PHYSICAL REVIEW B

CONDENSED MATTER AND MATERIALS PHYSICS

THIRD SERIES, VOLUME 57, NUMBER 4

15 JANUARY 1998-II

RAPID COMMUNICATIONS

Rapid Communications are intended for the accelerated publication of important new results and are therefore given priority treatment both in the editorial office and in production. A Rapid Communication in **Physical Review B** may be no longer than four printed pages and must be accompanied by an abstract. Page proofs are sent to authors.

Dimensional crossover of quantum nucleation processes in charge-density-wave phase slips

Noriyuki Hatakenaka NTT Basic Research Laboratories, Atsugi, Kanagawa 243-0198, Japan

Masato Shiobara, Ken-ichi Matsuda, and Satoshi Tanda Department of Applied Physics, Hokkaido University, Sapporo 060-8628, Japan (Received 10 July 1997; revised manuscript received 21 November 1997)

A phenomenological model for quantum phase slips of charge-density waves that takes into account the system-size effect is presented. The process of quantum nucleation leading to the phase slip changes from vortex-pair to vortex-ring creations as the external electric field increases, which is analogous to the evolution of a ripple in a rectangular water tank. The crossover field is determined by the system size. The present model describes a number of features observed in the nonohmic conductivity in TaS_3 at low temperature. [S0163-1829(98)52004-8]

Since the beginning of 1980, the applicability of quantum mechanics on a macroscopic scale has been tested through typical quantum effects. One such effect is macroscopic quantum tunneling (MQT), which has been established via quantum tunneling of the phase difference of Josephson junctions with small capacitance. It has been clarified that tunneling on a macroscopic scale is strongly affected by the macroscopic degrees of freedom. Later, MQT was verified in various macroscopic systems, including the domain wall in ferromagnets and the spin-density wave in (TMTSF)₂PF₆.

Although the charge-density waves (CDW's) are another type of macroscopic quantum phenomena, the issue of quantum tunneling in CDW's (Refs. 6 and 7) remains unsettled for two main reasons: at high temperatures quantum processes are probably masked by classical processes, and there is a lack of the evidence associated with macroscopic numbers of electrons. Zaitsev-Zotov recently made a breakthrough.⁸ He observed *temperature-independent* nonlinear conduction at low temperatures and that the electric current is transferred by portions of charge corresponding to displacement of the CDW's by a distance of the order of their wavelength. Although he tried to explain his experimental results in terms of quantum creep, they might be possibly explained by MQT of CDW phase slips. The interpretation of the results are still an open question. At least he

clarifies previously uncertain points and provides a physical picture of nonohmic conductivity at low temperatures, that is, the phase slip model.^{9,10}

Zaitsev-Zotov's most important discovery was to find a scaling form of nonlinear conductivity expressed as $\exp(-\operatorname{const}/\epsilon^2)$, where ϵ is an applied electric field. This form was a problem for the existing tunneling models^{6,7} with $\exp(-\operatorname{const}/\epsilon)$. To overcome this problem, Duan has proposed a homogeneous quantum phase-slippage model based on the vortex-ring creation in bulk CDW systems.¹¹ This successfully explains the scaling form. However, he implicitly assumes that the low-temperature conduction of thick samples proceeds by the same mechanism as in thin ones. Thus his model cannot explain the system-size effect. The low-temperature conduction in thick and thin samples of TaS₃ is similar, but there are important differences: 12 (a) The ln I(E,T) as a function of temperature does not level out for $T \ge 4.2$ K, whereas in thin samples it levels out at T ≤ 10 K. (b) At T < 10 K, the slope of the Arrhenius plot of the current, $\partial \ln[I(E,T)]/\partial(1/T)$, increases with a decrease in the electric field, E, whereas it decreases in thin samples.

In this paper, we consider the effect of system size on quantum nucleations mechanisms leading to phase slips in the CDW of TaS_3 . In particular, we make a possible conjecture to explain the experimental results in terms of the di-

mensional crossover of the quantum nucleation from a vortex pair to a vortex ring as a function of an applied electric field.

The slope of the Arrhenius plot of the current is thought to be created by the competition between classical and quantum processes. The first difference (a) might be understood by considering the electric field to be fixed. The tunneling process is temperature-independent, while the thermal activation process depends exponentially on temperature. The crossover from a classical to a quantum process occurs as the temperature is reduced past a certain point, and finally the tunneling process becomes dominant at low temperatures. The crossover temperature shifts to a higher temperature as the tunneling rate increases. One can easily imagine that the temperature-dependent current regions will shrink when the tunneling rate is high. We will show later that the tunneling rate in a thick sample is lower than that in a thin sample. In fact, Zaitsev-Zotov argued this in his experiment.⁸

