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PHASE DIAGRAM OF ANISOTROPIC TWO-DIMENSIONAL QUARTER-FILLED BAND OF INTERACTING ELECTRONS

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I. INTRODUCTION.

Ever since its original discovery(1), the magnetic field induced spin density wave (FISDW) transition observed in superconducting TMTSF salts has continued to be of considerable interest. This phenomenon has been studied most widely in the ambient pressure superconductor $(\text{TMTSF})_2\text{ClO}_4$, where for moderate magnetic fields $H > H_f$ a cascade of transitions to semimetallic SDW phases was observed(2). Until very recently, it was generally accepted that the theoretical explanation for the FISDW lied in the gradual one dimensionalization of the Fermi surface in the presence of the magnetic field and the accompanying nesting instability(3-7). These theories do not account for the more recently discovered very high field transition (VFHT) for $H > H_v$. In contrast to the predictions of the above theories the SDW transition temperature T_{SDW} actually decreases for $H > 17\text{T}$ and then goes to zero at $H = 25\text{--}30\text{T}$ (8,9). At present, there exists only one theoretical attempt to explain the VFHT(10). Within this model(10) the material is at the one dimensional limit for very high fields, and the competition between the equally strong Peierls and Cooper channels in this limit destroys the FISDW. The system is then an ordinary metal for $H > H_v$.

We believe that the very high field phase is *not* an ordinary metal. It is characterized by a high resistivity, (weakly) activated conductivity and vanishing Hall signal(9). All of these suggest that the system is a semimetal, and perhaps even a narrow gap semiconductor. In addition the system is diamagnetic in this region, which also suggests that its nature is different from simple metallic.

In the present paper we present preliminary work on a slightly different theoretical model. We believe that the model presented here describes charge transfer solids in general, and FISDW behavior in particular, better. In contrast to much of the existing theoretical work (see, however, reference 5), we emphasize the direct Coulomb interaction between the carrier holes. More importantly, we consider a spatial broken symmetry

neglected in theories of FISDW so far, -- the bond order wave (BOW), -- which competes with the SDW as the one dimensional limit is approached (11). Finally, the actual band filling, or more precisely, the number of holes ρ per molecular site, plays an important role in our model. This is a consequence of explicit inclusion of both on site and intersite Coulomb interactions(12,13). The very important role of ρ is missed in noninteracting models. While we agree with Yakovenko(10) that the very high field phase is near the one dimensional(1D) limit, we believe that the relevant competition is between the BOW and the SDW, and not between superconductivity and SDW.

The motivation for the present work, however, goes beyond explaining FISDW. We intend to show that the same theoretical description applies to the normal states of all organic superconductors as well as related materials with molecular cations that are similar to TMTSF structurally. Specifically, we have chosen (TMTTF)₂X and (BEDT-TTF)₂X, since we believe that similarities at the molecular and structural levels of these materials warrant the same basic theoretical model, albeit with different magnitudes of the various parameters. For example, it is our belief that the pressure induced spin-Peierls(or BOW in our nomenclature) to SDW transition(14,15), that has been observed in (TMTTF)₂PF₆, is a manifestation of the same basic dimensionality cross-over within the same model Hamiltonian.

II. THEORETICAL MODEL.

The model that we consider is the single band quasi-2D extended Hubbard model,

$$\hat{H} = \sum_i (t_x c_{i\sigma}^\dagger c_{i+x} + t_y c_{i\sigma}^\dagger c_{i+y} + \text{H.c.}) + \sum_i (V_x n_i n_{i+x} + V_y n_i n_{i+y}) + U \sum_i n_{i\uparrow} n_{i\downarrow} \quad (1)$$

where $c_{i\sigma}^\dagger$ creates a hole with spin σ at the molecular site i , $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ is a number operator, $n_i = \sum_\sigma n_{i\sigma}$, U is the on site Coulomb repulsion between two holes occupying the same molecular site. V_x and V_y are the intrachain and interchain intersite nearest neighbor repulsions, and t_x and t_y are intrachain and interchain hopping integrals, respectively. We do not consider a realistic lattice at this preliminary stage, but assume a rectangular lattice with variable anisotropy ($0 < t_y/t_x < 1$, $0 < V_y/V_x < 1$). The stack axis is chosen to be the x -direction. The implicit parameter ρ is very important in our theory, particularly at moderate to large t_y/t_x . We consider $\rho = 1/2$ only, although TMTSF salts may be characterized by weak incommensurability(16) arising due to band structure effects or incomplete charge transfer.

