

FIG. 1. Reduced plots of the pressure-dependent elastic constants of  $\text{KH}_2\text{PO}_4$  (KDP). Data points and dashed lines represent  $[f(p)/f_0]^2$ , solid lines are the elastic constants (including the corrections for sample dimensional changes).

quantity is  $C_{66}^{E}$ , with  $C_{66}^{P}$  being calculated, when possible, from existing piezoelectric and dielectric data. We have only attempted to evaluate  $C_{66}^{P}$  for KDP and dKDP and the procedure for this calculation will be discussed below. The agreement of the elastic parameters determined in this work with the previous measurements listed in Table II is reasonably good (generally within 1 or 2%). The present data are not considered accurate to better than about 1% (in absolute value), because crystal orientation was not held to a close tolerance and because bonding effect corrections were not made.

## B. High-pressure data

The data for the high pressure runs are shown in Figs. 1-4. For the pulse-superposition technique the experimentally measured quantity is the repetition rate oscillator frequency f as a function of pressure p.

In Figs. 1-4 the measured values of f(p) are indicated as discrete data points connected by smooth curves as visual guides. The following relations hold among f, the ultrasonic round trip transit time t, the ultrasonic path length l, the velocity v, the effective elastic constant C', and the volume V:

$$\frac{f(p)}{f_0} = \frac{t_0}{t(p)} = \frac{l_0 v(p)}{l(p) v_0} , \qquad (2)$$

$$\frac{C'(p)}{C_0'} = \frac{V_0}{V(p)} \left(\frac{l(p)f(p)}{l_0 f_0}\right)^2.$$
(3)

Here the zero subscripts indicate values at atmospheric pressure. In Figs. 1–3 the quantity  $f^2(p)/f_0^2$  is plotted for comparison of this quantity (which represents the high-pressure elastic constants without correction for dimensional changes of the sample) with the actual (corrected) elastic constants (see below). In Fig. 4 the ADP data are plotted as  $f(p)/f_0$ .



FIG. 2. Same as Fig. 1 for KD<sub>2</sub>PO<sub>4</sub> (dKDP).



FIG. 3. Same as Fig. 1 for RbH<sub>2</sub>PO<sub>4</sub> (RbDP).

The data in Figs. 1-4 show the important results of this series of experiments. Modes 1  $(C_{11})$  and 2  $(C_{33})$  show increases in velocity with increasing pressure which are typical of crystals with compressibilities of the same magnitude as those of the KDP-type crystals studied here, and therefore we conclude that the  $C_{11}$  and  $C_{33}$  elastic constants exhibit normal behavior. Modes 5 and 6 in ADP also exhibit normal pressure dependences, so that there is no reason to expect any anomalous behavior for the  $C_{12}$  or  $C_{13}$  elastic constants.

Modes 3  $(C_{44})$  and 4  $(C_{66})$ , on the other hand, show strikingly unusual pressure dependences in all four materials studied. These two modes, which are the two pure shear modes propagating along an *a*-axis and polarized either along the *c* axis or along the other *a* axis, exhibit strongly nonlinear behavior over the pressure range of the measurements.

For KDP and dKDP the modes show pressure dependences that appear nearly identical, with the anomalous mode velocities first increasing with increasing pressure, then bending over and decreasing with further increase of pressure. The maxima of the curves for the two materials occur at roughly the same pressures; for KDP the  $C_{66}$  mode maximum is at ~ 6 kbar compared to ~ 8 kbar for dKDP, while the  $C_{44}$ mode maximum is at ~ 11 kbar for KDP and ~13 kbar for dKDP. These results suggest immediately that deuteration has little, if any, effect on the anomalous elastic properties.

For RbDP (Fig. 3) the  $C_{44}$  mode repetition rate frequency is nearly independent of pressure up to ~6 kbar and subsequently begins to decrease with increasing pressure. The  $C_{66}$  mode for this material has a negative slope at all pressures and exhibits significant curvature as a function of pressure.

The results for ADP (Fig. 4) show a qualitative difference from the other three crystals in that the  $C_{44}$  mode decreases more rapidly with pressure than does the  $C_{66}$  mode, just the opposite of the behavior seen in Figs. 1–3. The  $C_{66}$  mode repetition frequency has a maximum at ~3 kbar. Again, both modes exhibit strongly nonlinear pressure dependences.

## C. Analysis of data

Although the data points in Figs. 1-4 illustrate the salient qualitative features of the elastic anomalies for the KDP-type crystals we have studied, it is nevertheless instructive to proceed further and use the data to actually determine the pres-



FIG. 4. Reduced repetition rate data  $f(p)/f_0$  for NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> (ADP).

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