Next, we discuss the electric field dependence of the slope. In general, both tunneling and thermal activation rates increase with an increasing external electric field. The slope decreases as ϵ increases if $\partial \Gamma_{\text{thermal}}/\partial \epsilon < \partial \Gamma_{\text{tunneling}}/\partial \epsilon$, where $\Gamma_{thermal}$ and $\Gamma_{tunneling}$ are thermal activation and tunneling rates, respectively. In the thick sample, this situation is supposed to be attained. On the other hand, in the thin samples, temperature-independent current has already been observed at lower electric fields. Then, the angle of the slope increases as the external electric field increases. This means above relation changes to $\partial \Gamma_{\text{thermal}}/\partial \epsilon$ $> \partial \Gamma_{tunneling}/\partial \epsilon$. This cannot be explained by monoscaling theory. An important clue to solving case (b) is also contained in Zaitsev-Zotov's experiments; the form of scaling seems to turn into $1/\epsilon^2$ from $1/\epsilon$ in an external electric field at 2 K (see the inset of Fig. 3 of Zaitsev-Zotov). Until now, all previous tunneling theories were considered not to be changed in their scaling form regardless of the electric field magnitudes. If we accept the change of scaling form as a function of an external electric field in one sample, it may be possible to attain $\partial \Gamma_{\text{thermal}}/\partial \epsilon > \partial \Gamma_{\text{tunneling}}/\partial \epsilon$. In what follows, we investigate the sample-size dependence of the tunneling rate and the change of scaling form as a function of an external electric field.

First let us consider how the scaling form changes as a function of an external electric field. The scaling form is determined by the type of nucleation process of a bubble leading to phase slips. Maki pointed out that the process is related to the anisotropy of the system; a phase vortex-pair process gives rise to a $1/\epsilon$ scaling form in a strongly anisotropic system, while a phase vortex-ring process leads to a $1/\epsilon^2$ scaling form in a more isotropic system. Unfortunately, the vortex pair and ring creations have been discussed separately, since anisotropy was previously thought to be a peculiar characteristic of the sample matter. In this case, the scaling form cannot be changed in one sample. Here we insist that this anisotropy depends on sample-size effect.

 TaS_3 is nearly isotropic within the xy plane perpendicular to the conduction direction because $\xi_x \sim \xi_y \sim 10$ Å. Thus we discuss how anisotropy results from such an isotropic situation. Suppose that you dropped one drop of water in the center of a rectangular water tank. It will create a ripple on the water surface that will spread through the whole water

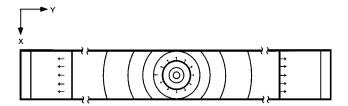


FIG. 1. The time evolution of a ripple created by one drop of water. A topological dislocation will grow in the same way.

tank, with time. At first, the ripple will spread in concentric circles (R = finite). After it reaches a long edge of the tank, the water wave front will be parallel to the edge (R = infinity) and will move toward another edge. This simple analogy demonstrates the continuous changes from ring to pair creations; that is the appearance of anisotropy from an isotropic situation. This is a key idea for the mechanism of change of nucleation processes in one sample. (See Fig. 1.)

Now let us apply this idea to the CDW system. The phase slips in CDW behave like water drops: a phase slip proceeds from a vortex ring to a vortex pair. In the imaginary-time path-integral formalism, the rate of quantum phase slips is expressed as

$$\Gamma = A \exp[-\Delta S_{\text{Eucl}}/\hbar], \qquad (1)$$

where $\Delta S_{\rm Eucl}$ is the difference between saddle-point action and metastable-state action. The action is basically described as

$$\Delta S_{\text{Fucl}} = -\alpha^{(D)} R^{D+1} \epsilon + \beta^{(D)} R^D, \qquad (2)$$

where D is a system dimension and R is the bubble radius. The first term of the right-hand side of Eq. (2) is volume energy of a bubble gained from an external electric field and the second is the surface tension energy of the bubble. The extremal of the action, $\Delta S_{\text{Eucl}}^{M}$, determines the tunneling rate since the prefactor A, originating from small fluctuations around the tunneling path, is less important. The action is estimated by using the radius expressed as

$$R_M = \frac{\beta^{(D)}D}{\alpha^{(D)}(D+1)} \frac{1}{\epsilon},\tag{3}$$

which is derived from $\partial \Delta S_{\text{Eucl}}/\partial R = 0$. R_M is inversely proportional to an external electric field. The larger ϵ is, the smaller R_M . Therefore, it is important to know whether R_M is larger than the system size. It determines the nucleation process: (i) $L_x < L_y < R_M$; phase slips occur coherently at whole cross sections. It is not necessary to create the phase slip center at the xy plane. (ii) $L_x < R_M < L_y$; the phase slip proceeds through the vortex-pair creations. (iii) $R_M < L_x < L_y$; the vortex-ring creation is dominant. In this way, the nucleation process changes from a vortex pair to a ring. The maximum action is given as

$$\frac{\Delta S_{\text{Eucl}}^{(D+1)}(R_M)}{\hbar} = \frac{1}{\hbar} \frac{\beta^{(D)}}{D+1} \left[\frac{\beta^{(D)}}{\alpha^{(D)}} \frac{D}{D+1} \right]^D \frac{1}{\epsilon^D} = \frac{\epsilon_0^{(D+1)}}{\epsilon^D}. \tag{4}$$