We have investigated numerically the phase diagram of the above theoretical model as a function of t_y/t_x and V_y/V_x . Before presenting our numerical results we discuss here the qualitative reasonings for our belief that Eq.(1) can reproduce the experimental behavior.

In the limit of $t_y = 0$ and $V_y = 0$, Eq.(1) has been widely studied as a model for $\rho = 1$ Mott insulators(11) as well as for conductors with arbitrary ρ (12,13). Because of the continuous nature of broken symmetry in the SDW, the SDW never occurs as a distinct phase. Rather, the dominant broken symmetry here is an unconditional $2k_F$ BOW(11,17) (which, depending upon the parameters, may also be accompanied by a $4k_F$ instability(17)). For $V_x = V_y = 0$ and $t_y = 0$, it has been shown analytically that the charge-spin decoupling that occurs in the large U Hubbard model leads to the magnetic part of H to be described by an isotropic Heisenberg spin Hamiltonian (18). Similar

results are expected to persist even for $V_x > 0$, although the expression for the Heisenberg exchange integral is expected to be quite different. It is known unambiguously that such a system exhibits a spin-Peierls (BOW) transition, and the SDW does not occur for any U, V_x . Experimental evidence for the spin-Peierls transition in charge transfer salts is common.

The above situation is expected to change as t_y is increased from zero. Existing nonzero t_y results are only for the $\rho = 1$ limit, where the consensus is that the BOW is destroyed in the presence of Coulomb interaction(19,21). Although these results are for $t_y = t_x$, we believe that the disappearance of BOW (and appearance of SDW) can occur at t_y/t_x considerably less than 1. Furthermore, we believe that the spin-Peierls to SDW transition should not be limited to $\rho = 1$ only but also should be seen in $\rho = 0.5$, where the BOW vanishes for $t_y > t_{y1}$. The special feature of $\rho = 0.5$ is that unlike $\rho = 1$, where the SDW gets progressively stronger with the increase of t_y , the SDW is weakened for t_y greater than an upper critical value t_{y2} . The reason for this is shown in Fig. 1. Due to the nonzero V_x and V_y , holes have a tendency to occupy alternate sites. For large enough t_y , the intrachain and interchain antiferromagnetic spin couplings can become comparable, and the resulting spin frustration (see Fig. 1) will destroy the SDW.

