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► To cite this version:

Jean Pouget, B. Hennion, M. Sato. Dynamics and charge density wave collective excitations of the blue bronze. *Journal de Physique IV Proceedings*, 1993, 03 (C2), pp.C2-215-C2-220. 10.1051/jp4:1993243 . jpa-00251326

HAL Id: jpa-00251326

<https://hal.science/jpa-00251326>

Submitted on 1 Jan 1993

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Dynamics and charge density wave collective excitations of the blue bronze

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Abstract

Recent neutron scattering investigations of the pretransitional dynamics and of the phase and amplitude excitations of the charge density wave (CDW) condensate of the blue bronze are reviewed. It is shown that the dynamics of the Peierls transition is dominated by the intrachain fluctuations. The key role of Coulomb forces in the intrachain deformation process of the phase of the CDW is also emphasized.

Introduction

Neutron scattering is one of the best experimental technique allowing the study of electron-phonon coupled systems exhibiting structural instabilities such as low dimensional conductors stabilizing CDW ground states. Earlier neutron studies of the one dimensional (1D) metal $\text{K}_2\text{Pt}(\text{CN})_4\text{Br}_{0.3}\cdot\text{H}_2\text{O}$ (KCP) have revealed that the CDW instability could give rise to a giant Kohn anomaly in the phonon spectrum at the wave vector $2k_F$ (k_F is the Fermi wave vector of the 1D electron gas) [1]. In contrast other families of CDW materials such as the 1D charge transfer salt TTF-TCNQ or the 2D transition metal dichalcogenides 2H-TaSe_2 and NbSe_2 revealed only a smooth Kohn anomaly not very critical in temperature and which peculiar behavior could be due to strong electron-electron or electron-phonon interactions.

The displacive nature of the pretransitional fluctuations as well as the collective excitations of the CDW condensate predicted by the standard theory of the Peierls transition could not be studied in detail because of the frozen dynamics and of the spreading out of the Peierls transition due to the intrinsic disorder of KCP. It's only recently that such an investigation could be performed in the blue bronze $\text{K}_{0.3}\text{MoO}_3$ [2, 3]. This bronze is an ordered quasi-1D conductor undergoing a 2nd order Peierls transition at $T_p = 183 \text{ K}$, temperature below which a 3D long range sinusoidal modulation of the lattice positions is observed at the critical wave vector $\mathbf{q}_p = (1, 2k_F, 0.5)$, where $2k_F$ is an incommensurate value close to 0.75 [4]. In the following we shall summarize the main results of the neutron scattering investigation performed at the Orphée reactor of the LLB-Saclay on a large single crystal of $\text{K}_{0.3}\text{MoO}_3$ of a volume of $\sim 5 \text{ cm}^3$

$T > T_p$: The Kohn Anomaly

The pretransitional dynamics of the Peierls transition of the blue bronze consists of the softening of an acoustic-like phonon branch at the wave vector $2k_F$ together with the critical growth in the vicinity of T_p of a central peak in frequency. Figure 1 shows a 3-D display of the Kohn anomaly just above T_p at 190 K. A damped harmonic oscillator analysis of the neutron scattering cross section clearly reveals the softening of the quasi-harmonic frequency (ν_K), but also an enhancement of the damping (γ_K) of the critical phonon branch at $2k_F$. The thermal dependence of these quantities is shown fig.2a. γ_K quantitatively accounts for the excess thermal conductivity observed above T_p in the blue bronze [5]. An analysis of the dispersion of the Kohn anomaly in the vicinity of its minimum ν_K for wave vectors parallel to the chain direction (q_b) according to the expression :

$$\nu_K^2(q_b) = \nu_K^2 + \nu_0^2 |q_b - 2k_F|^2 \quad (1)$$

allows, with $\nu_0 = 31 \pm 4 \text{ THz}\text{\AA}$ [6], to estimate at $18 \pm 2 \text{\AA}$ the intrachain thermal coherence length $\xi_{0//}$ near T_p ($\nu_0 = \nu_A \xi_{0//}$, where $\nu_A = 1.73 \text{ THz}$ is the frequency of the amplitude mode frequency at 0°K [7]).

