Weighing a single atom using a coupled plasmon–carbon nanotube system

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1. Introduction

Hybrid plasmonic nanostructures provide a promising strategy for the development of sensitive optical devices, via the interaction of the components of the hybrid system with the optical fields [1]. Surface plasmon can enhance the signals and amplify the field strengths in hybrid plasmonic nanostructures, which contributes to the new properties of nonlinear response and light propagation [2, 3]. Since 2008, we have investigated the light propagation in an artificial hybrid nanocrystal system, which consists of a metal nanoparticle and a semiconductor quantum dot (QD), driven by two optical excitations, i.e. the slow light effect and the nonlinear Kerr response enhancement [4, 5]. Recently, Fofang et al [6] and Cabrera-Granado et al [7] have used the same idea in the experiment and theory, respectively. Fofang et al reported the ultrafast optical dynamics of excitons in a system of strongly coupled excitons and plasmons, while Cabrera-Granado et al studied the slow-light performance of molecular aggregates arranged in nanofilms by means of coherent population oscillations. Both groups demonstrated that the surface plasmon enhances optical properties and results in a slow-light effect.

Nanomechanical systems (NMS) have also been actively studied for mass measurements, owing to their sensitivity to external perturbation [8–11]. NMS can use a cantilever, doubly clamped beam or carbon nanotube (CNT) as the sensing element, and the CNT is the best choice for single-atom detection because of its lower mass and higher quality factor. The mass of a CNT (∼10⁻²¹ kg) is typically at least four orders of magnitude lower than that of a state-of-the-art micromachined resonator (∼10⁻¹⁶ kg) [12, 13]; it changes significantly upon atomic adsorption, offering the possibility of weighing a single atom. Recently, CNT-based mass sensors have been realized experimentally using electrical detection. A spectral linewidth of 0.5–1 MHz has been achieved, which should allow the detection of 100–200 atoms [8, 12, 14]. In this article, we theoretically propose weighing a single atom with a hybrid CNT–plasmon system. We demonstrate that the single-atom detection using such a system is possible owing to the low CNT mass, spectral enhancement by the plasmon and the use of double optical excitation.

Although the techniques of ultralow-mass measurement are rather challenging, the basic principle remains simple [8, 12]. A harmonic oscillator model can be used to describe the motion of a doubly clamped CNT, having an effective mass \( m_\text{n} \), a spring constant \( k \) and a fundamental resonance frequency \( \omega_\text{0} = \sqrt{k/m_\text{n}} \) [12, 14]. The changes in the CNT mass \( \Delta m \) and resonance frequency \( \Delta \omega_\text{0} \) are related...
as follows:
\[
\Delta m = \left(\frac{m_n}{\omega_n}\right)\Delta \omega_n.
\] (1)

Thus, if one can detect the frequency shift of a resonator upon the addition of a particle, the mass of that particle can be calculated. Here it is assumed that \(\Delta m \ll m_n\), that is, the adsorbed mass is much smaller than the CNT mass. Equation (1) suggests that the detection of a single atom or a single molecule can be realized using a nanometer-sized CNT [12, 14].

2. Theory

In conventional mass sensing using electrical detection, the CNT must be suspended between two electrodes above a conducting plate, while a voltage is applied to them [12, 14, 15]. This method is limited by its frequency response and heating effects [16, 17]. The mass detection using double optical excitation is totally different. As shown in figure 1, in the presence of a strong pump field (frequency \(\omega_p\)) and a weak signal field (frequency \(\omega_s\)), a single gold metal nanoparticle (MNP) attached to the tip of a sharp optical fiber is positioned above the CNT. An atomic force microscope (AFM) is used to probe the tip and stabilize its distance to the CNT [18–20]. The MNP radius is \(a_0\) and \(R\) is the center-to-center distance between the MNP and CNT. In terms of electronic structure, a semiconducting CNT can be viewed as a graphene rolled into a cylinder. In the absence of a magnetic field, there is a single bright level \(E_{11}\) corresponding to the singlet direct exciton [21].