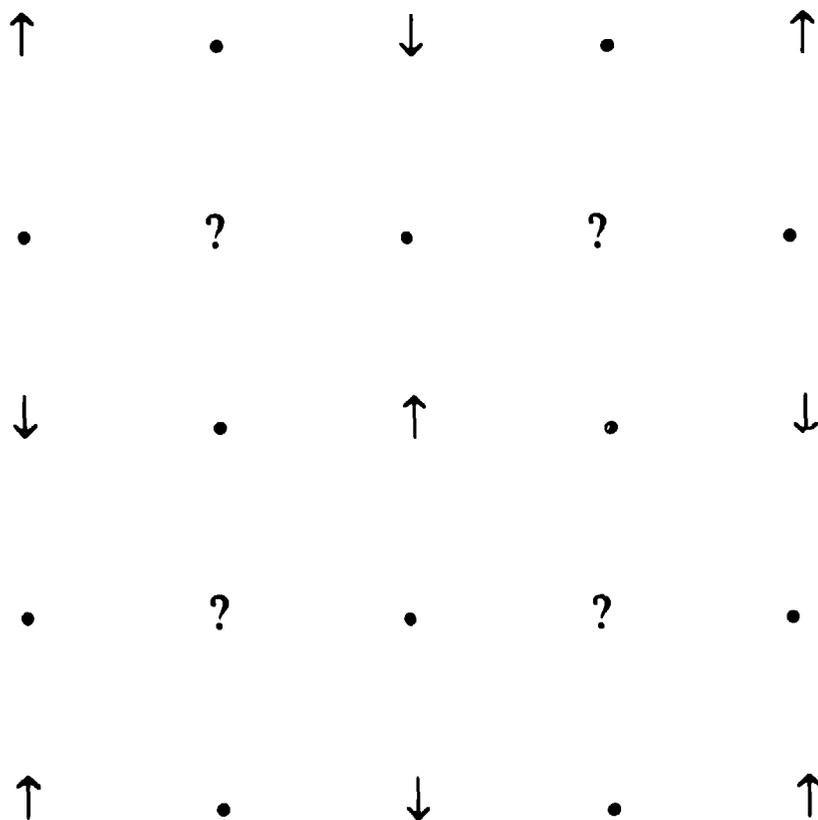


Fig. 1: Spin configurations for the 1/4-filled band. Note that because of the antiferromagnetic couplings between spins along \bar{x} , \bar{y} , and $\bar{x}+\bar{y}$ directions, spins at middle sites are frustrated.

Three distinct regions are then expected: $t_y < t_{y1}$, $t_{y1} < t_y < t_{y2}$, and $t_y > t_{y2}$. Generally

opening. The frustrated materials are in region 1, TMTSF's in region 2, and BEDT-TTF salt in region 3. TMTTF and TMTSF materials can further be modified by pressure (which increases two dimensionality) or magnetic field (which decreases two dimensionality), and crossover from one region to another has been observed. Note that for spin frustration to occur it is essential that V_x and V_y are explicitly included in the Hamiltonian, and the proper role of $\rho = 0.5$ is taken into account. The importance of considering the role of V_x and ρ has been emphasized before(12,13).

III. NUMERICAL RESULTS.

We present some of our numerical results in this section that give partial support to what have been said in the previous sections. We study the BOW and SDW phases within the Hamiltonian (1). The "structure factors" corresponding to the BOW and the SDW are defined as

$$\text{BOW}(q) = \frac{1}{N} \sum_{i,j} e^{iq(\tau_i - \tau_j)} (\langle B_i B_j \rangle - \langle B_i \rangle \langle B_j \rangle) ,$$

where $B_i = \sum_{\sigma} (c_{i\sigma}^{\dagger} c_{i\sigma} + \text{H.c.})$ is the bond order parameter, and

$$\text{SDW}(q) = \frac{1}{N} \sum_{i,j} e^{iq(\tau_i - \tau_j)} \langle (n_{i\uparrow} - n_{i\downarrow}) (n_{j\uparrow} - n_{j\downarrow}) \rangle .$$

Both these quantities were calculated explicitly in the 1D limit and then for varying anisotropy. In the 1D limit, $t_y = 0$ and $V_y = 0$, the results were obtained by a world line quantum Monte Carlo simulation on lattice of size $N = 128$ atoms at temperature $T = 1/32$ in units of $t_x = 1$ (for a t_x of 0.2 eV this would be about 70K which is higher than the temperatures of interest in FISDW but which is reasonably close to the spin-Peierls temperatures in many organic systems). Only the results for $V_x = 0$ are shown here. For nonzero but moderate V_x (V_x less than or equal to $2t$) the results are the same qualitatively (17). In Fig. 2 we plot $\text{BOW}(q)$ as a function of the wave vector q for several values of U . In all cases a peak at $q = 2k_F$, where k_F is the Fermi wave vector within single-particle model is clearly visible. Although the Coulomb repulsion suppresses the BOW here (unlike at $\rho = 1$, where an enhancement is seen for moderate U (11)), a logarithmic divergence as a function of N always exists, which implies that even for weak electron-lattice coupling a $2k_F$ BOW instability will occur, leading to a tetramerization in the present case.

