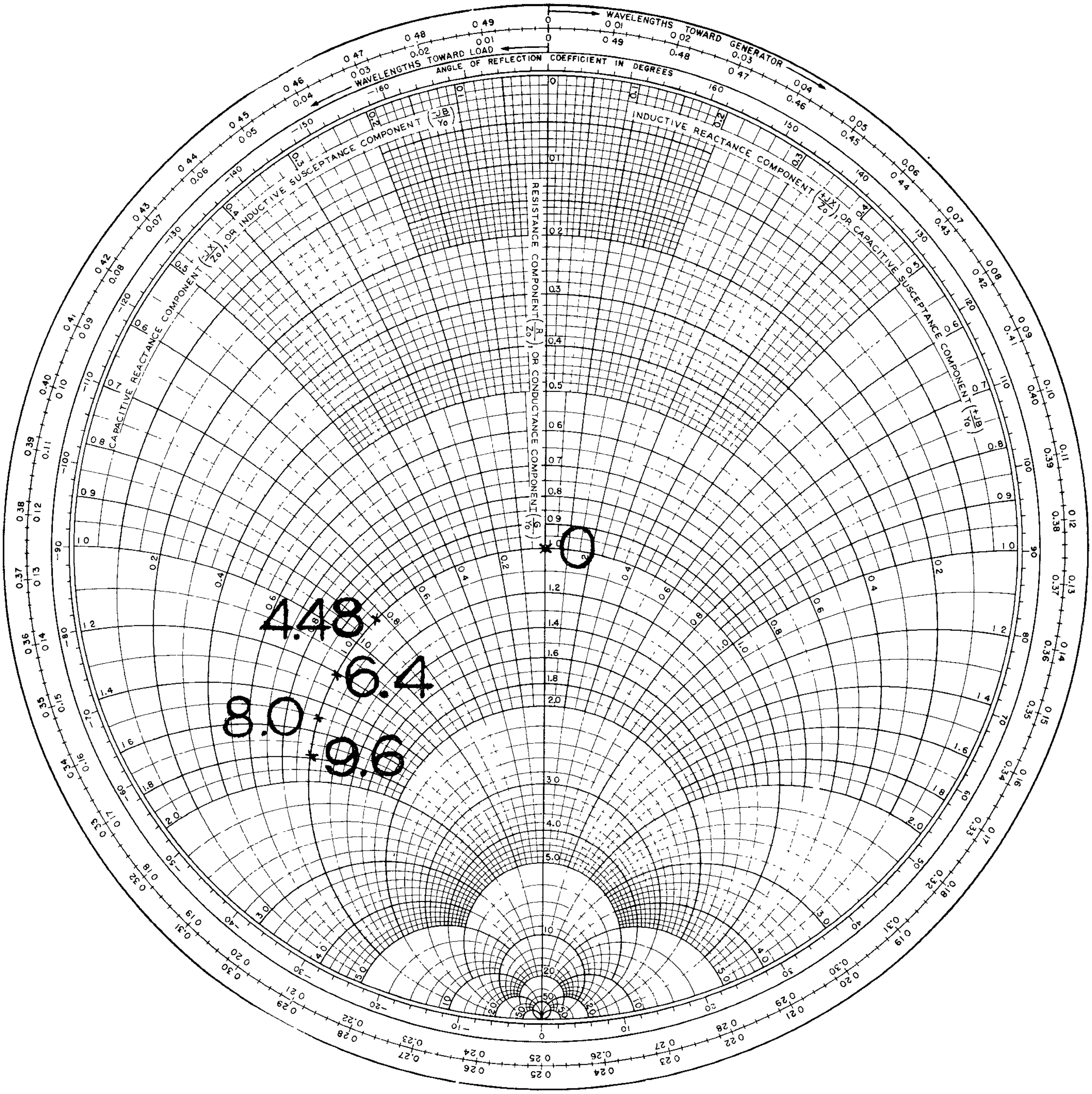


Fig. 3— $\frac{\text{Change of susceptance}}{\text{conductance}}$ of a specimen as a function of dc bias.

