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New Type of Soliton Solutions from a Landau Potential Describing the β - γ - δ -Transitions in $(C_3H_7NH_3)_2MnCl_4$

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Abstract: The incommensurate γ -phase of the perovskite-type layer structure compound $(C_3H_7NH_3)_2MnCl_4$ is sandwiched between two phases (β, δ) which have the same structure. The Landau potential describing this behaviour reveals besides the plane-wave solution also a new type of soliton solutions which differ from the solutions of the sine-Gordon equation.

Introduction

Perovskite-type layer structures of the formula $(C_nH_{2n+1}NH_3)_2MCl_4$ with short hydrocarbon chains ($n < 5$) are known to exhibit several structural phase transitions which are connected with reorientational jumps of the alkylammonium chains /1/. Within this family the compound $(C_3H_7NH_3)_2MnCl_4$ is a special case because of its complicated phase sequence with two incommensurate phases /2,3/. The different phases are denoted as $\alpha, \beta, \gamma, \delta, \epsilon$ and ζ .

$P2_1/n$	Inc.	Abma	Inc.	Abma	I4/mmm
$Z=6$	$\vec{q} \approx \frac{1}{3}\vec{b}^*$	$Z=4$	$\vec{q} \approx \frac{1}{6}\vec{a}^* + \vec{c}^*$	$Z=4$	$Z=2$
ζ	ϵ	δ	γ	β	α
110 K	165 K	344 K	396 K	443 K	T

The most interesting feature is the reentrant behaviour of the γ - δ -transition. It was shown by means of NMR-NQR that the β and the γ -phase have indeed the same structure /4/. They differ only in the saturation of the order parameter of the α - β -transition. This reentrant behaviour could be well described by a

Landau-type free energy /4/. It was shown that a plane wave modulation is an exact solution of the corresponding Euler equations. In this contribution we want to stress also soliton-like solutions in order to explain the observed types of x-ray satellite reflections.

Incommensurate Wave Vectors of the γ -Phase

An x-ray analysis of the γ -phase revealed three types of satellite reflections /2/:

- type A1: $\vec{q}_1 = \alpha \vec{a}^* + \vec{c}^*$, $\alpha \approx 0.17$, strong *)
 A2: $\vec{q}_2 = 2\alpha \vec{a}^*$, weak
 B : $\vec{q}_3 = \beta \vec{a}^* + \vec{c}^*$, $\beta \approx 0.05$, weak

$\vec{q}_1$ and $\vec{q}_3$ are zone-boundary vectors on the H line near the Y point, $\vec{q}_2$ is on the Λ line (notation according to ref.5). The type A2 satellites are obviously due to a higher harmonic of the type A1 modulation and are generated by a third-order anharmonic potential $V_3(Q_{q_1}^2 Q_{-q_2} + Q_{-q_1}^2 Q_{q_2})$. The origin of the B-type modulation is still an open question.

The commensurate part of the modulation ($=\vec{c}^*$) destroys the A-centering of the unit cell. The superspace group compatible with the A1 reflections is N_{111}^{Abma} /2/. The soft mode leading to this superspace group must transform according to the irreducible representation H_1 , which splits at the Y point into the one-dimensional representations Y_1^+ and Y_3^- having at the Γ point x^2 and x symmetry respectively. At the Y point there is no degeneracy of modes and therefore no Lifshitz invariant is allowed. At the other end of the H line, at the T point ($\frac{1}{2}\vec{a}^* + \vec{c}^*$), all modes are doubly degenerate and Lifshitz invariants can be formed. The mode softening with a wave vector close to the Y point is in our case due to a coupling of two modes with Y_1^+ and Y_3^- symmetry.

) $\vec{a}^$, $\vec{b}^*$, $\vec{c}^*$ are given for the A-centered unit cell.

