Enhanced superconducting and spin-density-wave ordering temperatures of gated quasi-one-dimensional organic superconductors

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Presented are the results of calculations suggesting that the quasi-one-dimensional organic superconductors $(TMTSF)_2X$ (where TMTSF represents tetramethyltetraselenafulvalene and X is PF_6 AsF_6, ClO_4, etc.) may show a substantial increase in their superconducting and spin-density-wave ordering temperatures when the Fermi level is raised through application of an electrostatic gating voltage. A rich behavior is observed, strongly dependent on the form of the superconducting order parameter, as the Fermi level approaches the Van Hove singularity at $k_a=0$. Included are predictions for the behavior of these materials under zero and moderate applied pressure. It is found that T_{SDW} as high as 50 K and superconducting T_c as high as 20 K may be achieved at optimal gate voltages of approximately 100 mV.

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I. INTRODUCTION

Since the discovery by Van Hove[1] in 1953 that quasiparticle densities of states $N(E)$ in one-, two-, and three-dimensional systems must necessarily contain singular points [where $N(E)$ diverges], much attention has been paid to the phenomenon of Van Hove singularities and their possible role in superconductivity. Given the BCS weak-coupling relationship[2] $T_c \sim 1/\sinh\{1/[VN(E_F)]\}$, with $N(E_F)$ the normal-state density of states at the Fermi level and V the attractive interaction, it is clear that enhancing $N(E_F)$ has the potential to greatly enhance T_c. As there is a close similarity between BCS weak-coupling theory and quasi-one-dimensional charge- and spin-density-wave (SDW) theory,[3] with $T_{\mathrm{SDW}} \sim 1/\sinh\{1/[UN(E_F)]\}$, where U is the repulsive Coulomb interaction, it is clear that a similar enhancement of T_{SDW} is possible if $N(E_F)$ is enhanced.

Several authors[4–6] have attempted to describe enhanced-temperature superconductivity (as observed in the cuprates) as being at least partly due to the Van Hove singularity in the two-dimensional (2D) square lattice quasiparticle dispersion $-2t[\cos(k_x)+\cos(k_y)]$ at $(k_x,k_y)=(\pi,0)$ and equivalent points, and simple numerical models[7] and some experimental data[8,9] offer support for this idea. While the role of this singularity in cuprate superconductivity is far from clear,[10] it is possible that this singularity could be partly responsible for the high transition temperatures observed in the cuprates.

This singularity, however, is strictly speaking a zero-dimensional singularity (although it is a saddle point). The dimensionality of this singularity necessarily limits the impact it can have on superconductivity on a two-dimensional Fermi surface, as is observed in the cuprates.

By contrast, quasi-one-dimensional materials, such as the organic superconductors $(TMTSF)_2X$ (where TMTSF represents tetramethyltetraselenafulvalene and $X=PF_6$, AsF_6, ClO_4, etc.),[11–15] have an extended one-dimensional Van Hove singularity at the center of the Brillouin zone [where $\cos(k_a)=1$], and have a quasi-one-dimensional Fermi surface paralleling this singularity, as depicted in Fig. 1. All that is then necessary, in this theoretical treatment, to substantially enhance T_c and T_{SDW} is a means of changing E_F so that the Fermi surface lies on or near the Van Hove singularity in the zone center. This can be accomplished by application of a gate voltage to the organic superconductor, as was done for a cuprate superconductor by Ahn *et al.*[16,17] The quasi-one-dimensional organic superconductors[15] $(TMTSF)_2X$ exhibit both superconducting and spin-density-wave order, albeit at different applied pressures. With the exception of one compound, $(TMTSF)_2ClO_4$, these materials undergo a spin-density-wave transition at ambient pressure around 10 K, while under pressure several of these materials become superconducting at temperatures of order 1 K. The author therefore explicitly considers the competition between spin-density-wave and superconducting (SC) order, by solving both the BCS gap equation and an analogous form for the transition temperature T_{SDW} of the spin-density wave, in order to describe under what gate voltages and temperatures the system is expected to be in a SDW or a SC state. For the rest of this paper, by "gate voltage" we actually mean the change in the chemical potential, noting that the two will differ in an actual gating experiment.