We envisage a suspended CNT where the center of exciton mass is localized via the spatial modulation of the Stark shift induced by a static inhomogeneous electric field. We analyze a tip electrode configuration that realizes an optically active nanotube quantum dot with an excitonic level spacing in the eV range; such spacing corresponds to a confinement length shorter than 10 nm. The quantum confinement is induced by the inhomogeneity in the field component along the CNT axis \(E_z\). In turn, the normal component \(E_{\perp}\) can be used to induce a tunable parametric coupling between the exciton and the flexural motion of the CNT [22]. A localized exciton with an effective size of about 2 nm is formed in a nanotube of 1.2 nm radius between its clamped ends [24–26], leading to a quantized energy spectrum in the longitudinal direction. In this case, the lowest-energy resonance of the clamped CNT corresponds to the fundamental flexural mode with the frequency \(\omega_n\), and the resonator is assumed to have a sufficiently high quality factor [22]. The eigenmode is described using a quantum harmonic oscillator with the Hamiltonian \(H_n = \hbar \omega_n a^\dagger a\), where \(a\) and \(a^\dagger\) are the annihilation and creation operators in the CNT, respectively.

The inset of figure 1 shows the excitonic energy structure in a CNT while dressing the plasmon modes and the vibration modes of CNT. This structure can be modeled as a two-level system consisting of the ground state \(|g\rangle\) and the first excited state (single exciton) \(|e_x\rangle\) and can be characterized using the pseudospin –1 operators \(\sigma^\pm\) and \(\sigma^z\). The Hamiltonian of this localized two-level exciton can be expressed as \(H_{ex} = \hbar \omega_{ex} \sigma^z\), where \(\omega_{ex}\) is the exciton frequency. Recently, the double optical excitation has become a hot topic, and experiments have been conducted to investigate the phenomena related to light propagation in mechanical-like systems, such as electromagnetically induced transparency, ultraslow light and stopped light [27–30]. The results suggested that dual optical excitation can produce novel effects, such as on-chip storage of light and ultrasensitive detection. In the simultaneous presence of a strong pump field and a weak signal field, the Hamiltonian of the total system can be written as [2, 31, 32]

\[
H = \hbar \omega_{ex} \sigma^z + \hbar \omega_0 a^\dagger a + \hbar \omega_s \eta \sigma^z (a^\dagger + a) - \mu (E_{QD} \sigma^+ + E_{QD}^* \sigma^-),
\] (2)

where \(\eta\) is the coupling strength between the CNT mode and exciton, \(\mu\) is the dipole matrix element of the exciton, and \(E_{QD}\) is the optical field acting on the localized exciton. In a frame rotating at the pump field frequency \(\omega_p\), the total Hamiltonian is given by

\[
H = \hbar \Delta_p \sigma^z + \hbar \omega_0 a^\dagger a + \hbar \omega_s \eta \sigma^z (a^\dagger + a) - \mu (\tilde{E}_{QD} \sigma^+ + \tilde{E}_{QD}^* \sigma^-),
\] (3)

where \(\Delta_p = \omega_{ex} - \omega_p\) and \(\delta = \omega_p - \omega_s\) are the frequency differences of the exciton and pump fields and pump and signal fields, respectively. \(\tilde{E}_{QD} = E_p + E_s e^{-i\delta t} + \frac{\omega_{ex}}{\omega_{ex}} E_{QD}^*\), \(\tilde{E}_{QD}^*\), \(\tilde{E}_{QD}^*\), \(\tilde{E}_{QD}^*\), and \(\tilde{E}_{QD}^*\) are the amplitudes of the background medium and exciton, respectively. \(E_p\) and \(E_s\) are the optical fields acting on the CNT and signal fields. The effective dielectric constants of the CNT and background medium are \(\epsilon_{eff} = \frac{2 \epsilon_{CNT}}{\epsilon_{CNT} + \epsilon_0}\), where \(\epsilon_{CNT}\) and \(\epsilon_0\) are the dielectric constants of the background medium and exciton, respectively. \(E_p\) and \(E_s\) are the optical fields acting on the CNT and signal fields.
field, respectively, and $S_p$ is the polar factor for electric field polarization; $S_e = 2$ when the polar direction is along the $z$-axis of the hybrid system. The dipole $P_{\text{MNP}}$ is related to the charge induced by the signal field. For a spherical particle (such as Au atom) with a radius much smaller than the wavelength of light, the electric field is uniform across the particle and the electrostatic (Rayleigh) approximation can be used. Then the $P_{\text{MNP}}$ is given by [2, 33]

