Brief Reports

Brief Reports are accounts of completed research which, while meeting the usual **Physical Review** *standards of scientific quality, do not warrant regular articles. A Brief Report may be no longer than four printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Homogeneous quantum phase slippage in bulk charge-density-wave systems

Ji-Min Duan

Department of Physics & Astronomy, Northwestern University, 2145 Sheridan Road, Evanston, Illinois 60208

(Received 14 May 1993)

We study the dynamic process of homogeneous quantum phase slippage in *bulk* charge-density-wave systems. A topological singularity, namely, a "vortex shell" in 3+1 dimensions is proposed as a trial wave function of the saddle-point field. The quantum tunneling rate is roughly $\Gamma \sim \exp(-\text{const}/\varepsilon^2)$ as a function of applied electric field ε. The physics is qualitatively different from previous works.

The charge-density wave (CDW) is an interesting ground state of matter.[1] Below the Peierls transition temperature, electrons that are otherwise in the normal metallic state condense into a CDW condensate which can be described by a complex order parameter, in common with other ordered states such as the spin-density wave and the superfluid, etc. The CDW is mainly a quasi-one-dimensional phenomena, with an intrinsic chain structure in, e.g., $NbSe_3$. The coherence length in the chain (z axis) direction is $\xi_z \sim 10^2$ Å, while in the perpendicular directions, $\xi_x, \xi_y \sim 10$ Å. Much interesting physics stems from this one dimensionality, such as, for example, charged solitons.[2]

Experimentally, one of the most interesting properties of the CDW condensate is nonlinear transport in an electric field ε applied parallel to the chain direction. In addition to a normal linear conductivity, a nonlinear conductivity is found[3] to scale as $\sim\exp(-\text{const}/\varepsilon)$. There are *two quantum-tunneling theories* trying to explain this phenomena. One paper by Maki[4] proposes quantum nucleation of the soliton-antisoliton pair in a one-dimensional space plus a one-dimensional imaginary time (1+1D). The other by Bardeen[5] deploys a Landau-Zener–type tunneling model. Both these theories rely on quasi-one-dimensionality and get a scaling form of the conductivity as $\exp(-\text{const}/\varepsilon)$. However, late experiments suggest that the nonlinear conductivity is *thermally excited.*[6] This discrepancy was reconciled in a quantitative theory by Ramakrishna *et al.*[7] by realizing that the length scale of the experimental samples is much larger than the coherence length and, hence, one must use the 3D Ginzburg-Landau free energy to formulate the corresponding theory. The essence of their model is a thermally excited dislocation loop (i.e., a vortex ring), similar to the one proposed by Langer and Fisher[8] in the context of superfluid ^{4}He.

However, the issue of quantum tunneling in a CDW remains unsettled. Theoretically, we are curious to know the corresponding quantum process for 3D CDW's. This issue is also of experimental interest, since at extremely low temperature thermal activation is presumably dominated by quantum tunneling. It is therefore desirable to estimate this quantum limit to see if it is experimentally feasible. In this paper we investigate the homogeneous quantum phase slippage in *bulk* CDW systems in the weak impurity potential limit, in the hope that the physics presented here is qualitatively correct, and that quantitatively the order-of-magnitude estimate of the tunneling exponent is not misleading.

To study the dynamical quantum process we need to consider the time variation of the order parameter. At low temperature the dynamics of the phase ϕ of the order parameter is believed to be important and there is a characteristic phason velocity c_0 along the chain direction.[2,4] There is also a collective mode for the order-parameter amplitude A with a characteristic velocity $2c_0/\sqrt{3}$ in the chain direction.[9] Together with the static 3D free energy,[7] we construct a total action S as a functional of the order parameter in 3D space plus 1D time as

$$S\{Ae^{i\phi}\}=\int_{-\infty}^{\infty} dx\,dy\,dz\,dt \left\{ \frac{A^2}{2}\left[\frac{K_z}{c_0^2}\left[\frac{\partial\phi}{\partial t}\right]^2 - K_x\left[\frac{\partial\phi}{\partial x}\right]^2 - K_y\left[\frac{\partial\phi}{\partial y}\right]^2 - K_z\left[\frac{\partial\phi}{\partial z}\right]^2\right] + \frac{1}{2}\left[\frac{3K_z}{4c_0^2}\left[\frac{\partial A}{\partial t}\right]^2 - K_x\left[\frac{\partial A}{\partial x}\right]^2 - K_y\left[\frac{\partial A}{\partial y}\right]^2 - K_z\left[\frac{\partial A}{\partial z}\right]^2\right] + \frac{\alpha}{2}A^2 - \frac{\beta}{4}A^4 - \frac{e\rho_s(A)}{Q}\varepsilon\phi - V_{\text{imp}}(\phi)\right\}. \quad (1)$$

0163-1829/93/48(7)/4860(4)/$06.00

Here, K_i $(i=x,y,z)$ are anisotropic elastic constants measuring the bending energies of the order parameter; α,β are the usual Ginzburg-Landau coefficients; e is the absolute charge of an electron; $\rho_s(A)$ is the number density of condensed electrons; $Q=2k_F$; and the impurity pinning potential $V_{\rm imp}(\phi)$ is periodic in ϕ.

