nature photonics
SUPPLEMENTARY INFORMATION
DOI: 10.1038/NPHOTON.2015.123

Observation of a Luttinger-liquid plasmon in metallic single-walled carbon nanotubes

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S1. Theory of 1D plasmon of Luttinger liquid in SWNT bundles.

Interacting electrons in one dimension behave as Luttinger liquid, which can be solved analytically through Bosonization. We follow the pioneering work of Kane et al[1] to derive the 1D plasmon properties of Luttinger liquid in carbon nanotubes.

The Bosonized representation of free electrons in a SWNT takes the form
$H_0 = \sum_{\alpha,\beta} \mathcal{H}_0(\theta_{\alpha\beta}, \phi_{\alpha\beta})$, and $\mathcal{H}_0(\theta, \phi) = \int dx \frac{\hbar v_F}{2\pi}[(\partial_x\theta)^2 + (\partial_x\phi)^2]$,
where the electronic density in a given channel is given by $\rho_{\alpha\beta} = \partial_x\theta_{\alpha\beta}/\pi$, and α and β are spin and valley indices, respectively.

For a SWNT with Coulomb interaction screened by an external metal shell of a radius R_s, the energy required to charge a unit length of nanotube is
$\mathcal{H}_{int} = e^2\rho^2 \ln\left(\frac{R_s}{R}\right)$. Here R is the nanotube radius and the total charge density $\rho = \sum_{\alpha,\beta} \partial_x\theta_{\alpha\beta}/\pi$. Since the interaction Hamiltonian involves only ρ, and we can introduce a new charge channel by $\theta_\rho = \sum_{\alpha,\beta} \theta_{\alpha\beta}/\sqrt{4}$ and $\rho = \sqrt{4} \cdot \partial_x\theta_\rho/\pi$, where 4 is the number of channels with the spin and valley degeneracy. The total Hamiltonian can be decomposed into three neutral channels and one charge channel, where the charge channel is described by

$$\mathcal{H}_\rho = \int dx \frac{\hbar v_F}{2\pi}[(\partial_x\theta_\rho)^2 + \left(\partial_x\phi_\rho\right)^2] + \int dx \left[\frac{4}{\pi^2}\left(\partial_x\theta_\rho\right)^2 e^2 \ln\left(\frac{R_s}{R}\right)\right] =$$
$$\int dx \frac{\hbar v_F}{2\pi}[(1 + \frac{4\cdot 2}{\pi\hbar v_F} e^2 \ln\left(\frac{R_s}{R}\right))(\partial_x\theta_\rho)^2 + \left(\partial_x\phi_\rho\right)^2].$$

Consequently, the collective charge excitation (i.e. 1D plasmon) in an individual SWNT propagates at a renormalized velocity v_ρ described by

$$v_\rho/v_F = 1/g = \sqrt{1 + (4\cdot 2e^2/\pi\hbar v_F)\ln(R_s/R)}.$$

In a nanotube bundle containing N SWNTs on top of a substrate, the interaction Hamiltonian for short-wave plasmon oscillation is modified in four aspects: (1) The number of conducting channels will be 4N; (2) the substrate screening leads to an effective dielectric constant of ε_{eff}; (3) potential energy for a periodic oscillating charge density has a cutoff length determined by the plasmon wavelength; and (4) the nanotube bundle has an effective diameter R_t different from R. From electrostatic calculation, we obtain an interaction Hamiltonian of

$$\mathcal{H}_{int} = \frac{e^2\rho^2}{\varepsilon_{eff}} \mathrm{K}_0\left(\frac{2\pi R_t}{\lambda_p}\right) = \frac{\hbar v_F}{2\pi}[\frac{4N\cdot 2}{\pi\hbar v_F}\frac{e^2}{\varepsilon_{eff}} \mathrm{K}_0\left(\frac{2\pi R_t}{\lambda_p}\right)](\partial_x\theta_\rho)^2,$$

where K_0 is the 0^{th} order modified Bessel K function. Note here we used an effective shell (with consideration of the capacitance) with diameter related to the plasmon wavelength.

