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Room temperature nonlinear mass sensing based on a hybrid spin-nanoresonator system*

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We present a room temperature nonlinear mass sensing based on a hybrid spin-nanoresonator system with the microwave pump–probe technique and the spin readout technique, which includes a single spin of nitrogen–vacancy (NV) center in diamond and a nanomechanical cantilever. The resonance frequency of the nanoresonator can be measured with the nolinear Kerr spectrum, and the parameters that influence the nolinear Kerr spectrum are also investigated. Further, according to the relationship between frequency shifts and variable mass attached on the nanoresonator, this system can also be used to detect the mass of DNA molecules with the nolinear Kerr spectrum. Benefiting from the single spin of the NV center in diamond has a long coherence time at 300 K, the hybrid system can realize room temperature mass sensor, and the mass response rate can reach 2600 zg/Hz.

Keywords: nonlinear mass sensing, nitrogen–vacancy center, nanomechanical resonator

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1. Introduction

In recent years, the research of nanomechanical resonators (NRs) to form hybrid quantum systems has been very active in various fields. Due to their unique properties of lower effective masses, higher resonant frequencies, and quality factors, NRs systems are widely used in quantum information processing[1–6] and nanoscale sensing[7–10] in biological and chemical contents. One of the widespread applications is the sensitive detectors, such as force and mass detecting via NRs including carbon nanotube resonators,[11] monolayer MoS_2 resonators,[12] and graphene nanoribbons.[13] However, mass sensors based on the NRs systems should be worked in a cryogenic environment. In 2008, room-temperature high quality factors NRs with frequencies about 1 MHz have been demonstrated.[14] Recently, a novel design of on-chip mechanical resonators can also exhibit fundamental modes with high frequency f and large quality factor Q at room temperature, and these resonators own low mechanical dissipation and large reflectivity.[15] Actually, the features of different NRs strongly depend on the material and fabrication technique,[16] which presents a possible experimental platform for observing massive quantum behaviors at room temperature.

Although the sensitivity of mass sensing using NRs has been achieved with a high mass response rate, most of these experiments have limited to cryogenic temperatures. In the past few decades, the interaction between a single electron spin and an NR has already been extensively investigated. The single electron spin possesses very long decoherence time even at room temperature and NRs also provide a suitable platform for detecting and controlling the states of single electron spin. Additionally, a mechanical resonator coupled to a single electronic spin has been demonstrated in different hybrid systems.[17–21] In the hybrid resonator systems, the single spin of an NV center in diamond is very promising, because it has a long coherence time at 300 K.[22,23] In combination with pump–probe scheme and spin readout techniques,[24] we propose a room temperature nonlinear mass sensing based on a hybrid spin-nanoresonator system.

As we know, there are three steps in classical mass spectrometry: ionization, separation, and detection.[25–27] However, if the particles cannot be ionized, they will not be measured in this way, and then an electric scheme for detecting the mass of particles was proposed. In the electric scheme, NRs systems were introduced and the voltage is applied to both ends of the NR, then the vibrator is broken under the excitation of current. When the measured particles land on the surface of the vibrator, the change of its natural vibration frequency is obtained by measuring the change of young's modulus of the nanomechanical vibrator. It is convenient to calculate the mass of the particles attached to the resonator according to formula (13) (in the following context). But the electric method will produce heating effects and energy loss in the process of measurement if the current into the NR is too strong.[28,29] In order to overcome the difficulties, we propose a scheme of nonlinear mass sensing in microwave domain by means of the spin–oscillator coupling system. A single electronic spin of an NV center in diamond can be polarized and detected, which exhibits outstanding coherence prop-

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erties at room temperature.[30,31] The degree of freedom of electron spin can provide an excellent quantum energy level structure.[32]

In this paper, we consider a hybrid quantum system where the electric spin associated with an NV impurity in diamond and an NR with a magnetic tip on the free end as a mass sensor, and theoretically propose a realizable way to measure the resonance frequency of the NR by the coupling system.[33–38] In the presence of a strong pump field and a weak probe field, the dynamic strain-mediated coupling between a single electronic spin of an NV center and a vibrational nanomechanical cantilever is analogous to the coupling of cavity and mechanical oscillator in optomechanical systems, the NV spin in magnetic field plays the role of the optical cavity.[39] This coupling system can exhibit excellent coherence properties even at room temperature.[40,41] In combination with a pump–probe technique, we put forward a nonlinear scheme to detect the mass in nanoscale at room temperature via a strong magnetic coupling between an electric spin qubit and a mechanical resonator.