Similar quantum Monte Carlo simulations for 2D lattices when ρ not equal to 1 are difficult currently because of the well known "negative sign" problem. This problem becomes even more severe for large Coulomb interaction and low temperature. In 2D therefore we have done exact calculations for a 4×4 lattice. Notice that this already gives huge Hamiltonian matrices for Eq. (1). The exact ground state wave function was calculated using a Lanczós diagonalization procedure. As we are restricted to a single lattice finite size scaling analysis is not possible and our conclusions are drawn from comparison with the results for the $U = V_x = V_y = 0$ case, for which the results are known analytically. Finally, the number of parameters that appear in Eq.(1) in 2D are rather large, so we assume $V_y/V_x = t_y/t_x$ in all our calculations. Since both the nearest neighbor hopping and Coulomb integrals are functions of intermolecular distances, this is not an unreasonable assumption. In any case, this restriction is not a limitation and can be relaxed later.

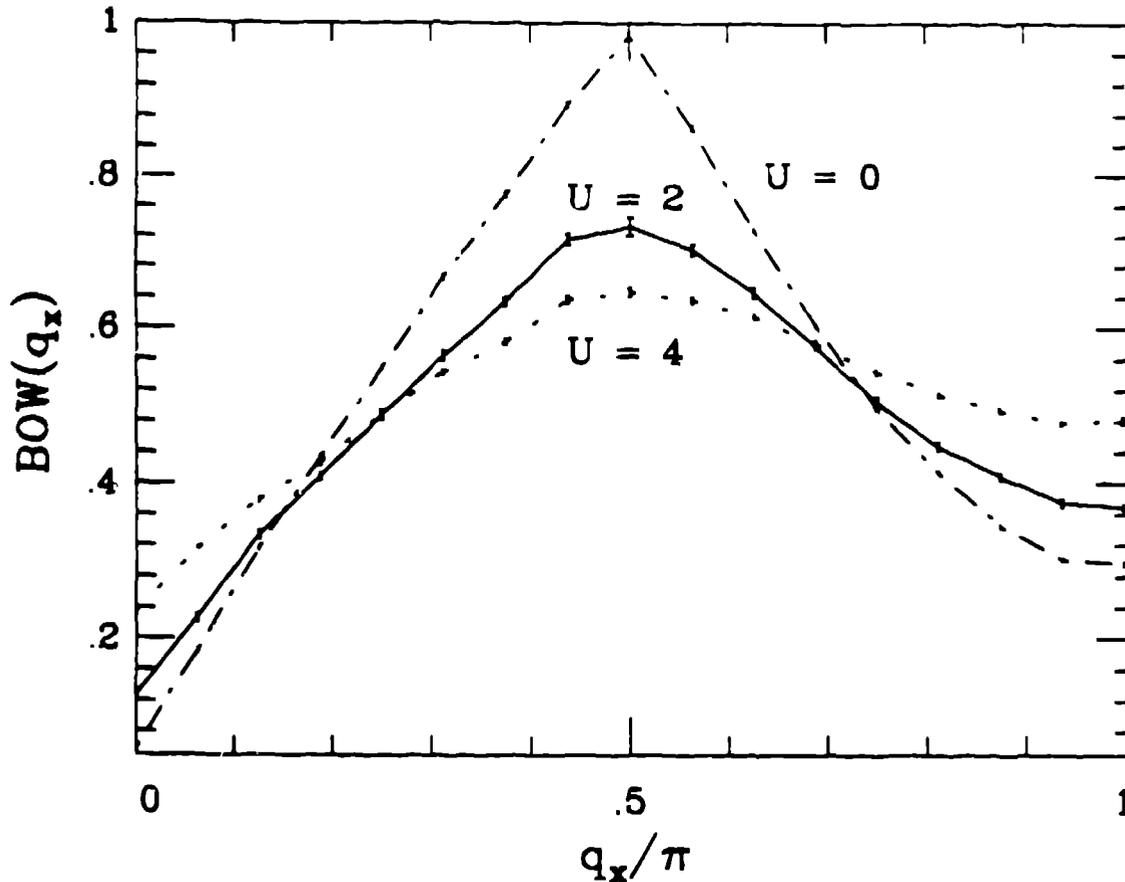


Fig. 2: Bond-Order-Wave Structure Factor for the 1D Hubbard Model.
 $N = 128$, $T = t_x/32$, and $U = 0, 2, 4$.