Given that the weak-coupling relationships for the condensation energy at $T=0$ for both orders are identical[3]

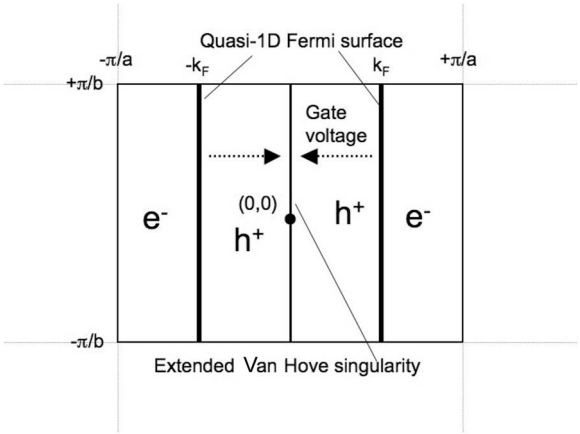

FIG. 1. Schematic diagram of the Brillouin zone and Fermi surface of a quasi-1D organic superconductor. As a gate voltage is applied, the Fermi surface moves inward toward the Van Hove singularity at $k_a=0$.

$[=-\frac{1}{2}N_0\Delta(0)^2_{\text{order}}]$, and that the temperature dependence of the gaps is essentially equivalent, it is simply assumed that, at given gate voltage, the system transitions to the order with the highest transition temperature and retains that order at all lower temperatures. This approximation[18] is most accurate when the two orders have substantially different transition temperatures, with a more complex behavior possibly applying right at transition points between the two phases. We do not consider such behavior here. The transition from SDW to pressure-induced superconductivity in these materials has been attributed[15,19] to decreased nesting due to increased t_b. Therefore we perform calculations of both order parameters for two scenarios: $t_b=0.08t_a$, corresponding to a material that exhibits SDW at 11.5 K at ambient pressure (we fix the Coulomb potential U to match $T_{\text{SDW}}=11.5$ K at zero gate voltage), and $t_b=0.11t_a$, corresponding to a material whose SDW instability is nearly absent at zero gate voltage and is overwhelmed by the SC instability. This second case represents $(\text{TMTSF})_2X$ under pressure, and also $(\text{TMTSF})_2\text{ClO}_4$, the only ambient-pressure TMTSF superconductor.[20,21]

II. CALCULATIONS: BCS WEAK-COUPLING THEORY AND SDW THEORY

In the TMTSF series, due to the dimerization,[15] the dispersion contains a $\cos(k_a a/2)$ term so that the extended Van Hove singularity lies at $k_a=0$, in the center of the Brillouin zone. This does not pose a problem, however, as in these materials the electrons lie on the outside of the Fermi surface. Formally speaking, in the BCS approximation T_c itself would diverge if $N(E)$ diverged at the Fermi level; however, this artifact is easily removed by the requirement of integration over the Brillouin zone, as well as the inclusion of a small but finite second hopping term t_b (=0.08 or 0.11), so that the dispersion acquires a slight 2D character. For the purposes of calculation we assume a rectangular 2D lattice with normal-state quasiparticle dispersion given by

$$\epsilon_k-\mu=2t_a\cos(k_a a/2)+2t_b\cos(k_b b)-\mu \tag{1}$$

with t_a and t_b the hopping coefficients in the indicated directions and μ the chemical potential, to be modified by a gate voltage. We take t_a as 250 meV.

In practice[15] the TMTSF salts have two bands, with the lower filled and the upper half filled so that $k_F=\pi/2a$ (for the moment ignoring the k_b dispersion). One reasonable assumption is that it is only the upper band that is active in the superconductivity. The full situation is somewhat more complex than this, as the lower band can be expected to contribute to the susceptibility χ_0, with a concomitant effect on T_{SDW}, and in the case of spin-fluctuation-mediated pairing this would then impact T_c. Since the bottom band is far from the Fermi surface, however (and the Fermi surface nesting largely determines T_{SDW}), inclusion of this multiband effect would likely add a nearly momentum-independent term to the susceptibility $\chi(\mathbf{q})$. Given that we ultimately choose U so that T_{SDW} in the ungated situation at $t_b=0.08$ matches the experimental value of 11.5 K, the main effect of including the lower band would be a renormalization of U, with the predictions for T_{SDW} at various gate voltages unlikely to change significantly.