Thermodynamic Potential and Discussion of the Euler Equations

A free energy explaining the reentrant behaviour of the γ - δ -transition is given in ref.4. A coupling of the order parameter with the density of the layers leads to renormalized Landau coefficients $\hat{A}(T)$ and $\tilde{B}(T)$ containing both linear and quadratic terms in $(T-T_0)$. Outside the γ -phase the density of layers must exhibit a linear term in the temperature dependence to provide the reentrant behaviour. The free energy density is thus given by:

$$g = g_0 + \frac{1}{2} \hat{A} \eta \eta^* + \frac{1}{4} \tilde{B} (\eta \eta^*)^2 - \frac{1}{2} \kappa \frac{d\eta}{dx} \frac{d\eta^*}{dx} + \frac{1}{2} \lambda \frac{d^2\eta}{dx^2} \frac{d^2\eta^*}{dx^2}, \quad \kappa, \lambda > 0$$

The plane wave $\eta = A e^{iq_0 x}$ ($A = \text{const.}$, $q_0^2 = \kappa/2\lambda$) is an exact solution of the resulting Euler equations. A more general solution can be obtained within a constant-amplitude approximation ($\eta = A e^{i\phi(x)}$, $A = \text{const.}$). In this case ϕ can be expressed in terms of elliptic integrals of the first kind. Introducing the function

$h = \frac{1}{q_0^2} \left(\frac{d\phi}{dx} \right)^2 - 1$ the Euler equation for the phase ϕ reads after one integration:

$$\left(\frac{dh}{dx} \right)^2 = 4 q_0^2 (h+1)(h^2 - \mu^2)$$

μ : integration const.

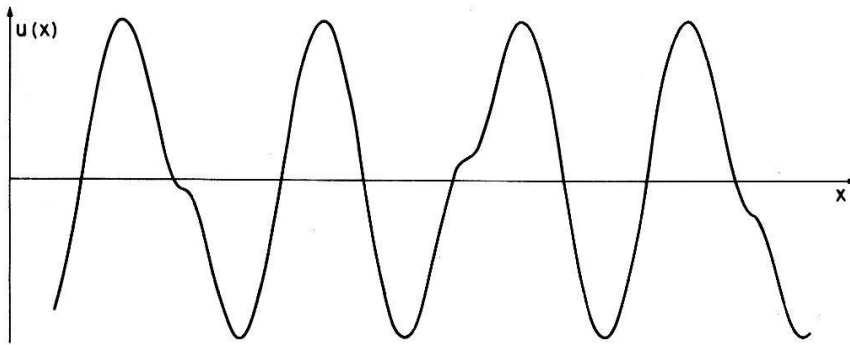


Fig. 1 Normalized atomic displacement $u(x) = \sin(\phi(x))$ for $k = \sin^{-1}(89^\circ)$.

The solution can be evaluated by the integral

$$\chi = \frac{1}{\sqrt{1+\mu}} \frac{1}{q_0} \int_0^{\varphi} \frac{d\varphi}{\sqrt{1-k^2 \sin^2 \varphi}}, \quad h = -1 + (1-\mu) \sin^2 \varphi, \quad k^2 = \frac{1-\mu}{1+\mu}$$

or written in a condensed form:

$$\phi = \int_0^x q_0 \sqrt{1+h(x')} dx', \quad h(x) = -1 + (1-\mu) \sin^2(\text{sn}(q_0 \sqrt{1+\mu} x))$$

In the plane-wave case, $h=0$, $\mu=0$ and $k=1$.

Some results are shown in Figures 1 and 2. In contrast to the solution of the sine-Gordon equation where only higher harmonics are obtained our equation leads also to "subharmonic" parts which would explain the B-type x-ray reflections.

By introducing h into the free energy $F = \int g dV$ one gets:

$$F = F_0 + V \left\{ \frac{1}{2} \left[\hat{A} + \lambda q_0^2 (-1 + H(k)) \right] A^2 + \frac{1}{4} \hat{B} A^4 \right\}$$

$$H(k) = 2 \langle h^2 \rangle - \frac{1-k^2}{1+k^2}$$

The average of h^2 is given by $\langle h^2 \rangle = \frac{\int_0^{\pi/2} h^2(\varphi) d\varphi}{\int_0^{\pi/2} d\varphi} = \frac{\int_0^{\pi/2} \frac{h^2(\varphi) d\varphi}{\sqrt{1-k^2 \sin^2 \varphi}}}{\int_0^{\pi/2} \frac{d\varphi}{\sqrt{1-k^2 \sin^2 \varphi}}}$

