Intertwined Solitons and Impurities in a Quasi-One-Dimensional Charge-Density-Wave System: In/Si(111)

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(Received 3 August 2018; published 10 January 2019)

Recent studies on a quasi-one-dimensional (quasi-1D) charge-density-wave (CDW) system, In atomic wires on Si(111), have brought intriguing issues on topological solitons in quasi-1D systems: the existence of few-atom-sized solitons, chirality of solitons, and the realization of logic exploiting the chirality switching. Using scanning tunneling microscopy and first-principles calculations, we show that the previously reported "short" phase-flip defects are In adatoms and thus have nonsolitonic nature, resolving the controversy over the existence of highly localized solitons. The observed "long" phase-flip and phase-slip defects are genuine solitons with and without chirality, respectively. While achiral solitons (phase-slip defects) can exist on the pristine CDW (8×2) surface, chiral solitons (phase-flip defects) cannot due to their breakage of 8×2 ordering. The chiral solitons can exist only when they are trapped by In adatoms and constitute a part of a *closed-loop* domain wall. The intertwinement of chiral solitons and In adatoms implies the limitations of the previously proposed logic utilizing soliton chirality, but it provides an opportunity to realize this by controlling the In-adatom defect.

DOI: 10.1103/PhysRevLett.122.016102

The concept of a soliton is relevant in many branches of physics, including hydrodynamics, fiber-optic communications, atomic physics, condensed-matter physics, and quantum-field theories [1]. In condensed matter, a soliton is an excitation that appears as a topological "defect" or "misfit" bridging two energetically degenerate ground-state configurations. Topological solitons have been described in various systems, ranging from ferroelectric and magnetic materials to electronic materials. Examples include dislocations in crystals, ferroelectric or magnetic domain walls, magnetic vortices, and skyrmions. In electronic materials, particularly in one dimension (1D) or in quasi-one dimension (quasi-1D), the solitons are related to Peierls-dimerized or charge-density-wave (CDW) ground states [2].

Topological solitons in 1D electronic materials were experimentally found in *trans*-polyacetylene [3,4], where they can be created by doping. They were shown to be mobile and responsible for the high electrical conductivity of organic materials [5]. Visualizations of the solitons using a scanning tunneling microscope (STM) were reported for a few quasi-1D CDW systems, including $NbSe_3$ [6] and self-assembled metallic wires on silicon surfaces [7–9]. Particular attention has been given to In atomic wires on Si(111) [10]. The presence of the solitons in this system and their dynamics have been reported previously [7,8], but there has recently been renewed interest [11,12]. Zhang *et al.* [11] observed a local phase-flip boundary with a length of a few atoms separating two energetically degenerate 1D CDW phases. They attributed it to a topological soliton. In a subsequent study, Kim *et al.* [12] classified the defects observed on an 8×2 CDW surface into two defects: *long* and *short* defects with phase slip or phase flip across the defect along the wire. Such defects were referred to as phase-slip defects (PSDs) and phase-flip defects (PFDs), respectively (see Fig. 1). The extended features (*long* PSD and PFD) were considered intrinsic topological solitons, whereas local features (*short* PSD and PFD) were not. The long PFD was shown to have chirality (right or left handedness) [13], and algebraic operations of topological information using chiral switching were proposed [14].

The solitonic nature of the localized, slow-moving short PSDs or PFDs was rejected based on a comparison with the extended and highly mobile solitons of polyacetylene [12,15]. However, it was controversial whether there must be an intrinsic requirement for soliton length and mobility in two systems of differing structures and materials [15,16]. In addition, the fact that most of the long PSDs/PFDs were observed in the presence of short PFDs [12–14] suggested a close association between them. This poses a question of whether the long PSDs or PFDs—the *presumed* intrinsic solitons—can exist by themselves even in the absence of other defects. All these issues should be resolved to understand the unique and unusual characteristics of the solitons in this quasi-1D In wire system.

In the present study, we resolve the controversy over the origin of the short PSD/PFD and uncover their relation to