Within a spin-fluctuation-mediated pairing scenario (see Sec. III A), this renormalization of U and increase of χ_0 would tend to decrease the pairing interaction, given the assumption that $U\chi_0$ at zero gate voltage would be unchanged, as our empirical approach would dictate. The increase would occur due to the form of the pairing interaction $V(\mathbf{k}-\mathbf{k}')=U^2\chi_0(\mathbf{k}-\mathbf{k}')/[1-U\chi_0(\mathbf{k}-\mathbf{k}')]$, with the extra power of U in the numerator. However, most of the superconductivity calculations in this paper do not assume a particular microscopic pairing potential, and the conclusion reached in Sec. III A—that gap symmetry would likely change due to gating in a spin-fluctuation pairing scenario—remains unchanged. For simplicity, therefore, we consider only the upper band.

Then within BCS weak-coupling theory the usual gap equation is given by

$$\Delta(\mathbf{k})=-\sum_{\mathbf{k}'}\frac{V_{\mathbf{k},\mathbf{k}'}\Delta(\mathbf{k}')\tanh(\beta E_{\mathbf{k}'}/2)}{2E_{\mathbf{k}'}}. \tag{2}$$

Here $\beta=1/kT$ and $E_{k'}=\sqrt{(\epsilon_{k'}-\mu)^2+\Delta^2_{k'}}$. $V_{k,k'}$ is the interaction causing the superconductivity. For simplicity we shall assume a separable interaction of the form $V_{k,k'}=-\langle f^2\rangle^{-1}V_0 f(k)f(k')$, and we shall make the corresponding assumption that $\Delta(\mathbf{k})=f(k)\Delta(T)$. Here $\langle f^2\rangle$ represents the average of f^2 taken over the entire Brillouin zone, with the maximum value of $f=1$. We will study the effects of changing the chemical potential on T_c for several order parameters potentially applicable to the quasi-1D organics: $f(k)=\cos(k_a)$, $\sin(k_a)$, $\cos(k_b)$, $\sin(k_b)$, the f-wave order parameter $\sin(2k_a)$, $\cos(2k_a)$, and the s-wave order parameter $f=1$. We note that there is evidence that some of the $(\text{TMTSF})_2X$ compounds[22] contain triplet order parameters, which would suggest the $\sin(k_a)$, $\sin(k_b)$ or $\sin(2k_a)$ gap functions, but the issue has not been definitively decided, so for completeness we include calculations for both even- and odd-parity order parameters. We perform the momentum sum by integrating over the entire Brillouin zone. Then the gap equation becomes

$$1/V_0=\langle f^2\rangle^{-1}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\frac{dk_a dk_b}{(2\pi)^2}f^2(k')\frac{\tanh(\beta E_{k'}/2)}{2E_{k'}}. \tag{3}$$

Finally, since we are interested in a possible raising of the transition temperature, the equation determining T_c [obtained by setting $\Delta(k')=0$ in the quasiparticle energy[23]] is as follows:

$$1/V_0=\langle f^2\rangle^{-1}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\frac{dk_a dk_b}{(2\pi)^2}f^2(k')\frac{\tanh[(\epsilon_{k'}-\mu)/2T_c]}{2(\epsilon_{k'}-\mu)}. \tag{4}$$

The corresponding equation determining T_{SDW} is[15]

TABLE I. V_0 and $T_{c,\max}$ values.

