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MANDELSTAM-BRILLOUIN AND ULTRASONIC INVESTIGATION OF CsH_2AsO_4 NEAR CURIE POINT

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All elastic stiffnesses of CsH_2AsO_4 have been measured at phase transition region by means of ultrasonic and Mandelstam–Brillouin methods. The behaviour of elastooptic coefficients p_{66} and p_{31} was studied also. An anomaly of p_{66}^E near T_o was found. The results are discussed on the basis of thermodynamic theory.

CsH_2AsO_4 undergoes a phase transition of the first order from the paraelectric $\bar{4}2m$ to ferroelectric $mm2$ phase at 143 K [1]. The crystal is isomorphous to other crystals of the KH_2PO_4 family. The ferroelectric transition in KH_2PO_4 has been investigated in detail.[1,2] The physical properties of CsH_2AsO_4 near Curie point have not been completely studied. Dielectric, elastic, thermal and electrooptic measurements of the crystals have been made.[3–5] Raman[6] and Mandelstam–Brillouin[7] scattering of light were investigated also. However, only some elastic stiffnesses of CsH_2AsO_4 have been measured so far. The behaviour of photoelastic coefficients near phase transition region has not been studied yet. Moreover, the results are inconsistent in many cases and differ considerably from sample to sample.[3,7] These circumstances obscure the understanding of the transition mechanism in CsH_2AsO_4. Therefore more complete and thorough measurements of properties near the Curie point are required.

Here we shall discuss the results of Mandelstam–Brillouin and ultrasonic investigations of CsH_2AsO_4 in the region of the ferroelectric phase transition. Samples from the same crystal were used in these experiments.

The scattered light was collected at 90° to the incident beam and analysed by Fabry-Perot interferometer IT-51. The spectra were excited with He-Ne laser LG-38 (λ = 6328 Å) and were detected with help of cooled photomultiplier and photon counting techniques.

The corresponding geometries of scattering for elastic and elasto-optic measurements are shown in Table I. Room temperature values of density (ρ = 3.43 g·cm^{-3} [3]) and refractive indices[8] were used for c_{ij} and p_{ij} calculations. In order to maintain constant electrical boundary conditions silver electrodes were applied to z-faces. The absolute precision of temperature measurement was ±0.02 K. The gradients on a sample were less than 0.1 K · cm^{-1}.

The Mandelstam–Brillouin spectra of CsH_2AsO_4 at several temperatures are shown in Figure 1. The elastic stiffnesses c_{11}, c_{33} and $c_{ef} = (c_{11} + c_{12})/2 + c_{66}$ were determined from the spectra.

In order to obtain all components of the elastic tensor, pulse ultrasonic method with 10 MHz frequency[9] was used. The stabilization of temperature and sample gradient was about ±0.01 K. The ultrasonic velocities versus temperature are shown in Figure 2. Good agreement between the elastic properties determined from the Mandelstam–

TABLE I

The geometries of light scattering in Mandelstam–Brillouin experiments

$\bar{K}_i$	$\bar{K}_s$	$\bar{Q}_{ph}$	$I \sim p_{ij}^2 n^8/\rho v^2(n+1)^4$	q_{light}	n
[$\bar{1}$10]	[110]	[100]	$p_{31}^2 n_c^8 (c_{11}(n_c+1)^4$	⊥	$n_c = 1.546$
[10$\bar{1}$]	[101]	[001]	$p_{13}^2 n_a^8/c_{33}(n_a+1)^4$	⊥	$n_a = 1.567$
[$\bar{1}$00]	[010]	[110]	$p_{31}^2 n_c^8/0.5(c_{11}+c_{12}+2c_{66})$	⊥	
[$\bar{1}$00]	[010]	[110]	$p_{66}^2 n_a^8/0.5(c_{11}+c_{12}+2c_{66})$	∥	

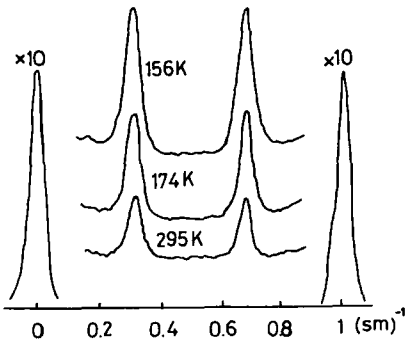

FIGURE 1 Mandelstam–Brillouin spectra of CsH_2AsO_4 with $x(y,x)y$ scattering geometry at several temperatures.

