

Band-Gap-Dependent Electronic Compressibility of Carbon Nanotubes in the Wigner Crystal Regime

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Electronic compressibility, the second derivative of ground-state energy with respect to total electron number, is a measurable quantity that reveals the interaction strength of a system and can be used to characterize the orderly crystalline lattice of electrons known as the Wigner crystal. Here, we measure the electronic compressibility of individual suspended ultraclean carbon nanotubes in the low-density Wigner crystal regime. Using low-temperature quantum transport measurements, we determine the compressibility as a function of carrier number in nanotubes with varying band gaps. We observe two qualitatively different trends in compressibility versus carrier number, both of which can be explained using a theoretical model of a Wigner crystal that accounts for both the band gap and the confining potential experienced by charge carriers. We extract the interaction strength as a function of carrier number for individual nanotubes and show that the compressibility can be used to distinguish between strongly and weakly interacting regimes.

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The Wigner crystal, an ordered crystalline lattice of electrons with extremely strong interactions, is one of the most fascinating regimes of solid-state physics [1]. One of the observables of this regime is the electronic compressibility κ , which is a reflection of the many-body interactions of the target system and can be obtained from $\kappa = (d^2E/dN^2)^{-1} = (d\mu/dN)^{-1}$, where E is the ground-state energy, N is the total electron number, and $\mu = dE/dN$ is the chemical potential of the system. The inverse compressibility κ^{-1} , in particular, corresponds to the amount by which the chemical potential must be raised in order to add an electron; small κ^{-1} indicates that the system easily accommodates additional electrons. Various studies have been conducted on compressibility (and quantum capacitance, which is directly proportional to compressibility) of quantum structures such as quantum wires, two-dimensional electron systems, mono- and bilayer graphene, etc. to explore the interactions in these systems [2–6]. When the density of states is constant and electron-electron (e - e) interactions are relatively weak, the compressibility of an electronic system is independent of the charge carrier density. These assumptions are violated, however, in the low-density regime, and the compressibility varies strongly with density. In particular, in the Wigner crystal regime, strong correlations between electrons are predicted to lead to a sharp decrease in κ^{-1} with decreasing density [2,7,8]. Previous studies have indeed observed a reduction in κ^{-1} at low densities in macroscopic (i.e., laterally unconfined)

structures [9–13]; this trend has been attributed to strong screening effects from a nearby metal gate, the presence of disorder in the system, or contributions from the exchange interaction. Unfortunately, for meso- or nanoscale systems, the downward trend in κ^{-1} is easily reversed by the effect of an electrostatic confining potential produced by gate and source or drain electrodes, which tends to push electrons into an even smaller spatial region as their density is reduced. To our best knowledge, suppression in κ^{-1} at low densities has never been reported in laterally confined quantum structures.

Suspended carbon nanotubes (CNTs) are a promising platform for investigating the effects of strong electronic correlations in one dimension. As a clean, interacting quantum system, electrons in a suspended CNT at low density [14] may be described as a Wigner crystal [15]. Indeed, experimental studies have confirmed fascinating magnetic and electronic properties of the Wigner crystal phase, such as their exponentially suppressed exchange energy [16], absence of excited energy states [17], and giant orbital magnetic moment [18]. These observations indicate that, despite more than a decade of studies on the 1D Wigner crystal, improvement in device fabrication and higher quality carbon nanotubes lead to the discovery of novel signatures that have not been revealed before. Very recently, Shapir *et al.* [19] have developed a technique to observe the Wigner crystal directly by imaging the charge density of the system in real space. Providing detailed

theoretical calculations, they showed that the Wigner crystal regime has one of the strongest e - e interactions in the solid state. The strength of interactions is usually parametrized by r_s , defined as the ratio of the Coulomb interaction between electrons separated by a distance r , e^2/r , to their typical kinetic energy $\hbar^2/(m^*r^2)$. The interaction strength can be written in terms of the effective Bohr radius a_B as $r_s \approx 1/(na_B)$, where n is the one-dimensional electron density. The effective mass m^* is proportional to the CNT band gap Δ (with $\Delta = 2m^*v^2$, where v is the Fermi velocity), so that increasing Δ leads to a larger r_s and a stronger role for interactions. Previous studies of the addition energy spectrum, capacitance, and compressibility of CNTs have mostly focused on noninteracting physics and the weak-interaction regime [14,20–22]. But the effect of the band gap on these quantities in the Wigner crystal regime has not yet been considered.