$$P_{\text{MNP}} = \gamma a_0^3 \left( E_p + E_e e^{-i\omega t} + \frac{S_0 P_{\text{QD}}}{\epsilon_{\text{eff}} R^3} \right),$$

where

$$\gamma = \frac{\epsilon_{\text{Au}}(\omega) - \epsilon_0}{2\epsilon_0 + \epsilon_{\text{Au}}(\omega)},$$

$$\epsilon_{\text{eff}} = \frac{2\epsilon_0 + \epsilon_{\text{Au}}(\omega)}{3\epsilon_0},$$

$$\epsilon_{\text{Au}}(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma_p)}.$$  

Here $\epsilon_{\text{Au}}(\omega)$ is the dielectric constant of the Au nanoparticle, $P_{\text{QD}}$ is the total polarization of a localized exciton in the CNT, $\omega_p$ is the bulk metal plasma frequency, and $\gamma_p$ is the frequency-dependent damping. The imaginary part of $\epsilon_{\text{Au}}$ determines the metallic losses [34, 35]. The dipole moment of the localized exciton is expressed via the off-diagonal elements of the density matrix: $P_{\text{QD}} = \mu \sigma^-$ [36]. The dipole approximation used here is reasonable when the distance $R$ is large and the exciton-plasmon interaction is relatively weak [33]. Therefore, the total optical field $E_{\text{QD}}$ experienced by the localized QD is $E_{\text{QD}} = A(E_p + E_e e^{-i\omega t}) + \mu \sigma^-$, where $A = 1 + (\gamma a_0^3 S_p/\epsilon_{\text{eff}} R^3)$ and $B = \gamma a_0^3 S_p^2/\epsilon_{\text{eff}} R^6$.

Applying the Heisenberg equations of motion for operators $\sigma^+$, $\sigma^-$ and $q$, and introducing the corresponding damping and noise terms, we derive the quantum Langevin equations as follows [37, 38]:

$$\frac{d\sigma^+}{dt} = -\Gamma_1 (\sigma^+ + 1) + i\Omega (A^+ \sigma^- - A \sigma^+) + \frac{\mu^2}{\hbar} [\sigma^+ \sigma^- (B - B^*)] + \frac{i\mu}{\hbar} (A^+ E_e e^{-i\omega t} - A E_e e^{i\omega t}),$$

$$(8) \quad \frac{d\sigma^-}{dt} = -[\Gamma_2 + i(\Delta_p + \omega_n \eta_q)] \sigma^- - 2i\Omega A \sigma^+ - \frac{2i\mu}{\hbar} A E_e e^{-i\omega t} - \frac{2\mu^2}{\hbar} \sigma^+ + \hat{F}_e, \quad (9)$$

$$\frac{dq}{dt} + \gamma_n \frac{dq}{dt} + \omega_n^2 q = -2\eta_0 \sigma^+ \sigma^- + \eta \hat{\xi}.$$  

Here $q = a + a^*$; $\Omega = \mu E_p/\hbar$ is the Rabi frequency of the pump field; $\Gamma_1$ and $\Gamma_2$ denote the exciton relaxation rate and the exciton dephasing rate, respectively; $\gamma_n$ is the decay rate of the CNT resonator due to the coupling to a reservoir of ‘background’ modes and the other intrinsic processes [16, 22]. $\hat{F}_e$ is the $\delta$-correlated Langevin noise operator, which has zero mean $\langle \hat{F}_e \rangle = 0$ and the correlation relation $\langle \hat{F}_e(t) \hat{F}_e^*(t') \rangle \sim \delta(t-t')$. The operator $\xi$ signifies the Brownian stochastic force, since the motion of a CNT resonator is affected by the thermal bath of Brownian and non-Markovian processes [37, 39]. The quantum effects on the resonator are only observed in the limit of high quality factor, that is, $Q = \omega_n/\gamma_n \gg 1$. The Brownian noise operator can be modeled as Markovian with the decay rate $\gamma_n$ of the resonator mode. Therefore, the Brownian stochastic force has zero mean value $\langle \xi \rangle = 0$ and can be expressed as [39]

$$\langle \hat{\xi}(t) \hat{\xi}(t') \rangle \approx \frac{\gamma_n}{\omega_n} \int \frac{d\omega}{2\pi} \frac{e^{-i\omega(t-t')}}{1 + \coth \left( \frac{\hbar \omega}{2k_B T} \right)}.$$  