The minimum of F with respect to k is obviously independent of A and coincides with the minimum of $H(k)$. This function is shown in Fig.3. It can be seen that the plane-wave solution ($k=1$) has a lower free energy than a solution with a space-dependent $\frac{d\phi}{dx}$. However, our free energy contains only the leading terms necessary to explain both, the incommensurability and the reentrant behaviour. A complete free-energy density up to the fourth order of

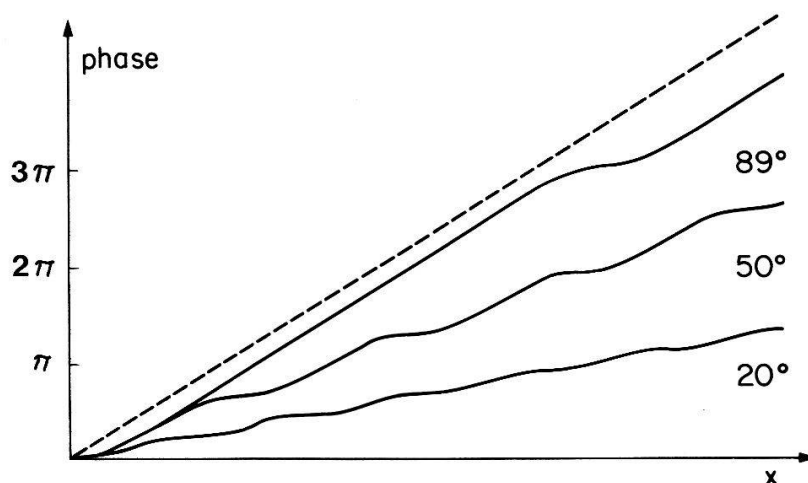


Fig. 2 Phase angle ϕ vs. x for different values of $\sin^{-1}(k)$. The dashed line corresponds to the plane-wave solution.

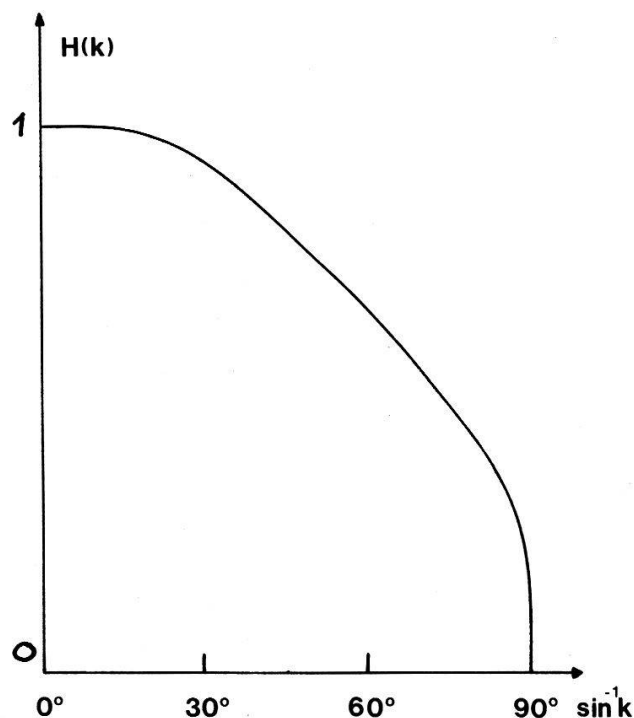


Fig. 3

The function H (as defined on the preceding page) vs. $\sin^{-1}(k)$.

the amplitude should also take into account the terms $\sigma \left(\frac{dn}{dx} \frac{dn^*}{dx} \right)^2$ and $\rho \left(\frac{d^2n}{dx^2} \frac{d^2n^*}{dx^2} \right)^2$. Especially the first of these terms with $\sigma > 0$ would favour a solution with $k < 1$, since with lower k the value of $\langle \frac{d\phi}{dx} \rangle$ is reduced.

The effect of a space-dependent amplitude can not be predicted since it would require the solution of two coupled strongly non-linear Euler equations. It was shown/6/, however, that the amplitude variations do not play an essential part in modulated structures of the β - K_2SO_4 family.

To conclude we can say that our Landau potential doesn't only describe the reentrant behaviour but also explains all kinds of the observed x-ray satellites. The crucial experiment to test our theory should be the measurement of the temperature dependence of the splitting and the intensity of the B-type reflections.

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