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Graphitic Electrical Contacts to Metallic Single-Walled Carbon Nanotubes Using Pt Electrodes

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ABSTRACT

We investigate electronic devices consisting of individual, metallic, single-walled carbon nanotubes contacted by Pt electrodes in a field effect transistor configuration, focusing on improvements to the metal–nanotube contact resistance as the devices are annealed in inert environments including ultrahigh vacuum. At moderate temperatures ($T < 880$ K), thermal processing results in high resistance contacts with thermally activated barriers. Higher temperatures ($T > 880$ K) achieve nearly transparent contacts. In the latter case, analytical surface measurements reveal the catalytic decomposition of hydrocarbons into graphene layers on the Pt surface, suggesting that improved electronic behavior is primarily due to the formation of an all-carbon nanotube–graphite interface rather than to the improvement of the nanotube–Pt one.

Carbon nanotubes have many outstanding electronic properties, and they are considered an attractive candidate to replace silicon in future field effect transistors (FETs) and copper in high-density integrated circuit interconnects.^{1,2} Despite experimental and theoretical progress in these research areas, problems remain in the understanding and control of the interfacial electrical resistance between nanotubes and metal electrodes.² Small-diameter, single-walled nanotubes (SWNTs) in particular have high contact resistances, even when they have metallic bandstructure and no interfacial Schottky barriers.³ These contact effects are known to be sensitive to interface chemistry, with the choice of contact metal and the diameter of the SWNT being equally important.^{4,5} Empirically, Pd and Pt electrodes provide excellent electrical contacts to many types of nanotubes, and recent experiments using these noble metals have observed the intrinsic properties of SWNTs,^{4,6} including exotic physics in uncontaminated, freely suspended SWNTs.^{7–9}

Here, we present an investigation into the properties of Pt-contacted SWNTs and show that the catalytic properties of Pt are of unexpected importance. Transparent electrical contacts occur following the heating of Pt surfaces, during which graphene and graphitic surface layers nucleate to coat the noble metal's surface. Experiments combining electrical transport measurements with complementary surface-chemical analysis techniques, including X-ray photoelectron spec-

troscopy (XPS), electrochemical cyclic voltammetry, and Raman spectroscopy, together prove that contact resistance improvements directly correlate with a graphitic carbonization of the important Pt-SWNT interface. All of the devices selected in this study are metallic and/or small band gap SWNTs in order to minimize the complicating effects of extrinsic dopants on the gate bias dependence. Even in this relatively straightforward case of Pt contacts to metallic SWNTs, the diameter- and metal-dependence of contact barriers remains an unresolved issue generally.

Experimental Results. Figure 1 shows the resistance of two SWNT devices cycled between room temperature and 920 K in an ultrahigh vacuum (UHV) apparatus described previously.¹⁰ Device resistance as a function of temperature and back gate bias $R(T, V_g)$ is measured in situ over multiple temperature ramps between 300 and 1050 K in order to observe both the reversible and irreversible changes that occur upon heating. Device 1 was ramped quickly at a rate of 1 K/s to 600 K and then 900 K with an intermediate cooling back to 300 K (Figure 1A). Device 2 was heated at a much slower rate of 1 K/min to allow thermally activated processes to approach equilibrium throughout the ramp (Figure 1B). Both devices exhibit 10- to 100-fold changes between their maximum and minimum $R(T)$, even though current–voltage measurements remained linear throughout the experiment. Initial and final resistances R_i and R_f are given in Table 1 for these devices and four others measured in a similar manner.

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Figure 1. (A) $R(T)$ of two SWNT devices, with arrows indicating the direction of heating and cooling. First anneal to 600 K and back to room temperature (black) leaves device 1 in a very high resistance state. Annealing to 900 K (red) removes the contact barrier for all subsequent temperature ramps (blue). Inset: Arrhenius plot of the contact resistance during the higher temperature anneal shows a change in E_a above 670 K. (B) Similar experiment on device 2 with a single, continuous anneal cycle. Inset: AFM topograph of device 1.