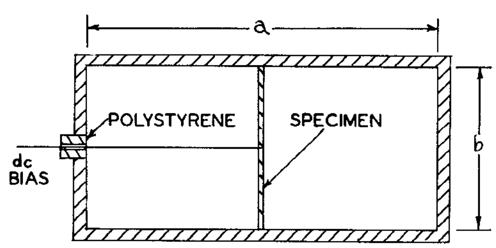
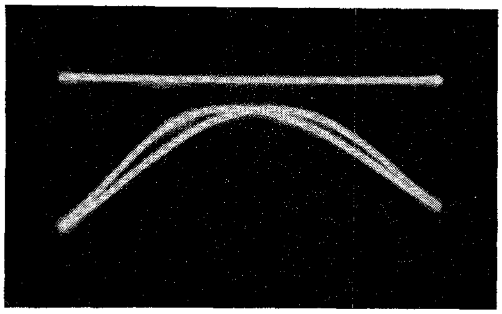
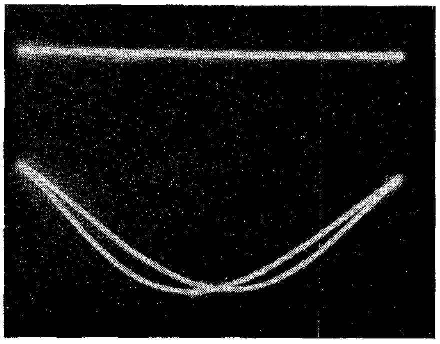


Fig. 4—Specimen holder.

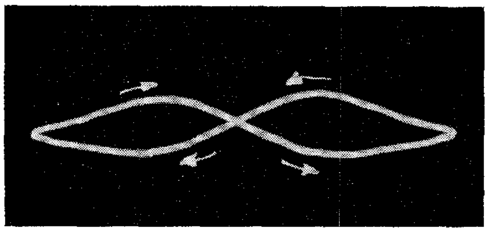


(a)

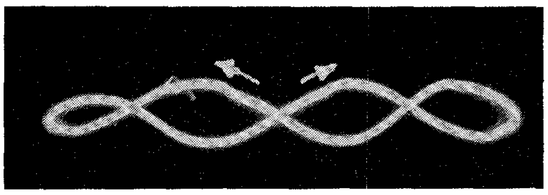


(b)

Fig. 5—Butterfly loops obtained with standing-wave detector at the position of the voltage. (a) Minimum voltage. (b) Maximum voltage.



(a)



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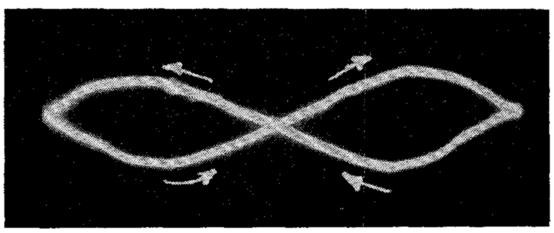


(c)

Fig. 6—Butterfly loops obtained with klystron. (a) Set at 9401. (b) Set at 9405.8. (c) Set at 9409.4 Mc.

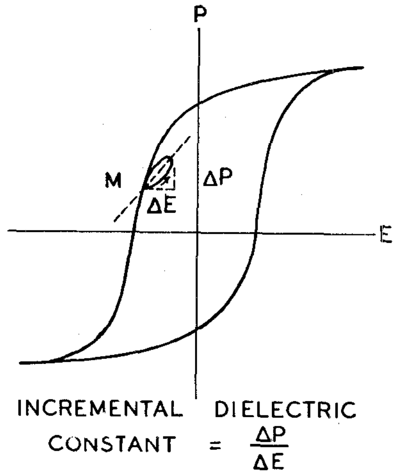
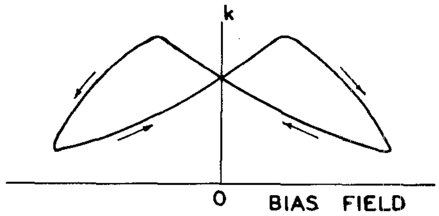
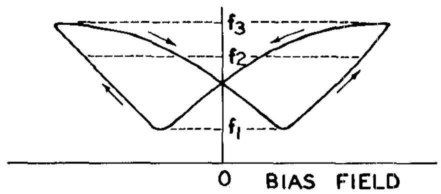