In this Letter, we have studied one-dimensional systems with different e - e interaction strengths, using long, suspended CNTs of various band gaps. We report two contrasting trends of enhancement and suppression of κ^{-1} . In CNTs with very large band gaps, we observe suppression of κ^{-1} at low densities and provide a theory to show how this trend can be produced by a Wigner crystal. Using this theory, we show that compressibility is sensitive to both band gap and confining potential of the nanotube, which provides insight into the electronic interactions in these materials.

Our CNTs are grown using chemical vapor deposition across a $\sim 2 \mu\text{m}$ wide trench on prefabricated substrates to eliminate disorder effects (see Supplemental Material [23] [26]). A pair of gate electrodes is at the bottom of the trench and $\sim 750 \text{ nm}$ below the contact electrodes. Figure 1(a) shows a schematic of the device.

We focus on the low-density regime of electrons or holes in clean CNTs, which clearly exhibit single-electron or -hole conductance peaks in the Coulomb blockade (CB) regime, down to the last electron (hole) at the conduction (valence) band edge. The charge carrier density of CNTs can be modulated using electrostatic gating. A high-resolution map of the differential conductance dI/dV as a function of gate voltage V_g and source-drain bias voltage V_{SD} is shown in Fig. 1(b) for $T = 1.5 \text{ K}$ and illustrates CB diamonds and a band gap of $\Delta \sim 25 \text{ meV}$ in nanotube CN1. Figure 1(c) plots the conductance of CN1 as a function of carrier number. The regularity of CB peaks in these data, as well as the electron interference patterns in our devices with more transparent contacts [27] made with the same procedure, indicates that our devices are high quality and defect-free. In Fig. 1(c), the CB peaks get closer going from low to high carrier number. Figure 1(d) shows similar data from another device (CN2, with $\Delta \sim 165 \text{ meV}$). The CB peaks in CN2 show the opposite behavior; i.e., the CB peaks spread further apart with increasing carrier number.

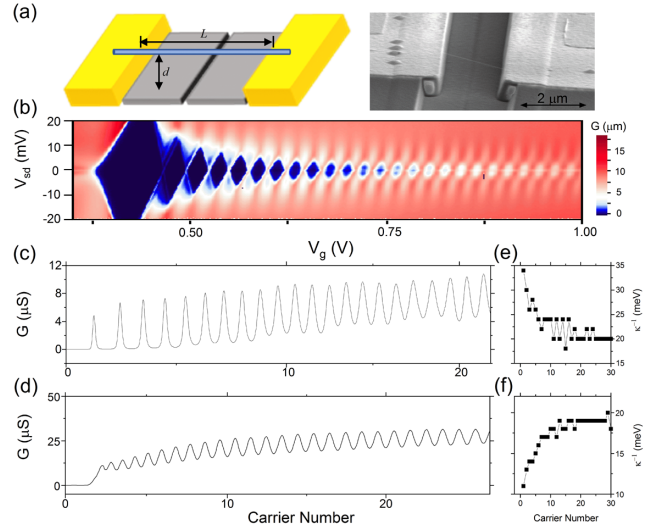


FIG. 1. (a) Schematic diagram (left) and SEM image of a device (right). Carbon nanotube is suspended over a $2 \mu\text{m}$ wide trench. The vertical spacing between trench and contact electrodes is $d = 760 \text{ nm}$. (b) Color scale plot of differential conductance versus gate voltage V_g and source-drain bias V_{SD} in CN1. Conductance G versus carrier number in (c) CN1 and (d) CN2. (e), (f) Inverse compressibility as a function of carrier number for the related device.

