Creation of Soliton Pairs by Electric Fields in Charge-Density–Wave Condensates[(a)]

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The creation probability of soliton-antisoliton pairs due to quantum-mechanical tunneling in the presence of an electric field in charge-density–wave condensates is calculated. This process gives rise to an additional electric conductivity strongly nonlinear in the electric field, which may account in part for the anomalous conductivity observed in $NbSe_3$.

In order to account for the electric conductivity of low-temperature phases [which are currently identified with a pinned Fröhlich charge-density-wave (CDW) condensate] in the interesting linear-chain conductors tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ) and $K_2Pt(CN)_4Br_{0.3}\cdot 3H_2O$ (KCP) an extremely elegant model has been proposed by Rice *et al.*[1] The model is based on the idea by Lee, Rice, and Anderson[2] (LRA) that in an ideal system a perfectly uniform Fröhlich CDW condensate has two degrees of freedom, namely the amplitude and the phase, and that in a weakly pinned system the phase mode (phason) has an extremely small energy gap. Rice, Bishop, Krumhansl, and Trullinger (RBKT)[1] then pointed out that in this circumstance the nonlinear mode (φ soliton) associated with the phase degree of freedom plays the important role in the electric conductivity at low temperatures. They derived an explicit expression for the electric conductivity in the low-temperature limit and found that it has an activated form in qualitative agreement with the observed conductivities of TTF-TCNQ[3] and KCP,[4] if the pinning frequencies ω_f of these systems are sufficiently small ($\omega_f \sim 10^2$ K).

In this Letter I study the same model in the presence of an electric field ϵ. The model Lagrangian density is then given by[1]

$$L(\varphi) = L(\varphi_0) + N_0\{\tfrac{1}{2}\varphi_t^2 - \tfrac{1}{2}C_0^2\varphi_x^2 - \omega_f^2 V(\varphi)\} - e^*\varphi\epsilon, \quad (1)$$

where $N_0 = N_s m^* q_0^{-2}$ and the other notation is the same as in Ref. 1. The last term in Eq. (1) describes the coupling of φ to the electric field ϵ along the x direction. This term arises from the partial integration of

$$e\,\delta N_s(x,t)\,V(x) = -eN_s q_0^{-1}\varphi_x V(x), \quad (2)$$

where $\delta N_s(x,t)$ is the excess electron density associated with φ_x. For definiteness I take in the following a pinning potential

$$V(\varphi) = N^{-2}(1 - \cos N\varphi),$$

so that in the absence of ϵ, the equation of motion reduces to a sine-Gordon equation.

In the presence of an electric field ϵ, the Lagrangian density (1) predicts that φ-soliton and anti-φ-soliton pairs are created by the quantum-mechanical tunneling process. This can be most simply visualized as follows. In φ space the potential $V = N_0\omega_f^2 V(\varphi) + e^*\epsilon\varphi$ is a tilted sinusoidal function (see Fig. 1). When φ is one of the local minima (say φ_0) of the potential, it is always possible that within a small space-time region φ tunnels from φ_0 to φ_{-1}, which has potential energy lower by $\Delta E = e^*(2\pi/N)\epsilon$. Such a tunneling process can be most easily studied within the pseudoparticle model.[5,6] Following 't Hooft[5] we consider a two-dimensional Euclidean space E_2 instead of the ordinary Minkowski space M_2 by letting $t = -i\tau$.

In this E_2 space the above tunneling process is represented[6] as formation of a small (two-dimensional) bubble with $\varphi = \varphi_{-1}$ in the matrix of the $\varphi = \varphi_0$ vacuum. Since the action A is isotropic in the $(C_0\tau, x)$ space, the most favorable bubble is circular with radius $R = [(C_0\tau)^2 + x^2]^{1/2}$. (Here I have assumed that the bubble appears at the origin, although the center of the bubble can be anywhere in the E_2 space.) If the radius of the bubble is larger than the soliton width[1] $d = C_0/\omega_f$, the

