smaller H values ($H < H_e$), the effective field experienced by the Mn^{++} ions is a vectorial sum of H and H_e, the mean spin direction of the magnetic molecule being no longer aligned along H.

To conclude, SFRS in semimagnetic semiconductors is a new giant magneto-optic effect, which could be expected. But it also reveals a powerful tool to study magnetic molecules through a direct process (spin flip), which is simply related to the magnetic moment of these complexes.

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[9] In addition, microscopic inhomogeneities are likely responsible for a part of the linewidth of SFRS. In the case of impurities, they are enhanced by the spatial localization (~50 Å) of the magnetization inducing the bound-electron spin splitting.

[10] Actually, the free electrons experience a magnetization averaged over their mean free path, of the order of 1000 Å.

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Fractionally Charged Excitations in Charge-Density-Wave Systems with Commensurability 3

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A theoretical study of topological excitations (kinks) in a one-dimensional one-third-filled Peierls system is presented. The charges associated with the kinks are found to be fractional $Q = \pm\frac{1}{3}e, \pm\frac{2}{3}e$. Calculations of the spatial widths and electronic structure of different types of kinks are carried out numerically. Possible applications to tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ) are mentioned.

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Recently, some novel physics have emerged from studying the nonlinear topological excitations in a simple one-dimensional coupled electron-phonon system.[1] It is known that in this model for the half-filled-band case (one electron per site), the lattice will undergo a commensurate Peierls distortion, i.e., dimerize, thereby opening a gap in the electronic spectrum at the Fermi surface, $k_\mathrm{F} = \pm\pi/2a$, where a is the mean lattice spacing. The dimerization is due to the energy lowering of the occupied electronic states being initially larger than the lattice strain energy, with the total energy reaching a minimum for two displacements $\pm u_0$. These two degenerate dimerization patterns are termed the A ($+u_0$) and B ($-u_0$) phases. B is simply a translation of A by one lattice spacing.

A topological soliton excitation or kink is formed by a domain wall separating regions of A and B phase material. Associated with each kink is a localized electronic state φ_0 located at the center of the gap. This state results from the removal of one-half of a state from the valence band and one-half from the conduction band in the vicinity of the kink. Since for topological reasons a soliton S and an antisoliton $\bar{S}$ must be formed at the same time, an integral number of states is removed from either band, as required for a Hermitian Hamiltonian. If the electrons were spinless and the gap-center state were empty, then because the deficit in the valence band is one-half of a state, the net charge of the kink would be $+\frac{1}{2}|e|$. This is related to the fermion-number-$\frac{1}{2}$ object discovered by Jackiw and Rebbi[2] in a relativistic field theory. The fractional charge is masked in polyacetylene by the spin degeneracy of the electron but leads to unusual charge-spin relations,[1] namely, charged kinks are spinless and neutral kinks are spin $\frac{1}{2}$ in contrast to the charge $\pm e$ and spin $\frac{1}{2}$ of electron and hole excitations in solids. In the one-third-filled-band case, however, the local charge deficit is no longer $\frac{1}{2}|e|$ per spin but rather $\frac{1}{3}|e|$ or $\frac{2}{3}|e|$ and one is left with a fractional charge for kinks $Q=\pm\frac{1}{3}|e|$, $\pm\frac{2}{3}|e|$. For commensurability n, the kinks have charge which is a multiple of $\pm e/n$.[3]

To perform quantative calculations, we consider the model Hamiltonian originally used for polyacetylene,[1]

$$H=-\sum_{ns}[t_0-\alpha(u_{n+1}-u_n)](c_{n+1,s}{}^\dagger c_{n,s}+\mathrm{H.c.})+\frac{K}{2}\sum_n(u_{n+1}-u_n)^2+\sum_n\tfrac{1}{2}M\dot{u}_n{}^2, \tag{1}$$

where u_n is the displacement of the nth unit from its equilibrium position, $c^\dagger$ is the electron creation operator, and M is the mass of one unit of the chain. For model calculations, it is convenient to take $t_0=2.5$ eV, $\alpha=4.81$ eV/Å, $k=17.4$ eV/Å^2, M = mass of CH. These values lead to a gap of 4.0 eV and a coherence length $\xi_2=2.75a$ in the dimerized case[4] (compared with a gap of 1.4 eV and $\xi_2\equiv 7a$ for actual polyacetylene).

