Heat Capacity of the 1D SDW system (TMTTF)$_2$Br and 2D CDW system 1T-TaS$_2$ in magnetic field

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Abstract. The equilibrium heat capacity of the 1D system (TMTTF)$_2$Br with spin density wave and the 2D system 1T-TaS$_2$ with charge density wave were investigated in the temperature range between 0.07K and 10K and in a magnetic field up to 10T. For both materials an additional contribution to the heat capacity was found below 1K, which is proportional to $T^{-2}$. This contribution is caused by low energy excitations with a broad spectrum of their relaxation time. The relaxation time is strongly temperature dependent and follows the Arrhenius law, i.e. the relaxation is a thermal activated process. For both materials the same absolute value of the $T^{-2}$ term and the same activation energy $E_a/k_B = 0.5K$ was found. In difference to (TMTTF)$_2$Br, where the heat capacity is strongly field dependent, the heat capacity of 1T-TaS$_2$ is constant up to 5T.

1. Introduction

Spin and charge density waves (SDWs and CDWs), which appears below the Peierls transition temperature of quasi-one-dimensional (quasi-1D) systems, are characterized by a spatial modulation of the electronic density (or spin) along the chains. The phase profile is the result of a compromise between the elastic energy that penalizes large phase gradients and the pinning energy that tends to fix the phase at the strong pinning centers. These two ingredients lead to metastability as the result of collective pinning. Collective pinning is, however, frozen below a glass transition of order 50K, as shown by dielectric susceptibility experiments [1]. At very low temperature, the residual degrees of freedom in a zero magnetic field correspond to the deformations at strong pinning defects. Larkin [2] and Ovchinnikov [3] have shown that a single strong pinning impurity leads to a bound state of an electron-like soliton and a hole-like antisoliton. This results in a potential with multiple minima, leading to slow relaxation in agreement with the very low temperature heat relaxation experiments [4]. R. Mélin et al [5,6] improved this model by taking into account the interaction between the soliton excitations created at different pinning centers, which provided interpretations of the strongly time-dependent $T^{-2}$ contribution to the heat capacity, the existence of magnetic field induced metastable phases and the strong magnetic field dependence of the heat capacity [7,8]. In this paper we present our experimental results for the equilibrium heat capacity of the commensurate 1D system with SDW (TMTTF)$_2$Br and the commensurate 2D system with CDW 1T-TaS$_2$. (TMTTF)$_2$Br is a quasi-1D conductor at room temperature, with a charge gap.
opening around 50K. It undergoes a spin density wave transition at 12K [9]. In 1T-TaS$_2$ the 2D CDW appears below 200K. This transition is accompanied by a Mott localization [10].

In the incommensurate 1D SDW system (TMTSF)$_2$PF$_6$ a strong influence of the magnetic field on the heat capacity was observed below 0.5K [11]. However, it was impossible to determine for this material the equilibrium heat capacity, since the upper limit of the relaxation time spectrum was too large (> 10$^4$s). From “waiting time experiments” we know that the relaxation time spectrum of commensurate systems is less extended [12,13,14] and so there is a better chance to get the equilibrium heat capacity.

2. Experimental Results and Discussion

The heat capacity measurements were performed with 140mg 1T-TaS$_2$ and 64mg (TMTTF)$_2$Br. Both materials are single crystals. The magnetic field was directed parallel to the c axis for 1T-TaS$_2$. The (TMTTF)$_2$Br sample contains a lot of small needles randomly orientated in the sample holder. The heat capacity was obtained from the temperature drift after switching on or off the heater with a constant power. Fig.1 shows this drift after switching off the heater at the equilibrium temperature $T_1 = 99$ mK to the new equilibrium temperature $T_0 = 92$ mK in different magnetic field.

Figure 1. The cooling $T(t) - T_0$ from 99 mK to 92 mK of the (TMTTF)$_2$Br sample after a constant heater power was switched off for different magnetic field.