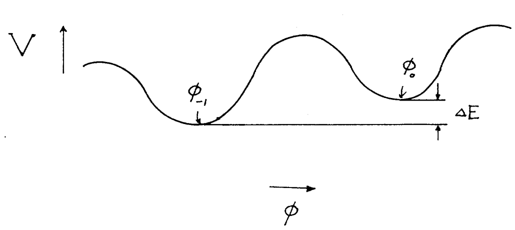

FIG. 1. Total potential V vs φ.

action associated with the bubble is given by[6]

$$C_0 A = -\pi R^2 \Delta E + 2\pi R S, \tag{3}$$

where the first term arises from the volume energy while the second term comes from the surface energy. The surface tension S is nothing but the soliton energy,

$$S = E_\varphi = 2(2/N)^2 N_0 C_0 \omega_f, \tag{4}$$

at least for a small ϵ.

The most probable tunneling takes place at the extremum of the action A, which is given by

$$\partial A/\partial R = 0, \text{ or } R = R_m = S/\Delta E = 4N_0 C_0 \omega_f / \pi e^* N \epsilon, \tag{5}$$

with

$$A_m = A(R_m) = \frac{\pi}{C_0}\frac{S^2}{\Delta E} = \frac{\frac{1}{2} N E_\varphi^2}{e^* C_0 \epsilon}. \tag{6}$$

According to Frampton,[6] the tunneling probability per unit space-time is then given by

$$P = \frac{C_0}{\pi R_m^2}\exp\left(-\frac{1}{\hbar A_m}\right) = \frac{4\pi C_0}{N^2}\left(\frac{e^*\epsilon}{E_\varphi}\right)^2 \exp\left(\frac{-\epsilon_0}{\epsilon}\right), \tag{7}$$

with

$$\epsilon_0 = N E_\varphi^2 / 2\hbar e^* C_0. \tag{8}$$

Physically the coefficient in front of the exponential follows from the consideration that in order that the local pair creation be independent, the elementary bubbles I have considered cannot overlap each other.

Finally the energy production per unit space-time is given by

$$W = 2E_\varphi P = \left(\frac{2}{N}\right)^2 \frac{C_0}{E_\varphi}(e^*\epsilon)^2 \exp\left(-\frac{\epsilon_0}{\epsilon}\right), \tag{9}$$

which can be rewritten in terms of the quantum-mechanical tunneling conductivity σ_{tun} in the presence of the electric field by $W = \sigma_{\text{tun}}\epsilon^2$. This yields

$$\sigma_{\text{tun}} = 2\pi\left(\frac{2}{N}\right)^2 e^{*2}\frac{C_0}{E_\varphi}\exp\left(-\frac{\epsilon_0}{\epsilon}\right). \tag{10}$$

Equation (10) predicts that in the system described by Eq. (1), there is an additional conductivity associated with the soliton pair production due to the electric field (of pure quantum-mechanical origin) and it is strongly nonlinear in ϵ. So far I have ignored what will happen to the soliton-antisoliton pairs thus created. In order to establish a stationary state in the presence of continual pair production, those pairs should be annihilated into phasons very efficiently by some relaxation mechanism, which is not included in Eq. (1), so that the creation process would be the bottleneck of the pair dissipation process. Within this limitation Eq. (10) gives the correct conductivity associated with the pair production.

In order to assess the importance of Eq. (10), it is useful to compare this with the conductivity due to the thermally activated φ solitons[1]:

$$\sigma_{\text{therm}} = \frac{4e^2\omega_f l}{(F_\varphi k_B T)^{1/2}}\left(\frac{n_s}{n}\right)^2 \exp\left(-\frac{E_\varphi}{k_B T}\right).$$

In the low-temperature limit, the ratio of the two conductivities is approximately given by

$$\frac{\sigma_{\text{tun}}}{\sigma_{\text{therm}}} = \frac{d}{l}\left(\frac{k_B T}{E_\varphi}\right)^{1/2} \exp\left(\frac{E_\varphi}{k_B T} - \frac{\epsilon_0}{\epsilon}\right). \tag{11}$$

This implies that when $E_\varphi/K_B T \gg \epsilon_0/\epsilon$ the tunneling term dominates the conductivity. Making use of accepted values[1] of E_φ, C_0, etc., for TTF-TCNQ and KCP, I conclude that the tunneling process is in fact insignificant in the temperature regime where the experiments are currently conducted.[3,4] However, at lower temperatures (say $T < 1$ K) or in large electric fields the predicted tunneling term should be seen in experiments in these linear systems if the model (1) has some validity.