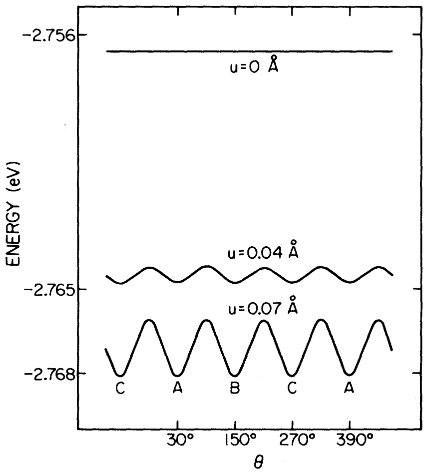

FIG. 1. Total energy per site plotted as a function of the phase angle θ for three different values of the amplitude of trimerization u.

Following Ref. 1, we plot in Fig. 1 the total energy per site as a function of the trimerization amplitude u and phase θ in a perfectly trimerized chain:

$$u_n=u\cos(\tfrac{2}{3}\pi n-\theta). \tag{2}$$

The three ground states A, B, and C are obtained by taking $u=u_0=0.07$ Å and $\theta-\frac{1}{6}\pi=0$, $\frac{2}{3}\pi$, and $\frac{4}{3}\pi$, respectively, as illustrated in Fig. 2, where dashed bonds mean shorter bond length than undashed bonds.

Notice the B-phase pattern is just the A-phase pattern displaced to the right by one unit, while the C-phase pattern is the A-phase pattern displaced to the right by two units or to the left by one unit. Also notice that in Fig. 1 there are two different condensation energy scales for going from A to B phase; the energy increase is about 0.002 eV per site for purely unwinding of the phase, while 0.011 eV for reducing the amplitude to zero as well. These two energy scales are important in determining the structure and energy of the kinks in the trimerized system.

For a perfectly trimerized chain, let the hopping integral associated with a short bond be $-(t_0+\delta)$ and for a long bond, $-(t_0-2\delta)$, where $\delta=\sqrt{3}\alpha u_0/2$, from Eqs. (1) and (2). The total charge on each site is given by the integral over occupied states of the imaginary part of the diagonal Green's function. One finds that the charge oscillates around the average value of $-\frac{2}{3}|e|$ with period of $3a$. For example, for the A phase the charges on sites 2 and 3 are equal and are smaller than that on site 1, by a fraction of order $3\delta/t_0$.

Knowing the degenerate ground states, we can study the structure of kinks connecting them. We distinguish two classes of kinks: type I, which

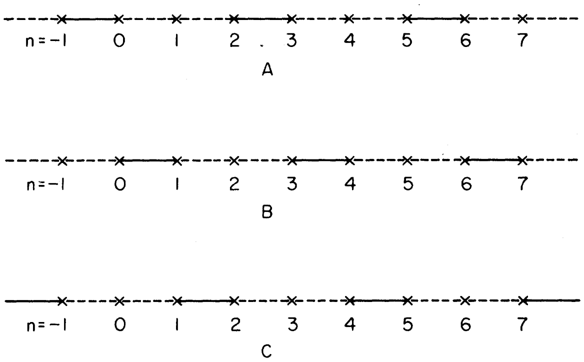

FIG. 2. The three degenerate ground states of a perfectly trimerized chain.