2. Model and theory

The physical model is shown in Fig. 1, where the hybrid system is composed of an NR with a magnetic tip at the free end and a single NV center. The coupling of a single electronic spin associated with an NV center and a magnetized mechanical resonator has been demonstrated in an experiment.[19] In our scheme, the magnetized cantilever of dimensions $(l,w,t)\approx(25,0.1,0.1)$ μm and frequency $\omega_{\rm n}/2\pi\sim$ 1 MHz is separated from the NV center by a distance $h\approx$ 10 nm. The strong magnetic coupling can be achieved via the electron spin and the motion of the NR.[20] In the simultaneous presence of a stronger pump field and a weak probe field, the magnetic field along the z axis is illustrated as well. The motion of the NR tip is along the z axis, and the quantized Hamiltonian can be expressed as[42] $H_{\rm n}=\hbar\omega_{\rm n}a^{\dagger}a$, where $\omega_{\rm n}$ is the intrinsic vibration frequency of the NR, $a^{\dagger}$ and a are the corresponding creation and annihilation operators, respectively.

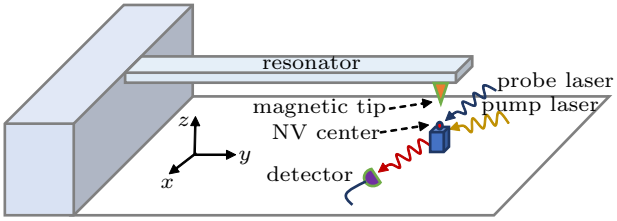


Fig. 1. Schematic diagram of a spin–oscillator system of a magnetized clamped-free NR coupled to the electronic spin associated with an NV center in diamond in the presence of a strong pump laser and a weak probe laser.

For a spin in the NV center of the diamond, we can take $|\uparrow\rangle=|m_{\rm s}=1\rangle$ and $|\downarrow\rangle=|m_{\rm s}=0\rangle$ as the spin-1 ground state of the NV central and ignore $|m_{\rm s}=-1\rangle$ state because it is far away from the other two states after adding the external fixed magnetic field.[18] Therefore, the electron spin in a single NV center can be regarded as an $S=1/2$ electron spin caused by an external magnetic field with a splitting frequency up to $\omega_{\rm e}\sim$ GHz. The Hamiltonian of the electronic spin qubit is $H_{\rm s}=\hbar\omega_{\rm e}S^{z}$, where S^{z} is the z component of the electronic spin operator, and another two operators S^{+} and S^{-} with the commutation relation $[S^{z},S^{\pm}]=\pm S^{\pm}$ and $[S^{+},S^{-}]=2S^{z}$ describing the electronic spin are also introduced. Here, the electron spin in a single NV center is analogous to a two level quantum dot (QD) at low temperature. Benefiting from advances in modern nanoscience and nanotechnology, QDs for their applications in photonics and optoelectronics have attracted tremendous attention. QD, as a simple stationary atom with the tunability of optical properties[43,44] exhibiting abundant physical phenomena of quantum confined systems, are attractive for they present optical responses in a widely tuned spectral range, which paves the way for numerous potential applications,[45] such as single photon sources,[46,47] quantum dot qubit,[48,49] and QD mechanical resonator.[50]

