

cause coupling of the two modes by the transducer at each reflection resulted in some mutual contamination. The mode 6 data were obtained by manual measurements of the frequency shift of the interference pattern because the on-line computer had failed. In spite of the incompleteness and poor quality of some of the data, they are sufficient to constrain the moduli derivatives fairly well because none of the larger relative errors (such as in mode 7, Fig. 1) were translated into large absolute errors.

The measured phase derivatives, with uncertainties estimated from the scatter in the data, are given in Table 4, along with the derived mode moduli derivatives, including corrections for length change and transducer phase shift. The transducer longitudinal and transverse impedances are  $15.2$  and  $10.3 \times 10^5 \text{ g cm}^{-2} \text{ s}^{-1}$ , respectively, and the logarithmic resonance frequency derivatives are  $1.51$  and  $-3.68 \text{ Mbar}^{-1}$  [21]. The only redundancy in these data is between modes 2 and 8. Since mode 2 is suspect, the mode 8 data were used, with fairly large uncertainty. The measured and derived single crystal moduli derivatives are included in Table 1. The estimated uncertainty of the derived quantities is about  $0.2$ – $0.3$ , based on the uncertainties of the measured derivatives given in Table 4.

Pressure derivatives of the isotropic aggregate bulk and shear moduli are also included in Table 1. These have been estimated from the derivatives of the Hashin-Shtrikman bounds [26,27], but it should be noted that these are not themselves bounds on the derivatives. It can be seen that the estimates of the shear modulus derivative are quite uncertain. The estimated isotropic aggregate derivatives are compared in Table 1 with measured derivatives on polycrystalline aggregates given by Rai and Manghnani [12]. The agreement is excellent.

## 6. Discussion

These results suggest that the two most notable features of the pressure derivatives of the elastic moduli of the rutile-structure oxides are also present in this fluoride analogue: the value of  $K'$  of  $5.1$  is fairly high, though not as high as in any of the oxides, and the value of  $c'_s = (c'_{11} - c'_{12})/2$  is negative.

The value of  $K'$  may not in fact be unusually high

for fluorides. The values of  $K'$  for LiF, NaF, KF (rocksalt structures) and  $\text{CaF}_2$  and  $\text{BaF}_2$  (fluorite structures) all are in the range  $4.9$ – $5.2$  [5]. The increasing values of  $K'$  through the sequence  $\text{MgF}_2$ ,  $\text{SnO}_2$ ,  $\text{GeO}_2$  suggest that larger values of  $K'$  may be associated with more covalent bonding. Thus stishovite may indeed have a quite high value of  $K'$ , close to  $7$  [5].

The long wavelength  $B_{1g}$  optic mode of rutile also has a negative pressure derivative [28,29]. It has been pointed out that the combined displacements associated with the  $B_{1g}$  optic mode and the  $c_s$  acoustic mode are the same as those required to transform the rutile structure into the  $\text{CaCl}_2$  structure [30]. Both of these "mode softenings" may thus be associated with an approaching instability of the rutile structure.

Theoretical predictions of the pressure derivatives of the elastic moduli were made by Stiefeler and Barsch [31] using a modified rigid-ion model including an effective ionic charge and first- and second-nearest neighbor central repulsive forces of the Born-Mayer type. Their predicted values are included in Table 1, and can be seen to be consistently substantially lower than the measured values. In analogous models of rutile-structure oxides, Stiefeler and Barsch [32] found that non-central forces had to be included in order to obtain a reasonable fit to data. The discrepancies for  $\text{MgF}_2$  suggest that some such modification of the model is also required in the case of fluorides.

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