Certain similar features are observed in all of the $R(T)$ characteristics, despite device-to-device differences in SWNT diameter d , channel length L , and R_i . As fabricated, the devices exhibit little temperature dependence below 400 K but become substantially noisier for $440 < T < 550$ K. Heating the devices above 500 K induces a dramatic change manifested in increased, high contact resistances with Arrhenius temperature dependences, i.e., $R_C(T) \sim \exp(E_a/kT)$,¹¹ with E_a being an effective energy barrier height exceeding 100 meV. Once converted to this high resistance intermediate state, devices can be cooled to room temperature or heated as high as 650 K with minimal changes to E_a , though new processes begin to change E_a at still higher temperatures (Figure 1A, inset). At and above 880 K, a second transition occurs in which $R(T)$ recovers a linear, metallic temperature dependence with a slope that is almost sample-independent (Table 1) and which agrees with previous measurements of the temperature-dependent, SWNT mean free path.¹² After heat treatment above 880 K, no further hysteresis is observed on subsequent thermal cycles up to 1200 K. We observe similar differences between initial and final device resistances for samples heated in a conventional tube furnace in flowing

Figure 2. (A) XPS spectra from 4 mm² area Pt films with (blue) and without (black) heat treatment to 920 K. For comparison of the carbon C 1s peaks, the two spectra are normalized to the Pt 4f peak. (B) Quantitative spectra acquired for a small window around the C 1s peak for the as-fabricated film (black), the identical film annealed to 920 K (blue), and the same annealed film after Ar sputtering in situ (gold).

argon, but all of the results described here are exclusively for devices treated in ultrahigh vacuum. We also note complementary findings in the literature for Pt electrodes contacting larger, double-walled nanotubes and annealed by joule heating.¹³

The behaviors at 400 and 500 K are likely dominated by the removal of surface physisorbates and chemisorbates from the SWNT system, notably N_2 ,¹⁴ H_2O ,¹⁵ and O_2 .¹⁶ Similar effects are observed using Ti and Pd electrodes.¹⁰ The higher temperature changes in device character, however, are unique to devices with Pt metal. Therefore, changes in surface chemistry were investigated ex situ using SWNT devices and identically prepared Pt films that had been cycled to the relevant temperature ranges.

XPS spectra in Figure 2 clearly indicate the formation of a surface carbon layer on the heat-treated Pt electrodes and films. Spectra from different samples, normalized to the Pt 4f peak, all show the expected Pt 4f, 4d, and 4p peaks, along with the signature of adsorbed oxygen. Samples heat treated to 920 K show a much larger C 1s peak near $E_b = 284$ eV than the as-deposited Pt. Argon ion sputtering (10 μ Torr at 3 keV) for 10 min reduces this carbon peak to below the level of residual carbon contamination on the as-prepared

Table 1. Initial and Final Device Characteristics

device	d (nm)	$L \pm 0.1$ (μ m)	R_i (k Ω)	R_f (k Ω)	R_{Cf}/R_{Ci}	E_a (meV)	$(dR/dT)/L$ (Ω/μ m K)	ΔG_i (%)	ΔG_f (%)
#1	2.0 ± 0.2	3.6	530 ± 40	48 ± 1	0.04	120	104 ± 3	8	52
#2	1.2 ± 0.1	1.8	4600 ± 500	130 ± 2	0.03	630	63 ± 4	<1	57
#3 ^a	1.7 ± 0.5	5.5	1300 ± 300	170 ± 1	0.10	220	82 ± 3	<1	7
#4 ^a	1.7 ± 0.5	7.8	2500 ± 200	250 ± 2	0.08	1300	71 ± 1	<1	11
#5 ^b	1.9 ± 0.1	2.6	920 ± 20	67 ± 1	0.05	—	—	23	85
#6 ^b	2.1 ± 0.2	4.7	480 ± 10	88.0 ± 0.5	0.11	—	—	47	83

^a Devices #3 and 4 are fabricated on two segments of the same SWNT. ^b Resistance measurements after exposure to air, instead of in situ in UHV.