Order parameter	V_0 (meV)	$T_{c,\max}$ (K)
s wave	65.6	17.4
$\sin(k_a)$	38.5	1.3
$\cos(2k_a)$	46.2	5.8
$\cos(k_b)$	64.7	19.9
$\sin(k_b)$	65.9	17.8

$$1 = U \sum_{0 \leqslant k_a \leqslant \pi, |k_b| \leqslant \pi} \frac{f_F(\epsilon_{\mathbf{k}}) - f_F(\epsilon_{\mathbf{k}-\mathbf{Q}})}{\epsilon_{\mathbf{k}-\mathbf{Q}} - \epsilon_{\mathbf{k}}}. \quad (5)$$

Here f_F represents the Fermi function at T_{SDW} and Q is the SDW ordering or "nesting" vector.

Taking $t_a = 2900$ K, we find $T_c = 3.72 \times 10^{-4} t_a$. Given a half-filled top band, the chemical potential μ is $2\cos[(1/2)\pi/2] = 1.414 t_a$, at zero gate voltage. Solving the T_c gap equation at $t_b = 0.11 t_a$ for the various order parameters gives the V_0 values shown in Table I (we have also included the maximum value of T_c under each scenario). Here V_0 has been fixed for each order parameter by demanding that T_c at zero gate voltage be 1 K.

We note immediately a variation of more than one order of magnitude in the predicted T_c values, along with substantial variation in the coupling constant V_0 producing these T_c's. [We note parenthetically that the $\cos(k_a)$ order parameter (not shown) has a predicted maximum T_c of over 300 K but requires a V_0 of over 200 meV, which we regard as highly unlikely. Similarly, the $\sin(2k_a)$ order parameter (not shown) has a predicted maximum T_c of over 100 K, shown in Fig. 2(b), but requires a V_0 of 117 meV, which we also regard as relatively unlikely.]

III. RESULTS AND DISCUSSION

In Figs. 2(a) and 2(b) we plot our results for T_c for the various superconducting order parameters, along with the SDW ordering temperature, for the two cases $t_b = 0.08$ and 0.11. We see immediately that for both cases T_{SDW} rises sharply as the Fermi surface approaches the Van Hove singularity, reaching a maximum of 49 K at 98 mV gate voltage for the $t_b = 0.08$ case and 29 K at 80 mV gate voltage for $t_b = 0.11$. For $t_b = 0.08$ the Fermi surface first touches the Van Hove singularity at a chemical potential of $1.84t$, or 106 mV gate voltage; for $t_b = 0.11$ this happens at $1.78t$, or 91 mV. The destruction of the SDW via reduced nesting, caused by an increased t_b, observed at zero voltage does not occur once the Fermi surface is moved via application of a gate voltage; in fact, the nesting properties of the Fermi surface increase as it approaches the Van Hove singularity, as can be seen from Fig. 3.

We note also that, for both cases, for most of the region between zero and 100 mV gate voltage, the SDW wins over the superconductivity, with the exception of the $\sin(2k_a)$ order parameter [shown in the inset of Fig. 2(b)]. For the $t_b = 0.08$ case more favorable to SDW, only one of the four most probable order parameters, $\sin(k_b)$, shows a Van Hove

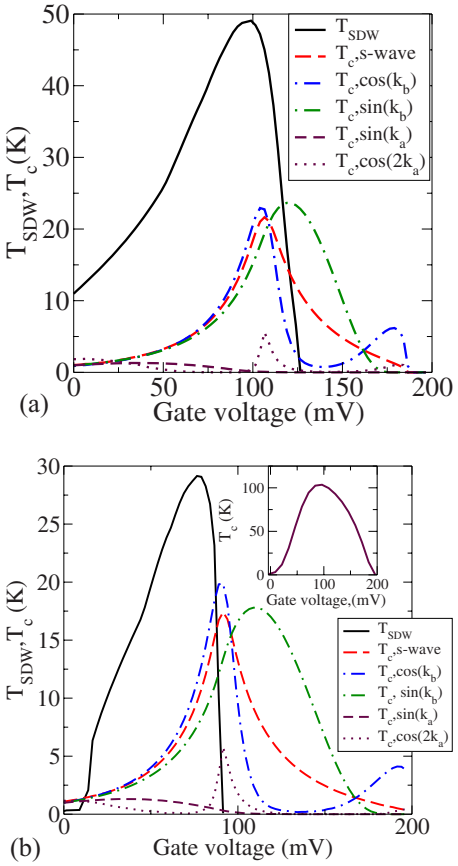

FIG. 2. (Color online) Superconducting and SDW transition temperatures as a function of gate voltage. (a) the case with $t_b = 0.08$, representing the TMTSF series at ambient pressure; (b) $t_b = 0.11$, representing the TMTSF series under pressure, as well as $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ at ambient pressure. Inset of (b): T_c vs gate voltage for $\sin(2k_a)$ order parameter.

