Spectral function of few electrons in quantum wires and carbon nanotubes as a signature of Wigner localization

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We demonstrate that the profile of the space-resolved spectral function at finite temperature provides a signature of Wigner localization for electrons in quantum wires and semiconducting carbon nanotubes. Our numerical evidence is based on the exact diagonalization of the microscopic Hamiltonian of few particles interacting in gate-defined quantum dots. The minimal temperature required to suppress residual exchange effects in the spectral function image of (nanotubes) quantum wires lies in the (sub-) Kelvin range.

After half a century of research, electrons in one dimension still attract attention as a paradigm of interacting behavior that deviates from Fermi liquid theory, exhibiting e.g. spin-charge separation or solid-like order\textsuperscript{1-3}. This impulse comes from recent experiments in systems with high aspect ratio—cleaved-edge overgrowth structures\textsuperscript{4-11} multiple-gate quantum wires\textsuperscript{12} carbon nanotubes\textsuperscript{13-14}—which are all effectively one-dimensional as their transverse and longitudinal degrees of freedom are decoupled. The refinement of such devices allows to easily reach the dilute regime of electron density yet minimizing the impact of disorder. At sufficiently low density, the Coulomb energy gain overcomes the kinetic energy cost of localization, hence electrons are expected to freeze their motion in space forming a regular array—a Wigner correlated solid\textsuperscript{15,16}.

So far, the evidence of Wigner localization has ultimately relied on the measure of the energy gap between ground and low-lying excited states, which vanishes in the dilute limit\textsuperscript{5,12,14}. This excitation energy decreases gradually from the liquid- to the solid-like regime, as an effect of both quantum fluctuations and samples’ finite size—systems often act as quantum dots (QDs) in the Coulomb blockade regime\textsuperscript{2,5,12-14}. Therefore, an alternative signature of the electron solid, directly related to the wave function, would be desirable. A possible observable is the momentum-resolved spectral function (SF)—the quasiparticle wave function square modulus in reciprocal space\textsuperscript{6,12}. Unpromisingly, it was predicted that the SF was qualitatively similar in both Wigner and non-interacting limits\textsuperscript{18} and that any distinctive structure of the SF was washed out by temperature\textsuperscript{19}.

In this Communication we demonstrate that the space-resolved spectral function of few electrons provides a clear signature of Wigner localization at temperatures above $T_{\text{ex}}$, that is the characteristic scale of exchange interactions. This fundamental observable may be accessed through scanning tunneling spectroscopy (STS)\textsuperscript{20,21}. Our exact diagonalization\textsuperscript{22-25} (ED) results show that the SF resembles the charge density for $T \gtrsim T_{\text{ex}}$, displaying $N$ peaks as the $N$th electron tunnels into a Coulomb blockaded QD already containing $N-1$ electrons. The peak-to-valley ratio of such image allows to assess directly the onset of Wigner localization. In sharp contrast, for $T \ll T_{\text{ex}}$ the SF is system-dependent and unrelated to $N$. Overall, the joint measurements of $N$ and of the SF are able to unveil the Wigner solid.

FIG. 1. (Color online) (a-c) SF signal $G$ vs $x$ at various $T$ for $N = 2 \rightarrow N = 3$. (a) Quantum wire (QW). The length unit is $\ell_{\text{QD}} = 23.9$ nm. (b) Carbon nanotube no. 1 (CNT1). $\ell_{\text{QD}} = 26.8$ nm. (c) Carbon nanotube no. 2 (CNT2). $\ell_{\text{QD}} = 17.9$ nm. (d) SF signal $G$ vs $x$ at $T = 10$ K for a CNT as $\hbar \omega_0$ is varied. The five curves, from red [light gray] to black, correspond to $\hbar \omega_0 = 5, 20, 60, 100, \infty$ meV, respectively. The length unit is $\ell_{\text{QD}}$ and the remaining parameters are those of CNT2. (e) $T_{\text{ex}}$ vs $\hbar \omega_0$ for QW, CNT1, and CNT2. The filled circles are the values pertinent to panels (a-c).

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The scenario reported in this Communication agrees with the theory of the ‘spin-incoherent’ Luttinger liquid. On the other hand, crucial approximations of this theory poorly reproduce key experimental features of systems with a moderate number of electrons, and most noticeably: (i) finite-size effects are prominent; (ii) the occurrence of the gap and band curvature through the effective mass and valley degeneracy, band curvature through the effective mass.