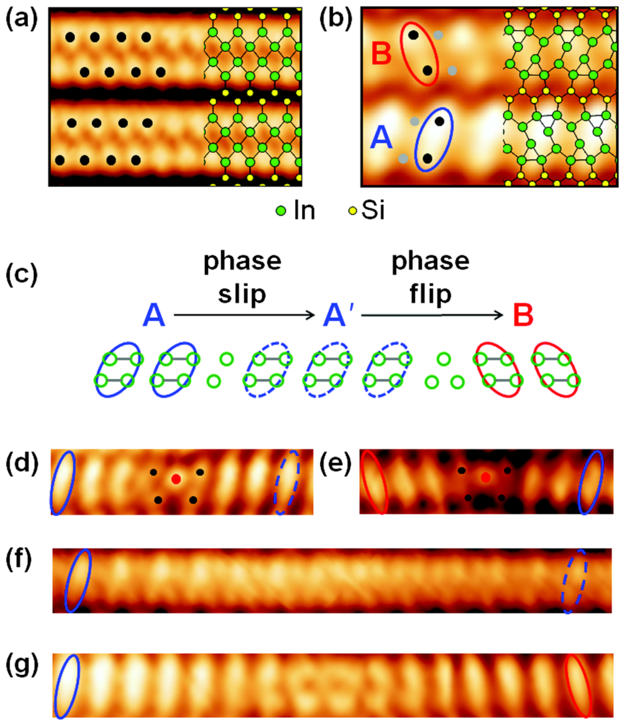

FIG. 1. STM images of an In/Si(111) surface with (a) 4×1 and (b) 8×2 structures taken at 300 K and 80 K, respectively. Both images were obtained with a sample bias voltage (V_s) of −1.5 V. (c) Schematic illustration of the phase slip and phase flip in a 4×2 wire. The ovals represent CDW features in two possible tilt orientations. (d)–(g) STM images of defects observed on the 8×2 surface, which are classified into two categories: *short* defects [(d) phase-slip defect (PSD) and (e) phase-flip defect (PFD)] vs *long* defects [(f) PSD and (g) PFD].

the chiral solitons in an In/Si(111)-(8×2) CDW system. The short PSDs/PFDs are caused by extra In atoms on the surface (i.e., In adatoms), and their solitonic nature is unambiguously discarded. The chiral solitons (long PFDs) do not exist on pristine surfaces and can only exist when trapped by adjacent In adatoms. They are topologically restricted to exist as part of a two-dimensional (2D) closed-loop domain wall that separates two 2D domains. This intertwinement of chiral solitons with In adatoms is rooted in the 2D nature of the CDW in the In/Si(111)-(8×2) system. In contrast, an untrapped achiral soliton (long PSD) exists and it can be much longer than that expected from the soliton theory in a 1D CDW system.

Figures 1(a) and 1(b) show STM images of an array of In atomic wires on a Si(111) surface with 4×1 (at 300 K) and 8×2 structures (at 80 K), respectively. In the 4×1 structure, each individual wire consists of parallel pairs of zigzag In chains. Below 125–130 K, both dimerization of the In atoms and shear distortion between the two chains occur to form tilted In hexagons in the 4×2-wire structure [17]. Two tilt orientations of the hexagons [*A* and *B* in Fig. 1(b)] alternate across the wires, and an 8×2 structure is formed. This period-doubling phase transition from 4×1 to 8×2 with band-gap opening was considered to be a CDW transition driven by Fermi-surface nesting in electronic structures of the metallic 4×1 surface [10]. Previous publications referred to the 8×2 structure as a CDW phase as a whole, but here, we call it a 2D CDW to avoid confusion with the 1D CDW in a 4×2 wire. Because of the unique double-chain structure with two possible tilt orientations, the phase in the 4×2-CDW wire can slip or flip across the defects [Fig. 1(c)]. The images of defects observed as PSDs or PFDs in previous publications [11,12] are shown in Figs. 1(d)–1(g).

Figure 2(a) shows a STM image, taken at 300 K, of an In/Si(111)-(4×1) surface prepared by a typical recipe to make a uniform, vacancy-free surface. Cooling this seemingly perfect surface down to 80 K resulted in an 8×2

