Solid State Communications, Vol. 10, pp. 887–889, 1972. Pergamon Press. Printed in Great Britain

EFFECT OF PRESSURE ON THE DIELECTRIC PROPERTIES OF A PARAELECTRIC MATERIAL*

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(*Received* 27 *January* 1972 *by A.A. Maradudin*)

Employing a self-consistent renormalization procedure for the treatment of the frequency spectrum of a model paraelectric, the essential features of the pressure dependence of the static dielectric constant of the incipient ferroelectric $KTaO_3$ are reproduced.

IN A PREVIOUS work[1] (hereafter referred to as I) self-consistent phonon techniques were applied to the treatment of the frequency spectrum of a model paraelectric. It was found that many of the qualitative features of the frequency spectrum of the paraelectric $KTaO_3$ could be reproduced, e.g., the temperature dependence of the mode coupling between the optic and acoustic branches of the same symmetry, as well as the temperature dependence of the soft zone center transverse optic mode frequency $\omega_{TO}(\Gamma)$. On the basis of an extension of that model, we examine in this note the effect of pressure on the frequency $\omega_{TO}(\Gamma)$. It is by now well established[2,3] that the temperature dependence of $\omega_{TO}^2(\Gamma)$ reflects a temperature dependence similar to that of reciprocal of the static dielectric constant ϵ^{-1}. This has been demonstrated experimentally for the paraelectric $SrTiO_3$ by Cowley[4] and for the paraelectric $KTaO_3$ by Shirane, *et al.*[5] Similarly, one expects the pressure dependence of $\omega_{TO}^2(\Gamma)$ to follow closely the pressure dependence of ϵ^{-1}. Indeed, our calculations of $\omega_{TO}^2(\Gamma)$ yield good qualitative agreement with recent pressure measurements be Abel[6] on the static dielectric constant of $KTaO_3$. In particular, the temperature T_1 at which ϵ^{-1} exhibits deviations from Curie–Weiss behavior increases with pressure. Furthermore, at fixed temperature $\omega_{TO}^2(\Gamma)$ is found to increase linearly with pressure up to a pressure P_1 beyond which deviations from linearity occur.

* This work was supported by the U.S. Atomic Energy Commission.

The calculations in I were based on a model crystal of the rocksalt structure, with a nearest neighbor anharmonic short range interaction. The dipolar interaction was between rigid anions and cations and was treated in the harmonic approximation. It was shown that the parameters entering into the model could be scaled in such a way that the long wavelength properties were rendered relatively independent of the details of the dipolar interaction, the masses of the two types of ions, and the atomic volume. Thus, it was argued that the qualitative behavior of the zone center soft mode would mirror a similar behavior in a more complicated lattice such as the perovskite structure. This is indeed the case for the pressure dependence of the dielectric properties as we demonstrate below.

Following the procedure introduced in I, we indicate briefly the self-consistent procedure to be followed in treating the pressure dependence of the paraelectric soft mode. We introduce a trial free energy per particle of the form

$$F_t = \frac{1}{N} \mathrm{Tr}[\rho_t (H + \beta^{-1} \ln \rho_t)], \qquad (1)$$

where

$$\rho_t = e^{-\beta H_t} / \mathrm{Tr}\,(e^{-\beta H_t}), \quad \beta \equiv (K_B T)^{-1}. \qquad (2)$$

The canonical density matrix (2) is based on a trial harmonic Hamiltonian of the form