PARTIAL REENTRANCE AND HIGH ORDER NESTING IN THE SPIN DENSITY WAVE INDUCED BY THE MAGNETIC FIELD

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ABSTRACT

A thermodynamic study of the splitting process for the transition lines of the Spin Density Wave induced by the magnetic field (FISDW) is presented. Experimental evidence for a partial reentrance of the metallic phase into the FISDW is given. Magnetocaloric effect and specific heat data are used to analyze the "fractional" transitions in the framework of the fractional quantized nesting model.

INTRODUCTION

In the past years a great deal of attention has been devoted to the FISDW states in the Bechgaard salts [1]. The low-field part of the phase diagram, as determined by several experimental investigations, is qualitatively well understood in terms of an interference effect between the two principal periodicities of these quasi-2D systems: the deviation from the perfect nesting of the SDW modulation and the magnetic wavelength related to the flux quantization. As a consequence the SDW wave vector is quantized so as to maintain the Fermi level in between two adjacent Landau bands, the transport properties are quantized and a cascade of phase transitions induced by the magnetic field is observed [1]. But as the temperature is decreased far below T_c these transition lines are not stable [2,3], and the phase diagram displays a fine structure for very well ordered samples [4]. Here this complexity will be investigated by means of the magnetocaloric effect.

THE PARTIAL REENTRANCE OF THE METALLIC PHASE

The specific heat experiments performed at constant temperature, as a function of the magnetic field, give a direct evidence for the partial reentrance of the metallic phase in between two adjacent quantized FISDW phases. As the field is increased, the specific heat displays first at the threshold field a positive jump (Fig. 1), on going from the metallic state to the first SDW ; then a rounded peak in the SDW; then a negative jump toward the metallic state value, followed for a small field range by a plateau at this value; finally a large positive jump as the system enters the next SDW, and a series of anomalies, marking the cascade of field-induced phase transitions. Specific heat investigation at constant field confirms this phenomenon; in a given SDW the critical temperature T_c is not monotonic: it reaches a maximum, and then decreases as the field gets closer to the reentrance tricritical point. Above this point T_c displays a sudden increase.

Confirmation of this partial reentrance behavior is given by the magnetocaloric experiments performed at constant temperature below the tricritical point. The $(\partial M/\partial T)_B$ vs B curve at 0.425K displays a series of reversible jumps corresponding to the transitions between SDWs (Fig. 2). The jumps are negative, meaning that the electronic system lowers its entropy by entering each ordered state. But this general behavior is not observed in the whole field range: the entropy variation becomes positive just below the first critical fields (dashed areas on Fig. 2a).

This is an indication for a negative slope of the T_c vs B curve, showing that the gap value, and consequently the order parameter, does not change monotonously across a given phase: it increases first and then decreases toward a minimum, for each critical field B_n separating two adjacent SDWs. This reflects the competition between the two neighboring order parameters.

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Moreover, the gap value undergoes a discontinuity at B_n. As indicated by the T_c behavior, it is clear that the gap is much more developed just above B_n.

Such partial reentrances are observed at each transition between SDW sub-phases. The reentrance depth decreases with increasing field up to 8 teslas, where it finally vanishes [5]. Furthermore, the depth at a given tricritical point increases as the electronic mean free path is increased by reducing the cooling rate. This behavior must be compared with the similar influence of the cooling rate on the total reentrance of the metal observed at about 27 teslas [1].

