Energy relaxation in disordered charge and spin density waves

R. Mélin\textsuperscript{1,a}, K. Biljaković\textsuperscript{2}, and J.C. Lasjaunias\textsuperscript{1}

\textsuperscript{1} Centre de Recherches sur les Très Basses Températures (CRTBT)\textsuperscript{b}, B.P. 166, 38042 Grenoble Cedex 9, France
\textsuperscript{2} Institute of Physics, 10 001 Zagreb, P.O. Box 304, Croatia

Received 8 September 2004 / Received in final form 21 December 2004
Published online 30 March 2005 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2005

Abstract. We investigate collective effects in the strong pinning model of disordered charge and spin density waves (CDWs and SDWs) in connection with heat relaxation experiments. We discuss the classical and quantum limits that contribute to two distinct contribution to the specific heat (a \( C_v \sim T^{-2} \) contribution and a \( C_v \sim T^{0} \) contribution respectively), with two different types of disorder (strong pinning versus substitutional impurities). From the calculation of the two level system energy splitting distribution in the classical limit we find no slow relaxation in the commensurate case and a broad spectrum of relaxation times in the incommensurate case. In the commensurate case quantum effects restore a non vanishing energy relaxation, and generate stronger disorder effects in incommensurate systems. For substitutional disorder we obtain Friedel oscillations of bound states close to the Fermi energy. With negligible interchain couplings this explains the power-law specific heat \( C_v \sim T^{\alpha} \) observed in experiments on CDWs and SDWs combined to the power-law susceptibility \( \chi(T) \sim T^{-1+\alpha} \) observed in the CDW a-TaS\(_3\).

PACS. 71.45.Lr Charge-density-wave systems – 05.70.Ln Nonequilibrium and irreversible thermodynamics – 63.50.+x Vibrational states in disordered systems – 75.30.Fv Spin-density waves

1 Introduction

There exist many examples of systems showing slow relaxation and ageing: spin glasses [1,2], disordered dielectrics [3–5], supercooled liquids [6], etc. Charge density waves (CDWs) and spin density waves (SDWs) \cite{7–10} show “interrupted ageing” \cite{11}, meaning that there exists an upper bound \( \tau_{\text{max}} \) to the relaxation times. The protocol of ageing experiments in CDWs and SDWs is the following: the system is equilibrated at the temperature \( T \) (one waits for a time longer than \( \tau_{\text{max}} \)). At time \( t = 0 \) the temperature is changed from \( T \) to \( T + \Delta T \) where \( \Delta T > 0 \) is very small compared to \( T \). The temperature is kept constant until the waiting time \( \tau_w \) where it is brought back to \( T \). The heat flows between the CDW or SDW sample and the cold source are recorded as a function of time. Ageing in the thermal response takes place even for very small values of \( \Delta T \). Since very small temperature variations are applied in the experiment it is reasonable to suppose that the size of the correlated objects does not evolve in time and that the thermal response is due solely to the variation in the population of metastable states. This can be contrasted with the coarsening dynamics where the size of correlated domains increases with time.

In a recent work \cite{12} we applied the idea of dynamical renormalization group \cite{13,14} to calculate the spectrum of relaxation times of a model of disordered CDW or SDW \cite{15–24}, including interactions among bisolitons. A drawback of this approach \cite{12} is that we supposed a coarsening dynamics following a quench from high temperature, a situation that is not realized in experiments, and we were not able to address the waiting time dependence of the relaxation time spectra. One goal of the present article is to address these issues that were left open in our previous work \cite{12}, and to put on a microscopic basis the random energy-like (REM-like) trap model that was proposed in reference \cite{12}, and inspired from trap models developed for glasses and spin glasses \cite{11,25}.

More specifically we show here that heat relaxation experiments can be described by assuming two types of defects (strong pinning and substitutional impurities), corresponding to the “classical” limit where the CDW or SDW is viewed as a classical elastic medium with bisolitons generated by strong pinning impurities distributed at random, and to the “quantum” limit where solitons due to substitutional disorder interact quantum mechanically by excitations of the gaped background. The existence of two effects is in agreement with the experimental observation...
that the low temperature out-of-equilibrium specific heat can be decomposed into three contributions: (i) the $C_v \sim 1/T^2$ tail of a Schottky anomaly at very low temperature (typically for $T \lesssim 100 \pm 300$ mK; the upper bound depends on the amplitude of the 1/$T^2$ contribution); (ii) a $C_v \sim T^\alpha$ power-law specific heat with $\alpha \simeq 0.3 \div 1.2$ at intermediate temperatures ($0.1 \lesssim T \lesssim 1$ K); and (iii) the “trivial” contribution of phonons $C_v \sim T^3$ at high temperature ($T \gtrsim 1$ K). By Schottky anomaly we mean that the equilibrium specific heat of a two-level system with energies $E_0$ and $E_0 + \Delta E$ is given by

$$C_v(T) = \frac{(\Delta E)^2}{4T^2} \frac{1}{\cosh^2(\Delta E/2T)}$$

having a maximum (called a Schottky anomaly) at $T_{\text{max}} \simeq 0.416 \Delta E$. The specific heat is approximately equal to $C_v \simeq (\Delta E)^2/4T^2$ in the large temperature tail.