(11)

Following standard methods from quantum optics, we derive the steady-state solution of equations (8)–(10) by setting all the time derivatives to zero. They are given by

$$\langle \delta \sigma^+ \rangle = -\frac{\gamma_n^2}{\omega_n^3} \left[ \langle \hat{F}_e \rangle - \frac{\gamma_n^2}{\omega_n^2} \right] \quad \text{and} \quad \langle \delta \sigma^- \rangle = \frac{2\mu^2}{\hbar} \langle A E_e e^{-i\omega t} \rangle,$$

(12)

$$\langle \delta \sigma^+ \rangle = -\frac{\gamma_n^2}{\omega_n^3} \left[ \langle \hat{F}_e \rangle - \frac{\gamma_n^2}{\omega_n^2} \right] \quad \text{and} \quad \langle \delta \sigma^- \rangle = \frac{2\mu^2}{\hbar} \langle A E_e e^{-i\omega t} \rangle,$$

(13)

$$\langle \delta q \rangle = \frac{\mu^2}{\hbar} \langle A E_e e^{-i\omega t} \rangle,$$

(14)

$$\langle \delta q \rangle = -\frac{\gamma_n^2}{\omega_n^3} \left[ \langle \hat{F}_e \rangle - \frac{\gamma_n^2}{\omega_n^2} \right] + \frac{2i\mu}{\hbar} \langle A E_e e^{-i\omega t} \rangle,$$

(15)

We further make the ansatz [40]:

$$\langle \delta \sigma^+ \rangle = \sigma_s e^{-i\omega t} + \sigma_{-} e^{i\omega t}, \quad \langle \delta \sigma^- \rangle = \sigma_s e^+ e^{i\omega t} + \sigma_{-} e^{-i\omega t}, \quad \langle \delta q \rangle = q_s e^{-i\omega t} + q_{-} e^{i\omega t}.$$  

Upon substituting this approximation into equations (13)–(15) and considering only the lowest order in $E_p$, but all the orders in $F_p$, we finally obtain the linear optical susceptibility in the steady state as follows:

$$\chi_{\text{eff}}^{(1)}(\omega) = \frac{\mu}{\epsilon_0 E_p} \sigma_s = \frac{\mu^2}{\epsilon_0^2 \hbar \Omega^2} \chi^{(1)}(\omega) = \Sigma \chi^{(1)}(\omega),$$

(16)
Here $\Sigma = \mu^2/(\epsilon_0 h \Gamma_2)$, and the dimensionless linear susceptibility is given by

$$\chi^{(1)}(\omega_n) = \frac{2 A^2 d w_0 - A \phi c w_0}{c^2 \phi - c \delta_0 - 2 d^2 A^2 - 4 i d B_{R0} A w_0},$$

where

$$c = \Delta_\rho 0 - \omega_{00} \eta^2 w_0 + B_{R0} w_0 - i(1 - B_{R0} w_0),$$

$$d = \Delta_\rho 0 - \omega_{00} \eta^2 w_0 + B_{R0} w_0 + i(1 - B_{R0} w_0),$$

$$\phi(\delta_0) = \left[ 2 \Omega_0^2 (c - 2 i B_{R0} w_0) (\omega_{00} \eta^2 w_0 \zeta + A d - B^2_{R0} A w_0) \right]^{1/2}$$

$$\zeta (\omega_n) = \frac{\omega_{00}^2}{\omega_{00}^2 - \delta_0^2 - i \gamma_{00} \delta_0},$$

where $w_0 = 2 \omega_{00}^2 \delta_0$, $\delta_0 = 2i \Delta_\rho / \Gamma_2$, $\Omega_0 = \Omega / \Gamma_2$, $\omega_{00} = \omega_{00} / \Gamma_2$, $\Delta_\rho 0 = \Delta_\rho / \Gamma_2$, $\Gamma_1 0 = \Gamma_1 / \Gamma_2$, $B_0 = \mu^2 B / (h \Gamma_2)$, $B_{R0} = \text{Re}(B_0)$ and $B_{R0} = \text{Im}(B_0)$. From equation (17), we can see that the $R$-dependence of the linear optical susceptibility originates from the $R$-dependent expressions of $A$ and $B$. In this case, the center-to-center distance $R$ will not change $E_p$ or $E_n$, but it will alter the exciton–plasmon interaction while the Au cluster on the AFM tip approaches the CNT.