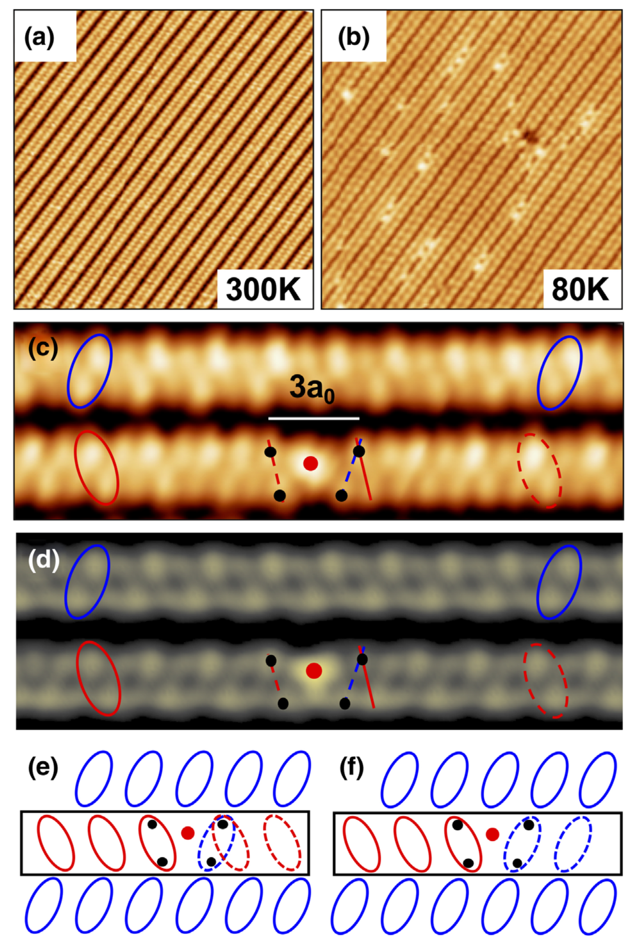

FIG. 2. STM images of an In/Si(111) surface prepared by a typical recipe [images taken at 300 K ($V_s = -1.5$ V) (a) and at 80 K ($V_s = 1.7$ V) (b)]. While the 4×1 surface at 300 K appears perfectly uniform and defect-free, the 8×2 surface at 80 K reveals many bright protrusions (In adatoms). Comparison of the experimental STM image ($V_s = -2.3$ V) of a short PSD (c) and the theoretical simulation ($V_s = -2.2$ V) of an isolated In adatom (d) on an 8×2 surface. Schematic of interwire registries created by an 8×2-preserving short PSD [boxed in (e)] and an 8×2-breaking short PFD [boxed in (f)].

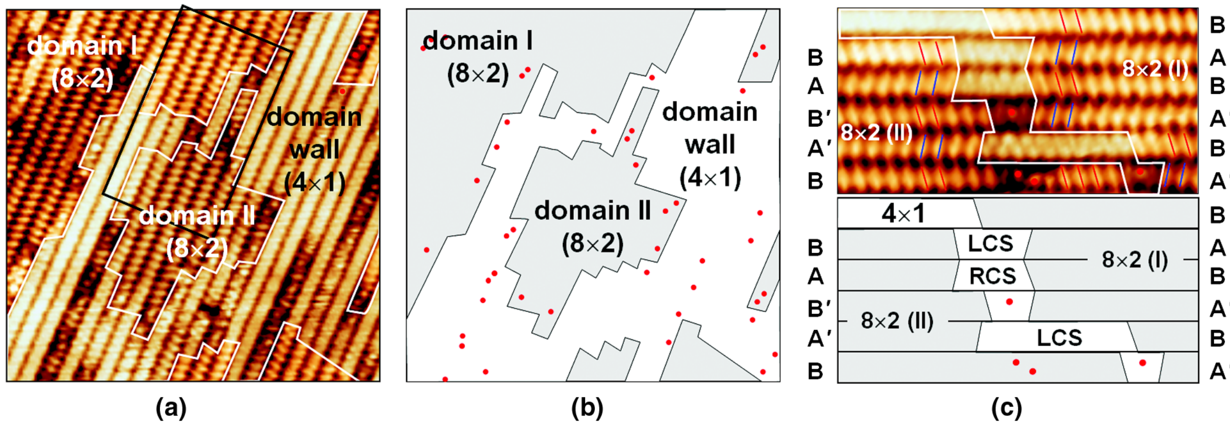

FIG. 3. (a) STM image of an In/Si(111)-(8 × 2) surface rich with In adatoms taken at 96 K ($V_s = -1.0$ V) and (b) its schematic illustration (In adatoms are designated by red dots). A 2D domain wall (enclosed by lines) consists of In adatoms (short PFDs), chiral solitons (long PFDs), and 4 × 1 segments. It forms a closed loop and separates topologically two 8 × 2-CDW domains (I and II). (c) Magnified STM image (top) and its schematic (bottom) of a boxed area, a part of the closed-loop domain wall in (a). The two 8 × 2 domains (I and II) are separated by phase-flipping segments; chiral solitons [right chiral soliton (RCS) and left chiral soliton (LCS), as designated according to the schemes from Ref. [13]], In adatoms (red dots), and 4 × 1 segments.

surface where a number of bright features appeared [Fig. 2(b)]. The bright features on the 8 × 2 surface are the short PFDs and PSDs [shown in Figs. 1(d) and 1(e)]. These defects were largely removable by annealing the surface at higher temperatures, which created more vacancies. In contrast, they could be produced by additional In deposition on the surface. (see Fig. S1 [18]). These observations indicate that the short PFDs and PSDs are In adatoms. Typically, such In adatoms form without being recognized when a vacancy-free 4 × 1 surface is prepared. At room temperature, they are invisible in STM images because they move fast [23]. Upon cooling, the In adatoms can be imaged by STM as their movements slow down. The fact that the short PFDs and PSDs are In adatoms is firmly confirmed by the good agreement between the experimental STM image of the short PSD [Fig. 2(c)] and the theoretical simulations of an In adatom on an 8 × 2 surface [Fig. 2(d)]. The adsorption structure of an In adatom is presented in Fig. S2 [18]. This identification ends the debate on the origin of the short PFD and PSD: they are not intrinsic solitons, but extrinsic defects (In adatoms).