maximum that is not subsumed by the SDW. For the $t_b = 0.11$ case, the Van Hove peaks for three of the four order parameters fall just outside the SDW curve, so that, depending on the order parameter of these materials, one may ob-

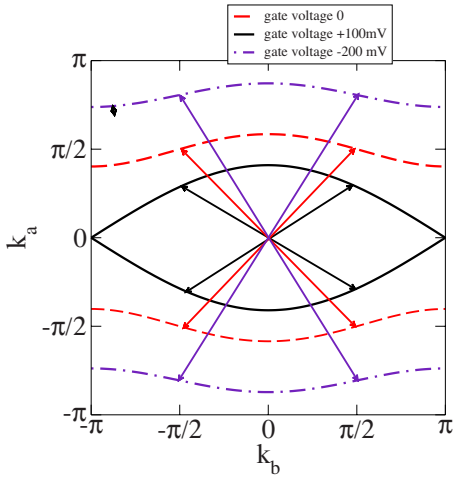

FIG. 3. (Color online) Fermi surfaces for the indicated gate voltages, with the nesting vectors indicated.

serve a SDW-superconductor transition around 100 mV. We note, however, that if the $\sin(k_a)$ order parameter is chosen by the system, the SDW will completely dominate the superconductivity and no dramatic SDW-superconductor transition will occur. This may be more likely given that this order parameter has the lowest V_0 of all order parameters modeled. We note, however, that there is evidence[24] that the TMTSF materials have line nodes on the Fermi surface, and the $\sin(k_a)$ order parameter is nodeless in the ungated situation. The matter will ultimately be decided by a gating experiment.

The main T_c curves generally show relatively sharp maxima rather near the point at which the Fermi surface just touches the Van Hove singularity, with the exception of the $\sin(k_b)$ curves, which exhibit broader maxima situated at a voltage slightly above this point. The reason for this is that this order parameter vanishes at $(k_a,k_b)=(0,\pm\pi)$, the points at which the Fermi surface first touches the Van Hove singularity. The maximum is therefore shifted higher in voltage, so that the Fermi surface no longer touches the points where the order parameter vanishes. The Fermi surface approaches the Van Hove singularity, resulting in a greater effective $N(E_F)$, and reaches regions of larger $\sin(k_b)$, increasing the effective coupling constant, but also shrinks in size; the competition between these three effects yields the broad maximum shown in both plots. By contrast, the $\cos(k_b)$ maximum is rather narrow and situated near 90 mV for the $t_b=0.11$ case and 100 mV for the $t_b=0.08$ case, the points where the Fermi surface just touches the Van Hove singularity at $(k_a,k_b)=(0,\pm\pi)$. Both the order parameter (i.e., $|f|$) and effective $N(E_F)$ are maximum at this point, yielding the narrow peaks shown in Fig. 2. One unusual feature of this order parameter is the secondary peak situated at 180 mV for $t_b=0.08$ and 190 mV for $t_b=0.11$. This feature occurs at a gate voltage such that the Fermi surface is very small and mainly encloses the point (0,0). At this point $\cos(k_b)$ reaches its maximum value, partly counteracting the decreased Fermi surface size, yielding the broad maximum seen in Fig. 2.