The population inversion $(w_0)$ of the localized exciton is determined using the equation

$$(w_0 + 1) [(1 - B_{R0} w_0) - (\Delta_\rho 0 - \omega_{00} \eta^2 w_0 + B_{R0} w_0)^2] + 2 \Omega_0^2 A^2 w_0 = 0.$$  

3. Results and discussion

For a clear demonstration of the single-atom detection, we choose a realistic CNT resonator coupled to an Au nanoparticle and the following set of parameters: $(\Gamma_1, \Gamma_2, \eta, a_0, R, \mu, \epsilon_0, \epsilon_n, \gamma_{00}) = (0.6 \text{ GHz}, 0.3 \text{ GHz}, 0.17, 2.5 \text{ nm}, 18 \text{ nm}, 40 \text{ D, 1, 6, 10/60})$ [2, 22, 33, 34]. The effective mass of a CNT $m_n = 1580 \text{ zg}$ ($1 \text{ zg} = 10^{-21} \text{ g}$), which corresponds to the vibrational frequency $\omega_n = 725 \text{ MHz}$, the quality factor $Q = 1000$, and the decay rate $\gamma_n = \omega_n / Q = 8 \times 10^4 \text{ Hz}$ [41].

Figure 1 shows our proposed setup of the plasmon-enhanced mass sensing in the presence of a strong pump laser and a weak signal laser. The two optical fields are applied at the center of the nanotube resonator, which is the position of the localized exciton. Detailed information on the double optical excitations can be found in [27–30].

According to equation (16), we first radiate a strong pump beam on the hybrid MNP–CNT system, and then detect the absorption spectrum of the second beam as a function of signal-exciton detuning $\Delta_s$, with $\Delta_\rho = 0$ (figure 2(a)). The red curve in figure 2(a) contains two peaks at both sides of the resonance, which correspond to the vibrational frequency of CNT. Particularly, for the vibrational frequency $\omega_n = 725 \text{ MHz}$, the spikes will appear at $\pm 725 \text{ MHz}$. These two peaks represent the resonance amplification and absorption of the vibrational mode of the CNT, respectively, and this behavior is different from that of a traditional atomic system [40]. When dressing with the vibrational modes of a CNT (vibrational modes are treated as phonon modes), the original energy levels of exciton $(|ex\rangle$ and $|g\rangle$) split into dressed states $|ex, n\rangle$ and $|g, n\rangle$, where $|n\rangle$ denotes the number states of the nanotube resonance mode. The left amplified peak signifies the transition from the lowest dressed level $|g, n\rangle$ to the highest dressed level $|ex, n + 1\rangle$ by the simultaneous absorption of two pump photons and emission of an $\omega_p - \omega_n$ photon. This process can amplify a wave at $-\omega_n$. The central peak corresponds to the vibration-induced stimulated Rayleigh resonance due to a transition from the lowest dressed level $|g, n\rangle$ to the dressed level $|ex, n\rangle$. The right absorption peak shows a usual resonance modified by the AC-Stark effect.

**Figure 2.** (a) Absorption signal as a function of $\Delta_s$. The parameters used are $\Delta_\rho = 0$, $\gamma_n = 8 \times 10^5 \text{ Hz}$, $\eta = 0.17$, $\omega_n = 725 \text{ MHz}$ and $\Omega^2 = 0.01 \text{ (GHz)}^2$. The insets show the corresponding transitions and energy levels. (b) Enlarged view of the right peak when the surface plasmon is involved or not in the absorption. The signal bandwidth is about 75 kHz when the distance between the Au MNP and CNT is 18 nm.
Figure 2(b) shows an enlarged view of the right peak when plasmon is involved or not in the absorption. It is obvious that the plasmon contribution sharpens and enhances the signal, thereby increasing the sensitivity of mass detection. The signal bandwidth can be optimized by varying the distance $R$ between the Au MNP and the CNT resonator. The red curve in figure 2(b) corresponds to a bandwidth of $\sim 75$ kHz, demonstrating that the proposed mass sensor has a sufficient spectral resolution for the single-atom detection.