The magnetic field generated by the magnetic tip of the NR is $|\boldsymbol{B}_{\rm tip}|\simeq G_{\rm m}\hat{z}$, where $G_{\rm m}$ is the magnetic field gradient and $\hat{z}=a_0(a^{\dagger}+a)$ is the position operator, the magnetic field is proportional to the position operator. So the interaction between electron spin and NR can be expressed by the Hamiltonian[20,51] $H_{\rm s-m}=\hbar g_0(a^{\dagger}+a)S^{z}$ and $g_0=a_0g_{\rm s}\mu_{\rm B}G_{\rm m}/\hbar$ is the coupling strength, where $a_0=\sqrt{\hbar/2m\omega_{\rm n}}$ is the amplitude of zero-point fluctuations with resonator mass m, $g_{\rm s}\simeq 2$ is the Lande g factor, and $\mu_{\rm B}$ is the Bohr magneton. As the strong pump microwave field and the weak probe microwave field act on simultaneously the spin–oscillator coupling system, the electron spin in the system interacts with them through spin flipping.

We use the classical method to deal with the microwave fields, then the Hamiltonian of coupling the electronic spin with the pump field and the probe field can be written as[52,53] $H_{\rm s-p}=-\mu E_{\rm pu}(S^{+}{\rm e}^{-{\rm i}\omega_{\rm pu}t}+S^{-}{\rm e}^{{\rm i}\omega_{\rm pu}t})-\mu E_{\rm pr}(S^{+}{\rm e}^{-i\omega_{\rm pr}t}+S^{-}{\rm e}^{{\rm i}\omega_{\rm pr}t})$, where μ is the electric dipole moment of the electronic spin, $E_{\rm pu}(E_{\rm pr})$ is the intensity of microwave field, and $\omega_{\rm pu}(\omega_{\rm pr})$ refers to the frequency of the pump field (probe field).

In the rotation coordinate transform with pump field frequency $\omega_{\rm pu}$, we can obtain the whole Hamiltonian of the system as[54]

$$\begin{aligned}H\;=\;&\hbar\Delta_{\rm pu}S^{z}+\hbar\omega_{\rm n}a^{\dagger}a+\hbar g_0(a^{\dagger}+a)S^{z}\\&-\hbar\Omega(S^{+}+S^{-})-\mu E_{\rm pr}(S^{+}{\rm e}^{-{\rm i}\delta t}+S^{-}{\rm e}^{{\rm i}\delta t}),\end{aligned}\quad(1)$$

where $\Delta_{\rm pu}=\omega_{\rm e}-\omega_{\rm pu}$ is the detuning between the electron spin and the pump field, and $\delta=\omega_{\rm pr}-\omega_{\rm pu}$ is the probe–pump detuning. For simplicity of calculations, we change the intensity of pump field $E_{\rm pu}$ into the Rabi frequency of the pump

field $\Omega = \mu E_{\rm pu}/\hbar$. The following equations can be obtained by solving the above Hamiltonian with the Heisenberg motion equations[55]

$$\frac{{\rm d}S^z}{{\rm d}t} = -\Gamma_1(S^z+\frac{1}{2})+{\rm i}\Omega(S^+-S^-) + \frac{{\rm i}\mu E_{\rm pr}}{\hbar}(S^+{\rm e}^{-{\rm i}\delta t}-S^-{\rm e}^{{\rm i}\delta t}), \quad (2)$$

$$\frac{{\rm d}S^-}{{\rm d}t} = -[\Gamma_2+{\rm i}(\Delta_{\rm pu}+g_0N)]S^- - 2{\rm i}\Omega S^z - \frac{2{\rm i}\mu E_{\rm pr}}{\hbar}S^z{\rm e}^{-{\rm i}\delta t}, \quad (3)$$

$$\frac{{\rm d}^2N}{{\rm d}t^2}+\gamma_{\rm n}\frac{{\rm d}N}{{\rm d}t}+\omega_{\rm n}^2N = -2\omega_{\rm n}g_0S^z, \quad (4)$$

where Γ_1 (Γ_2) is the relaxation rate (decoherence rate) of the two-level electron spin in the NV center, $\gamma_{\rm n}$ represents the decay rate of NR. For simplicity of process of derivation, we describe the resonator amplitude $N = a^\dagger + a$.

