FIZIKA VOLUME 10 SUPPLEMENT 2, 1978
Proceedings of the VI Yugoslav Symposium of the Physics of Condensed Matter, Kruševac, September, 18-22, 1978

PECULIARITIES OF COHERENT CRITICAL NEUTRON SCATTERING IN HYDROGEN-BONDED FERROELECTRICS

S.Stamenković, R.B.Žakula
The "Boris Kidrič" Institute of Nuclear Sciences-Vinča,
Laboratory of Theoretical Physics

V.L.Aksenov
The Joint Institute for Nuclear Research, Dubna, SSSR

Abstract. On the basis of the dynamical pseudospin-phonon model the peculiarities of the coherent critical neutron scattering in hydrogen-bonded ferroelectrics are analysed. The experimental conditions are predicted in terms of the soft-mode parameters, temperature and momenta of the initial neutrons to observe such specific quasielastic neutron scattering.

In the recent paper of two authors[1)] the differential cross section for the slow neutron scattering by KDP-type ferroelectrics has been derived in the complete consistency with the VSK-model[2)] and the "unique fitting"[3)] of the corresponding experimental results. For this purpose the dynamics of the neutron scattering by the soft-mode is of a particular interest.

The peaks in emission (-) and absorption (+) intensities of scattered neutrons are determined by the energy conservation law

$$\frac{h^2p'^2}{2m} - \frac{h^2p^2}{2m} = \pm E_{\vec{q}+\vec{g}} \qquad (1)$$

Here $\vec{q}=\vec{p}-\vec{p}'$ is the scattering vector, m-the neutron mass and $\vec{g}$- the reciprocal lattice vector. By neglecting anisotropic effects (due to dipolar interaction) the soft-mode of the Cochran type is given in the form[2)]

$$E_q^2 = P|T-T_c|+Qq^2 \qquad (2)$$

(P different above and below T_c)

In the case of the quasielastic scattering $p' = p + \eta$, $\eta \gtrless 0$, and from (1) and (2), up to η^2 terms, for every scattering angle θ (closed by $\vec{G} = \vec{g} + \vec{p}$ and $\vec{p}'$) and for the scattering of both types (+,-) one obtains two autgoing neutron momenta

$$p'_{1,2} = p + \eta_{1,2} = p + \frac{Gp_1^2}{p_1^2 - p^2}\left[\cos\theta - \frac{p}{G} \pm \sqrt{(\cos\theta - \cos\theta_1)(\cos\theta - \cos\theta_2)}\right]; \quad (3)$$

$$\cos\theta_{1,2} = \frac{1}{Gp_1^2}\left[p^3 \pm \sqrt{(p^2 - p_1^2)(p^4 - p_2^4)}\right]. \quad (4)$$

As one can easily see these momenta are restricted by the conditions

$$p < p_{1,2}; \quad \left| p^3 \pm \sqrt{(p^2 - p_1^2)(p^4 - p_2^4)} \right| < p_3^3; \quad (5)$$

$$p_1 = \sqrt{b}, p_2 = (bG^2 + a|T - T_c|)^{1/4}, p_3 = (bG)^{1/3}; \quad a = pm^2/h^4 \quad b = Qm^2/h^4.$$

The sign of $\eta_{1,2}$ determines the type of scattering ($\eta_{1,2} > 0$, (+); $\eta_{1,2} < 0$, (-)) in a cone of the width θ_1 round and along the vector $\vec{G}$. A similar physical situation, but at T~0, occurs in magnon[4] or polarization-waves[5] systems. The case $p = p_1$ is a specific one so then only one solution is possible

$$p' = p + \eta = p + \frac{p_1^2 + G^2 - 2p_1 G\cos\theta + p_1^{-1} a|T - T_c|}{2(G\cos\theta - p_1)}. \quad (6)$$

Here, for $p_1 = G\cos\theta$, there is no singularity in fact, as in η^3 order approximation one obtains again two solutions, $\eta_{1,2} = \pm \sqrt[3]{a|T - T_c|/p_1}$. The scattering inside a θ_2-cone is also possible but as predominantly of the "backwards" type it is not of special interest.

In a further analysis a small parameter $\varepsilon=a|T-T_c|$ and the ratio $\lambda=b/G^2$, $\lambda>0$, are introduced. Then from (4), (5) and (6) seven characteristic ingoing momenta appear (as functions of the model parameters (P,Q), ε and the intensity of the reciprocal lattice vector $|\vec{g}|$):

$$p_1 = \lambda^{1/2}G, p_2 \simeq \lambda^{1/4}G(1+\varepsilon/4\lambda G^4), p_3=\lambda^{1/3}G \,, \tag{7}$$

$$p_{4,5} \simeq G(1+\alpha\varepsilon^{1/2}+\ldots), p_6 \simeq G\alpha\varepsilon^{1/2}, p_7 \simeq G(1+\varepsilon/2\lambda G^4); \alpha=(B^{-1}+G^{-2})^{1/4}/G^{1/2}$$

while the characteristic values of λ are:

$$\lambda_o \simeq 1+\varepsilon/G^4; \quad \lambda_1 \simeq 1+3\varepsilon/G^4. \tag{8}$$

Possible intervals for p and θ as well as the scattering types (+,-) are given in the table.