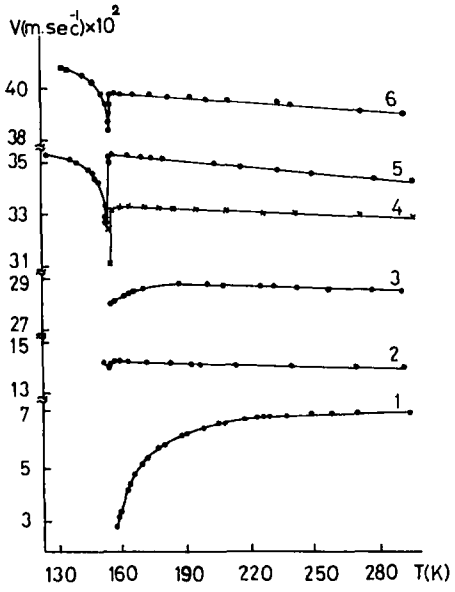

FIGURE 2 Temperature dependences of the velocities of longitudinal and transversal waves in directions: [100] [010] (1), [001][100] (2), [110][110] (3), [101][101] (4), [001][001] (5) and [100] [100] (6).

Brillouin and ultrasonic experiments was obtained. Hence, there is no acoustic dispersion in CsH_2AsO_4 up to 10 GHz frequency. Considerable changes of some velocities at the Curie point are clearly seen. The value of shear velocity determining c_{66} decreases when $T \to T_0^+$ with a tendency towards zero.

The ultrasonic measurements enable us to estimate the difference of transition temperature T_0 and the Curie–Weiss temperature of clamped sample T_c^x as $T_0 - T_c^x = (19 \pm 1)$ K. The difference of Curie–Weiss temperatures for free and clamped states is $T_c^X - T_c^x = (18 \pm 2)$ K. The Curie constant D

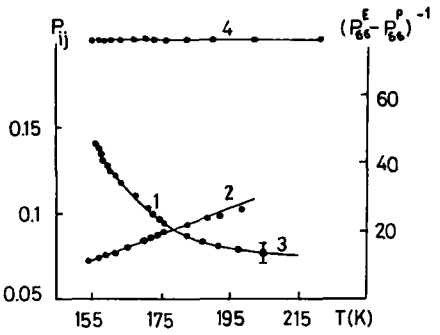

FIGURE 3 Temperature behaviour of elastooptic coefficients: P_{66}^E (1), $(p_{66}^E - p_{66}^P)^{-1}$ (2) and p_{31}^E (3).

equals $66 \cdot 10^{11}$ K·cm²·dyn^{-1}. These estimates are in agreement with Garland's data.[3]

The temperature behaviour of elastooptic coefficients p_{66} and p_{31} for electrically shorted and free samples is shown in Figure 3. These values, within error limits, do not depend on electrical boundary conditions. The value of p_{66} at room temperature was determined from the relation

$$\frac{I_2}{I_1} = \frac{n_c^8 p_{31}^2}{n_a^8 p_{66}^2} \tag{1}$$

For p_{31} the spectra of fused quartz and CsH_2AsO_4 crystal were recorded under the same experimental conditions. The formula

$$\frac{I_{fq}}{I_{cr}} = \frac{(n^8 p_{12}^2)_{fq}(c_{11} + c_{12} + 2c_{66})}{2(n_c^8 p_{31}^2)(c_{11})_{fq}} \tag{2}$$

where I—maximal values of the Mandelstam–Brillouin intensities was used. The values of elastooptic coefficients at room temperature are: $p_{31}^E = 0.2 \pm 0.005$; $p_{66}^E = 0.065 \pm 0.007$.

The elastooptic coefficient p_{66}^E increases when $T \to T_0^+$ and just above T_0 it equals 0.14. Large Rayleigh scattering does not give the opportunity to measure p_{66}^E in the neighbourhood of T_0. It follows from thermodynamics[10] that

$$p_{66}^E = p_{66}^P + m_{63} a_{36} \varepsilon_3^x \tag{3}$$

where m_{63} is electro-optic constant, a_{36}—piezoelectric coefficient, ε_3^x—clamped dielectric permeability. Hence, the increase of p_{66} can be explained by ε_3^x anomaly. Really, the $|(p_{66}^E - p_{66}^P)^-$ dependence follows Curie–Weiss law at 30° interval above T_0. The estimates show that in this case

$T_0 - T_c^x = (21 \pm 2)$ K. The Curie constant D^* equals 2.16. The estimate of p_{66}^P with available data on m_{63} and a_{36}[3,5] shows that this value is equal to 0.04. Thus the behaviour p_{66}^E can be described approximately by the equation

$$p_{66}^E = 0.04 + \frac{2.16}{(T - 135\ \text{K})} \tag{4}$$

The transition temperature T_0 of our CsH_2AsO_4 samples determined from Mandelstam–Brillouin and ultrasonic experiments is equal to 155.5 K.

The elastooptic coefficient p_{31} does not depend on temperature (Figure 3). It also does not change with external electric fields up to 2000 $V \cdot cm^{-1}$.

Thus, the values of $(T_0 - T_c^x)$ and the temperature regions where the Curie–Weiss law for proper quantities is fulfilled are nearly the same from the Mandelstam–Brillouin and ultrasonic measurements.

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