In order to solve Eqs. (2)–(4), we make the ansatz,[44]

$$S^z = S_0^z + S_1^z{\rm e}^{-{\rm i}\delta t}+S_{-1}^z{\rm e}^{{\rm i}\delta t}, \quad (5)$$

$$S^- = S_0 + S_1{\rm e}^{-{\rm i}\delta t}+S_{-1}{\rm e}^{{\rm i}\delta t}, \quad (6)$$

$$N = N_0 + N_1{\rm e}^{-{\rm i}\delta t}+N_{-1}{\rm e}^{{\rm i}\delta t}. \quad (7)$$

Substituting Eqs. (5)–(7) into Eqs. (2)–(4) and upon working to the lowest order in $E_{\rm pr}$, but to all orders in $E_{\rm pu}$, we can obtain S_1 and S_{-1}, which correspond to the linear and nonlinear susceptibilities, respectively. Here, in this work, we are more interested in the nonlinear susceptibility, and we can obtain the nonlinear susceptibility as $\chi_{\rm eff}^{(3)}(\omega_{\rm pr}) = \mu S^-(\omega_{\rm pr})/(3E_{\rm pu}^2E_{\rm pr}) = \Sigma_3\chi^{(3)}(\omega_{\rm pr})$, where $\Sigma_3 = \mu^4/(3\hbar^3\Gamma_2^3)$, and $\chi^{(3)}(\omega_{\rm pr})$ is given by

$$\chi^{(3)}(\omega_{\rm pr}) = \frac{-[\omega_0\Omega+{\rm i}S_0(\Gamma_2-{\rm i}\Delta_2)]\Pi_1\Gamma_2^3}{\{(\Gamma_2+{\rm i}\Delta_1)[(\Gamma_1+{\rm i}\delta)(\Gamma_2-{\rm i}\Delta_2)-{\rm i}\Omega\Pi_2]+{\rm i}\Omega\Pi_1(\Gamma_2-{\rm i}\Delta_2)\}\Omega^2}, \quad (8)$$

where

$$N_0 = -{\rm g}_0\omega_0/\omega_{\rm n},$$
$$\Delta_1 = \Delta_{\rm pu}+\delta+g_0N_0, \quad \Delta_2 = \Delta_{\rm pu}-\delta+g_0N_0,$$
$$\Pi_1 = -{\rm i}(g_0S_0X^*+2\Omega), \quad \Pi_2 = {\rm i}(g_0S_0^*X^*+2\Omega),$$
$$S_0 = \frac{\Omega\omega_0}{g_0^2\omega_0/\omega_{\rm n}-\Delta_{\rm pu}+{\rm i}\Gamma_2}, \quad X = -\frac{2\omega_{\rm n}g_0S_1^z}{\omega_{\rm n}^2-\delta^2+{\rm i}\delta\gamma_{\rm n}},$$
$$S_0^* = \frac{\Omega\omega_0}{g_0^2\omega_0/\omega_{\rm n}-\Delta_{\rm c}-{\rm i}\Gamma_2}.$$

The imaginary and real parts of $\chi^{(3)}(\omega_{\rm pr})$ indicate the nonlinear absorption and the Kerr coefficient, respectively.

In addition, in Eqs. (5)–(7), S_0^z, S_0, and N_0 are the steady-state values, which have the following relationship:

$$\Gamma_1(S_0^z+1/2)-{\rm i}\Omega(S_0^*-S_0) = 0, \quad (9)$$

$$({\rm i}\Delta_{\rm pu}+\Gamma_2)S_0^z+{\rm i}g_0N_0S_0+2{\rm i}\Omega S_0^z = 0, \quad (10)$$

$$\omega_{\rm n}^2N_0+2\omega_{\rm n}g_0S_0^z = 0. \quad (11)$$

They together determine the population inversion ($\omega_0 = 2S_0^z$) of the spin, and we can find that ω_0 is determined by the following equation:

$$\Gamma_1(\omega_0+1)\left[\Gamma_2^2+\left(\Delta_{\rm pu}-g_0^2\frac{\omega_0}{\omega_{\rm n}}\right)^2\right]+4\Omega^2\omega_0\Gamma_2 = 0. \quad (12)$$