Fig. 7—Incremental dielectric constant.



(a)



(b)

Fig. 8—(a) Variation of dielectric constant *k* with biasing field. (b) The expected variation of the resonant frequency of a cavity loaded with a material of (a) characteristics.

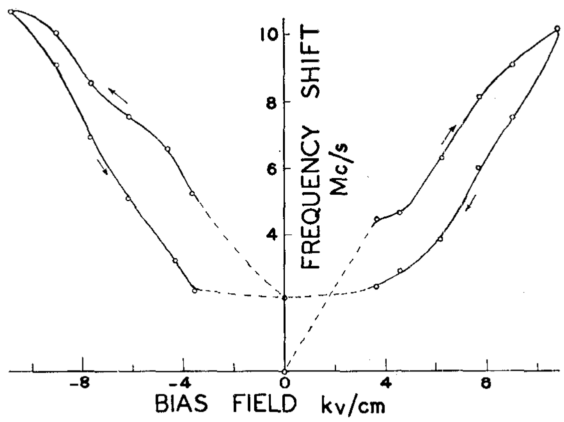


Fig. 9—Shift of the resonant frequency of a loaded cavity as a function of the dc bias applied to the specimen.

domain alignment or rotation processes (as in the case for magnetic substances) one is led to contradiction with experimental data."[1] He proposed a model for polycrystalline ferroelectric materials and his model satisfactorily explained his experimental data.

Diamond's model provides a qualitative explanation for the observed anomalous behavior of the specimens mentioned earlier. Consider that we are working very close to or lower than the Curie temperature (T_c). Due to an applied field E the effective Curie temperature T_c^* [18] will change so that $T_c^* = T_c + \gamma E$. The factor γ is a constant of proportionality and will be assumed constant for simplicity. For an applied electric field of 10 kv/cm the change in the Curie temperature is of the order of 13°K (γ for single crystal barium titanate is about 1.3°K/kv/cm). The dielectric constant, as shown in Fig. 8 [18], has a sharper fall below the Curie temperature than above it. Fig. 10 shows a plot of two curves, similar to those in Fig. 8 [18], whose Curie temperatures differ by an equal amount from the operating temperature. The effective Curie temperature of the specimen increases for positive applied bias (assume it to be at B) and decreases for negative applied bias (consider it to be at A). The dielectric constant of the specimen at the operating temperature will be at D and C for increasing and decreasing bias voltages, respectively. This analysis indicates that the incremental permittivity, for the same magnitude of the biasing field, should be higher for decreasing bias irrespective of the frequency of measurement. The general shape of the dielectric constant vs temperature curve for Davis and Rubin [2] is similar to that of Diamond [18]. For single crystal [19] barium titanate the dielectric constant decreases sharply below the Curie temperature and rather slowly above it. More work is necessary to give a satisfactory quantitative explanation of the anomalous behavior of the author's specimens.

For application of ferroelectric materials to microwave devices, one would avoid hysteresis and work slightly above the Curie point. The simple experiments described above could be utilized for visual observance of the nonlinearity of the specimen using ac biasing, and for quantitative measurements using dc biasing.

To obtain the microwave equivalent of a low-frequency hysteresis loop the following experiment can be performed. Let the specimen (Fig. 2) be subjected to a high microwave field. The reflected wave, collected by a directional coupler and a fraction of the incident wave, can be fed to two pairs of deflecting plates of a microwave oscilloscope [20] resulting in butterfly loops similar to those in Fig. 5. Table I is a collection of results of experiments at microwave frequencies on ferroelectric materials.