While the calculations were done for several different parameter values we present the results for $U = 3$, $V_x = 1$ (in units of $t_x = 1$), which are representative. Instead of showing the full set of results for the complete (q_x, q_y) subspace we present the results for $q_y = \pi$ only. This is because the correlation functions along (q_x, π) are the ones that are relevant for describing the spin-Peierls and SDW transitions in two dimensions. In Fig. 3 we have plotted $BOW(q_x)$ as a function of the anisotropy. It is clear that the BOW is progressively weakened as t_y and V_y are increased. This is what we expected, based on our earlier $p = 1$ results. What is more interesting is the behavior of the SDW, as seen in the plots of $SDW(q_x)$ for the same set of parameters in Fig. 4. Unlike the BOW, the SDW is enhanced initially, since the peak at $2k_F = \pi/2$ becomes more pronounced as t_y increases from $t_y = 0.2$ to $t_y = 0.5$. However, with further increase in t_y , the SDW amplitude *decreases*, until at $t_y = 1$, where its behavior becomes indistinguishable from single particle behavior, indicating a vanishing of the SDW due to the spin frustration discussed above. Qualitatively, this behavior is exactly what we claimed in section II., although the actual spin frustration here seems to become relevant for relatively large t_y .

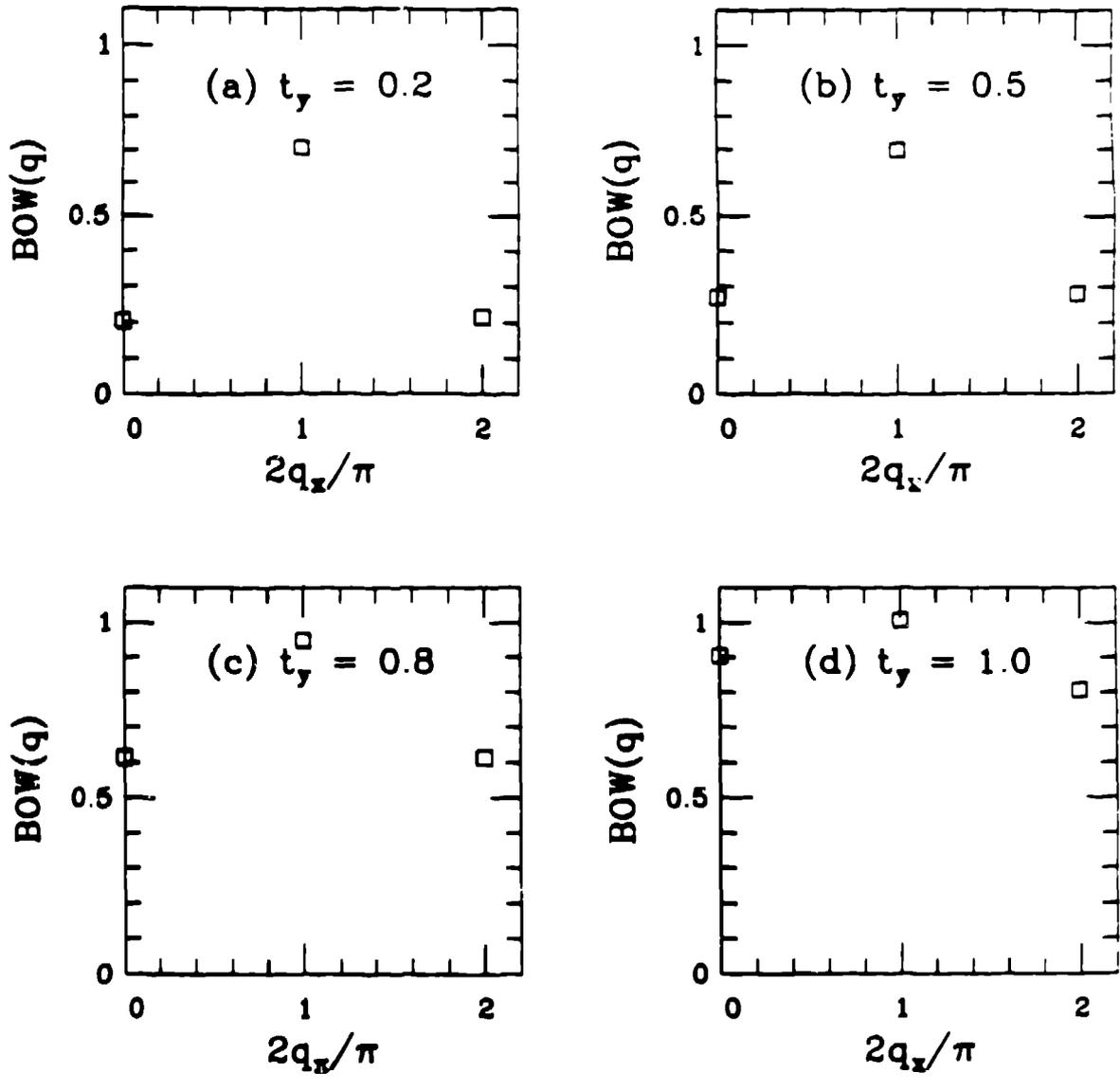


Fig. 3: Bond-Order-Wave Structure Factor $BOW(q_x)$ as function of the anisotropy. (see the text)
 $U = 3$, $V_x = 1$, and $t_x = 1$.