The compressibility of the nanotube can be obtained from gate voltage spacing between the neighboring CB peaks in the transport data converted to energy: $\delta_N = E_{N+1} - 2E_N + E_{N-1} = \kappa^{-1}$ see, e.g., [28,29], using $\mu = \alpha e V_g$, where gate voltage lever arm $\alpha = V_c/V_g$, and V_c is the height of the rhombic pattern in the $G(V_g, V_{SD})$ diagram [30]. Figures 1(e) and 1(f) plot the extracted value of κ^{-1} as a function of carrier number for CN1 and CN2. The alternating pattern in some parts of the plots arises from filling the subsequent orbital states with two electrons having opposite spins [14,20]. In CN1 ($\Delta \sim 25 \text{ meV}$), κ^{-1} is higher at low densities. This trend of addition energy has been reported previously and explained using a single-particle picture [14,20,21]. Because of the small effective mass of CN1, the energetics in this device has been considered to be dominated by a classical charging energy and the quantum kinetic energy. It is worth noting that the device imaged by Shapir *et al.* [19] with $\Delta = 45 \text{ meV}$ has similar energetics to CN1 and was found to be a Wigner crystal. On the other hand, we observe the opposite trend in CN2 with $\Delta \sim 165 \text{ meV}$; in this device, κ^{-1} is suppressed at low densities. In contrast to CN1, the effective mass of CN2 is large and the energetics are more likely to be dominated by Coulomb interactions. Correspondingly, the electronic compressibility of a Wigner crystal may follow a different trend in samples with such large gaps.

It is desirable to vary the band gap parameter to study its effect on κ^{-1} . One way to do this in a continuous manner is by applying an external magnetic field (B) parallel to the

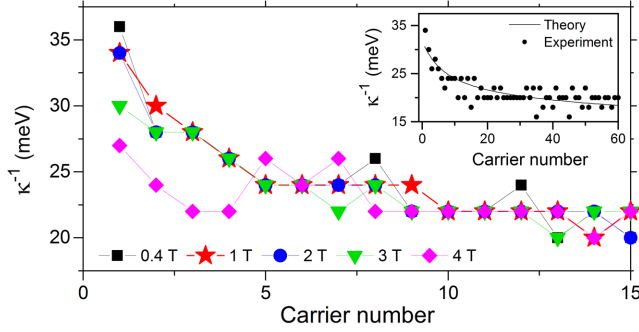


FIG. 2. Effect of magnetic field on κ^{-1} of CN1 for a range of magnetic fields, from $B = 0.4$ T, where the band gap reaches its minimum, to 4 T. (Inset) Comparison of theoretical (solid line) and experimental (dots) results of κ^{-1} as a function of carrier number for CN1.

axis of the tube [31–33]. This is particularly applicable to CN1, which has a small band gap at $B = 0$ and can display field-dependent energetics. Figure 2 shows κ^{-1} in CN1 as the magnetic field is varied from $B = 0.4$ to $B = 4$ T. The minimum band gap Δ_{\min} is obtained at 0.4 T and at higher fields the gap increases at a rate of ~ 2.5 meV/T. As the band gap is increased, κ^{-1} is observed to decrease at lowest densities.

To study the suppression in κ^{-1} at low densities, similar to CN2, we examine a range of different samples with appropriately large band gaps. Figure 3(a) shows the measured κ^{-1} as a function of carrier number in five devices (CN2–CN6) with band gaps ≥ 150 meV. In all of these samples, we observe the same trend as in CN2, meaning that in these tubes κ^{-1} is suppressed by going to low densities.

To understand behaviors of κ^{-1} at low densities, we propose an interacting model in which we calculate the ground-state energy E of a system having N electrons using the Hamiltonian $H = \sum_i K_i + \sum_{i \neq j} V(r_{ij}) + \sum_i U(r_i)$, where K_i is the kinetic energy operator for electron i , $V(r_{ij})$ is the interaction energy between two electrons separated by a distance r_{ij} , and $U(x)$ is the potential energy of an electron at position x due to an external electric potential. In the case of $U(x) = \text{constant}$, electrons are arranged with a uniform (voltage-dependent) density n along a line of length L_{eff} , so that the total number of electrons in the system is $N = nL_{\text{eff}}$. In the Wigner crystal limit, the electrostatic energy E_{el} of the system can be approximated by that of a classical collection of point charges with regular spacing $1/n$. In the limit where L_{eff} is much longer than the distance d to the gate electrode, $E_{\text{el}} = N \sum_{i=1}^{\infty} V(i/n)$, where the interaction energy $V(r)$ is given by the gate-screened Coulomb repulsion $V(r) = [e^2/(4\pi\epsilon_0)]\{(1/r) - [1/\sqrt{r^2 + (2d)^2}]\}$. At low electron densities $nd \ll 1$, the typical interaction energy becomes that of a dipole-dipole interaction, $V(1/n) \sim e^2 n^3 d^2 / (4\pi\epsilon_0)$. This rapid vanishing of V with n at low density