We assume the QD confinement potential along $x$ to be harmonic, $V_{QD}(x) = m_0^2x^2/2\hbar^2$ since this is the generic low-energy form for gated QDs embedded in quantum wires (QWs) and semiconductor carbon nanotubes (CNTs), as in Refs. 5, 8–12. We consider $N$ electrons in a QW interacting through a screened Coulomb interaction $V(x, x') = e^2/4\pi\epsilon_0\epsilon_r[(x - x')^2 + \lambda^2]^{-1/2}$, with $\lambda$ being a short-range cutoff and $\epsilon_r$ the dielectric constant. The CNT Hamiltonian is more complex, due to the presence of valleys $K$ and $K'$, spin-orbit coupling, inter- and intra-valley interactions. The interaction potential interpolates between Coulomb and Hubbard-like behavior: see Ref. 23 for details. In all cases we diagonalize the Hamiltonian in the space spanned by the Slater determinants built by filling with $N$ electrons the lowest 60 spin-orbitals in all possible ways.

The outcome of the ED consists in the first excited $N$-body states, $|N, i\rangle$, and their energies $E_N^i$. The SF for a given initial state at $T = 0$, $A_{N-1, i}(x, \omega)$, is

$$A_{N-1, i}(x, \omega) = \sum_j \langle N, j | \hat{\Psi}^\dagger(x) | N-1, i \rangle^2 \delta(\omega - E_j^N + E_i^{N-1})$$

with $\hat{\Psi}^\dagger(x)$ being the operator creating an electron at $x$ and $\hbar\omega$ its resonant tunneling energy, whereas the ground-state charge density is $g(x) = N^{-1}\langle N, 0 | \hat{\Psi}^\dagger(x) \hat{\Psi}(x) | N, 0 \rangle$. Ideally, the STS differential conductance at vanishing temperature and bias, $dI/dV$, is proportional to $A_{N-1, 0}(x, \varepsilon_F/b)$, with $\varepsilon_F$ being the Fermi energy of the STS tip. If the thermal broadening $k_B T$ is larger than the typical energy spacings $\hbar\omega$, $dI/dV$ is proportional to

$$G = \int d\omega \left[ -\frac{\partial f(\hbar\omega)}{\partial \omega} \right] A_T(x, \omega),$$

where $f(\cdot)$ is the Fermi distribution function and $A_T(x, \omega)$ is the finite-temperature SF,

$$A_T(x, \omega) = \frac{1}{Z} \sum_i e^{-\beta E_i^{N-1}} A_{N-1, i}(x, \omega),$$

with $\beta = 1/(k_B T)$ and $Z = \sum_i \exp(-\beta E_i^{N-1})$. In the following we tune $\varepsilon_F$ appearing in $f(\cdot)$ to match the $N-1 \rightarrow N$ transition between ground states.

To immediately grasp the key results of this Communication, consider in Fig. 1(c) the SF signal $G$ induced by the tunneling of the third electron into a realistic CNT (here labeled CNT2) at various temperatures. At low temperature, $T \ll T_{ex} = 0.017$ K, the signal shows two peaks that resemble the charge distribution of three Wigner-localized electrons [cf. $g(x)$ in Fig. 1(b)]. The peak-to-valley ratio at $T \gg T_{ex}$ directly measures the degree of spatial localization. This is apparent in Fig. 1(d), as the impact of few-body correlations is reduced by increasing the QD confinement energy $\hbar\omega_0$ and hence the ratio of kinetic to Coulomb energy. Whereas for strong correlations (red [light gray] curve) the three peaks are well resolved since electrons separately localize in space, as the interaction is turned off $G$ becomes featureless (black curve for $\hbar\omega_0 = \infty$), clearly discriminating between Wigner and weakly-interacting regimes.

Also the magnitude of $T_{ex}$ points to electron correlation, the lower the temperature $T_{ex}$, the stronger the localization. $T_{ex}$ varies significantly for typical QWs and CNTs as a function of device parameters, like $\hbar\omega_0$. In Fig. 1(d) the increase of $\hbar\omega_0$ quenches correlations and amplifies the effects of Fermi statistics, raising $T_{ex}$. For realistic parameters [cf. circles in Fig. 1(d)], we find that $T_{ex}$ may be as low as 10 mK in some semiconducting CNTs. On the other hand, $T_{ex}$ is one-two orders of magnitude higher in quantum wires (QWs), $T_{ex} \sim 1$ K, as an effect of the different impact of screening.