The basic principle of mass sensing is to detect the frequency shift of oscillators caused by additional particles attached to the NR. The NR can be described by a simple harmonic oscillator model with an effective mass M, a spring constant k, and a basic resonance frequency $\omega_{\rm n} = \sqrt{k/M}$. Additional particles landing on the surface of the NR will increase the effective mass of the resonator, which will decrease the resonance frequency at the same time. Mass measurement is based on monitoring the change (the frequency shift $\Delta\omega_{\rm n}$) of the resonance frequency $\omega_{\rm n}$. The relationship of the frequency shift $\Delta\omega_{\rm n}$ and the additional mass Δm can be described as

$$\Delta m = \frac{2M}{\omega_{\rm n}}\Delta\omega_{\rm n} = R\Delta\omega_{\rm n}, \quad (13)$$

where R is the mass response rate.[56,57] In the process of mass measurement, we assume the additional particles are uniformly distributed on the surface of the NR, and increasing mass does not affect the spring constant k of the oscillator. In practice, the position of the particles landing on the NR affects the resonance frequency. When the particles are attached to the position with the largest vibration amplitude, the resonance frequency is the largest.[58] Therefore, we will attach particles to the free end of NR to achieve the maximum NR frequency deviation, which not only fully takes into account the effect of the additional position, but also keeps the spring constant unchanged.

3. Results and discussion

In what follows, we will present numerical results to illustrate how the nonlinear Kerr spectrum can be used to implement the effective mass measurement in the spin–oscillator composite system. Here we use feasible experimental parameters:[14,19,20] the vibration frequency of the cantilever $\omega_{\rm n} = 1.0$ MHz, the spin–oscillator coupling strength $g_0 = 10$ kHz, the quality factor of the NR $Q = 10^6$, the decay rate $\gamma_{\rm n} = \omega_{\rm n}/Q = 10^{-3}$ kHz. The eigen-spin life T_2 has been

observed in nitrogen–vacancy in the range of 1 ms–10 ms, and we chose spin life $T_2 = 1$ ms corresponding to $\Gamma_2 = 1.0$ kHz. The experiments can be done at room temperature by using the setup of the mass sensor shown in Fig. 1. Then, we introduce how to weigh the mass of added DNA molecules on the surface of NR by the pump–probe technique.

The most important thing of mass sensing is detecting the frequency shift of NR when adding particles are landed on the surface of NR. Therefore, how to obtain the original frequency (without anything particles on the surface of NR) of NR resonator is very crucial. Here, we present a nonlinear scheme to determine the original frequency of NR with Kerr spectroscopy. Figure 2 shows the Kerr coefficient $\mathrm{Re}[\chi^{(3)}(\omega_s)]$ as a function of the probe–spin detuning Δ_s for three different vibrating frequencies of NR. The two sharp peaks (the red lines) indicating the fundamental resonance frequency $\omega_n = 1.0$ MHz exactly locate at $\Delta_{pr} = \pm 1.0$ MHz. To verify its accuracy, two additional resonance frequencies with frequencies $\omega_n = 0.8$ MHz and $\omega_n = 1.2$ MHz are also demonstrated in Fig. 2 corresponding to the black and blue curves locating at $\Delta_{pr} = \pm\omega_n$. It is clear that the horizontal coordinate magnitude of the two sharp peaks (the blue lines and the black lines) of the Kerr coefficient is the same magnitude of the vibration frequency of NR. The physical principle is due to mechanically induced coherent population oscillation making quantum interference between the two optical fields and NR via the electron spin when the probe–pump detuning is equal to the vibration frequency of NR. Thus, keeping the detuning of the pump–spin $\Delta_c = 0$ and scan the probe frequency across the spin-flip transition frequency in the spectrum, then we can obtain the vibrational frequency of NR from the nonlinear Kerr spectrum.