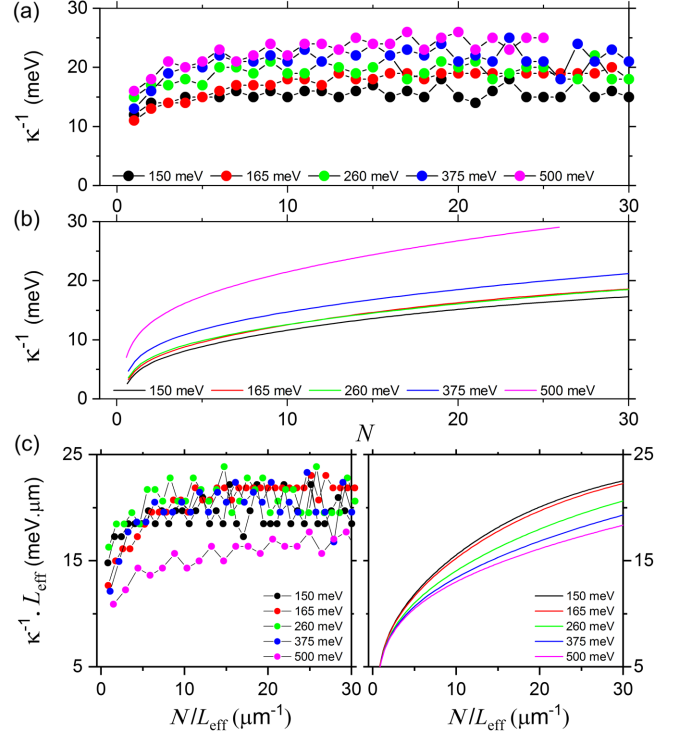


FIG. 3. Experimental data (a) and theoretical results (b) of κ^{-1} as a function of carrier number (N) for nanotubes with different band gaps. (c) Experimental (left) and theoretical (right) results converted to κ^{-1} times L_{eff} as a function of charge density.

implies that the electrostatic cost of inserting an additional energy decreases with decreasing concentration in cases where the electron density is uniform.

In the limit where $e-e$ interactions dominate over the quantum kinetic energy of electrons and the system adopts a Wigner crystal-like arrangement, the kinetic energy can be treated as a perturbation. In this situation, electron wave functions have little spatial overlap with each other and one can approximate the kinetic energy via a description where each electron is confined into a box of width $1/n$, such that neighboring electrons have no wave function overlap. Total kinetic energy is therefore given by the number of electrons multiplied by ground-state energy of a 1D particle in a box. We describe the kinetic energy of an electron via the relativistic dispersion relation $\epsilon(p) = \sqrt{(vp)^2 + (\Delta/2)^2} - \Delta/2$, where v is the Fermi velocity, p is the electron momentum, and the band gap is $\Delta = 2m^*v^2$. Note that, at low electron densities with small p , $\epsilon(p)$ reduces to the familiar form of $\epsilon(p) \cong p^2/(2m)$. Our approximation of a Coulomb-dominated electron state is justified when $V(1/n)$ is much larger than the typical kinetic energy scale $\epsilon(p = \pi\hbar n)$. At low electron densities, this inequality is satisfied in the usual limit of large r_s , $na_B \ll 1$. In our experiments, a_B is no larger than ~ 15 nm, while our CNT lengths are of order $2 \mu\text{m}$, so our approximation is justified when there are fewer than ~ 100

electrons in the system. The inverse compressibility is then $\kappa^{-1} = (1/L_{\text{eff}})d\mu/dn$. A detailed expression for μ is given in the Supplemental Material [23]. Our theoretical results for κ^{-1} , illustrated in Fig. 3(b), have the same trend and magnitude as our transport data for CN2–CN6. These modeling results demonstrate that our theory achieves an expected explanation for the behavior of κ^{-1} in the large band gap devices.