From (1) we can define anisotropic coherence lengths $\xi_i=\sqrt{K_i/\alpha}$ and a homogeneous condensate amplitude $A_\infty=\sqrt{\alpha/\beta}$. For later convenience, we will from now on work in dimensionless coordinates in which the order-parameter amplitude $f=A/A_\infty$ $(0\le f\le 1)$ and space variables are scaled by coherence lengths, and imaginary time τ $(\tau=it)$ is scaled by the "coherence time" ξ_z/c_0,

$$x'=x/\xi_x,\quad y'=y/\xi_y\ ,$$

$$z'=z/\xi_z,\quad \tau'=\tau/(\xi_z/c_0)\ .$$

The Euclidean action is

$$S_{\rm Eucl}=\frac{\alpha A_\infty^2}{2}\frac{\xi_x\xi_y\xi_z^2}{c_0}\times(S_{\rm phase}+S_{\rm ampl}+S_{\varepsilon\ \rm field}+S_{\rm imp})\ , \tag{2a}$$

where the contributions from phase, amplitude, electric field, and impurity potential are, respectively,

$$S_{\rm phase}=\int d^4r' f^2\left[\left[\frac{\partial\phi}{\partial\tau'}\right]^2+\left[\frac{\partial\phi}{\partial x'}\right]^2+\left[\frac{\partial\phi}{\partial y'}\right]^2+\left[\frac{\partial\phi}{\partial z'}\right]^2\right]\ , \tag{2b}$$

$$S_{\rm ampl}=\int d^4r'\left[\frac{3}{4}\left[\frac{\partial f}{\partial\tau'}\right]^2+\left[\frac{\partial f}{\partial x'}\right]^2+\left[\frac{\partial f}{\partial y'}\right]^2+\left[\frac{\partial f}{\partial z'}\right]^2-f^2+\frac{f^4}{2}\right]\ , \tag{2c}$$

$$S_{\varepsilon\ \rm field}=\int d^4r'\frac{2e\rho_s(A)}{Q\alpha A_\infty^2}\varepsilon\phi\ , \tag{2d}$$

$$S_{\rm imp}=\int d^4r'\frac{2}{\alpha A_\infty^2}V_{\rm imp}(\phi)\ . \tag{2e}$$

From (2a) and (2c), one can see that $-\alpha A_\infty^2/4$ is the homogeneous bulk condensation energy density, while the factor $\xi_x\xi_y\xi_z^2/c_0$ at the right-hand-side (RHS) of (2a) is clearly the 3+1D coherence volume.

The quantum phase slippage rate is

$$\Gamma=F\exp(-\Delta S_{\rm Eucl}/\hbar)\ , \tag{3}$$

where the prefactor F originates from small fluctuations around the tunneling path and is less important. In this paper we concentrate on the exponent where $\Delta S_{\rm Eucl}$ means the difference between saddle-point action and metastable-state action, both of which are extremals of $S_{\rm Eucl}$, i.e., satisfying the condition that the functional variation of $S_{\rm Eucl}$ vanishes. The metastable state is just a homogeneous plane-wave state along the chain direction, while the saddle-point state, in principle, must be solved from a 3+1D (equivalent to 4D) nonlinear equation, which turns out to be difficult. So we use trial wave-function method to construct the picture of this topological soliton.

We assume that the saddle-point picture is a linear superposition of the homogeneous metastable state and a localized excitation centered at, e.g., the origin. This local excitation must have the topology of slipping the phase by 2π. A vortex ring has just this topology.[8] So, we propose a "vortex shell" in the $x'y'\tau'$ space as the new topological singularity (see Fig. 1), with a radius R ($\gg 1$) and a thickness $\sim O(1)$. Within the thickness of the wall, f drops to zero while outside the wall (both inside and outside the shell) $f\cong 1$. The circulation direction points to the unit vector $\mathbf{e}_{\varphi'}$ of the azimuthal angle φ' in the $x'y'$ plane.