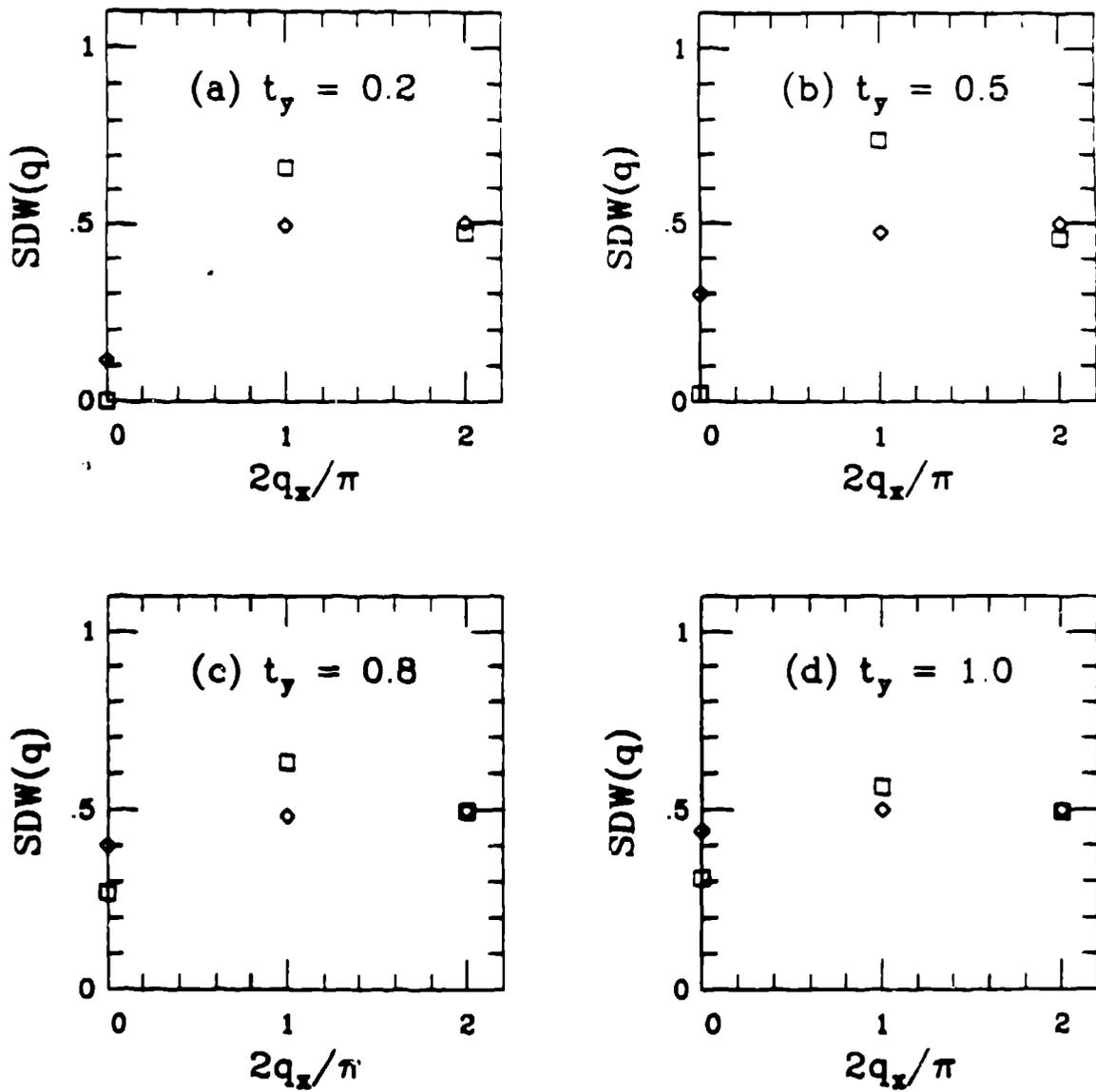


Fig. 4: Spin-Density-Wave Structure Factor $BOW(q_x)$ as function of the anisotropy.(see the text)
 $U = 3, V_x = 1, \text{ and } t_x = 1$;(squares)
 $U = 0, V_x = 0, \text{ and } t_x = 1$;(diamonds)

The rapid destruction of the BOW for even small nonzero t_y and the destruction of the SDW in the other limit of large t_y clearly suggests three distinct regions. Quantitative comparisons with results of simulations for such a small system would be meaningless, and any critical t_y obtained from our calculation would be too large. We therefore do not attempt quantitative comparisons at this preliminary stage. The emphasis is rather on qualitatively explaining the behavior of all three families of materials, $(\text{TMTTF})_2\text{X}$, $(\text{TMTSF})_2\text{X}$, and $(\text{BEDT-TTF})_2\text{X}$ within the same theoretical model.

Within our model ambient pressure $(\text{TMTTF})_2\text{X}$ lies in the $t_y < t_{y1}$ region. Since both bonding and Coulomb interactions are larger along the stacking axis than along the interchain direction, the lattice is expected to be more compressible along the latter. Pressure therefore increases t_y until the region $t_{y1} < t_y < t_{y2}$ is reached and the spin-Peierls to SDW transition occurs. Further increase in pressure increases T_{SDW} , which, however, reaches a maximum and then begins to decrease. Note that the peak in T_{SDW} is expected in our model, -- this is the region where spin frustration starts to be relevant. Experimentally, it is currently not clear whether there is a narrow region of coexistence between the spin-Peierls and the SDW phases. Due to the strong finite size effects in our numerical simulation, we are unable to resolve this issue theoretically.

The effect of magnetic fields on TMTSF is opposite to that of pressure in that the anisotropy increases with H . For $H < H_f$ and $t_y > t_{y2}$, there is no SDW due to spin frustration. For $H > H_f$ the increase in anisotropy takes the system into the region $t_{y1} < t_y < t_{y2}$, where SDW is the characteristic. The current model is too simplistic to give the cascade of SDW transitions. (This would be investigated later.) However, the VFH is expected in our model. In the extreme 1D limit $t_y < t_{y1}$, the BOW phase occurs at low temperatures. This phase is then *not* a metal, and we expect semimetallic behavior (or even weakly semiconducting behavior), in agreement with the transport measurements(9). Note that the observed diamagnetism is associated with the spin-Peierls phase.

Finally in $(\text{BEDT-TTF})_2\text{X}$ the anisotropy is considerably weaker and t_y is much larger than t_{y2} . The absence of SDW in these materials is then expected from our model. Because of the smaller t_y in the sulphur-based materials, as compared to the selenium based materials, a tendency to charge density wave may be expected within model Hamiltonian Eq.(1), and this may explain the experimentally observed tendency to localization in these substances.

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