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The crossover field ϵ_{cr} can be estimated by setting $R_M \simeq L_x/2$ as

$$\epsilon_{cr} = \frac{2}{L_x} \frac{\beta^{(D)} D}{\alpha^{(D)} (D+1)}.$$
 (5)

To evaluate the sample-size-dependent tunneling rate and the crossover field, we recall the previous theories. ¹³ Since the amplitude coherence length of the CDW is orders of magnitude smaller than typical sample dimensions, the CDW phase slip should be three dimensional (3D). Starting from a 3+1D phase Hamiltonian, one can obtain the action

$$\Delta S_{\text{Eucl}} = A_0 \int \prod_{\mu=0,1,2,3} dx^{\mu} [(\partial_{\mu} \Delta \phi)^2 + 2(\partial_{\mu} \Delta \phi \cdot \partial_{\mu} \phi)],$$
(6)

where x^{μ} is one component of four-dimensional space-time coordinates: $x^0 = \tau$, $x^1 = x$, $x^2 = y$, and $x^3 = z$, $\Delta \phi$ is the phase difference from the metastable state $(\phi_{\rm m})$ defined by $\Delta \phi = \phi - \phi_{\rm m}$, and A_0 is constant. We ignore the impurity potential because the temperature-independent behavior of the current at fixed ϵ is observed only in a pure sample. In the 1+1D case, the action is given by

$$\Delta S_{\text{Eucl}}^{(1+1)} = A_0 \left(L_x \frac{v_F}{v_a} \right) \int \prod_{\mu=0,2,3} dx^{\mu} [(\partial_{\mu} \Delta \phi)^2 + 2(\partial_{\mu} \Delta \phi \cdot \partial_{\mu} \phi)], \qquad (7)$$

where v_F and v_a are the Fermi velocities for z and x directions, respectively. The factor $L_x v_F / v_a$ means physically the length of the vortex lines and shows the sample-size effect (the thicker the sample, the bigger the action). Thus the tunneling rate decreases as the sample size increases. This elucidates case (a).

Next, we discuss the crossover field to explain case (b). To do this, it is necessary to compare the action in 1+1D with that in 2+1D. Unfortunately, the parameters $\epsilon_0^{(1+1)}$ and $\epsilon_0^{(2+1)}$ may depend on many factors and are difficult to estimate using the existing theories. Instead of the theoretical values, we have used the experimental data. Zaitsev-Zotov fitted his data as $I^{(1+1)} = I_0^{(1+1)} \exp[-\epsilon_0^{(1+1)}/\epsilon]$ and $I^{(2+1)} = I_0^{(2+1)} \exp[-\epsilon_0^{(2+1)}/\epsilon^2]$. The former is a good fit for lower fields, while the latter is good for higher ones. Using the values $I_0^{(1+1)} = 0.015$ Å, $\epsilon_0^{(1+1)} = 900$ V/cm, $I_0^{(2+1)} = 1.5 \times 10^{-7}$ Å, and $\epsilon_0^{(2+1)} = 17433$ V²/cm², we obtained 43.8 V/cm as the crossover field. This is almost consistent with the field where the slope appears in Fig. 2 of Zaitsev-Zotov. The current $I^{(2+1)}$ is indeed smaller than $I^{(1+1)}$. This means that at low temperatures where the thermal activation is really excluded the quantum phase slip rate is slowing down as the field increases. This may cause the situation that makes a slope in I-(1/T) curves: $\partial \Gamma_{\text{temmal}}/\partial \epsilon > \partial \Gamma_{\text{tunneling}}/\partial \epsilon$.

In summary, we have discussed the quantum phase slips of charge-density waves in TaS_3 , taking into account the system-size effect. The phase slip process has been changed by increasing the external electric field from a phase vortex pair to a phase vortex ring. The present model describes a number of features observed in the nonohmic conductivity in TaS_3 at low temperatures. Our model will be tested by observing the crossover field predicted by Eq. (5) through samples with various dimensions.

We would like to thank Kazumi Maki, Sergei V. Zaitsev-Zotov, Takashi Sambongi, Hiroshi Matsukawa, Hiroyuki Miyake, Nobuo Matsumoto, and Kazuhiko Yamaya for valuable discussions. We also thank Mariko Uchitani for her kind assistance. One of us (S.T.) is grateful for his financial support through a Grant-in-Aid for Scientific Research from the Ministry of Education, Science, and Culture of Japan.

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