This vortex shell, in fact, describes the dynamic process of quantum nucleation of a vortex ring, since a snapshot at fixed τ' corresponds to a vortex ring of unit vorticity in real space (see Fig. 2): (a) at $\tau'=-R$ a singular point appears at the center of the $x'y'$ plane where $f=0$; (b) for $-R<\tau'<0$ the singular point evolves into an expanding vortex ring; (c) at $\tau'=0$ the ring stops to expand; (d) for $0<\tau'<R$ the ring shrinks; and (e) finally, at $\tau'=R$, the ring converges back into a singular point and disappears. This picture is similar to that of Ref. 8 (see Fig. 1 of Ref. 8). And, in common with all nucleation processes, there is a critical shell size R_c (to be determined later) which corresponds to the saddle point. For $R<R_c$ the shell collapses into itself, while for $R>R_c$ it expands to infinity and hence slips the phase by 2π.

Mathematically, the vortex shell is described by

$$\mathbf{K}(\mathbf{r}')=2\pi\mathbf{e}_{\varphi'}\delta(\sqrt{x'^2+y'^2+\tau'^2}-R)\delta(z')\ . \tag{4}$$

Its Green's function is

$$\mathbf{P}(\mathbf{r})=-\frac{1}{4\pi^2}\int d^4r'\frac{\mathbf{K}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|^2}\ , \tag{5}$$

which satisfies $\partial_\mu\partial^\mu\mathbf{P}(\mathbf{r})=\mathbf{K}(\mathbf{r})$. All vectors in Eqs. (4)

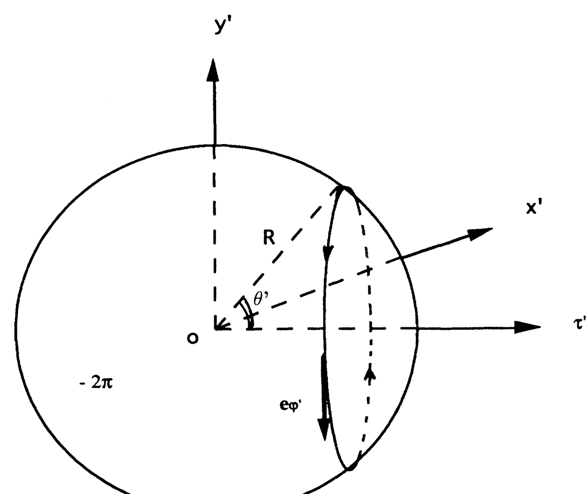

FIG. 1. A vortex shell in $x'y'\tau'$ space. -2π indicates the 2π phase loss inside the shell.

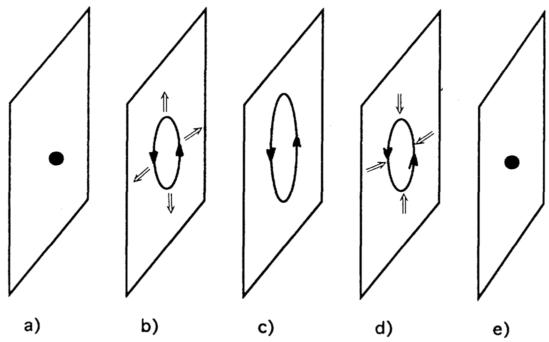

FIG. 2. Snapshots of the vortex shell at different imaginary times. Only the $z'=0$ plane is drawn here.

and (5) are 4D, but **K** and **P** have only two components since $\mathbf{e}_{\varphi'}$ is in the $x'y'$ plane; δ's are δ functions; $\mu=(x',y',z',\tau')$ and the repeated index means summation. Since $\mathbf{e}_\mu \cdot \mathbf{e}_\nu = \delta_{\mu\nu}$, we have $\partial_\mu = \partial^\mu$, etc.

In 3D, the phase ϕ obeys $\nabla^2\phi=0$ outside the singular regions (say, cores of vortex lines) so one can find a vector potential **v** (corresponding to our **P**) such that the gradient of the phase is equal to the curl of the vector potential $\nabla\phi=\nabla\times\mathbf{v}$. In 4D the correspondent of $\nabla\times\mathbf{v}$ is a two-index antisymmetric tensor[10] $\mathbf{B}_{\mu\nu}=\partial_\mu P_\nu-\partial_\nu P_\mu$ and $(\partial_\mu\phi)^2 \leftrightarrow B^2_{\mu\nu}/2=(\partial_\mu P_\nu)^2-\partial_\mu P_\nu \partial_\nu P_\mu$. Using partial integration and the property that surface integration vanishes at infinity for our local excitation, we have $\int B^2_{\mu\nu}/2=-\int[\mathbf{P}\cdot(\partial_\mu\partial_\mu\mathbf{P})+(\partial_\mu P_\mu)^2]$. From (4) and (5), one can show $\partial_\mu P_\mu=0$. Using $\mathbf{e}_\varphi\cdot\mathbf{e}_{\varphi'}=\cos(\varphi-\varphi')$, we find the contribution of the vortex shell to the phase action (2b):

