Eur. Phys. J. B **28**, 185–186 (2002)
DOI: 10.1140/epjb/e2002-00214-x

THE EUROPEAN PHYSICAL JOURNAL B

Comment on

Low temperature specific heat of blue bronze $K_{0.30}MoO_3$, by J. Odin, J.C. Lasjaunias, K. Biljaković, K. Hasselbach, and P. Monceau

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Eur. Phys. J. B **24**, 315 (2001)

Received 21 February 2002
Published online 19 July 2002 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2002

Abstract. Here we comment on a recently published paper on the presence of a *phason* contribution in the low temperature heat capacity data of the charge-density-wave compounds $K_{0.3}MoO_3$ and $(TaSe_4)_2I$. We have shown that the anomaly in the C_P/T^3 data reported by Odin *et al.* is straightforwardly interpreted in terms of low energy phonon modes resulting from the peculiar topology of these compounds.

PACS. 71.45.Lr Charge density wave systems – 65.60.+a Thermal properties of amorphous solids and glasses: heat capacity, thermal expansion, etc. – 63.50.+x Vibrationalstates in disorderd systems

The understanding of the low temperature specific heat in low dimensional compounds undergoing a Peierls phase transition is the object of intense study and the paper by J. Odin *et al.* [1] intends to shed light onto this problem. Below the Peierls phase transition temperature (T_P), the electronic ground state is characterized by a, typically incommensurate, charge-density-wave (CDW) of period $2k_F$, with k_F the Fermi wave vector, and the appearance of an atomic lattice distortion of the same periodicity in order to locally neutralize the CDW. Two new collective excitations, fundamentally related to the incommensurability, develop below T_P: a low frequency, acoustic-like excitation corresponding to the phase of the modulation slowly varying in space and time (the so-called *phason*) and a higher frequency, optic-like mode corresponding to slowly varying amplitude variations (the so-called *amplitudon*). These modes are peculiar to the wider class of incommensurate modulated systems which includes CDW compounds. They have been observed in a certain number of compounds, but not in all incommensurate systems studied so far, the reason for that being not yet completely understood.

In their paper, Odin *et al.* [1] report on low temperature specific heat measurements on two *canonical* CDW compounds: the blue bronze $K_{0.3}MoO_3$ and $(TaSe_4)_2I$. Among the features described, two are of very special interest:

– Below ≈0.5 K, the appearance of very low energy excitations with a sub-linear temperature dependence characterized by a non-exponential heat relaxation and aging effects. This contribution is reminiscent of the disorder present in amorphous materials and is ascribed to two-level systems tunnelling states. In CDW systems this disorder is interpreted as to originate in the random pinning of the CDW by impurities. This contribution has been studied by authors over the last 15 years.
– Between 1–20 K, a deviation of the phonon T^3–law manifested through a bump in the C_P/T^3 *vs.* T plot. This deviation has been consistently observed in several CDW compounds and its interpretation is the object of this Comment.

Some years ago we raised the point that low dimensional electronic compounds are generally associated to structures of marked low dimensional, anisotropic elastic properties [2,3]. Transverse acoustic modes with very small sound velocity, acoustic modes with extended sections of (nearly) flat dispersion, low energy optic modes etc., are typical signatures that one can find in most of these compounds. Under these circumstances one expects that the lattice contribution to C_P deviates very rapidly from the *usual* Debye behavior ($C_P \propto T^3/\theta_D^3$). The "conventional wisdom" invoked by the authors to determine the range

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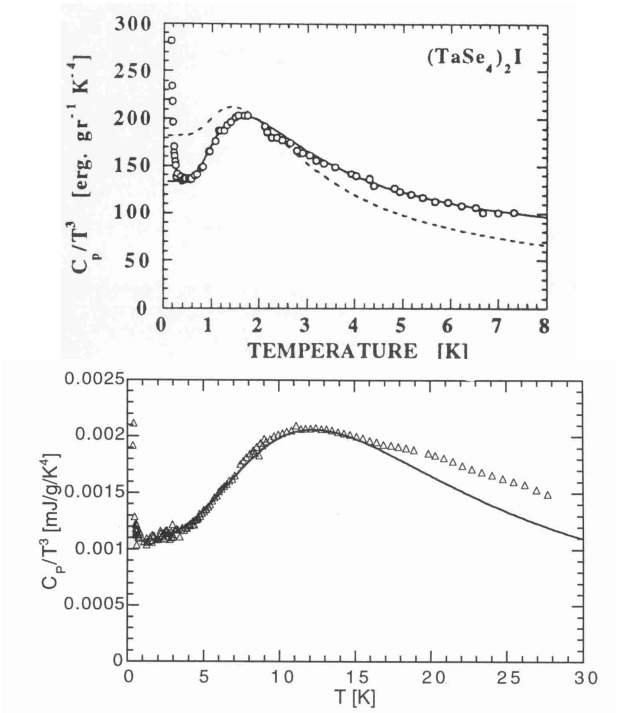