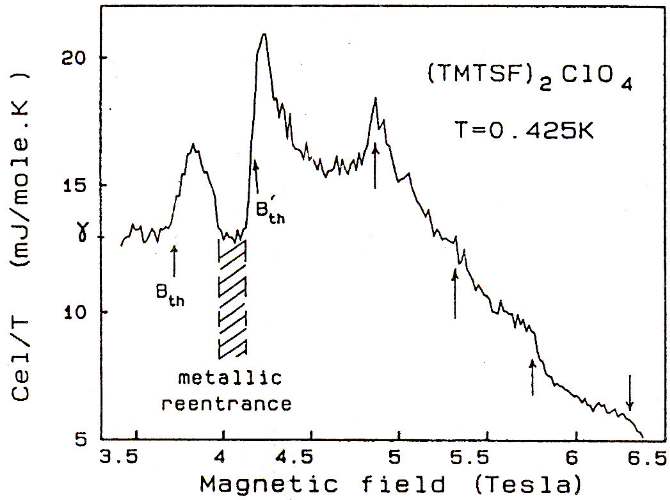

Figure 1: Electronic specific heat, in molar unit, of a 2 mg single crystal of $(TMTSF)_2ClO_4$ (cooling rate: 1.3 K/hour), as a function of the field.

ANALYSIS OF THE SPLITTING PROCESS

Simultaneous measurements of both specific heat, C_B, and magnetocaloric effect, $(\partial M/\partial T)_B$, allow us to build up a complex phase diagram for the very well relaxed (1.3 K/h) perchlorate compound. Firstly, sharp peaks in C_B and $\partial M/\partial T$ are associated with the transitions between "integer" SDWs (at B_n, for the n to n-1 transition; vertical lines on Fig. 2). They correspond to the main plateaus in the Hall effect data [6], and to the paramagnetic jumps in the magnetization [7].

Secondly, as the temperature is lowered, a fine structure emerges from the "integer" lines. New anomalies appear inside a given SDW. Some of them are clearly related to the negative Hall plateaus observed in the very well relaxed $(TMTSF)_2ClO_4$ [3], and in $(TMTSF)_2PF_6$ under pressure [8]. In particular it must be pointed out that these additional Hall anomalies exhibit a ternary periodicity (two positive plateaus are followed by a negative one). At the highest temperature, a similar ternary behavior is fairly well observed on the magnetocaloric curves, but as the temperature is lowered each sub-anomaly splits into sub-sub-anomalies in an iterative process, leading to a large number of sub-phases, and thus to a higher order periodicity.

Furthermore the splitting process follows a general rule: a sub-line, close to a parent one, only grows on its high-field side. But whereas this splitting just leads to an asymmetric shape for the two first $\partial M/\partial T$ anomalies (the low-field side is much more abrupt than the high-field tail, Fig 2a), it leads for the next ones to a striking experimental fact: in a doublet the high-field anomaly is always larger than the low-field one, as it is indicated by arrows on Figure 2b.

This property is particularly surprising for the doublets arising from the "integer" transitions. In such a case the smallest anomaly appears to coincide with the main specific heat anomaly, i.e. with the integer transition from n to n-1 SDW phases; it means that the largest anomaly is the "fractional" one, which involves a larger entropy than the "integer" one.

The asymmetric shape of the $\partial M/\partial T$ anomalies (Fig. 2a) cannot be understood as resulting from the discontinuity of the gap at the SDW-SDW transition: for a given temperature, the gap being much more developed on the high-field side, one would expect a sharper variation of the entropy above B_n, in complete disagreement with the observed behavior. An other explanation is more likely that the splitting process requires a well ordered state, with a well developed order parameter. This would be the reason why the splitting is richer for the high-field phases, where the T_c's are higher. In such a case, the low-field asymmetry would result from the stacking above B_n of a high density of unresolved transitions.

