Supplementary Material for Detection of single nanoparticles using the dissipative interaction in a high-Q microcavity

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I. Gold nanorod induced cavity loss and mode shift

The linewidth of a cavity represent the cavity energy loss, $\gamma/\omega = \delta U/U$, with U the total energy in the cavity, ω the angular frequency, δU the energy loss in the cavity, and γ stands for the total linewidth. Therefore, $\gamma = P/U$, where P is the total loss power. For a gold nanorod, both of the scattering loss power P_s and absorption loss power P_a contribute to the total loss, $P = P_0 + P_s + P_a$, where P_0 is the intrinsic cavity loss power.

Under dipole approximation, the scattering loss power can be expressed as $P_s = \mu_0\omega^4|p|^2/12\pi v$, where μ_0 is the permeability of free space, $p = \varepsilon_0\varepsilon_m\alpha E$ stands for the dipole momentum, and v is the speed of light in the medium. The absorption loss power can be written as $P_a = \varepsilon_0\omega\mathrm{Im}[\varepsilon]|\beta|^2|E|^2V_p/2$, where ε_0 is the permittivity of free space, ε represents the particle permittivity, β denotes the root-mean-square enhancement of the electric field inside the nanorod, and V_p is the particle volume. The total energy can be calculated by $U = \varepsilon_0\varepsilon_c|E_{\max}|^2V_c/2$, where ε_c is the cavity permittivity, $E_{\max}$ denotes the maximum electric field intensity in the cavity and $E = f(\vec{r})E_{\max}$, and V_c stands for the cavity mode volume. Thus, the gold nanorod induced loss is written as $\gamma = \gamma_s + \gamma_a$, where γ_s and γ_a are scattering induced loss and absorption induced loss respectively, and can be expressed as

$$\gamma_s = \frac{\varepsilon_m^{5/2}\,\omega_c^4 f^2(\vec{r})|\alpha|^2}{6\pi c^3\varepsilon_c V_c}$$

and

$$\gamma_a = \frac{|\beta|^2 f^2(\vec{r})V_p\omega_c\mathrm{Im}[\varepsilon]}{\varepsilon_c V_c},$$

where ε_m is the medium permittivity, ω_c represents the angular frequency of cavity mode, $f(\vec{r})$ denotes the cavity mode function and α is the particle polarizability.

Under perturbation theory [1], the backscattering coupling strength, contributing to the mode shift, can be written as

$$g = -\frac{\mathrm{Re}[\alpha]\omega\varepsilon_m f^2(\vec{r})}{2\varepsilon_c V_c}.$$

II. Wavelength depended nanorod polarizability

Since the polarizability of a nanorod shape does not have an analytical formula, to better understand the physics inside, we study a gold ellipsoid nanoparticle here instead, which have very similar property comparing with the nanorod. In Fig. S1, we plot wavelength dependence of mode shift induced by a nanorod and an ellipsoid with the same volume. The cavity response, i.e., the mode shift, of those two kind of nanoparticles is almost the same, which verifies the feasibility of our treatment.

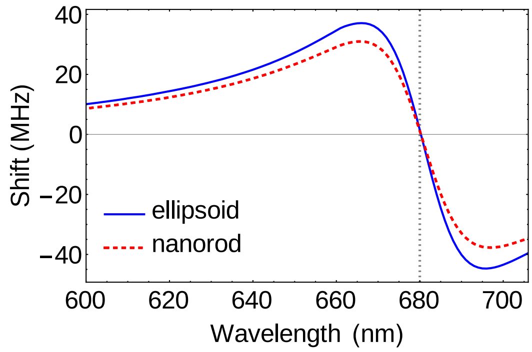

Fig. S1. Comparison of the mode shift induced by a gold ellipsoid (blue solid curve) and a gold nanorod (red dashed curve) with the same volume.

For an ellipsoid nanoparticle, the polarizability can be written as

$$\alpha = \frac{\varepsilon - \varepsilon_m}{\varepsilon_m + (\varepsilon - \varepsilon_m)L} V_p,$$

where ε and ε_m are the permittivity of the particle and the medium, respectively. The parameter L $(0 < L < 1)$ corresponds to the shape of the ellipsoid (from prolate spheroid $(L < 1/3)$ to sphere $(L = 1/3)$, then to oblate spheroid $(L > 1/3)$). The parameter L is set as 0.113 to make the SPR wavelength lay at 680 nm (Fig. S1).

Here, we plot $\mathrm{Re}[\alpha]/V_p$ vs. $\mathrm{Re}[\varepsilon]/\varepsilon_m$ and $\mathrm{Im}[\varepsilon]/\varepsilon_m$ in Fig. S2(a). The red dashed curve marks the case of $\mathrm{Re}[\alpha] = 0$. The blue dash-dotted line marks the SPR condition of $\mathrm{Re}[\varepsilon_m + (\varepsilon - \varepsilon_m)L] = 0$, corresponding to $\mathrm{Re}[\varepsilon/\varepsilon_m] = 1 - 1/L$. It is noted that $\mathrm{Re}[\alpha]$ does not have to be zero at the SPR condition, see the dashed-dotted line and the dashed curve in Fig. S2(a). In our experiment, however, the gold material has the permittivity shown in the solid white curve (at wavelength 400 ~ 800 nm from right to left) in Fig. S2(a). At the SPR wavelength, ~680 nm, $\mathrm{Re}[\alpha]$ approaches to zero. We extract data of $\mathrm{Re}[\alpha]$ on the white curve and plot it in Fig. S2 (b), which is very similar to the trend of the mode shift in Fig. 5. When $\mathrm{Re}[\alpha]=0$, the dipole moment and the electric field have a $\pi/2$ phase difference, so the coupling energy between the particle and the cavity is zero, resulting in zero mode shift.