$\cos\theta$	$n_{1,2}$	sign	$\lambda > \lambda_o$	$\lambda \leqslant \lambda_o$
$\frac{p}{G} \leq \cos\theta \leq 1$	n_1	+	$p_4<p<G$; $p_6<p<p_5$	$p_6<p<p_1$
		-		
	n_2	+	$p_4<p<G$; $p_6<p<p_5$	$p_6<p<p_1$
		-		
$-1 \leqslant \cos\theta < \frac{p}{G}$	n_1	+	$p_1<p<p_2$	
		-	$p_4<p<p_1$; $p_6<p<p_5$	$p_6<p<p_1$
	n_2	+		
		-	$p_4<p<p_2$; $p_6<p<p_5$	$p_6<p<p_1$
$\cos\theta_o = \frac{p}{G} \leq 1$	n_1	+	$p_4<p<G$; $p_6<p<p_5$	$p_6<p<p_1$
		-		
	n_2	+		
		-	$p_4<p<G$; $p_6<p<p_5$	$p_6<p<p_1$

For small intervals $\lambda_o<\lambda<\lambda$ and $1<\lambda<\lambda_o$ possible p are then in the interval $p_6<p<p_5$.

For a characteristic scattering angle $\theta=\theta_o$, i.e.

$$\cos\theta_o = \frac{(p/G)^2-\cos\theta_1\cos\theta_2}{2(p/G)-(\cos\theta_1+\cos\theta_2)} \tag{9}$$

there is only one p´ in two cases

$$p/G<\cos\theta\leqslant 1, \qquad \eta_1\neq 0, \quad \eta_2=0;$$
$$-1\leqslant\cos\theta<p/G, \qquad \eta_2\neq 0, \quad \eta_1=0; \tag{10}$$

if $\cos\theta=\cos\theta_1$, $\cos\theta_2\neq p/G$, one finds the characteristic values $\eta_1=\eta_2$, while in the case $\cos\theta_{1,2}=p/G$ a pure elastic scattering occurs. For the special case $p=p_1$ the scattering is characterized by the conditions

$$\eta>0, \quad p_1/G<\cos\theta\leqslant 1; \tag{11}$$
$$\eta<0, \quad -1\leqslant\cos\theta<p_1/G;$$
$$\eta=0, \quad \cos\theta=(p_1^4+p_1^2G^2+\varepsilon)/2p_1^3G\leqslant 1.$$

From the definition (4) it is obvious that the sacattering is possible for all angles θ. But if θ_1 is small enough then the scattering at small angle occurs, which is of the most interest in experiments. Such a scattering develops when $p\simeq p_3=p_1$, $p\simeq p_3=p_2$, $p\simeq p_1=p_2=p_3$, $p\simeq 0$* (under condition $p_1=G$) as well as when $p\simeq p_4$, p_5, p_6. In all these cases with $p=p_1$ the scattering is possible only for $\lambda<1$ but for every ε.

From the general expressions for the neutron scattering by the coupled (hibridized) proton-optic phonon ("soft") mode[1] the angular differential cross section in the neighbourhood of the recirpocal lattice vector $\vec{g}$ becomes

$$\left(\frac{d\sigma}{d\Omega}\right)^{\mp}_{coh}=N\sum_{j=1,2}\left[F_{gp}(E_{kj}^2-\omega_{kj}^2)+F_{gi}(E_{kj}^2-\varepsilon_{kj}^2)\right]\frac{n(E_{kj})+1/2\mp 1/2}{E_{kj}A_{kj}}$$
$$\frac{(p+\eta_j)^2|\eta_j|}{|p^2\eta_j-b(p-G\cos\theta)|}; \quad k_j^2=\frac{1}{b}(p^2\eta_j^2-\varepsilon). \tag{12}$$

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*Actually[6] $p>2\pi/aN^{1/3}$

The form-factors F_{kp} and F_{ki} stand for proton (p) and heavy ion (i) scattering quadratic amplitudes as well as for the peculiar Debye-Waller factors, respectively, and together with the quantity $A_k=E_k^+-E_k^-$, the pure proton (ω_k) and the phonon (ε_k) modes are given in ref. 1 (see also ref. 2). Note that in the case $p=p_1$ the following substitution holds

$$\frac{(p_1+\eta_{1,2})|\eta_{1,2}|}{|p_1^2\eta_{1,2}-p_1^2(p_1 - G\cos\theta)|} \Rightarrow \frac{(p_1+\eta)^2}{p_1^2}; \qquad (13)$$

$$k_{1,2}^2 \Rightarrow k^2=\eta^2-\varepsilon/p_1^2$$

On the basis of the present analysis the soft-mode parameters can be determined directly from the corresponding experimental results. For the real systems in the fluctuating spectra of the order ε a central peak appears in addition to the soft-mode peak at T_c and $k\to0$. However, for real systems one should include the anisotropic effect due to dipolar interactions. Therefore, the obtained angular intensity (12) should be improved by taking into account the width of the central peak as a function of a frequency dependent dynamical susceptibility of the system[7)]

To conclude, the peculiarities predicted by the present analysis are quite general and should hold for all systems (say, magnon)[8)] in which any kind of structural phase transition could occur.

REFERENCES:

1. V.L. Aksenov, S. Stamenković, FTT 19. 1366 (1977) (in Russian)

2.

2. J.Villain, S.Stamenković, Phys.Stat.Sol. 15, 585 (1966) K.K.Kobayashi, J.Phys.Soc. Japan 24, 497 (1968)

3. R.J.Elliott, S.R.P. Smith, A.P. Young, J.Phys. C4, L. 317 (1970)

4. S.V. Maleev, ZhETF 33, 1010 (1957) (in Russian)

5. S.Stamenković, J.Low Temp. Phys. 9, 47, 485 (1970)

6. R.J.Elliott, R.D. Lowde, Proc.Roy.Sos. 230, 46 (1955)

7. V.L.Aksenov, Dynamics of Hydrogen-Bonded Crystals, to be published in proceed of National Conf. Cond. Matter, Alushta, USSR (1978)

8. E.A.Ginzburg, I.Achieser, Phys, Lett. 37A, 63 (1971) J.Todorović, Solid State Commun. 21, 919, (1977).