$$\Delta S_{\text{phase}}(R)=-\int d^4r\,\mathbf{P}\cdot\mathbf{K}=\pi R^2 C \tag{6}$$

and

$$C=\int_0^\pi d\theta\int_0^\pi d\theta'\int_0^{2\pi}d\varphi\frac{\sin\theta\sin\theta'\cos\varphi}{1-\cos\theta\cos\theta'-\sin\theta\sin\theta'\cos\varphi} \quad (>0)\,.$$

The symbol C contains divergence which precludes an exact estimate. In principle, one can regulate such divergence by using the cutoff $1/R$ so that C is a function of $\ln R$. After integration over φ, we find that the leading divergent term in C is $2\pi\ln^2 R$, and for practical nucleation processes we assume $C\sim O(1)$.[7,8]

The order-of-magnitude estimate of amplitude action is easy, since f only varies appreciably within the hard core on the vortex shell; in the z' direction the ring thickness is $\sim O(1)$, and the integrand at the RHS of (2c) is $\sim O(1)$. These give us

$$\Delta S_{\text{ampl}}(R)\approx 4\pi R^2 \tag{7}$$

which is basically the surface area of the shell, and is comparable to ΔS_{phase} [see (6)].

The electric-field action [Eq. (2d)] is also obtainable, since the principal role of the topological vortex shell is to lose the exact 2π phase inside the shell. The integration of the phase change outside the shell cancels due to antisymmetry and gives a null result. So, $\Delta S_{\varepsilon\text{ field}}$ is proportional to the volume of the shell,

$$\Delta S_{\varepsilon\text{ field}}(R)\cong-\frac{(4\pi)^2 e\rho_s(A_\infty)}{3Q\alpha A_\infty^2}\varepsilon R^3\,. \tag{8}$$

An estimate for impurity action [Eq. (2e)] is hard because we do not have an explicit form of $V_{\text{imp}}(\phi)$. But since the phase change is 2π inside the shell and it decays to zero quickly outside the shell as the distance from the origin increases, and $V_{\text{imp}}(\phi)$ is periodic in ϕ, we know that only those impurities with their space time close to and outside the shell contribute significantly, hence, $\Delta S_{\text{imp}}(R)\sim R^2$, which is the same as the phase and amplitude terms. For the weak impurity potential ΔS_{imp} is qualitatively unimportant and we will neglect it.

Using Eqs. (6)–(8) and Eq. (2a), we get

$$\Delta S_{\text{Eucl}}(R)\approx\frac{\alpha A_\infty^2}{2}\frac{\xi_x\xi_y\xi_z^2}{c_0}\times\left[4\pi R^2 C'-\frac{(4\pi)^2 e\rho_s(A_\infty)}{3Q\alpha A_\infty^2}\varepsilon R^3\right], \tag{9}$$

where $C'=1+C/4$ is of order unity. From (9) one can see that $\Delta S_{\text{Eucl}}(R)\sim R^2$ for small R, and $\Delta S_{\text{Eucl}}\sim -R^3$ for large R (see Fig. 3). Similar to other nucleation problems, there is a competition between the surface energy (R^2 term) and the bulk energy (R^3 term) of the vortex shell. The critical-shell size R_c is determined from $\partial[\Delta S_{\text{Eucl}}(R)]/\partial R=0$,

$$R_c=\frac{C'Q\alpha A_\infty^2}{2\pi e\rho_s(A_\infty)}\frac{1}{\varepsilon}\,. \tag{10}$$

Note that R_c is inversely proportional to ε. Inserting (10) back into (9), and from (3) we get the quantum-tunneling rate

$$\Gamma\approx F\exp\left[-\frac{\alpha A_\infty^2}{2\hbar}\frac{\xi_x\xi_y\xi_z^2}{c_0}\left[\frac{\varepsilon_0}{\varepsilon}\right]^2\right], \tag{11}$$

where $\varepsilon_0=C'^{3/2}Q\alpha A_\infty^2/(\sqrt{3\pi}e\rho_s)$ is a constant with the dimension of an electric field. This scaling form is qualitatively different from those of previous quantum-tunneling theories.[4,5]

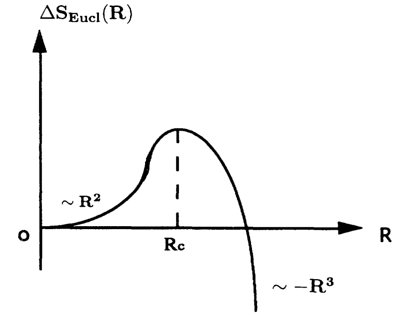

FIG. 3. $\Delta S_{\text{Eucl}}(R)$ as a function of shell radius R.