Zhang *et al.* [11] reported the short PFD as a standard soliton that is localized at the scale of a few atoms in this system. As for the occasionally observed short PSDs, the phase slip was attributed to disturbances from fuzzy fluctuating segments near the localized solitons [16]. However, we highlight that most of the observed In adatoms appear as short PSDs rather than PFDs. Within a 4 × 2 wire, an isolated In adatom has a nearly mirror-symmetric structure, and the phase flip across the defect might be favored. However, the phase flip in a single 4 × 2 wire would break the ground-state 8 × 2 ordering (alternating CDW orientations in adjacent wires), which is hindered by the energy cost due to a sizable interwire coupling. Therefore, the original CDW orientation is preserved, but its phase slips by π (corresponding to a $1a_0$ shift in distance) across the defect along the 4 × 2 wire [Fig. 2(e)]. This makes an isolated In adatom appear as a short PSD.

As in the case of short PFDs, the strong interwire coupling prohibits the presence of isolated long PFDs. Short or long PFDs can be formed under the special conditions where the neighboring wires also have phase-flipping segments. Figure 3 shows such a situation. The STM image of an In/Si(111)-(8 × 2) surface in Fig. 3(a) shows that not only short PFDs and long PFDs but also 4 × 1 segments can be phase-flipping segments. Each phase-flipping segment separates the two 1D CDWs with different tilt orientations in a 4 × 2 wire [see the magnified image in Fig. 3(c)]. Maintaining the 8 × 2 ordering within each 2D CDW domain, the phase-flipping segments are laterally connected to each other and form a *closed* loop. This *closed* loop topologically disconnects an inner 8 × 2 domain from the outer 8 × 2 domain. Thus, it forms a *2D domain wall* between the two different 2D CDW domains [24].

We confirm that the long PSDs and PFDs are topological solitons, as interpreted in earlier studies [7,8,12]. Previous analysis of the soliton chirality revealed that the long PFD has chirality (chiral soliton), but the long PSD does not (achiral soliton) [13]. Note that chiral solitons have never been observed in isolation. All observed chiral solitons were trapped by In adatoms or neighbored by other phase-flipping features in 2D closed-loop domain walls [25]. This implies that chiral solitons are intrinsically nonexistent on an 8 × 2 surface. Their formation can only be induced by the presence of In adatoms.

Strong coupling of the chiral solitons with the In adatoms is clearly demonstrated in Fig. 4. Figure 4(a) shows two STM images over the same area taken at different times

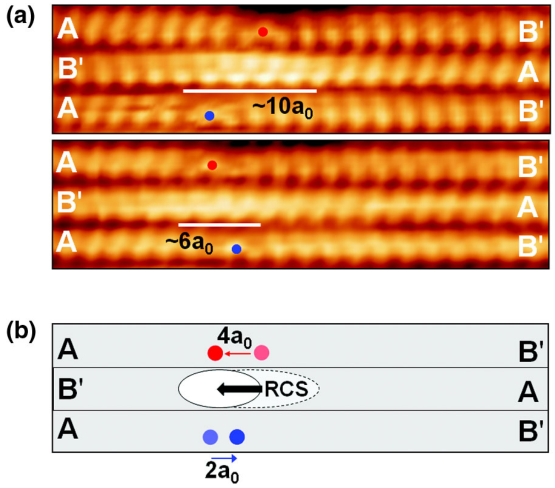

FIG. 4. (a) Sequential STM images ($V_s = -0.5$ V) and (b) schematic showing temporal motion of a chiral soliton [underlined in the middle wire in (a)] coupled with In adatoms (in the upper and lower wires).