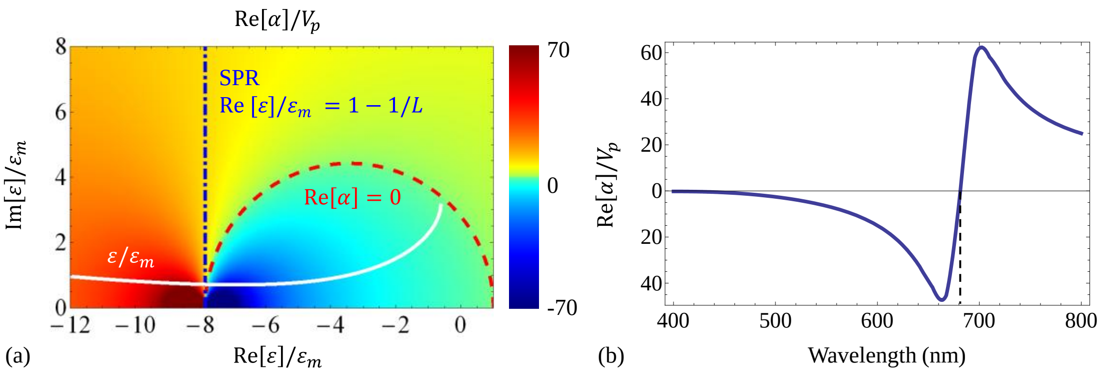

Fig. S2. (a) $\mathrm{Re}[\alpha]/V_p$ as a function of $\mathrm{Re}[\varepsilon]/\varepsilon_m$ and $\mathrm{Im}[\varepsilon]/\varepsilon_m$ for an ellipsoid nanoparticle. (b) $\mathrm{Re}[\alpha]/V_p$ as a function of wavelength when the particle material is gold.

Additionally, if one treats the SPR as an antenna (2-level system), a same behavior will be obtained [2].

The table below summarizes the experimental and theoretical mode shift and linewidth change at 680 nm and 635 nm respectively.

Table S1. Comparison of experimental results and theoretical predictions.

	Mode shift (MHz)		Linewidth change (MHz)	
Probe wavelength	680 nm	635 nm	680 nm	635 nm
Experimental average	6.5	11.2	13.5	10.2
Theoretical prediction	1.1	16.8	132.6	17.2

III. Three-dimensional finite-element-method simulation

We use a similar method as described in Ref. [3], and only simulate a half wavelength slice of the cavity using COMSOL 4.3a RF module. The 15 nm × 42 nm gold nanorod is oriented at the axial direction of the cavity and is placed 2 nm away from the microtoroid (major: 100 μm, minor: 6 μm) surface (Fig. S3(a)). We only use finer mesh near the nanorod and the cavity mode in order to save the resource and time. The refractive index of the gold nanorod is from Ref. [4]. The field distributions of the TE fundamental mode in the cavity and in the nanorod at 680 nm, which is on SPR, are shown in Fig. S3(b)-(d). The field enhancement factor β inside the nanorod ranges from about 5 to 20, and the average enhancement is about 13.

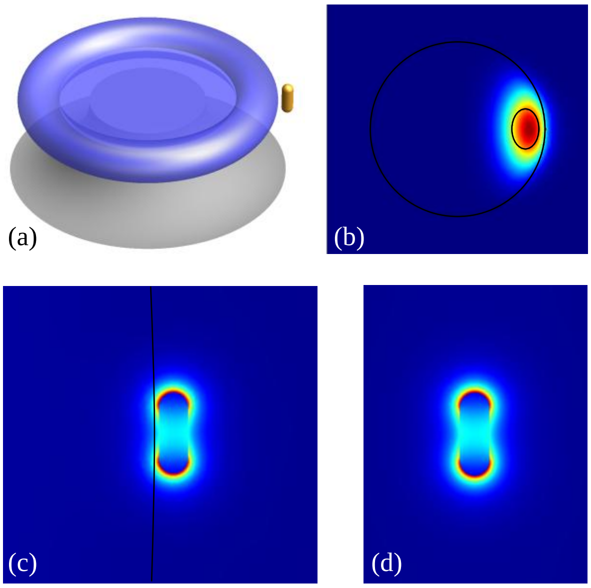

Fig. S3. 3-D FEM simulation using COMSOL. (a) Schematic setup, nanorod is at axial direction and with a 2 nm gap from the cavity surface. (b) TE fundamental mode of the cavity, the black circle represents the cavity boundary, the black ellipse inside is for finer meshing at the mode area. (c) and (d) are nanorod's electric field distribution in the azimuthal plane and the radial plane, respectively.

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