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Sub-Kelvin transport spectroscopy of fullerene peapod quantum dots

Pawel Utko^{a)} and Jesper Nygård

Nano-Science Center, Niels Bohr Institute, University of Copenhagen, Universitetsparken 5, DK-2100 Copenhagen, Denmark

Marc Monthieux and Laure Noé

Centre d'Elaboration des Matériaux et d'Etudes Structurales (CEMES), UPR A-8011 CNRS, B.P. 94347, 29 rue Jeanne Marvig, F-31055 Toulouse Cedex 4, France

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The authors have studied electrical transport properties of individual C₆₀ fullerene peapods, i.e., single-wall carbon nanotubes encapsulating C₆₀ molecules. Their measurements indicated power lawlike temperature dependencies of linear conductance similar to those for empty nanotubes. At temperatures below 30 K, peapod devices behaved as highly regular individual quantum dots showing regular Coulomb blockade oscillations. Signatures of Kondo physics appeared at the lowest measurement temperature of 315 mK. © 2006 American Institute of Physics.

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Carbon nanotubes encapsulating fullerene molecules,¹ the so-called fullerene peapods, have recently attracted interest as potential building blocks for nanoelectronics. Their prospective applications include data storage devices,^{2,3} single electron transistors,^{4,5} spin-qubit arrays for quantum computing,⁶ nanopipettes,⁷ and nanoscale lasers.⁸ A detailed review on peapod synthesis can be found in Ref. 9, whereas Ref. 10 provides an overview on their structural and electrical properties.

The fullerene presence inside a peapod is expected^{11–13} to modify the electronic band structure of the nanotube, thus affecting its electric properties. Moreover, the incorporation of fullerene molecules represents an interesting route to modify the one-dimensional electron system of the nanotubes¹⁴ or to introduce an interplay with mechanical degrees of freedom (fullerene motion) in nanotube quantum dots. To investigate such regimes and coherent transport phenomena, it is important to establish the electrical transport characteristics of peapods at the lowest temperatures.

Here, we investigate transport properties of individual C₆₀ peapods at temperatures down to 315 mK, where quantum effects dominate the device characteristics. In particular, we demonstrate that the temperature dependencies of the linear conductance indicate a power lawlike behavior similar to that for empty nanotubes. At low temperatures ($T < 30$ K), our devices behave as highly regular individual quantum dots showing regular Coulomb blockade oscillations. Signatures of Kondo physics appear at the lowest measurement temperatures as observed independently in Ref. 5, the only other report on peapod transport below 1.8 K.

Fullerene peapods used in our study are based on single-wall carbon nanotubes (SWNTs) grown by arc discharge. Their diameter distribution $d = 1.3–1.6$ nm (as determined by Raman spectroscopy) is appropriate for subsequent filling with C₆₀ molecules. The empty nanotubes were supplied by NANOCARBLAB as a purified material (80% pure grade). The purification procedure consisted of multistep heating of the raw material in air and soaking in HNO₃, followed by a microfiltration step. Thanks to the purification process, the

as-received SWNTs were already opened without the need for any further treatment. The tubes were then filled with C₆₀ fullerenes (98% purity, INTERCHIM) via the vapor phase method,⁹ in an evacuated and sealed ampoule, at 500 °C for 24 h. The resulting peapods were subjected to an 800 °C heat treatment under dynamic vacuum for 1 h, in order to remove the excess fullerenes.

The high structural quality of our peapod material was confirmed by a suite of complementary techniques: Transmission electron microscopy (TEM) imaging revealed a uniform filling of SWNTs, see Fig. 1(a). A quantitative analysis by x-ray diffraction, similar to that in Ref. 15, indicated a state-of-the-art 90%–95% content of peapods in the bulk material. In addition, signatures of the encapsulated C₆₀ fullerenes were found in Raman spectroscopy.

For electrical transport measurements, peapods were deposited on a SiO₂ surface by wetting it with a sonicated peapod suspension in dichloroethane. Individual tubes and bundles were located using atomic force microscopy (AFM). Only those with a diameter of 1.3–2 nm (as determined by AFM) were selected for further processing. An appropriate pattern of Pd/Au (15 nm/45 nm) electrodes was then fabricated by means of electron-beam lithography. The electrodes were spaced by about 400 nm, see Fig. 1(b). A highly

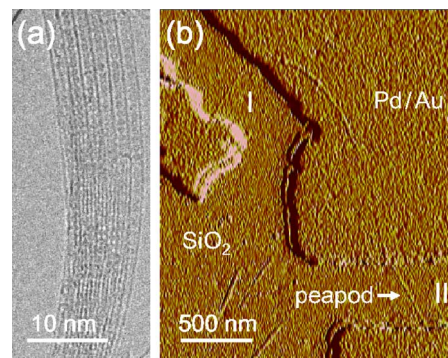


FIG. 1. (Color online) (a) High-resolution TEM image of C₆₀ fullerene peapods. (b) AFM micrograph of a fullerene peapod contacted with Pd/Au microelectrodes. Two sections (I and II) of the same peapod can be probed independently.