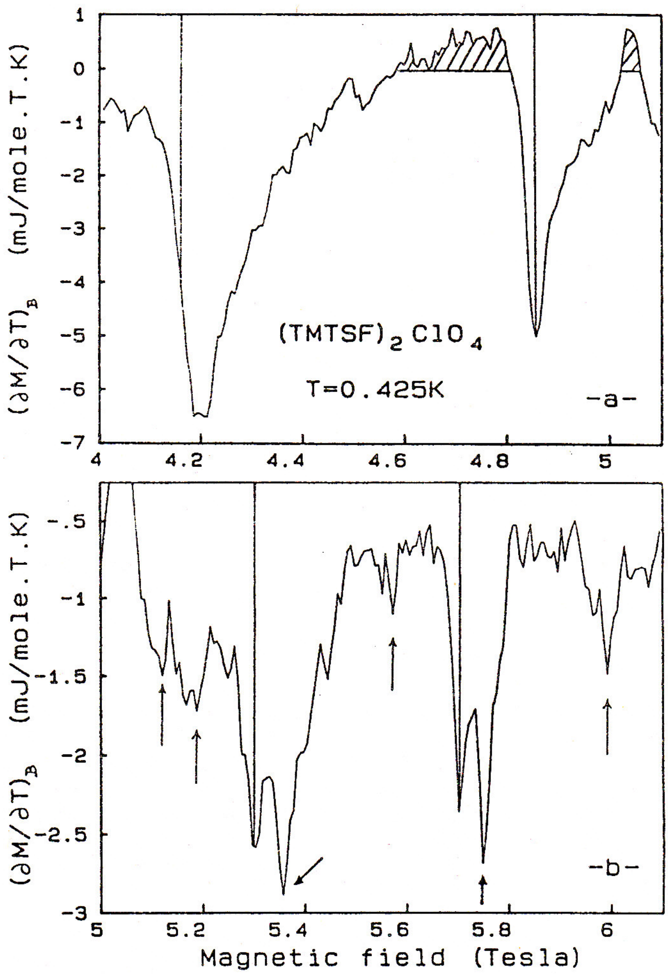

Figure 2: Magnetocaloric effect plotted versus the magnetic field. Vertical lines feature "integer" transitions between FISDWs, dashed areas represent positive entropies associated with partial reentrances, and arrows display the fractional doublets.

ARE WE FACED WITH ACTUAL PHASE TRANSITIONS ?

The issue is whether these splitted anomalies correspond or not to real phase transitions. Because of thermodynamic considerations, this is probably the case for most of them. Firstly, the specific heat displays a set of anomalies in the same field ranges as the $\partial M/\partial T$ anomalies (Fig 1). Secondly, part of them exhibit a variation in temperature, at variance with a mere Landau diagram, with even a <u>negative</u> slope for some ones. Thirdly, the sub-lines seem to separate from the main ones at some given temperature, and not to simply emerge at fixed field from a thermal "blurring". To support this point, it can be remarked that the high-temperature width of a given anomaly is too narrow to simply resulting from the thermal broadening of the low temperature doublet. Furthermore these ones separate with a finite slope.

On the other hand, some transitions occur at almost constant field. Nevertheless, the precise splitting point is difficult to determine experimentally because there are too many anomalies to be properly followed in temperature. Thus there is no clear evidence of the existence of sub-tricritical points where doublets appear, except maybe for the main doublets arising from the "integer" transitions: in this case the high-field sub-line may be extrapolated toward some point of the integer line [9].

A HIGH ORDER NESTING

These features can be qualitatively interpreted within the framework of the fractional nesting model [10-12]. "Fractional" transitions are believed to result from an interference effect of three periodicities: the lattice periodicity a along the chains coming in addition to SDW's and field's ones. Such as the Hofstadter's case of Bloch electrons in a rigid 2D lattice [13], this set of periodicities opens a complex family of gaps in the electronic spectrum [14]. But the issue here is not simply a Landau-Hofstadter quantization: the FISDW arises from a real phase transition and the Peierls gap opened at the Fermi level must be self-consistently taken into account. Moreover, as indicated by our calorimetric measurements, the strong coupling limit appears to be relevant in the Bechgaard's salts [5,15]. The gap value is thus greater than T_c and comparable with the cyclotron energy: it means that the distance between Landau gaps is of the same order of magnitude as the gap values. As a consequence, these gaps cannot be treated independently, especially far below T_c. In a high-order perturbation treatment involving an intermediate coupling, the fractional quantized nesting model invokes umklapp processes to produce a high order nesting in an extended zone scheme, leading to a fractional nesting in the first Brillouin zone [10]. The model's basic hypotheses are in a good agreement with the experimental observation that the splitting process takes place in the field regions where the order parameter is best developed, that is, for the SDW phases of low n (at the highest fields) and for a given n, on the high-field part of a doublet.

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