Microwave losses in ABO$_3$ type perovskites

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Abstract. Using Green's function method, microwave lossess were theoretically calculated for BaTiO$_3$, SrTiO$_3$ and KTaO$_3$ ferroelectric perovskites. In microwave range, an increase in frequency is followed by an increase in the dielectric loss. In the paraelectric phase, the dielectric loss decreases with increasing temperature showing the Curie-Weiss behaviour of the tangent loss.

Keywords. Ferroelectrics; perovskites; soft-mode; anharmonicity; microwave-loss.

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1. Introduction

The investigation of dielectric properties provides an important approach to the understanding of intra- and-inter molecular interactions, modes of motion and confirmational changes in the macromolecules. The temperature and frequency-dependence of the dielectric loss is the subject of considerable interest due to their extensive use in optical communication, memory display, temperature control devices, ceramic industry, etc. It is well known that several interesting temperature-dependent properties of ferroelectrics result from the temperature-dependence of the low lying transverse optic mode of vibration (Cochran 1969). Microwave losses in displacive ferroelectrics (BT, ST etc.) have been reported experimentally (Rupprecht and Bell 1961, 1962; Rupprecht et al 1961). Above the phase transition temperature, the results of the loss measurements can be represented by the temperature and frequency-dependence of the microwave loss tangent ($\tan \delta$) as

$$(T - T_c)\tan \delta = \omega(\alpha + \beta T + \gamma T^2),$$

where the parameter $\alpha$ depends strongly on the defect concentration. Parameters $\beta$ and $\gamma$ are third and fourth order anharmonic interaction terms which are temperature-independent but vary linearly with the frequency. Rupprecht and Bell (1961) found that in cubic SrTiO$_3$, the field independent loss tangent goes through a minimum at about 170 K with a much steeper slope on the low temperature side of the minimum than on the high-temperature side. The temperature-dependence of the loss tangent for the damping process is expressed as

$$\tan \delta = \gamma/(\Omega^2 - \omega^2) \approx \gamma/\Omega^2 \sim 1/(T - T_c).$$

Impurity scattering provides the sharp rise of the loss tangent on the low-temperature side of the curve. The microwave frequency was taken from 21–22 GHz.
We have measured experimentally (Baluni and Naithani 1986a) the dielectric constant and the dielectric loss in BaTiO₃ and theoretically (Baluni and Naithani 1986b; Panwar et al. 1989) calculated these properties in ferroelectric solids but have made no numerical calculations. The damping of the microwaves in a pure crystal for which \( \epsilon = 0 \), is due to the damping of the polarization mode by anharmonicities of the lattice vibrations. Temperature-dependence of the loss tangent is a reflection of the temperature-dependence of the polarization mode frequency which is given as

\[ \Omega \sim (T - T_c)^{\delta}. \]  

Microwave loss obeys the Curie-Weiss law. This may be taken as a direct evidence for the temperature-dependence of the polarization mode frequency. At transition temperature, the frequency of the soft mode tends to zero and the lattice displacement associated with this mode becomes unstable. This explains the anomalous behaviour of the dielectric loss near the phase transition. The earlier studies (Benedict and Durand 1958; Lurio and Stern 1960) on BaTiO₃ crystals observed relatively high microwave loss above the Curie temperature. The losses were found to be varying linearly with the frequency. Silverman-Joseph (1963) described a Hamiltonian for displacive ferroelectrics and studied the loss tangent for SrTiO₃ and found that coefficients \( \beta \) and \( \gamma \) and hence \( \tan \delta \) vary linearly with the frequency \( \omega \). SrTiO₃ (Rupprecht and Bell 1961), CaTiO₃ (Lurio and Herrington 1958), KTaO₃, KTaO₃: NaTaO₃ (Agrawal and Rao 1970) exhibit the same behaviour. Lurio and Herrington (1958) have explained that the loss tangent (in the low frequency range at RT) is proportional to the frequency in the range 3–36 GHz. Davis and Rubin (1953) have investigated the loss tangent of certain mixtures of Ba₅Sr₁₋₅TiO₃ at 35 GHz frequency above and below the Curie temperature. Tani (1969) had explained the critical slowing down of the damping constant in displacive ferroelectrics from the standpoint of the irreversible thermodynamics. In place of tangent loss description, only temperature-dependent damping constant is described. Vinogradov (1963) observed that \( \tan \delta \) for inorganic crystals with ideal lattice, is weakly dependent on frequency in the region of \( 10^5 \) to \( 10^8 \) Hz. West (1987) measured the electrical losses (\( \tan \delta \)) for BaTiO₃ ceramic at frequencies up to 2 GHz. The 1 MHz dissipation factor was found to be 0.025. At RT, \( \tan \delta \) is almost constant in the frequency range 0.001–10 MHz and in 10 MHz to \( 10^3 \) MHz, \( \tan \delta \) increases linearly with frequency.

Udagawa (1981) had studied the dielectric dispersion in tetragonal BaTiO₃ at RT using one oscillator dielectric function mainly due to soft TO phonons but tangent loss is not studied. Vogt (1982) has not calculated \( \tan \delta \) but he estimated the relative damping for cubic BT from hyper Raman spectrum. Freuly and Lazay (1971) have reported temperature-dependence of the Brillouin-Raman spectrum of BaTiO₃ using the approximated complex susceptibility. Balagurov (1970) studied the attenuation of critical vibrations and dielectric losses in displacive ferroelectrics theoretically. Naithani (1980) and Panwar et al. (1989) have expressed dielectric losses theoretically with no numerical estimation.

In the present work, we have theoretically studied the functional dependence of the microwave loss tangent on parameters such as temperature and frequency, using Green's function method (Zubarav 1960) in pure anharmonic ferroelectric (BT, ST and KT) crystals with the help of Pytte's (1970) modified Hamiltonian considering anharmonic effects up to fourth order and have compared the results with that of others.