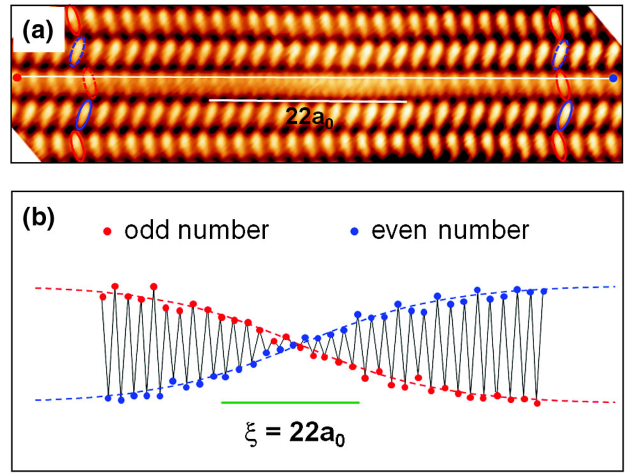

FIG. 5. (a) STM image ($V_s = -0.5$ V) of an achiral soliton (long PSD) on an 8×2 surface at 80 K (a full-size image is presented in Fig. S4 [18]). Note that the achiral soliton is well isolated without In adatoms in its vicinity. (b) STM height profile (dots) of the achiral soliton along the white line in (a) together with its best fit by a trial fitting function, $Z(x) = Z_0 \tanh[(x - x_0)/\xi]$ (red and blue lines). Only locally maximal and minimal heights at positions of na_0 (n: integer) were chosen to remove noises and slowly varying background shapes. The fitting function mimics the soliton as an envelope for the lattice modulation, although the detailed correct functional form is unknown for the soliton in this system. A characteristic length of $\xi = 22 \pm 2a_0$ was obtained by fitting.

(more images showing the temporal changes are presented in Fig. S3 [18]). The three 4×2 wires in the images are part of a 2D domain-wall region separating two 8×2 CDW domains on both the left and right sides. A chiral soliton in the middle wire is trapped by In adatoms in the upper and lower adjacent wires. As depicted in Fig. 4(b), the chiral soliton moves by following the movement of the In adatoms. The apparent size of the chiral soliton also changes as the relative positions of the two In adatoms are altered. All these observations indicate that the formation, dynamics, and size of the chiral soliton are strongly governed by the In adatoms. In other word, the chiral solitons are intertwined with the In adatoms.

Differing from the chiral soliton, an isolated achiral soliton can exist on the 8×2 surface without breaking the 8×2 ordering, as shown in Fig. 5(a). Notably, the isolated achiral soliton is clearly imaged without being trapped by any defects in the vicinity. This contrasts with the previous notion that untrapped solitons move fast and are thus hard to image at ~80 K [12,13]. In addition, the characteristic length (ξ) of the achiral soliton in Fig. 5(a) contrasts with previous works [12,13]. Previously, $\xi \sim 9a_0$ was claimed to be the intrinsic soliton size by showing that it agreed with the theoretical estimation based on an analogy with the case of polyacetylene [12]. However, the characteristic size ($\xi \sim 22a_0$) determined from the height profile in Fig. 5(b) is much larger than that determined in the previous work.

The discrepancy in soliton size between the observations in this work and the estimates based on simple 1D soliton theory [3] originates from the difference in dimensionality. The quasi-1D In wire array is basically a 2D system, although previous theoretical studies ignored this 2D nature for simplicity [13,26]. The formation and characteristics of the solitons in this 2D CDW system are expected to differ from those in purely 1D systems. More comprehensive theoretical treatments should consider the interwire interaction, which plays a crucial role in the 2D nature of this system.

Lately, algebraic operations utilizing the chiral switching of the solitons were proposed for the logic of future devices [14]. The proposal was based on the observation of the change of the handedness of a chiral soliton (from left to right and vice versa). However, we point out that the observed chiral solitons were trapped between two In adatoms in the same wire. The chiral switching was attributed to the merging of the chiral soliton with an achiral soliton whose passage was implied by an odd-a_0 hopping of the trapping defects [13,14]. Based on the chiral soliton's dependence on the In adatoms, we propose a more natural and direct interpretation: it is the simple odd-a_0 hopping of the trapping In adatom that triggers the chirality switching of the soliton.

We highlight that chiral solitons can exist only as part of the 2D closed-loop domain wall, being trapped by In-adatom defects. This indicates that the In adatom is the key element in realizing the proposed logic [14]. Thus, finding ways to manipulate In adatoms to form the chiral solitons and control their switchings is a prerequisite to realizing any logic that exploits the soliton chirality.

This work was supported by the Basic Science Research Program (No. 2010-0012231) and Mid-career Researcher Program (No. 2017R1A2B2003928, No. 2015R1A2A2A01005564) through the National Research Foundation of Korea (NRF) funded by the Korean government. The calculations were done using supercomputing resources supported by the Supercomputing Center/Korea Institute of Science and Technology Information (No. KSC-2016-C2-0060).

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