For short time $t < t_{in}$ we observe an inner relaxation process due to the broad relaxation time spectrum of low energy excitations (LEE) in these materials. The parameter $t_{in}$ corresponds to the end of this inner relaxation and is proportional to the upper limit of relaxation time spectrum $\tau_{max}$. For long enough time $t > t_{in}$ the LEE are in a quasi-equilibrium with the phonons and the equilibrium heat capacity can be obtained from the relaxation time $\tau$ and the heat link of the sample: $c_p = \tau/R_{hl}$, where $\tau$ is determined from $T(t) - T_0 = (T_1 - T_0)exp(-t/\tau)$, subtracting the heat capacity of addenda. The heat link was calculated from $R_{hl} = (T_1 - T_0)/U_h I_h$, where $U_h$ and $I_h$ are the constant voltage and current of the sample heater. For low temperature and high magnetic field the inner relaxation time $t_{in}$ becomes very long. For example, at 92mK and 7T the relaxation of the internal degrees of freedom is finished after 6750s and only the last 0.3mK yields the information about the equilibrium heat capacity. Nevertheless, the very small scattering of
the temperature allows us to register the relaxation of this $0.3mK$ during $20000s$. The parameter $t_{in}$ is a property of the material and cannot be reduced. Therefore we need in these experiments a $\tau$ and as the consequence a heat link so large as possible ($R_{hl}(0.1K) \approx 10^{7} K/W$). Since fluctuations of the parasitic power leads to fluctuations of the temperature, we need also a very small and stable parasitic power. In our experiments this fluctuations are less than $1pW$ over all the measuring time up to $50000s$, including the magnetic field experiments up to $10T$.

The measured heat capacity for both samples at $92mK$ as function of magnetic field are given in Fig.2. The heat capacity of (TMTTF)$_2$Br is strongly field dependent. At high field the heat capacity is proportional to $B^2$. After a first field treatment $B > 5T$ the zero field value of the heat capacity increases more than 3 times, i.e. the sample is in a new field induced metastable phase. The heat capacity of 1T-TaS$_2$ is nearly field independent up to $5T$. The fit yields a $B^2$ term 100 times smaller than for (TMTTF)$_2$Br (see Fig.2).

In Fig.3 is shown the heat capacity as function of temperature for different magnetic field. At high temperatures we observe the $T^3$ term of the phonons, while at low temperatures an additional $T^{-2}$ term dominates, i.e. we observe the high temperature part of a Schottky anomaly. Since the maximum of this term was not found, we can only calculate the upper limit of the Schottky energy $E_s/k_B < 52mK$ and the lower limit of the number of LEE $N > 3.7 10^{24}/mol$, which corresponds to about 6 LEE per unit cell. An other surprising result is the excellent agreement of the Schottky contribution in zero field for both very different materials.

This agreement we find also in the parameter $t_{in}$, which is the same for both materials in zero field and follows the Arrhenius law:

$$t_{in}(B,T) = \tau_0(B) \exp(E_a/k_BT),$$

(see Fig.4). Following, the relaxation time is not caused by tunnelling like in glasses, but by a thermal activated process. In addition, the thermal activation energy $E_a/k_B = 0.5K$ is nearly field independent. The activation energy is in an excellent agreement with the result of "waiting time experiments". In these investigations the relaxation time spectrum was fitted by two Gaussian distributions centered on two characteristic values $\tau_{fast}$ and $\tau_{slow}$. Both parameters follows the Arrhenius law with exactly the same activation energy $E_a/k_B = 0.5K$ [13,14].
The parameter $t_{in}$ strongly depends on magnetic field for (TMTTF)$_2$Br and is constant up to 5T for 1T-TaS$_2$. It is seen from Fig.4 that $t_{in}$ also increases remarkably (by a factor of 3) in the field induced metastable phase of (TMTTF)$_2$Br. For 1T-TaS$_2$ this phase was not observed.

3. Conclusions
For both materials, for the 1D SDW system (TMTTF)$_2$Br and the 2D CDW system 1T-TaS$_2$ the heat capacity contains a $T^{-2}$ contribution. This contribution is strongly time dependent for $t < t_{in}$. The parameter $t_{in}$ follows the Arrhenius law and becomes very large values at low temperatures. The thermal activation energy $E_a/k_B$ for both materials is 0.5K. The main difference between the two systems was found for the magnetic field dependence: while the $T^{-2}$ term of the heat capacity and $t_{in}$ is strongly field dependent for (TMTTF)$_2$Br, they are up to 5T constant for 1T-TaS$_2$.

4. References
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