Turning to other systems, we find that conductivity of the form (10) appears already to have been observed[7] in $NbSe_3$, which is considered another linear-chain-type conductor in addition to TTF-TCNQ and KCP already mentioned. However, in order to apply Eq. (10) directly to $NbSe_3$, there are serious difficulties. The experimental data by Monçeau *et al.*[7] on $NbSe_3$ near 123 and 54 K require that $E = 0.63$ and 0.15 K, respectively. This implies that in the CDW condensate of $NbSe_3$ the pinning potential should be extremely small, $\omega_f \sim 1$ K, and the soliton width extremely long, $d(=C_0/\omega_f) \sim 10^{-4}$ cm. More seriously, however, this means, that there is an overwhelming number of thermally excited solitons in the above temperature regime, which should give rise to practically the normal conductivity.[1] Therefore before Eq. (10) is applied to the conductivity of $NbSe_3$, we have to understand why the thermally activated solitons do not contribute significantly to the conductivity. Furthermore, in the presence of other solitons, the production probability of the soliton-antisoliton pair should be certainly modified, a circumstance of which further clarification is certainly desirable.

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Bremsstrahlung Resonances and Appearance-Potential Spectroscopy near the 3*d* Thresholds in Metallic Ba, La, and Ce

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The high polarizability of the 3*d* shells in Ba, La, and Ce leads to a dynamically screened $E_g \rightarrow 4f$, $\hbar\omega$ electron-photon coupling, having pronounced resonances due to intermediate $3d^9 4f^2$ levels. We qualitatively and even quantitatively reproduce the asymmetric bremsstrahlung intensity resonances experimentally observed by Liefeld, Burr, and Chamberlain.

In recent years there has been much interest shown in electron-excited soft-x-ray emission spectra as a means for investigating densities of states in solids and also as a tool for surface studies of more applied character. Whenever the energy of the incident electron is far away from core-level excitation thresholds the bremsstrahlung emission can be described as a one-electron process. In appearance-potential spectroscopy (APS), however, one is interested in varying the incident electron energy in the very threshold region and the dynamics of the core electrons can then become extremely important. Liefeld and co-workers[1,2] have experimentally demonstrated that for La and Ce in the M_α, M_β (3*d*) region the variations of the x-ray emission is due to a huge resonance in the bremsstrahlung "background" rather than to onset of characteristic soft-x-ray emission. In this Letter we study the resonant bremsstrahlung process in the 3*d* threshold region of Ba, La, and Ce[1,2] and compare with APS data for La.[3-5]

For incident electron energy E well below the 3*d* ionization threshold the bremsstrahlung process is well represented by the one-electron process shown in Fig. 1(a). The experiments of Liefeld, Burr, and Chamberlain[1] indicate that the incident electron can drop into a quite sharp 4*f*-like empty level, about 2 eV wide and 5.5 eV above

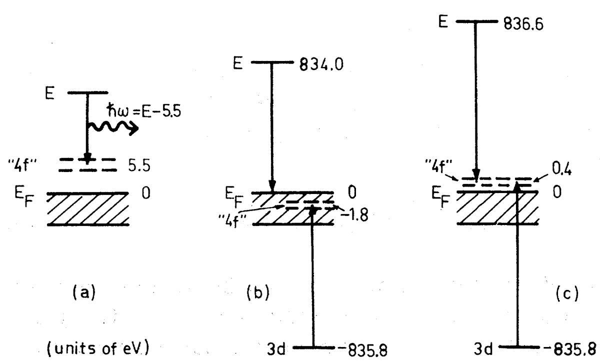

FIG. 1. "One-electron" band pictures with different, effective 4*f* levels for (a) the direct, nonresonant bremsstrahlung process; (b) the onset of characteristic $M_\alpha (M_\beta)$ line emission; (c) the nonradiative transitions to the resonant intermediate $3d^{-1}4f^2$ level. Note that a one-electron picture really is not valid.