Fig. 1. Low temperature specific data for $(TaSe_4)_2I$ (top) and $K_{0.3}MoO_3$ (bottom) in the standard representation of C_P/T^3 *vs.* T. Top: the dashed line is a calculation based on *weighted* neutron VDOS (no adjustable parameter) and the solid line represents the same calculation using a model that includes the acoustic branches. The parameters for this model (sound velocities, energy of the flat mode, etc.) are extracted from inelastic neutron scattering results [2]. Bottom: the line is the result of an analogous model of the lowest energy branches in $K_{0.3}MoO_3$ [3]. The deviation beyond 17 K is attributed to the remainder of phonon states not considered in the model.

of validity of the cubic regime (down to $T \approx \theta_D/20$) is of no help in this case. One has to carefully examine the phonon density of states or very cautiously compare the thermodynamic behavior of a parent compound that does not exhibit the sought behavior. Low temperature thermodynamic properties above 1 K are described by the vibrational density states (VDOS) alone, and although the Peierls phase transition carries on a softening of a mode at $2k_F$ no appreciable change in the measured VDOS between above and below T_P has been observed in the neutron scattering experiments [2,3]. We have shown that the specific heat above 1 K in the CDW compounds $(TaSe_4)_2I$, $K_{0.3}MoO_3$ and KCP is well reproduced by the *weighted* neutron VDOS extracted through inelastic neutron scattering measurements and by a calculation of the low energy VDOS taking into account the lowest energy (acoustic and optic) branches [2,3]. The two methods offer no adjustable parameter, only known quantities. Figure 1 reproduces the specific heat calculation based on *normal* phonon modes for $(TaSe_4)_2I$ and $K_{0.3}MoO_3$. A detailed description of the calculation can be found in [2,3] and references therein.

The contribution of the *phason* modes to the specific heat has been calculated by Boriack and Overhauser [4] and an excess of the low temperature specific heat for different CDW compounds ($(TaSe_4)_2I$, $K_{0.3}MoO_3$, etc.) has been interpreted accordingly by Odin *et al.* (see references in [1]). However, the intrinsic dynamical behavior of the incommensurate (super-) structure scales with the electron Fermi velocity and is very localized in q-space. The result is that the *phason* contribution to the VDOS amounts to a very small fraction of modes when compared with the *normal* phonons and therefore the signature of the *phason* dynamics in the specific heat data should be negligibly small. This conclusion is supported by the absence of a noticeable *phason* contribution in the neutron measurements on the *weighted* VDOS [2,3] between scans taken below and above T_P.

Indeed, the fit of the specific heat data in terms of Boriack's equation yields an excellent agreement. However, in their final interpretation the authors have completely disregarded the by far dominating role of low energy *normal* phonon excitations which, by themselves, account very well for the experimental data (see Fig. 1) without any free parameter. We believe that the authors' idea for a global interpretation of the low temperature specific heat data in term of *phason* pinning and glassy dynamics is very appealing. However, we have shown that the bump in C_P/T^3 in the heat capacity data originates from low energy anisotropic phonons modes, unrelated to the incommensurability and/or the CDW state. Therefore we think that authors' assessment lacks of firm experimental evidence. Incidentally, we find the parameter $\eta = \sqrt{\eta_1\eta_2} \approx 0.04$ (the anisotropy of the *phason* velocity [5]) ≈8 times larger than in their calculation as well as some inconsistencies as to the definition of the Debye temperature for the *phason* modes. With this value of η the estimation for the *phason* contribution (15–20% of the amplitude of the bump in C_P/T^3) given in [1] is difficult to follow. Finally, during the course of elaborating this Comment we came across another paper [6] on the low temperature thermodynamic properties of the organic chain conductor $(TMTSF)_2AsF_6$, and where a residual phonon contribution to C_P has been found once a *regular* Debye behavior has been subtracted. This residual contribution fits the Boriack-Overhauser equation [4] and has been interpreted as *phason* mode having a vanishingly small gap. As above, we believe that in the absence of inelastic neutron scattering data of the low energy excitations one should be cautious in interpreting the observed bump on C_P/T^3 (or other type of anomalies) in terms of *phason* modes. Whether or not it is possible to observe a *phason* contribution to the specific heat in other incommensurate compounds, is still a matter of controversy.

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