The next step is tracking the frequency shift of the CNT resonance before and after injecting the species to be detected. While landing on the CNT, these species will change its total mass and vibrational frequency. On the basis of the relationship $\Delta m = (2m_n \Delta \omega_0)/\omega_0$, by measuring the frequency of the CNT resonator before and after landing, we can determine the mass of the deposited nanoparticle. In traditional mass spectrometry, the analyzed atoms have to be ionized in the first instance and their charge needs to be known before the mass extraction. Difficulties in ionizing some atoms results in the uncertainty of the mass value report. Such process is unnecessary in the all-optical mass sensing. When a single atom is deposited on the CNT surface, the spectral peaks representing the resonant frequency of CNT shift according to the equation $\Delta m = (2m_n \Delta \omega_0)/\omega_0$. Following Chin et al [14], we select Xe as an example. For a single Xe atom, which has the mass $\Delta m = 0.22 \text{ zg}$, the red curve in figure 3 shifts by $\Delta \omega_0 = 23$ kHz. The inset in figure 3 shows a linear relationship between the frequency shifts and the number of atoms landing on the resonator, revealing that the single-atom weighing scheme is also suitable for the detection of atomic clusters. The slope gives the mass responsivity [12] of the resonator $\Re = (2m_n/\omega_0)^{-1} = 2.3 \times 10^{-28}$ Hz $\cdot$ g$^{-1}$, which is five orders of magnitude higher than that in conventional mass measurement using electrical detection. It should be noted that the proposed technique can also be applied to weighing chemically active atoms chemisorbing on the CNT since the deposited atoms only change the vibrational frequency of the CNT via equation eq. (1). A localized exciton with an effective size of about 2 nm radius [24–26] of the resonator is produced by a static inhomogeneous electric field. Here the size and electronic structure of the confined exciton are determined using the external voltage. The chemically active atom is smaller than 1 nm, whereas the CNT length is about 120 nm [22]. Therefore, the perturbation of the electronic state of the CNT while adsorption of a chemically active atom is hard to detect if the inhomogeneous electric field is modulated and the adsorbed site is far away from the location of the exciton in the CNT. In this case, the hybridization of electronic states of the CNT induced by chemisorbing active atoms can be neglected. Furthermore, the gold MNP used in our experiment has a radius of only 2.5 nm and is positioned right above the localized exciton. Therefore, adsorption of chemically active atoms far away from the localized exciton and the gold MNP should not affect the plasmons.

The mass sensing using a CNT as an intermediary has high sensitivity owing to the low mass and high quality factor of the CNT. In our theoretical calculation, noise processes are neglected, such as the thermomechanical noise generated by the internal loss mechanisms in the resonator and the adsorption–desorption noise from residual gas molecules in the resonator packaging, which will limit the mass sensing performance of the CNT [14]. Between these two noise types, the thermal noise of the mechanical motion of the CNT is the dominant component. It can be reduced by carrying out the mass measurement at low temperatures in vacuum, e.g. 5 K and 1 Torr [12, 14]. Meanwhile, the mass resolution limits imposed by the thermomechanical noise can be given using the expression $\Delta m \sim (m_n/Q) \times 10^{-3\text{DR}/20}$, where DR represents the dynamic range, which demonstrates that small effective mass and high quality factor $Q$ of the resonator are crucial for improving the mass resolution [42].

4. Conclusions

We have proposed an all-optical technique of weighing a single atom using a hybrid system of plasmon and a CNT resonator, driven by two optical fields. The mass of atoms attached to the CNT surface can be measured conveniently and precisely via the frequency shift in the absorption spectrum. Heating and energy loss characteristic for electrical detection can be avoided in an all-optical technique. Particularly, the spectral linewidth can be on the order of kilohertz, which is approximately five orders of magnitude less than that in conventional electrical measurements, allowing single atom to be weighed.

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