^{a)}Electronic mail: pawel@fys.ku.dk

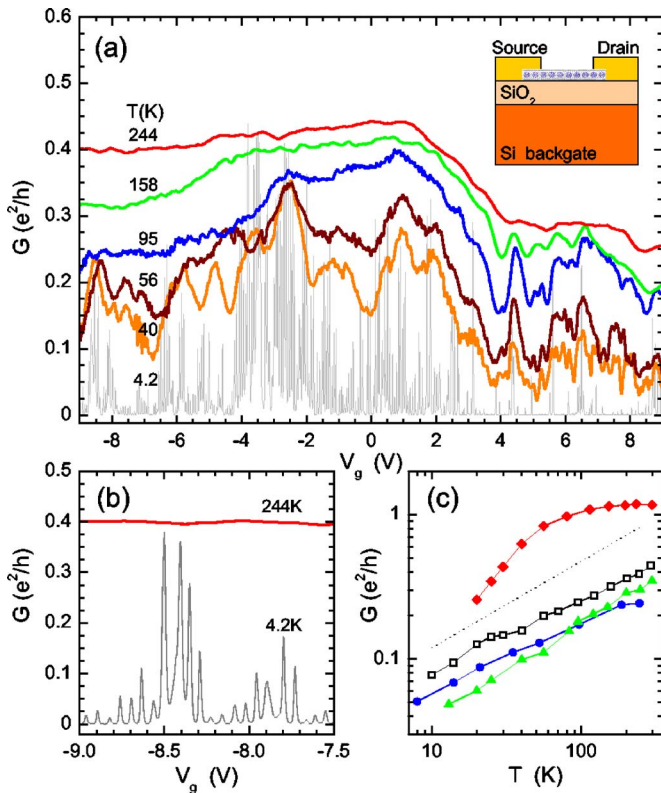


FIG. 2. (Color online) (a) Linear conductance G vs gate voltage V_g at fixed temperatures from 4.2 to 244 K, measured for section I in Fig. 1(b). Inset shows a schematic sample layout. (b) Closer view on Coulomb blockade oscillations observed for the same device at 4.2 K. (c) Linear conductance G vs temperature T for four different samples. Open squares indicate the data points for the same device as in (a) and (b). The dotted line indicates $G \propto T^{0.6}$ for comparison.

n -doped Si substrate, situated beneath the 500-nm-thick SiO_2 layer, served as a backgate. A schematic sample layout is shown in the inset of Fig. 2(a).

In this study, we present data for four peapod devices that were found to be conducting at room temperature and were further investigated at temperatures down to 315 mK. The experiments at $T \geq 4.2$ K were performed in a ^4He Dewar. Those at $T < 4.2$ K were carried out in a ^3He refrigerator with a base temperature of 315 mK. The differential conductance dI/dV was measured with an excitation voltage of 30–100 μV , using standard lock-in techniques.

Figure 2(a) shows a representative plot of linear conductance $G = dI/dV|_{V=0}$ for a peapod device, measured as a function of backgate voltage V_g . The measurements were performed at different temperatures from 4.2 to 244 K. A rather weak gate-voltage dependence at high temperatures ($T > 200$ K) indicates metallic properties of the device. The reduction in G around $V_g \approx 2$ V for this sample may indicate the presence of a small band gap.¹⁶ When the temperature is lowered, the average conductance decreases steadily until around $T \approx 30$ K, where discrete Coulomb blockade (CB) peaks appear in the $G(V_g)$ characteristics. This is shown in more detail in Fig. 2(b), for $T = 4.2$ K. Here, single electron charging dominates the electric transport.

The spacing between neighboring Coulomb peaks remains fairly constant along the whole gate-voltage range in Fig. 2(a), indicating that the device is dominated by a single quantum dot. Pronounced variations in peak height reflect the coupling of particular electronic states in the dot to the

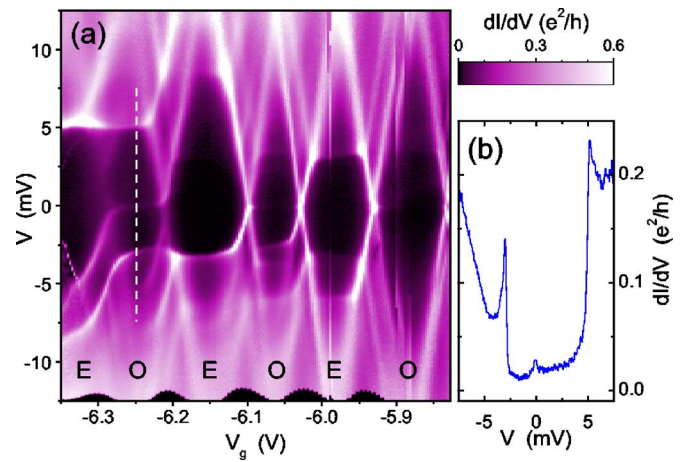


FIG. 3. (Color online) (a) Color-coded plot of differential conductance dI/dV vs gate voltage V_g and bias voltage V , measured at $T = 315$ mK. The odd (O) and even (E) occupancies of the dot are derived from the alternating presence of zero-bias Kondo resonances. (b) dI/dV vs V at a fixed gate voltage $V_g = -6.25$ V indicated by the vertical dashed line in (a).

source and drain leads. We have observed a similar CB behavior for three out of four tested samples, those with a room-temperature conductance of $G_{\text{RT}} \approx 0.3\text{--}0.5e^2/h$.