In principle, each tunneling event leads to a lower energy state. The energy difference per unit volume can be calculated from the electric-field term on the RHS of (1) by noting that after the tunneling phase in half-space, $z>0$ loses 2π and that for $z<0$ remains unchanged. This energy is dissipated through a phenomenological tunneling conductivity $\sigma_{\rm tun}$: $\sigma_{\rm tun}\varepsilon^2/2=2\pi\Gamma e\rho_s\varepsilon/2Q$. Using (11), we get

$$\sigma_{\rm tun}\approx\frac{2\pi e\rho_s}{Q\varepsilon}F\exp\left[-\frac{\alpha A_\infty^2}{2\hbar}\frac{\xi_x\xi_y\xi_z^2}{c_0}\left[\frac{\varepsilon_0}{\varepsilon}\right]^2\right]. \qquad (12)$$

Now we estimate the tunneling exponent for $NbSe_3$. From Ref. 7 we have $K_xA_\infty^2=6.2\times10^{-13}$ J/m, $K_yA_\infty^2=2.24\times10^{-13}$ J/m, $Q\cong10^{10}$ m^{-1}, and $\rho_s\cong10^{27}$ m^{-3}. Besides,[1] $c_0\cong10^4$ m/s and $\xi_z\sim10^2$ Å. So we have

$$\sigma_{\rm tun}\sim\Gamma\sim\exp\left[-\left[\frac{3\times10^7}{\varepsilon}\right]^2\right],$$

where ε is in units of V/m. This tunneling rate is too small to be detected experimentally since the applied field is usually[1] $\varepsilon<10^4$ V/m. In order to observe this quantum effect, one ideally needs $\varepsilon>3\times10^6$ V/m, but this electric field is strong enough to destroy the CDW condensate.

Finally, we remark on the method we use in evaluating $S_{\rm phase}$ [i.e., Eq. (2b)]. Presumably, $S_{\rm phase}$ is the action of the "Higgs boson" field ϕ with a single degree of freedom. We identify it with the action of the corresponding "pseudoelectromagnetic field" $B_{\mu\nu}$ with the "vector potential" P_μ. Note that in doing so we do not introduce extra degrees of freedom, since there are only two nonzero components of P_μ which are constrained to each other by the "Lorentz gauge" $\partial_\mu P_\mu=0$.

In summary, we propose a topological soliton, i.e., a 3+1D vortex shell as the saddle-point picture of quantum phase slippage in bulk CDW systems. This quantum effect is unlikely to be observed experimentally. The physics discussed here and the conclusions are qualitatively different from that of the relevant quantum theories proposed earlier.[4,5]

We are indebted to Sungkit Yip for stimulating discussions. We also would like to thank Anupam Garg, Tony Leggett, and Mike Stone for a discussion.

---

[1]For a recent review on CDW's, see G. Gruner, Rev. Mod. Phys. **60**, 1129 (1988).

[2]M. J. Rice, A. R. Bishop, J. A. Krumhansl, and S. E. Trullinger, Phys. Rev. Lett. **36**, 432 (1976).

[3]P. Monceau *et al.*, Phys. Rev. Lett. **37**, 602 (1976).

[4]K. Maki, Phys. Rev. Lett. **39**, 46 (1977).

[5]J. Bardeen, Phys. Rev. Lett. **42**, 1498 (1979).

[6]J. C. Gill, J. Phys. C **19**, 6589 (1986).

[7]S. Ramakrishna, M. P. Maher, V. Ambegaokar, and U. Eckern, Phys. Rev. Lett. **68**, 2066 (1992).

[8]J. S. Langer and M. E. Fisher, Phys. Rev. Lett. **19**, 560 (1967).

[9]P. A. Lee, T. M. Rice, and P. W. Anderson, Solid State Commun. **14**, 703 (1974).

[10]3D is a special case where $\partial_\mu P_\nu-\partial_\nu P_\mu$ is a vector.