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SOFT PHONON AND CENTRAL PEAK IN RbCaF₃

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Résumé. — Nous présentons brièvement quelques résultats concernant les premières expériences de diffusion inélastique des neutrons qui ont permis de mettre en évidence un pic central et un soft phonon près de la transition structurale à 193 K dans RbCaF₃.

Abstract. — We briefly present some results of preliminary neutron scattering experiments showing unambiguously the coexistence of a central peak and a soft phonon near the 193 K phase transition in RbCaF₃.

1. Introduction. — In the fluoperovskite family into the general formula AMF₃, up to now only KMnF₃ has been known for its structural phase transition due to MF₆ octahedra rotations [1]. Nevertheless ionic radius and force constants investigations allowed us to forecast the possibility of such rotations in the fluoperovskites containing M ions large enough to determine the dimensions of the cell and leave the A ions free to move with respect to the octahedra [2]. Indeed both X-ray diffraction and Raman scattering experiments have shown that RbCdF₃, TlCdF₃ and RbCaF₃ undergo a transition from a cubic (O₃) to a tetragonal (D₁₄) space group respectively at 124 K, 191 K and 193 K [2, 3, 4, 5]. These transitions are due to the condensation of a vibrational mode associated with the R point of the cubic Brillouin zone similar to the 105 K phase transition in SrTiO₃ and to the 186 K phase transition in KMnF₃.

In the quadratic phase, the temperature dependence of the soft mode may be studied by Raman scattering experiments [2], [7, 8]. As this is no longer possible in the cubic phase, inelastic neutron scattering experiments are helpful to understand the behaviour of this mode [6]. Owing to the great absorption of neutrons by cadmium such experiments are impossible on RbCdF₃ and TlCdF₃ but may be carried out on RbCaF₃. The first measurements performed at Oak Ridge [9] confirmed that the transition arises from the condensation of an R₂₅₅ zone boundary phonon. We briefly present in this paper some new results showing that as in e.g. SrTiO₃, this phonon softening is accompanied by the appearance of a central peak in RbCaF₃.

2. Experimental. — We have used a good single crystal (volume 2 cm³, mosaic spread less than 3' of arc) grown by the Bridgman-Stockbarger technique. Preliminary measurements were carried out on the EL 3 Saclay reactor, from which a rigid ion model was constructed and dynamical structure factors calculated.

Energy scans at the R point of the Brillouin zone were performed on the triple-axis spectrometer IN 2 at the I.L.L. High-Flux Reactor. The incident neutron energy was fixed at 4.9 meV from a pyrolytic graphite (002) monochromator. Higher-order contamination was reduced to a negligible level using a Ge (111) analyser combined with a cooled beryllium filter. The crystal was mounted in a liquid helium cryostat assuring a stability of temperature better than 0.03 K.

In order to perform the proper resolution corrections we have measured the curvatures of the dispersion surface in the vicinity of the R₂₅₅ mode respectively in the R-X, R-M and R-Γ' directions. The calculations are in progress and we just present here a summary of the most important rough data.
3. Results. — The critical temperature $T_c$ was determined by monitoring the intensity of a $(\frac{1}{2} \frac{1}{2})$ R point as a function of temperature. The results (Fig. 1) are consistent with our X-rays measurements giving $T_c = 193 \pm 1$ K.

![Graph showing temperature dependence of elastic scattering at $(\frac{1}{2} \frac{1}{2})$ R point.](image1)

**Fig. 1.** Temperature dependence of the elastic scattering at the $(\frac{1}{2} \frac{1}{2})$ R point.

Figure 2 shows high resolution constant-$Q$ scans at the $(\frac{1}{2} \frac{1}{2})$ R point performed successively for $T - T_c = 31$, 22, 19 and 8 K. This figure reveals two important features:

- the $R_{25}$ soft phonon,
- a narrow central peak whose linewidth is that of the instrumental resolution. We may notice that this component already appears at 22 K above $T_c$.

Better statistic will be necessary in order to decide whether or not a quasi-elastic contribution is contained in the rather broad central component which lie at the bottom of the central peak (scans at 212 K and 215 K).

In order to measure the anisotropy of the correlation between octahedra we have done constant $\omega = 0$ scans at 224 K, 215 K, 212 K and 201 K.

![Graph showing high resolution constant $Q$ scans at $(\frac{1}{2} \frac{1}{2})$ for T = 224 K, 215 K, 212 K and 201 K.](image2)

**Fig. 2.** High resolution constant $Q$ scans at $(\frac{1}{2} \frac{1}{2})$ for $T = 224$ K, 215 K, 212 K and 201 K.

![Graph showing constant $\omega = 0$ scan along a line joining M and R points.](image3)

**Fig. 3.** Constant $\omega = 0$ scan along a line joining M and R points.
scans in the vicinity of the \((\frac{1}{2} \frac{1}{2} \frac{1}{2})\) R point respectively in the \((0 0 Q)\) and \((3 \xi \xi 0)\) directions. Contrary to NaNbO₃ recently studied by F. Denoyer et al. [10], for RbCaF₃, even at 80 K above \(T_c\), the elastic intensity is mainly restricted to the vicinity of the R point (Fig. 3). Further experiments near other R points are required to separate the \(T_s\) and \(T_2\) phonons contribution from the quasielastic critical scattering.

4. Conclusion. — From these first measurements, we conclude that RbCaF₃ with its well separated underdamped soft phonon and central peak shows a behaviour very similar to SrTiO₃. These results are very promising because RbCaF₃ seems to be the first fluoperovskite for which a full determination of the soft phonon and central component parameters as a function of temperature may be possible. Further experiments are planned in the near future and should provide a better understanding of these features.

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References