Figure 2(c) summarizes our observations, showing a temperature dependence of the linear conductance G for all four investigated peapod devices. The data points, plotted here in a log-log scale, were collected in the gate-voltage ranges for which G showed no or little dependence on V_g at high temperatures. The results for the three samples with $G_{\text{RT}} < 1e^2/h$ are consistent with a power law dependence $G \propto T^\alpha$, closely resembling reports on Luttinger liquid behavior in tunneling through closed carbon nanotube quantum dots.^{17,18} Notably, we find that the exponent $\alpha \approx 0.6$ seen for our peapods is similar to those observed for empty SWNTs. This is in contrast to theoretical studies¹⁴ predicting a smaller interaction parameter for fullerene peapods g_p as compared to that for empty nanotubes g_t . Thus assuming an approximate relation¹⁴ $1/g_p^2 = 2/g_t^2 - 1$ and $g_t \approx 0.28$ for metallic SWNTs,¹⁷ a correspondingly larger exponent $\alpha = (g_p^{-1} - 1)/4 \approx 1.0$ is expected for peapods, but not observed here. The highly conducting sample ($G_{\text{RT}} \sim 1e^2/h$) shows a weaker suppression of G at high temperatures, in accordance with other data on open nanotube devices.¹⁸

Figure 3(a) shows a Coulomb blockade behavior at $T = 315$ mK. The color-coded conductance dI/dV is plotted with respect to both the gate voltage V_g and the source-drain bias V for a different sample than in Fig. 2(b). The regular structure of CB diamonds once again indicates that the device behaves as a single quantum dot. Moreover, the length scales¹⁸ determined from both the charging energy $U \approx 8\text{--}12$ meV and the level spacing $\Delta E \approx 2\text{--}3$ meV are consistent with the electrode separation of ~ 400 nm. These results imply that our peapods exhibit ballistic electron transport over the entire segment between the two electrodes, i.e., the elastic mean free path is at least 400 nm. Thus the nanotube openings, which allowed filling with C_{60} fullerenes, do not hinder clean electron transport over extended distances in our material.

The horizontal lines inside the Coulomb diamonds in Fig. 3(a) are signatures of cotunneling events. In particular, the lines seen at zero bias for every second diamond can be attributed to Kondo effect^{19,20} which exists for odd occu-

pancy (O) of the quantum dot, i.e., when it holds an unpaired spin. This is a definite proof that the device behaves as a single dot with coherent electron transport between source and drain electrodes. The horizontal lines at finite V indicate inelastic cotunneling processes²¹ leaving the dot in an excited state. Pronounced peaks observed in $dI(V)/dV$ characteristics at some of these bias thresholds [Fig. 3(b)] could suggest that Kondo correlations enhance also these resonances in nonequilibrium transport.²²

Theoretical studies¹¹⁻¹³ predict dramatic changes in electronic properties of the tubes upon fullerene encapsulation. Such changes would involve hybridization and corresponding shifts of electronic states of SWNTs and C_{60} fullerenes. This could explain, for example, modifications in the local electronic structure of the nanotube observed in scanning tunneling microscopy experiments on semiconducting peapods.²³ A similar scenario was also considered by Yu *et al.*⁴ to explain features seen in low-temperature electrical transport characteristics of semiconducting peapods. Their measurements, performed at 1.8 K, showed a fairly irregular structure of CB diamonds indicating the presence of multiple quantum dots in series. This was attributed to additional tunneling barriers due to the C_{60} -induced modulation of the electronic band structure of the nanotube.⁴ We note, however, that such a behavior could also result from a mere presence of impurities or defects in the tube.

In contrast, we have shown here that devices based on C_{60} -filled nanotubes can behave as highly regular individual quantum dots. The Kondo effect, a hallmark of coherent transport, is present at sub-Kelvin temperatures, in accordance with a recent study on similar quantum dots by Quay *et al.*⁵ where a different source of peapod material was used. At higher temperatures, the power lawlike $G(T)$ dependencies are similar to those for empty tubes, contradicting the theoretical predictions¹⁴ that a modified interaction parameter in peapods should change this behavior. Hence, our experiments indicate that the electron system of the nanotubes is less perturbed by the encapsulated fullerenes than expected, in line with recent photoemission experiments.²⁴

Having established the excellent overall transport characteristics of peapod quantum dots, the future work could involve studying (by means of detailed transport spectroscopy) the consequences of interactions between nanotubes and C_{60} molecules. In particular, the interplay with mechanical degrees of freedom of the encapsulated fullerenes could be probed.

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