Figure 2 shows $g(x)$ and the zero-temperature SF up to five electrons for a typical wire QD. In the ED we chose $\lambda = 5$ nm, bulk GaAs parameters, and $\hbar\omega_0 = 2$ meV as single-particle energy spacing, providing a characteristic QD length $\ell_{QD} = (\hbar/m_0\omega_0)^{1/2} = 29.3$ nm. As it is seen from the spread of $g(x)$ along the axis (black curves in Fig. 2), this corresponds to a typical size of $\approx 200$ nm for $N = 5$, which is comparable to the size $L_{loc}$ = 230 nm of the QD in the dilute limit of Ref. 3 (Table I). The profiles of $g(x)$ point to the partial localization of the $N$ electrons as $N$ peaks emerge from a featureless liquid droplet. On
The SFs appearing in Fig. 2 are similar to those obtained in the absence of interaction (dashed lines). In the non-interacting limit the SF is the square modulus of the orbital occupied by the $N$th electron that enters the QD.\textsuperscript{17} Such orbital has zero, one, one, and two nodes for $N = 2, 3, 4, 5$, respectively, since electrons fill in each orbital level twice due to Kramers degeneracy. As the symmetries of the quantum states do not change in the considered range of interaction, no qualitative differences are seen for the interacting SFs.\textsuperscript{26}

In the following we discuss two examplar CNT cases. Figure 3 is the analogue of Fig. 2 for the CNT device investigated in Ref. 3 (here labeled as no. 1). The ED parameters ($\hbar \omega_0 = 8$ meV, $\epsilon = 3.5$, radius $R = 3.6$ nm) were chosen in order to reproduce the measured chemical potentials, as detailed in Ref. 25. Apart from length renormalization ($\ell_{\text{QD}} = 26.8$ nm), charge densities (black curves in Fig. 3) are similar to those of the QW (Fig. 2). On the contrary, the zero-temperature SFs for the CNT (solid red [gray] curves in Fig. 3) are drastically different from those of Fig. 2, being all nodeless except for the $N = 4$ $\rightarrow$ $N = 5$ transition. This trend is qualitatively similar to the non-interacting filling sequence (dashed lines in Fig. 3), as each CNT level is four-fold degenerate in the absence of spin-orbit coupling, due to both spin and valley degeneracies. The spin-orbit interaction splits the multiplet into two doublets (here separated by $\Delta E_{\text{SO}} = 0.367$ meV) but leaves the spin-orbitals unchanged. Since $\Delta E_{\text{SO}}$ remains the energy scale of the low-lying excitations even in the presence of interactions in the sample no. 1 (Refs. 3 and 25), the SFs of Fig. 3 are unaffected by temperature for $T \ll \Delta E_{\text{SO}}/k_B \sim 4$ K [cf. Fig. 1(b)].

The SFs of Figs. 3(a), (b), and (c) display $N$ peaks as the $g(x)$’s, and placed approximately in the same locations. This genuine effect of interaction, reminiscent of the partial Wigner localization occurring in the QD, takes place also in the QW [Fig. 4(a)] and it has been observed for the tunneling of the second electron into elongated self-assembled InAs QDs.\textsuperscript{27}

The second CNT QD that we study experiences stronger interactions than the first one, as an effect of the smaller energy spacing ($\hbar \omega_0 = 5$ meV), dielectric screening ($\epsilon = 2.5$), radius ($R = 1$ nm). In terms of parameters, the CNT QD no. 2 lies in the middle between the devices investigated in Refs. 8 and 9 (see Ref. 25 for their placement in a phase diagram). As it is shown in Fig. 4 (black curves), the peak-to-valley ratios of charge densities are about twice as large as those in Figs. 2 and 3, hence electrons are strongly localized.

In this Wigner regime the low-lying excited states are easily thermally populated and are highly degenerate. The reason is that exchange interactions between localized electrons are suppressed ($T_{\text{ex}}$ is only 17 mK), therefore each electron may flip its spin independently from the others at low energy cost.\textsuperscript{28} This is true also for the isospin—the orbital angular momentum along $x$ labelling valleys K and K’. The overall result is that the many excited states which differ only in the (iso)spin value are almost degenerate. This situation is illustrated in Fig. 4(a), where we depict the excited-state ladders for $N = 2$ and $N = 3$. In the ultimate Wigner limit one expects the ground state to be exactly $4^N$ degenerate, as each electron may flip its spin and isospin in four different ways. The effect of spin-orbit interaction (here $\Delta E_{\text{SO}} = 1.32$ meV) is to split the ground-state multiplet into equally

FIG. 3. (Color online) CNT $g(x)$ of $N$ electrons (black curve) and SF at $T = 0$ for the $N - 1 \rightarrow N$ ground-state transition (solid red [gray] curve) vs $x$ for $N = 2$ (a), 3 (b), 4 (c), 5 (d). The dashed curves are the non-interacting SFs, rescaled by a factor 1/6. $A_{N-1,0}(x, \omega)$ is integrated over a tiny energy range around $E_N^0 - E_{N-1,0}^0$. The length unit is $\ell_{\text{QD}} = 26.8$ nm.