For completeness, we note that we have not included the effects of possible strong correlation (i.e., large Hubbard $U \gg t$), or Eliashberg-type strong-coupling effects in these calculations. Based on work performed for the cuprates,[25] the Eliashberg effects would tend to reduce the impact of the Van Hove singularity on T_c. There is evidence in the quasi-two-dimensional organic superconductors for such strong-coupling effects, based on the observed $\Delta(T=0)/T_c$ ratio,[26,27] as well as strong-correlation effects, with the Hubbard U of the order of the bandwidth $\sim 4t$.[28–33]

However, the evidence for Eliashberg-type strong coupling in $(TMTSF)_2X$ is less convincing. Two independent measurements[21,34] of the gap ratio $\Delta(T=0)/T_c$ reported values of 2.3 and 1.9, respectively, and while these values are enhanced from the BCS s-wave weak-coupling value of 1.76 they fall rather nearer the 2.14 weak-coupling d-wave value. It is worth noting[35] that a large number of order parameters share this d-wave value; in our case, the $\cos(k_a)$, $\cos(k_b)$, and $\sin(k_b)$ gap functions would fall into this category. In any case, given an anisotropic order parameter, these data would appear to suggest that the TMTSF superconductivity is not far from the weak-coupling limit. Finally, we note that, when the integration regime is not limited to a narrow band around the Fermi surface, but is extended over the whole Brillouin zone, as in Ref. 23, enhanced values of Δ_0/T_c are possible even within a weak-coupling formalism.

We also note that there is a wide range of Hubbard U estimates for the quasi-1D organics; a survey of published work[36–40] reveals U/t_a to be taken as low as 1.7 (Ref. 40) and as high as 7.[36] In our own calculations, the fitting of the SDW ordering temperature 11.5 K at zero gate voltage for $t_b=0.08$ yields $U=0.8t_a$, while a calculation in the reference work of Ishiguro[15] yielded $U=1.4t_a$. Given these wide disparities in the community in the actual value of U, we think it best to acknowledge that inclusion of the Coulomb repulsion may impact the effect of the Van Hove singularity on T_c, but that the actual size of this effect is unknown. Finally, we note that unconventional order parameters, as considered here, have real-space pair wave functions that vanish at the origin,[41] where the Coulomb repulsion is maximum, reducing the impact of this repulsion.

TABLE II. Effective coupling constants: spin fluctuation scenario.

Order parameter	$V_g=0$	$V_g=90$ mV
s wave	1.78	2.42
$\sin(k_a)$	0.159	−0.070
$\cos(k_a)$	−0.478	0.209
$\sin(2k_a)$	0.159	−0.070
$\cos(2k_a)$	0.274	−0.467
$\cos(k_b)$	−0.020	−0.081
$\sin(k_b)$	0.0065	0.027

A. Effective coupling constants and order parameter symmetry change

In an attempt to shed more light on the superconducting order parameter symmetry, and effects of gate voltage, in the TMTSF series, we have computed effective coupling constants (in arbitrary units) for each of the order parameters considered, based on the projection method described in Ref. 42 and assuming a spin-fluctuation interaction potential[13] of the form $V(\mathbf{k}-\mathbf{k}')=U^2\chi_0(\mathbf{k}-\mathbf{k}')/[1-U\chi_0(\mathbf{k}-\mathbf{k}')]$, with χ_0 the susceptibility defined in Eq. (6) below and U the on-site Coulomb potential. Note that a factor of $-1/3$ applies for the triplet, odd-parity order parameters. Since we determined T_c via a Brillouin-zone average in the previous section, the coupling constants have been similarly determined via a Brillouin-zone average, as opposed to the Fermi-surface average considered in Ref. 42. Finally, in order to calculate these constants it was necessary to use a somewhat smaller U ($0.5t_a$ instead of $0.8t_a$) than in the previous calculation, to avoid the intercession of a spin-density-wave instability phase in the calculation. As described previously, we have not considered the coexistence or competition problem between the SDW and superconductivity.[18] In Table II are presented the results of these calculations for two gate voltages

V_g, with negative coupling constants indicating attractive interactions.

As one would expect, in this scenario the *s*-wave channel is highly repulsive. The complex behavior of the other effective coupling constants, with several reversing sign as the gate voltage moves the Fermi level near the Van Hove singularity, suggests that, if in fact spin fluctuations are the source of the pairing interaction, order parameter symmetry changes as a function of gate voltage can be expected. Table II suggests that the $\cos(k_a)$ order parameter would be favored in the ungated situation, but that this would change to the $\cos(2k_a)$ order parameter as voltage is applied. This presumes, of course, that the superconductivity is not dominated by the SDW instability.