Figure 3. CVs of Pt films in 1 M H₂SO₄ before annealing (black), after annealing to 550 K (red), and after annealing at 920 K (blue). The current density j/A is scaled by the electrolyte potential ramp rate dV_{RE}/dt .

film, as shown in a narrow scan around the C 1s peak (Figure 2b). Quantitative analysis determines the heat-treated sample to have four times more surface carbon than the as-fabricated film.

To determine the physical extent of the surface carbon coverage, electrochemical cyclic voltammograms (CVs) were acquired in 1 M H₂SO₄. Clean Pt displays two pairs of peaks arising from the redox behavior of the Pt atoms and from H adsorption and desorption processes, and a comparison of H peak heights from different samples allows a direct determination of accessible Pt surface. Our as-fabricated Pt films exhibit four clean CV peaks and reach steady state after fewer than four cycles at modest oxidizing potentials (1.4 V vs SHE), indicating minimal surface contamination.

Figure 3 shows representative CVs of Pt films with and without heat treatments. On films heated to 550 K, the H processes are mostly unchanged but the Pt redox currents are reduced by approximately 50%. Furthermore, 10 or more cycles are necessary to reach a steady state, after which the Pt activity has recovered to 80% of its initial value. This variation indicates the presence and gradual oxidation of a chemically resistant contamination layer on the Pt surface. On films heated to 920 K, H adsorption currents are reduced by at least 90%, indicating that the carbon coating is nearly continuous. Moreover, the peaks are shifted to higher potentials and the Pt peaks are almost completely absent, indicating that the carbon film is not merely continuous, it is also chemically inert. This CV behavior is stable and unchanged for many cycles, so long as V_{RE} is maintained less than 1.8 V (vs SHE). The inactivity of the surface within this potential window is characteristic of C–C bonds and compares well with the threshold voltages for the onset of lattice damage for the oxidation of crystalline graphite in H₂SO₄.¹⁷

Evidence for graphitization is also observed in the Raman spectra of these samples, as depicted in Figure 4. As-fabricated films and films heated to 550 K are both featureless and flat over the 1000–3000 cm^{−1} range when compared to the clean Pt reference. Upon heating to 920 K, samples begin to exhibit weak peaks at 1604 and 1378 cm^{−1}, corresponding to the G and D bands characteristic of sp² bonding and aromatic ring structures.¹⁸ These Raman features confirm that the surface carbon is graphitic. Due to the high reflectivity of the Pt surface, extensive background scattering limits the

Figure 4. Raman spectra of Pt films with background subtracted as deposited and annealed at 550, 920, and 1050 K in UHV in the region of interest near the Raman bands of graphene. Traces are offset for clarity. The graphitic G, 2D, and D bands are labeled.

signal-to-noise ratio of the observed Raman bands. Thus, even though recent research has determined the dependence of Raman peak shapes and positions on the number of graphene layers, similar analysis is not yet possible on these Pt films. Heating to 1050 K helped to sharpen the G band feature and resolve the higher frequency 2D band, but did not allow reliable peak fitting for layer counting. We note that standard methods of graphene peeling and exfoliation have been attempted on these surfaces without successful removal of any Raman detectable graphene layers. This failure may indicate that the graphitic coating consists primarily of single graphene layers well-adhered to the Pt.

Finally, intermittent contact atomic force microscopy (AFM; Nano-R, Pacific Nanotechnologies) has been unable to clearly resolve any added thickness from these carbon films. The root mean square surface roughness of as-fabricated films is 0.8 nm, and while this value does not change appreciably at low temperatures, it increases approximately 25% after heating to 920 K in vacuum and by an additional 50% when heating with oxygen to remove the carbon. Such changes limit the resolution of quantitative topographical comparisons to about 1.0 nm; in other words, the thickness of the graphitized carbon films is no greater than 1.0 nm but cannot be further resolved. Improving upon this situation will require atomic resolution imaging of the device interfaces, perhaps using cross sectional transmission electron microscopy (TEM), as is standard in the semiconductor industry. A full investigation, both before and after high temperature treatment, could unambiguously determine the atomic structure of the interfaces. Unfortunately, imaging and cross sectioning single graphitic layers, especially layers adhered to metal substrates, is an outstanding technical challenge that remains unmet in the graphene community, and it has not been completed in this work.