FIG. 4. (Color online) SF signal $G$ vs $x$ at $T = 0.1$ K (dashed curve) and $T = 0.5$ K (solid red [gray] and green [light gray] curve) for the CNT QD no. 2. The green [light gray] curve is the non-interacting case. The following $N - 1 \rightarrow N$ transitions are considered: (a) $N = 2$, (b) $N = 3$, (c) $N = 4$, (d) $N = 5$. $g(x)$ for the $N$-body ground state (black curve) is plotted for comparison. The length unit is $\ell_{\text{QD}} = 17.9$ nm.
with regards to both the number of peaks and their relative intensities. The variations are dictated by the symmetries of initial and final states involved in the tunneling transition. For example, the fundamental transition $A \rightarrow a$ between two- and three-electron ground states displays two peaks located at opposite positions (red [light gray] curve), whereas all other depicted SFs have three peaks each. This shows that the SF at $T = 0$ may greatly deviate from the charge-density profile, corroborating the findings of Figs. 2(b) and 3(b). On the other hand, the locations of the maxima of curves in Fig. 3(c) coincide.

By statistically averaging the SFs of low-lying transitions, as those shown in Fig. 5(c), one obtains the finite-temperature signal $G$ [cf. Eq. (1)]. Figure 4 shows the pattern of $G$ at $T = 0.1$ K (dashed curves) and $T = 0.5$ K (solid red [gray] curves) for transitions $N - 1 \rightarrow N$ up to $N = 5$. The small dependence of $G$ on the temperature exhibited in Figs. 4(c-d) [see differences between dashed and solid red (gray) curves] is a finite-size effect due to the form of the potential $V_{\text{QD}}(x)$, as the electron density slightly increases with $N$ (Ref. 23). Remarkably, $G$ has a regular behavior as a function of $N$ already at $T = 0.5$ K, systematically displaying $N$ peaks of comparable heights whose positions are close to (but non coinciding with) the locations of the maxima of $g(x)$ (black curves).

The pattern of $G$ shown in Fig. 4(b)—exhibiting high peak-to-valley ratio—is peculiar of the Wigner regime and should be contrasted with the featureless, non-interacting profile (green [light gray] curves). Indeed, the low-lying excited states, as those of Fig. 4(a), have all roughly the same orbital wave function modulus, similar to the vibrational wave function of nuclei of polyatomic molecules. The differences among orbital states, as well as those among SFs [cf. Fig. 5(c)], originate from the different nodal surfaces. Since the weight is mainly localized around the equilibrium positions of electrons, nodeless interstitial regions may hardly be distinguished from nodal regions. Therefore, the statistical average of excited states shows a regular trend, linked to the positions of localized electrons.

At sufficiently high temperatures, the SF signals $G$ of all investigated devices behave similarly. This is shown in Fig. 4 for the tunneling transition $N = 2 \rightarrow N = 3$. Above their respective temperatures $T_{\text{ex}}$, highlighted as circles in Fig. 4(d), all $G$ profiles exhibit three peaks of similar height: see the curves for $T = 4, 10, 0.1$ K in Figs. 4(a), (b), (c), respectively. The different peak-to-valley ratios of these curves measure the degree of Wigner localization, the lower $T_{\text{ex}}$, the higher the ratio.

At $T \gg T_{\text{ex}}$ the central peak of the QW signal is depleted again [red (light gray) curve for $T = 30$ K in Fig. 4(a)], whereas CNT profiles [red (light gray) curves in Figs. 4(b) and (c)] remain stable well above 50 K. This change is due to the excitation of the energy scale associated to charge. In fact, the charging energy of the QW, estimated as the energy difference between the resonance energies of the first two electrons, is 51.1 meV, that is comparable with 30 K, whereas CNT charging energies are much larger ($\sim$ 20 meV).

In conclusion, we have shown that the spatial depen-
dence of the spectral function provides a clear fingerprint for Wigner localization, as the temperature overcomes the energy scale of exchange interactions. This temperature is low enough in both semiconducting carbon nanotubes and quantum wires to make scanning tunneling spectroscopy feasible. This effect has not been seen in past experiments, likely due to metallic screening, as well as to the presence of disorder and scattering from boundaries. We hope our prediction may stimulate further work along this path.

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