We have not attempted to model the effect of gate-voltage-variable coupling constants within the BCS theory presented earlier, as Table II might suggest, due to the uncertainty surrounding the pairing mechanism in the quasi-1D organics. We merely intend here to suggest the possibility of a gap symmetry change occurring with gating.

B. T_{SDW} and nesting effects

One point of observation regarding Fig. 2 is that, while both the SDW and superconducting instability are enhanced when the Fermi surface approaches the Van Hove singularity, this is not the only factor at work. In the t_b=0.11 plot, for example, the SDW becomes stronger than the superconductivity at gate voltages as low as 15 mV, and outstrips the superconductivity substantially over the next 70 mV, which cannot be explained as a simple density-of-states effect. The missing effect is that of the strength of the SDW instability at nesting vector **Q**, which can be quantified by the susceptibility χ:

$$\chi(\mathbf{Q}) = \sum_{\mathrm{BZ}} \frac{f(\epsilon_{\mathbf{k}-\mathbf{Q}}) - f(\epsilon_{\mathbf{k}})}{\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{Q}}}. \tag{6}$$

Figure 3 suggests that the nesting is increased in strength as the gate voltage increases toward 100 mV, and a quantitative calculation bears this out. Depicted in Fig. 4 is a plot of the maximum susceptibility observed by gate voltage [this maximum was typically observed near the vector $(2k_F, \pi)$ where $k_F = 2\cos^{-1}(\mu/2)$]. We see that the nesting increases as the gate voltage approaches 100 mV. A closer examination of Fig. 2 reveals that the SDW t_b=0.11 curve falls off significantly more steeply near 100 mV than the t_b=0.08 curve. It is this difference in slope that is largely responsible for the superconducting T_c peaks lying outside the SDW curve. In general, given the similarity between BCS superconductivity and SDW theory, one would naively expect that the Van Hove singularity has similar effects on both T_c and T_{SDW}, so

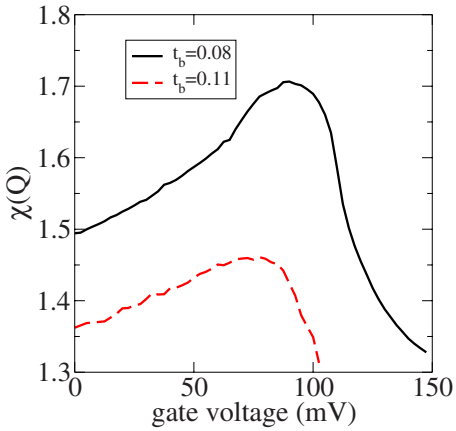

FIG. 4. (Color online) Susceptibility at the ordering vector **Q** for the t_b=0.08 and 0.11 cases.

that this behavior appears somewhat unusual. This difference in slope cannot be attributed solely to density-of-states effects, but arises from the decreased nesting associated with the larger t_b, as depicted in Fig. 4.

IV. CONCLUDING REMARKS

We note that, in the interests of simplicity and conciseness, we have modeled the TMTSF system as having a T_c of 1 K for t_b=0.11 and T_{SDW} of 11.5 K for t_b=0.08. These represent averages, and specific compounds have different values; $(TMTSF)_2FSO_3$, for example, has a T_c of 3 K under pressure,[15] so that for voltages with the Fermi surface near the Van Hove (VH) singularity, the superconductivity may win in this compound. The basic sequence, however, of T_{SDW} increasing as gate voltage is increased, then falling sharply at the VH singularity and superconductivity potentially appearing, should be unchanged. We are aware that performing a gating experiment on a sample under pressure could be very difficult,[43] but we also believe that the interest of such an experiment justifies substantial efforts to perform it.

To summarize, we here present results suggesting that electrostatic gating of quasi-one-dimensional organic superconductors $(TMTSF)_2X$ may result in a significant enhancement of the SDW and superconducting transition temperatures, due in part to the placement of the entire Fermi surface near an extended Van Hove singularity. We await with great interest the results of such a gating experiment.

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