Discussion. The well-developed surface science of Pt crystal surfaces agrees with and supports many of the experimental observations described above. Pt is known to be catalytically active at high temperatures, and at high temperature Pt selectively dehydrogenates the residual hydrocarbons present in most vacuum environments, including UHV.^{19,20} Pt does not form any known carbides or carbon alloys, so this chemical process leads to a surface uniformly coated with carbidic carbons (C_xH_y) at temperatures in the range 430 < T < 770 K.²¹ At still higher temperatures, T >

800 K, catalytically assisted graphitization of these carbons occurs.²² Similar chemical processes should be able to occur in any inert, nonoxidizing environment that contains a sufficient source of hydrocarbons. The synthesis of single-layer graphene is self-limiting on Pt, Ir, and Rh single crystals due to the catalytic mechanism at work, contrary to the multilayer synthesis that occurs via precipitation or decomposition on surfaces of Ni, Ru, or SiC.

The XPS, CV, and Raman characterization presented in this work are all consistent with a model in which catalytic decomposition forms a carbidic carbon soot on the Pt at low temperatures and a chemically inactive graphene coating at higher temperatures. The particularly novel finding is that these chemical changes occur in concert with dramatic changes in SWNT device properties. A rapid resistance degradation is observed at 440 K in the slow, quasi-equilibrium thermal scan depicted in Figure 1B, matching the temperature for catalytic carbonization of the Pt surface. Likewise, the transition to transparent contacts appears to be completed at 880 K in the slow scan, matching the lower bound for graphitization of the surface carbons. Even though SWNT devices are known to be highly sensitive to the surface chemistry of the SWNT–metal interfaces, past work on “ideal contacts” achieved with Pd or Pt contact electrodes have not previously considered the possible roles of chemisorbed carbons.^{4,6–9} In fact, the irreversible changes described here suggest that the entire advantage of Pt surfaces is due to its graphitization, and the electronic benefits of a SWNT–graphene interface.

Simpler annealing processes are the more conventional and accepted explanation for improved SWNT–metal contacts, and the mobility of Pt atoms at high temperatures might play a role in the observed measurements. Nevertheless, we argue here that annealing plays only a minimum role. First, simple annealing cannot explain the appearance and disappearance of a highly resistive, thermally activated contact barrier in the $440 < T < 880$ K temperature range. Second, Pt is a refractory metal that in bulk does not begin to anneal until 1300 K,²³ well above the experimental range used here. Third, much of the research that ascribes benefits to the annealing of SWNT–metal contacts^{24–28} has relied almost exclusively on electrical measurements before and after treatment, without the insights of in situ measurement or complementary surface chemical analysis.

To further check our conclusion that Pt annealing plays a minimal role in the SWNT devices, simple Pt wires were fabricated ($30 \text{ nm} \times 20 \text{ } \mu\text{m} \times 4.3 \text{ mm}$ long) and heated in UHV in parallel with the SWNT devices. Some irreversible resistivity decreases in the 350–420 K range occur due to the relaxation of structural imperfections in the microcrystalline films (see Supplemental Figure S1 in Supporting Information),^{29–31} and these may correlate with changes in the SWNT devices at the same temperatures. However, the Pt films never show nonmetallic behavior nor any effects in the 850–950 K temperature range. Finally, it is noted that heating can cause thick Ti adhesion layers to migrate along Pt film grain boundaries, increasing film resistivity and degrading substrate adhesion.³¹ However, our films retain

adhesion to 920 K and do not increase in resistivity over the temperatures studied here, so the underlying Ti adhesion layers are not believed to play significant roles.