[1] See Diamond [18], p. 909.

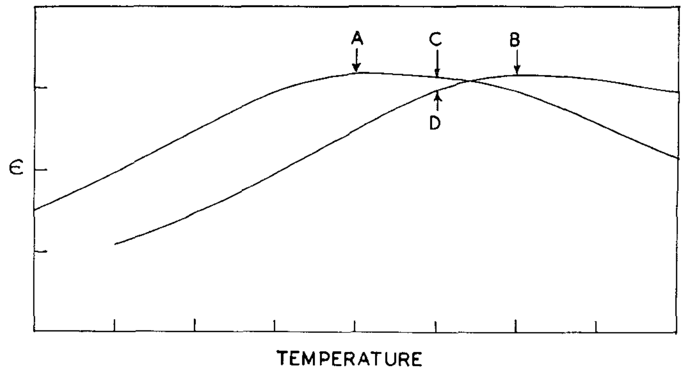


Fig. 10—Assumed variation of permittivity with temperature for two different Curie temperatures (A and B) differing by equal amount from the temperature of measurement (C and D).

TABLE I

Material	Relative Dielectric Constant	Loss Tangent	Measured at		Relaxation Frequency kMc
			Frequency kMc	Temperature °C	
Single crystal[3] $BaTiO_3$	2000 (ϵ_{11})	~0.15	24.0	20°	
TGS[4]	1200 (peak)	1.75 (peak)	2.0	47.8°	<2.0
Ceramic[7] $BaTiO_3$	600	~0.3	3.0	25°	
Hot-pressed[7] $Cd_2Nb_2O_7$	435	0.069	4.0	26°	
73 per cent $BaTiO_3$ −27 per cent $SrTiO_3$[21]	3350	~0.16	12.4	21°	~50
KH_2PO_4[6]	15 (ϵ_c)	0.003	9.2	−160°	
Single crystal[22] $SrTiO_3$	~1000	0.0013 (22 kMc)	7.65	−150°	>35
KD_2PO_4[23]	~40	0.2 (2 kMc)	1.0	−60°	<1.0

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The Reflecting Beam Waveguide

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Summary—In this paper a type of beam waveguide which uses appropriately shaped metal reflectors instead of dielectric lenses as the phase correcting devices is described. A theory has been developed which, subject to certain restrictions, describes the modes of this type of beam waveguide and predicts a loss of the order of of 0.01 db per iteration.

A reflecting beam waveguide comprising eight aluminum reflectors has been investigated at a wavelength of 4 millimeters. The measured loss per iteration is approximately 0.015 db which is in good agreement with the theoretical value. The cross-sectional electric field distribution has also been measured and found to be in satisfactory agreement with the theory.

It is shown that the reflecting beam waveguide is a practical system for the transmission of power at submillimeter wavelengths.

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Introduction

In recent years, the beam waveguide analyzed by Goubau and his co-workers [1], [2] has been considered as a possible transmission system for wavelengths from the low millimeter to the optical region of the spectrum. Basically, the beam waveguide consists of dielectric lenses or phase correctors, each having a focal length f, which are separated by a distance $a=2f$ as shown in Fig. 1. At each lens the phase distribution of a signal propagating along the beam waveguide is corrected to compensate for diffraction effects due to the finite aperture of the beam waveguide. The confocal spacing keeps these diffraction losses at a minimum for a given lens diameter. The losses on this type of waveguide are due to the diffraction losses cited above, to absorption losses within the lenses, and to reflection and scatter losses at the surfaces of the lenses.

For reasonable lens diameters and spacings the diffraction losses can be kept quite small in the millimeter and submillimeter region of the spectrum. However, it is difficult to obtain a dielectric phase corrector for which both the absorption and reflection losses are small at