leads from A to B, B to C, or C to A as one moves from left to right; and type II, which leads from A to C, C to B, or B to A with increasing n. To determine the charge of a type-I kink, consider an infinite chain of pure A phase. Suppose that one deforms the lattice displacement pattern by maintaining the left-hand portion of the chain $n \to -\infty$ in the A phase ($\theta = \frac{1}{6}\pi$) while $\theta - \frac{1}{6}\pi$ increases to $\frac{2}{3}\pi$, $\frac{4}{3}\pi$, and $\frac{6}{3}\pi$ as one moves to the right of three widely spaced points n_1, n_2, and n_3, respectively. Since the phase shift is 2π for $n \gg n_3$, the system returns to the A phase for large n, with type-I kinks centered at n_1, n_2, and n_3. The shape of the kinks is irrelevant to this argument so long as the kinks have finite width. Because of the 2π phase shift in the bonding pattern, relative to the initial perfect A phase, during the deformation processes a total charge $-2|e|$ flows past a Gaussian surface located far from the kinks.[3] From charge conservation and the fact that symmetry ensures that the charge of the three type-I kinks are identical, it follows that the primitive charge of a type-I kink is $+\frac{2}{3}|e|$. This simple result agrees with that of Green's-function calculations as well as numerical calculations on chains of finite length. Thus, stable elementary excitations of fractional charge can occur as a consequence of ground-state degeneracy.

A similar argument shows that the primitive charge of a type-II kink is $-\frac{2}{3}|e|$ since the phase shift for $n \to +\infty$ is now -2π and a charge $-2|e|$ is accumulated in the region containing the three kinks. It is natural to identify type-II kinks as the antiparticles of type-I kinks, i.e., $K_{BA} = \overline{K}_{AB}$, etc. Let the ordered pairs AB, BC, and CA be denoted by 1, 2, and 3, respectively. Then, without disturbing the ground state at large distance, one can create $K_i\overline{K}_i$ pairs or the triplets $K_1K_2K_3$ and $\overline{K}_1\overline{K}_2\overline{K}_3$. One cannot help but notice the analog with the quark structure of mesons and hadrons. In this regard, related relativistic field-theory studies are being pursued by Goldstone and Wilczek.[5]

We have investigated the electronic structure of sharp kinks using Green's-function methods for an infinite chain and numerical calculations for finite chains with a number of sites of order 80. As illustrated in Fig. 3(a) for type-I kinks, there is a localized state φ_l in the upper half of the lower gap and symmetrically a state φ_u in the lower half of the upper gap. The state in the lower gap results from precisely one-third of a state being removed from the lowest energy band and two-thirds of a state being removed from the middle band. The symmetrical situation occurs for the state in the upper gap. If φ_l is unoccupied, the state deficit of the lower band leads to a kink charge of $\frac{2}{3}|e|$, as deduced above. In this charge state, all spins are paired so the kink spin is 0. If φ_l is singly occupied, the kink has charge $-\frac{1}{3}|e|$ and spin $\frac{1}{2}$. Finally, if φ_l is doubly occupied, the charge is $-\frac{4}{3}|e|$ and the spin is 0.

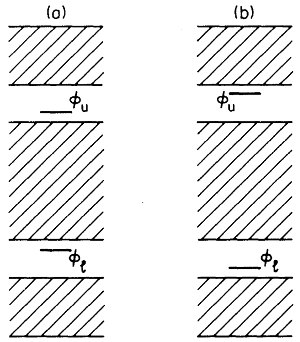

FIG. 3. Gap states associated with (a) a sharp type-I kink and (b) a sharp type-II kink.

For type-II kinks, the situation is similar. As shown in Fig. 3(b), the localized states φ_l and φ_u are now below and above the gap centers, respectively. Calculations show that precisely two-thirds of a state is removed from the lowest band for this type of kink so that if φ_l is unoccupied, the kink has charge $+\frac{4}{3}|e|$ and spin 0. For φ_l singly occupied the charge is $+\frac{1}{3}|e|$ and spin is $\frac{1}{2}$ while for φ_l doubly occupied the charge is $-\frac{2}{3}|e|$ and spin is 0, consistent with the above argument.