Instead, we conclude that the primary cause of the electronic changes is due to the Pt carbidization and graphitization, and the remaining discussion addresses the apparent role of these added carbon species at the Pt–SWNT interface. At 920 K, the graphitic domains remain nanometer-sized²² and yet the electronic modification is substantially complete, suggesting that very small quantities of graphitic carbon are sufficient for the effect. Empirically, metals make the poorest connections to SWNTs having small diameters (e.g., $< 1.5 \text{ nm}$), and the role of the nanocrystalline graphite may simply be to bridge the relevant interface with additional surface area. Extending the wave function overlap between the Pt electrode and coherently organized carbon may be sufficient to improve the electrical transparency of the interface. First principles modeling could address this hypothesis on planar devices and, in fact, has been used to understand electronic measurements between a nanotube and a graphite crystal surface³² and between two graphene layers.³³ Unfortunately, the experimental geometry here presents a much more complex case where the SWNT pierces the Pt surface rather than laying flat on it. Because graphitic nanocrystallites nucleate and grow conformally on the Pt surface,²² the contact interface between graphene and SWNT cannot benefit from simple, planar stacking. For the graphene to either partially wrap onto the SWNT or to covalently hybridize with it, symmetry-breaking, nonhexagonal rings or other defects would have to be incorporated, and these mechanisms would certainly depend on the SWNT diameter. All of these difficulties complicate efforts to model the interface precisely. The lower temperature device behaviors, such as the formation of an electronic barrier, may in fact be evidence of intermediate chemical modifications where the SWNT sidewall comes into contact with catalytically decomposed, reactive carbon and hydrocarbon fragments at the Pt interface.

The CVD conditions typically used for SWNT synthesis almost certainly coat prefabricated Pt electrodes with graphene, as revealed in the Raman spectra (Figure S4 in Supporting Information) of Pt films annealed in a CVD growth furnace. This fact suggests that recent reports of low contact resistances in similarly fabricated devices have probably employed SWNT–graphene interfaces rather than simply SWNT–Pt ones.^{7–9} Where previous work grew SWNTs on top of Pt electrodes, the measurements in Figure 1 use clean, bare Pt that is deposited after SWNT growth and never exposed to the carbon CVD processes. The difference in fabrication is critical, because it allows the direct observation of the transition from one type of contact to the other. Subsequent heating is clearly responsible for the interface improvements, and Table 1 shows the graphitized interface is 14 times less resistive on average than those incorporating as-fabricated Pt. The final contact resistance values, though still averaging more than double h/e^2 , compare favorably to other measurements of SWNTs on substrates, and measurements down to 77 K observe device resistances as low as

24.5 k Ω , indicative of transparent contacts. Contact resistance remains somewhat higher for small diameter SWNTs (<1.5 nm), consistent with other reports,^{3–5} but the limited data set should not be overinterpreted. Future work will need to investigate diameter dependence more systematically before the mechanisms involved can be clearly understood.

After graphitization, measurements also begin to resolve modest gate dependences in “metallic” SWNTs equivalent to those described in recent articles.⁸ Even though initially selected with little or no gate sensitivity, devices formed from small-diameter, metallic SWNTs all exhibit some gate dependence after the elimination of their high contact resistances. This dependence, expressed as a percentage change in the total conductance ΔG , is included in Table 1 for devices before and after annealing. Scanning gate microscopy confirms the gate sensitivity to be distributed along the length of the SWNT after annealing, rather than being solely at the electrode interfaces (Figure S2 in Supporting Information). Furthermore, in no case have we observed an increase that is inconsistent with the initial degree of gate dependence, e.g., as might occur if the SWNT were itself being chemically modified by the heat treatment. We have also carefully considered the possible effects of Pt migration and contamination along SWNT conductors but have been unable to topographically observe any evidence of surface Pt after these heat treatments. This fact suggests that the primary mechanism responsible for changing the apparent gate dependence is the elimination of high contact resistance. Once this resistance has been removed, the underlying gate dependence of the SWNT channel^{7–9} is more clearly resolved.