Fig. 1. Neutron-scattered intensity in function of the intrachain wave vector component q_b and of the frequency ν for the reciprocal positions $(1, q_b, 0.5)$ at 190 K (from ref [2]). The arrow shows the excitation branch giving rise to the amplitude mode below T_p

$T < T_p$: The CDW Collective Excitations

The dispersion of two phonon branches, quite well decoupled in frequency and corresponding to the collective excitations of the amplitude and of the phase of the CDW, has been measured below T_p in the

Fig. 2 a) Temperature dependence of the quasiharmonic frequency ν_i and the frequencies $\nu_i \pm \gamma_i$ at $2k_F$ for the Kohn anomaly ($i = K$), the amplitude mode ($i = A$) and the phase mode ($i = P$) of the blue bronze (from ref [2])
 b) Schematic variation of these quantities obtained from the numerical simulation of the dynamics of the Peierls chain of ref [13].

vicinity of q_p satellite reflections. The amplitude mode has an optic-like dispersion (fig. 3a) which is predicted to behave, for q_b close to $2k_F$, as :

$$\nu_A^2(q_b) = \nu_A^2 + 1/3 \nu_0^2 |q_b - 2k_F|^2 \quad (2)$$

The analysis of the experimental data according to the relationship (2) gives, within experimental errors, the same ν_0 as for $T > T_p$ [6]. Neutron [2] and Raman [7] investigations agree perfectly on the values of the quasi-harmonic frequency (ν_A) and of the damping (γ_A) of the amplitude mode at q_p . Fig. 2a shows that ν_A softens only weakly upon heating and does not vanish at T_p , in contrast with the predictions of the mean-field theory of the Peierls transition [14].

The phase mode has an acoustic-like dispersion (fig. 3b) until 100 K, the lowest temperature investigated. The phason velocity exhibits a large anisotropy which is in the vicinity of T_p close to that of the correlation lengths involved in the CDW amplitude fluctuations above T_p [3]. In the temperature range investigated the transverse velocity of the phase mode (respectively of $16 \pm 3 \text{ THz}\text{\AA}$ and $3.6 \pm 0.6 \text{ THz}\text{\AA}$ along the $2a^* + c^*$ and $2a^* - c^*$ directions respectively) remains constant within experimental errors, while the longitudinal velocity of the phase mode shows a considerable stiffening upon cooling (fig. 4a).

Fig. 3 Dispersions along q_b of the amplitude mode at 160 K(a) and of the phase mode at 175 K (b) in the vicinity of $2k_F$ (from ref. [2])

High resolution measurements [2] show that the phason response is overdamped near q_p , as generally expected [8]. A damped oscillator analysis of its ν dependence [2] leads to a small gap whose frequency $\nu_p = 0.2 \pm 0.1\text{ THz}$ agrees quite well with that of a resonance found in optical conductivity measurements and ascribed to the oscillatory response of the pinned collective phase mode of the blue bronze [9].

Coulomb Forces and Intrachain CDW Phase Deformations

The enhancement of the longitudinal phason velocity upon cooling, shown fig. 4a, can be ascribed to the thermal reduction of the screening of the Coulomb potential involved in the intrachain phase deformation process by the carriers excited through the 3D Peierls gap, $\Delta(T)$. In that case, it is predicted that the phason velocity v_ϕ should vary like [10] :

Fig. 4 a) Thermal evolution of the phase mode velocity in chain direction (from ref. [3])
 b) Electron-hole condensate densities f_s^p and f_s^A obtained from the experimental measurements of v_ϕ and v_A respectively (taken from ref [6]). The continuous line represents $f_s(T)$ given by the expression (5).

$$v_\phi^2 \equiv v_o^2 [1 - f_s^p(T)] \quad (3)$$

$$\text{where } f_s(T) = [1 - \frac{T\Delta'(T)}{\Delta(T)}]^{-1} \quad (4)$$

is the electron-hole condensate density [11]. Figure 4b gives $f_s^p(T)$ deduced from the most accurate determination of v_ϕ and the value of v_o previously quoted. It is interesting to remark that these f_s^p values can be accounted for by the expression (4) in which the thermal dependence of the 3D order parameter $\Delta(T)$ is taken from the square root of the $\mathbf{q}_p$ satellite intensity, $I(T)$:

$$f_s(T) = \left[1 - \frac{1}{2} \frac{\partial \text{Log} I(T)}{\partial \text{Log} T}\right]^{-1} \quad (5)$$

$f_s(T)$ thus obtained from $I(T)$ measured in ref. [12] is represented fig. 4b.

Intrachain Dynamics

The mean field theory of the Peierls transition predicts [14] that the amplitude mode frequency ν_A should behave in temperatures as :

$$\nu_A^2 \equiv \nu_A^{\circ 2} f_s^A(T) \quad (6)$$

where $f_s(T)$ is the electron-hole condensate density of the Peierls chain. Figure 4b shows that although f_s^A and f_s^P merge together at low temperature, f_s^A deduced from the square of ν_A / ν_A° remains finite at T_p . By continuity fig. 1 shows that the collective excitations of the amplitude of the CDW can still be observed above T_p . Upon heating these excitations progressively merge into the damped Kohn anomaly. This behavior is qualitatively that expected when the pretransitional dynamics is dominated by the intrachain fluctuations, as illustrated by fig. 2b which shows, on the basis of numerical calculations of the dynamics of the Peierls chain [13], how the 2kF response of the phase and the amplitude degrees of freedom behaves in temperature .

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