In addition to the Coulomb interactions between electrons, the electric potential difference between the gate and source or drain electrodes creates an external potential that may significantly affect the compressibility of the system. To model this effect, we assume that electrons reside in the minimum of a potential well described generically by $U(x) = [e^2/(4\pi\epsilon_0)][(x^2/D_1^3) + (x^4/D_2^5)]$. The position x is defined with respect to the location of the potential minimum, and the value of the electrostatic potential at this minimum can be set to zero. The length scales D_1 and D_2 define the strength of the potential. In the presence of such a confining potential, the electron density varies with position x , with electrons being more densely spaced at $x = 0$ and more sparsely spaced at larger distances from the minimum of the confining potential [7].

The inset of Fig. 2 compares the result of our theoretical calculation (solid line) and the measured data (dots) for CN1. Our fitted parameters D_1 and D_2 agree with the estimated value from electrostatic calculations in Ref. [7]. It can be seen that the theory matches very well with the experiment, implying that the confining potential plays an important role in the enhancement of κ^{-1} at low density for this tube. The larger values of κ^{-1} with decreasing N suggest that electrons are pushed together by the confining potential, so that L_{eff} of the device increases with increasing N . We have also calculated κ^{-1} in the presence of a magnetic field by adding a field-tunable gap [29] Δ_B to the nonvanishing gap Δ_{min} , and in the presence of a confining potential, we are able to derive a change in κ^{-1} as a function of B that is qualitatively similar to our experimental results (see Supplemental Material Fig. S2 [23]). The opposite trend of κ^{-1} observed in the larger band gap devices (CN2–CN6) suggests that the confining potential plays a weaker role in those devices, compared to CN1.

Obtained values of L_{eff} and a_B for individual tubes from our theory are presented in Table S1 of the Supplemental Material. The values of r_s range from ~ 3 (for small band gap and large N) to ~ 450 (for large band gap and small N). Large r_s is consistent with our initial assumption of the Wigner crystal regime [19] and justifies our estimate that contact interactions are negligible [34] in our large band gap tubes.

Interactions in the low-density regime are stronger in tubes with larger $\Delta \times L_{\text{eff}}$. In order to eliminate the effect of L_{eff} and present compressibility dependence on band gap of CNTs, results of Figs. 3(a) and 3(b) are illustrated in Fig. 3(c) in terms of $\kappa^{-1}L_{\text{eff}}$ as a function of density.

Levitov and Tselik [8] had previously theorized that large band gap tubes with slowly increasing κ^{-1} (with increasing density) are more strongly interacting. According to our measurements and calculations in Fig. 3(c), our tubes with larger band gap reach the constant κ^{-1} regime slower than CNTs with smaller band gap, which is consistent with Ref. [8]. Overall, our devices show the same behavior as our model, indicating that e - e interactions are stronger in the low-density regime of nanotubes with larger band gaps, causing κ^{-1} to grow with density.

The observed compressibility behavior by itself is not proof of a Wigner crystal. Previous works have explained a similar suppression of compressibility as a function of density, though not in a laterally confined structure, based on the exchange interaction in a uniform gas ($r_s = 0$) model [10,12]. The observed behavior in our devices could also be described using the simple model of a uniform electron gas with exchange interaction (presented in the Supplemental Material [23]). However, given the overwhelming evidence for Wigner crystallization from other experiments [16–19] in the parameter space of our devices, we can safely suggest our observed compressibility behavior as a probe of interaction strength of 1D Wigner crystals. Future studies will incorporate independent control of the band gap and confining potential.

In summary, we studied the effect of interactions on electronic compressibility of carbon nanotubes with different band gaps. We showed that contact interactions are not negligible in tubes with smaller band gaps and their compressibility can be tuned by applying an external magnetic field. For stronger (weaker) interactions, inverse compressibility decreases (increases) in the limit of low density in the Wigner crystal regime. In devices with suppression of addition energy at low density, tubes with larger band gaps reach the noninteracting regime at larger densities compared to tubes with smaller band gaps. Our theoretical modeling suggests that we are in a regime of relatively large r_s , and our data are consistent with a theoretical model of a Wigner crystal in a soft confining potential.

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The authors declare no competing interests.

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