Even though the electronic modification is finished at 920 K, changes in surface structure continue up to much higher temperatures. For example, graphitic nanodomains agglomerate and grow at temperatures above 920 K,²² a process which ultimately allows the faint resolution of graphene’s 2D band at 1050 K (Figure 4). Also, the maxima of the G (1604 cm^{–1}) and 2D (2720 cm^{–1}) bands are both upshifted in Figure 4 from their positions in graphite crystals. This effect is consistent with the expectations for epitaxial layers of sp² carbon grown on metal surfaces, like those observed on Ni³⁴ and Ru.³⁵ In principle, upshifts can be caused by doping³⁶ or elevated temperature,³⁷ though the UHV processing and low laser fluences used here make these mechanisms unlikely. Finally, individual Pt atoms certainly become more mobile at higher temperatures, and previous TEM work has observed relevant, high-temperature migration processes.³⁸ No residual metal remains upon cooling a SWNT, though, since Pt does not generally wet graphitic surfaces. An interesting exception to this rule can occur around defects, which serve to nucleate metal nanoparticles and fix them for observation at room temperature.³⁸ In some of our devices, Pt decorations are in fact observed after heat treatments to temperatures above 1000 K.

In conclusion, transparent contacts to SWNTs are reliably fabricated using Pt metal, but the key to low resistance appears to be the catalytic carbonization and graphitization of one or more graphene layers on the Pt surface. The initial

carbonization dramatically worsens contact resistance, resulting in thermally activated barriers of 100 meV or more. Graphitization occurs at and above 880 K, and on Pt is unavoidable under any conditions that are simultaneously compatible with SWNTs. Despite being intuitively appealing and theoretically tractable, little research has addressed graphene contacts to SWNTs, and the possibilities certainly deserve further theoretical and experimental exploration.

Experimental Methods. SWNT devices are fabricated using standard chemical vapor deposition methods and a molecular Fe₃₀Mo₈₄ nanocluster^{39–41} as growth catalyst. The initial catalyst is diluted in ethanol, spun directly onto thermally oxidized Si wafers, and then oxidized and reduced as described in previous reports.⁴² An ethanol carbon source and a deposition temperature of 940 °C result in long, well-isolated SWNTs with diameters ranging from 1.2 to 2.2 nm. Electrodes spaced 2–8 μ m apart were defined on top of the SWNTs by conventional optical photolithography using a lift-off resist (LOR 1A, Microchem) followed by an electron beam deposition of 30 nm of Pt. A minimal $\sim$ 0.8 nm Ti adhesion layer, likely consisting of a mixture of Ti and TiO₂, was kept thin to enhance Pt–SWNT contact and is not believed to affect the electrical properties.⁴³ Devices were inspected by AFM to determine the SWNT diameter d and channel length L values tabulated in Table 1 and to ensure each device consisted of a single SWNT.

XPS spectra are acquired using an ESCALAB MK II photoelectron spectrometer in an independent UHV system equipped with a Al/Mg twin anode X-ray source and a 150 mm hemispherical electron energy analyzer. Spectra were taken using Mg K α radiation (1253.6 eV) at a base pressure of 1×10^{-9} Torr. The constant analyzer energy mode was used throughout, and constant pass energies for survey scans and high-resolution scans were 100 and 20 eV, respectively.

Electrochemical CVs are acquired in 1 M H₂SO₄ on patterned Pt contact pads (surface area of 0.13 mm²) and continuous films (1.52 mm²). A 99.9% Pt foil and wire (Sigma-Aldrich) are used as counter electrode (CE) and reference electrode (RE), respectively. Both CE and RE are precleaned before each measurement in sulfuric piranha solution, followed by sonication in IPA and deionized water. Testing against a standard calomel reference electrode ensured stability of the RE, with errors estimated to be $\leq \pm 10$ mV.

Raman spectra were recorded using a 405 nm continuous-wave diode laser operating on TEM₀₀ mode (Newport Corp., Excelsior 405-050) as the excitation source. The laser is attenuated down to 2 mW and focused on the sample with an aspherical lens (NA = 0.5, f = 8 mm). The backscattered radiation is collected through the same lens and passed through a confocal spatial filter and a pair of notch filters (Semrock, NF02) prior to dispersing in a $1/4$ m polychromator and recording with a CCD array. The effective instrument resolution was 10 cm^{–1}, calibrated using dimethyl sulfoxide as the Raman scatterer. A Pt film, electrochemically cleaned in 1 M H₂SO₄, is used to record a background spectrum subtracted from the spectra in Figure 4.

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Supporting Information Available: Four figures include additional control measurements, Raman spectra, and scanning gate microscopy. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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