To obtain the minimum energy configuration of different types of kinks we use the equation-of-motion method used by us in Ref. 4. We start from some conveniently chosen configuration for a finite chain at rest, let it evolve according to the equation of motion in time, setting the velocities equal to zero after each time step. Each step lowers the total energy of the system until it approaches the minimum energy configuration. Numerical results for chains composed of approximately 80 groups suggest that the relaxed configuration depends sensitively on the occupancy n_l of the localized state φ_l of the kink, in contrast with the dimerization case. Kinks with $n_l = 0$ or 2 tend to relax with pure phase distortion and fixed amplitude, with the energy of the localized state moving up or down in the gap until it approaches the band edge. On the other hand, for $n_l = 1$, the energy of φ_l is located near the center of the gap

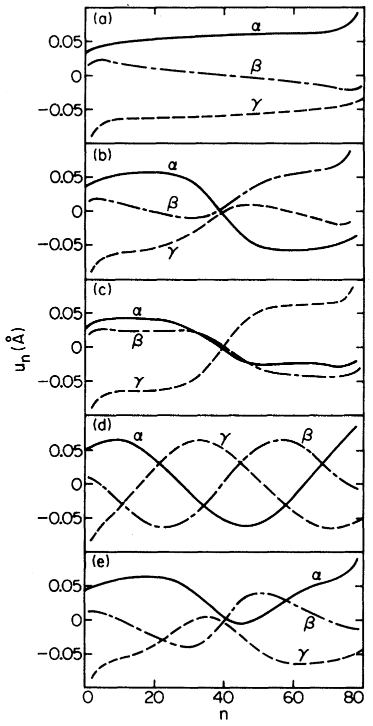

FIG. 4. Minimum energy displacement patterns u_n for finite chains with N_s sites and N_e electrons: N_s = 81, 79, 80, 81, and 81, and N_e = 54, 53, 53, 56, and 55 in (a)–(e), respectively.

for the minimum energy kink configuration and relaxation takes place primarily by modulation of the amplitude of the charge-density wave. For example, Fig. 4(a) shows the relaxed configuration of a chain of 81 sites and 54 electrons, i.e., exactly one-third-filled band. α is the line connecting the u_{3n} displacements for the first group in each unit cell with $n = 0, 1, \ldots, 26$. β and γ are the corresponding curves for the displacements of the second and third groups, u_{3n+1} and u_{3n+2}.

Figure 4(b) is the relaxed configuration of a chain of 79 sites and 53 electrons. These two numbers are chosen to produce a half-occupied type-I kink. Compared with Fig. 4(a), we see the end configurations are about the same; therefore, we conclude that the transition region intrinsic to the kink occurs within about 15 sites, $2\xi_3 \simeq 15a$, and $\xi_3 \simeq 7a$. It is interesting to note that this number agrees with that calculated from the usual relation $\xi = \hbar v_F/\pi\Delta$, where $2\Delta = 4\delta \simeq 1.2$ eV is either of the gaps in Fig. 3.

Figure 4(c) shows the corresponding result of a half-occupied type-II kink, while Fig. 4(d) is the result of adding two extra electrons to the chain in Fig. 4(a). Instead of creating two kinks as doping in polyacetylene does, the period of the displacement pattern decreases by the right amount so that there is a new state close to the valence band to accommodate the extra electrons. We might conjecture that if we have a chain long enough, we would see one of type I plus another of type II, both fully occupied. The fact that we do not see this in Fig. 4(d) is because the coherence length in this case is comparable or longer than the chain itself. Finally, Fig. 4(e) is the relaxed configuration for a half-occupied polaron. Again, the localized state is located at the center of the gap.

We conclude that stable excitations of fractional charge should exist in quasi-one-dimensional systems, with commensurability 3 being particularly favorable. Tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ) under 19 kbar pressure[6] is such a case since it is observed to have a commensurable charge-density wave of period $3a$.

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