Following reference [23], the contribution (i) is interpreted in terms of two-level systems due to strong pinning impurities. The contribution (ii) is interpreted as midgap states interacting through Friedel oscillations. Friedel oscillations of a single impurity were probed directly by X-ray diffraction experiments in reference [26]. Another evidence in favor of the coexistence of strong pinning and substitutional impurities is that the CDW compound o-TaS$_3$ can be doped by Nb, a substitutional impurity. This changes only the amplitude of the $C_v \sim T^\alpha$ contribution, but leaves unchanged the $C_v \sim 1/T^2$ contribution [27], suggesting that the power-law contribution is related to substitutional disorder. Even though not affected by substitutional disorder we do not interpret the $C_v \sim 1/T^2$ as a property of the pure compound. A nuclear hyperfine interaction can be excluded from the systematic study of many different CDW compounds [7]. We thus relate the $C_v \sim 1/T^2$ contribution to strong pinning impurities [23], even though the microscopic nature of these impurities is not well understood experimentally (see Ref. [28] for a study of ESR spectroscopy in o-TaS$_3$).

The commensurate organic spin-Peierls compound (TMTTF)$_2$PF$_6$ showing slow relaxation [29] contrasts with the inorganic spin-Peierls compound Cu$_{1-x}$Zn$_x$GeO$_3$ [30–33] showing antiferromagnetic ordering. We argue that the difference lies in the different nature of disorder. Substitutional disorder relevant to Cu$_{1-x}$Zn$_x$GeO$_3$ is qualitatively different from strong pinning impurities in CDWs and SDWs. The spin-Peierls compound Cu$_{1-x}$Zn$_x$GeO$_3$ has a fast dynamics, with a “microscopic” time presumably comparable to the one of spin glasses ($\tau_0 \simeq 10^{-12}$ s) whereas in CDW and SDW compounds we have $\tau_0 \simeq 1$ sec for the thermally activated process. This indicates that rather different mechanisms are at work, identified here are substitutional or strong pinning disorder. We generalize to the incommensurate case the model of substitutional disorder introduced in reference [34]. In this model the solitons are due to domain walls between two degenerate ground states since the impurity site can be removed from the chain (see Ref. [34] and Sect. 3), therefore leaving randomly distributed domain walls in the chain from which the impurities sites have been removed. The specificity of this model (as opposed to the strong pinning model) is that there are no metastable states of bisolitons like in the strong pinning model. The substitutional impurities do not contribute to the slow dynamics of bisolitons in the strong pinning limit but are expected to contribute to collective pinning of the phase of the density wave. However we consider here temperatures much lower than the glass transition temperature [35,36] so that interactions among solitons are the only remaining collective effects.

For substitutional disorder we find interactions among solitons due to Friedel oscillations. We obtain similarly to reference [34] a power-law specific heat $C_v(T) \sim T^\alpha$ and a susceptibility $\chi(T) \sim T^{-1+\alpha}$ in agreement with existing experiments on the CDW o-TaS$_3$ [27].

The article is organized as follows. In Section 2 we investigate a classical model of collective effects in a disordered CDW. Quantum effects are investigated in Section 3. Final remarks are given in Section 4.

2 Classical limit (strong pinning impurities)

2.1 Hamiltonian

Let us start with a classical model of disordered CDW [15–24]. To derive the 1D projection of the Hamiltonian of the phase of the CDW in the mean field approximation we follow the recent review by Brazovskii and Nattermann [24] and consider a system of coupled chains with a phase $\varphi_n(y)$ in chain $n$ ($y$ is the coordinate along the chain axis).

$$H = \frac{\hbar v_F}{4\pi} \sum_n \int dy \left( \frac{\partial \varphi_n(y)}{\partial y} \right)^2$$

$$+ \sum_{n,m} w_{m,n} \int dy \left[ 1 - \cos(\varphi_n(y) - \varphi_m(y)) \right]$$

$$- \sum_{n,i} V_i^{(n)} \left[ 1 - \cos (Qy_i^{(n)} + \varphi_n(y_i^{(n)})) \right],$$

where $\varphi(y)$ is the phase of the CDW Landau-Ginzburg order parameter along the chain $n$, and $Q = 2k_F$ is the wave vector of the CDW. Assuming dilute impurities we suppose that the chain $n = 0$ with $\varphi_0(x) = \varphi(x)$ is coupled to neighboring chains with $\varphi_n(x) = 0$. We arrive at the effective 1D Hamiltonian

$$H = \frac{\hbar v_F}{4\pi} \int dy \left( \frac{\partial \varphi(y)}{\partial y} \right)^2 + w \int dy \left[ 1 - \cos(\varphi(y)) \right]$$

$$- \sum_i V_i \left[ 1 - \cos (Qy_i + \varphi(y_i)) \right].$$

Metastable states due to the competition between the commensurate potential and the pinning energy were first