The change in Coulomb interaction energy leads to a Luttinger parameter described by

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$$1/g = \frac{v_p}{v_F} = \sqrt{1 + (\frac{8Ne^2}{\varepsilon_{eff}\pi\hbar v_F})\mathrm{K}_0(2\pi R_t/\lambda_p)} \approx \sqrt{1 + (\frac{8Ne^2}{\varepsilon_{eff}\pi\hbar v_F})\ln{(\lambda_p/2\pi R_t)}} \quad . \quad (1)$$

It nicely reproduces the experimentally observed quasi-quantization of the plasmon propagation speed in small nanotube bundles containing different number of metallic SWNTs.

S2. Determine the effective dielectric constant ε_{eff}.

We need an effective dielectric constant in the model. The dielectric constant is a result of screening by the dielectric substrate. We determine the effective dielectric constant by calculating the capacitance of the system for a nanotube with and without a substrate.

We determine the effective dielectric constant ε_{eff} due to the substrate screening using numerical simulation based on finite element methods (with the software COMSOL Multiphysics). In the simulation, we assumed that the charges are distributed on the nanotube shell, and the nanotube is separated from the BN substrate by 0.35 nm due to Van der Waals interactions. We obtain an ε_{eff} of 2.1 at 10.6 µm and an ε_{eff} of 1.0 at 6.1 µm, where the in-plane and out-of-plane dielectric constants of bulk hexagonal BN are 8.7 and 4.1 at 10.6 µm, and 0.7 and 1.6 at 6.1 µm, respectively[2].

Figure 4c is from the result of this COMSOL simulation.

S3. Determine the effective radius of a nanotube bundle composed of N SWNTs.

The model described is for an individual nanotube. However a bundle consists of multiple nanotubes could also be considered with this model. As shown in the equation, an effective radius is needed. We used COMSOL to determine the effective radius of the bundle by calculating the total capacitance of the system.

We calculated the capacitance of closely packed N-tube bundles using finite element simulations, as shown in Fig. S1 (red circles). In the simulation, we assume each SWNT has a radius of R=0.8 nm, and the tubes are closely packed with a van der Waals separation of 0.35 nm. We also assumed that the nanotube bundle is screened by an outer metal shell of 300 nm radius. This change of capacitance in nanotube bundles containing N SWNTs can be well approximated by introducing an effective bundle radius of $\sqrt{N}R$ (black squares).

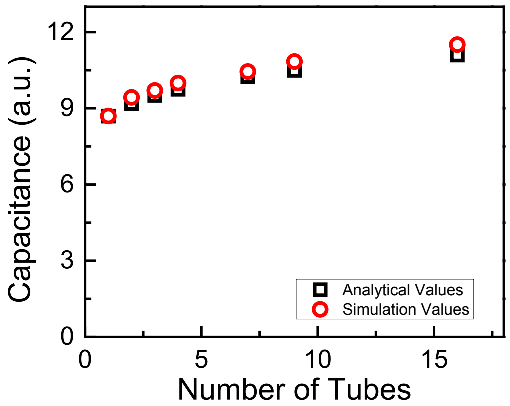

Fig. S1: The effective radius of a nanotube bundle. The red circles show capacitance of N-tube bundles as a function of N based on finite element simulation. The behavior can be well approximated by an effective radius of $\sqrt{N}R$ for the N-tube bundle (black squares). The discrepancy is smaller than 5%.

S4. Determination of individual single-walled carbon nanotube chirality

Optical transitions in individual single-walled carbon nanotubes are obtained by a recently developed polarization-based single tube spectroscopy[3]. Typically single-walled carbon nanotubes are characterized by one or two optical transitions. These transitions are compared with values in the previously established database, which tabulates all the transition energies for different chiralities in the visible spectral range[4]. Usually only one or two candidates are identified after this process. Finally we could use the diameter information (e.g. from sample preparation, Raman radial-breathing mode or AFM) to exclude the unlikely chiralities.

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