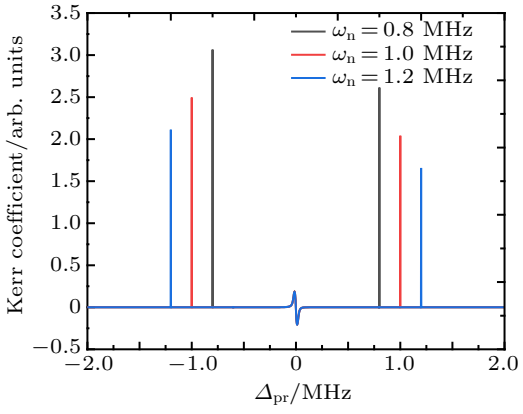


Fig. 2. The optical Kerr coefficient as a function of the detuning of the probe field from the exciton resonance with three vibrational frequencies of NR $\omega_n = 0.8$ MHz, $\omega_n = 1.0$ MHz, and $\omega_n = 1.2$ MHz for $\Omega^2 = 81$ (kHz)^2, $\Delta_c = 0$, $\gamma_n = 10^{-3}$ kHz, and $g_0 = 40$ kHz.

To explore the Kerr effect more carefully, we also present the optical Kerr coefficient $\mathrm{Re}\,\chi^{(3)}_{\mathrm{eff}}$ as a function of probe–spin detuning $\Delta_{pr} = \omega_{pr} - \omega_e$ with $\Delta_c = 0$ for several different coupling strengths and decay rates. An important parameter of the nanomechanical hybrid system is the coupling strength g_0. The Kerr coefficients for three different coupling strengths are shown in Fig. 3(a), where the blue line ($g_0 = 30$ kHz) is the shortest and the black line ($g_0 = 40$ kHz) is the highest. Figure 3(b) shows the details of the three lines in the left of Fig. 3(a). We can easily observe that the larger the coupling strength is, the higher the optical Kerr coefficient peak is. On the other hand, the effect of the vibration lifetime of NR in the nonlinear Kerr spectrum is shown in Fig. 3(c). The vibration lifetime plays an important role in the quantum phenomenon. In Fig. 3(d), it is obvious that the peak height of the Kerr coefficient increases with the increase of the vibration lifetime ($\tau = 1/\gamma_n$). The oscillator lifetime (the blue line $\gamma_n = 5$ Hz) is so small that the Kerr coefficient is close to zero. Besides, although the vibration life of the red line is only two times that of the black line, the height of the black line is outstanding. The result is not unexpected, because the phenomenon of optical Kerr effect is caused by the coupling between spin quit and NR, the strong coupling makes it more obvious. The larger enough oscillator lifetime is one of the important reasons for the system can be performed at room temperature. The results indicate that the coupling strength and oscillator lifetime are both especially important parameters in such a hybrid system.

Owing to the excellent properties of the spin–oscillator system, *i.e.*, the strong coupling strength and the long quantum coherence time, this hybrid system can be considered as an ultrasensitive mass sensor, which can be implemented at room temperature. When implementing the mass sensing, the first step is the determination of the frequency of the hybrid NR system. The method to measure the resonator frequency has given in Fig. 2, then we can implement the mass sensing. We will introduce the nonlinear Kerr effect of the hybrid NR system to carry out the mass sensing. For example, we weigh the mass of DNA molecule, the particles to be measured are functionalized, double-stranded DNA (dsDNA) containing 1587 base pairs, the single mass of dsDNA is $m_{\mathrm{DNA}} \approx 1659$ zg (1 zg $= 10^{-21}$ g). We attach DNA molecules on the free end of NR and measure the altered frequency of NR. The mass of DNA molecules will be obtained by the linear relationship of the frequency-shift and mass-variation ($\delta m_{\mathrm{DNA}} = (2M/\omega_n)\delta\omega$, where M is the effective mass of NR, $M_{\mathrm{NR}} = 1.3 \times 10^{-12}$ g, $\delta\omega$ is the changed frequency). In Fig. 4, we first add 10 DNA molecules on the free end of NR, there is a frequency shift $\delta\omega_1 = 6.38$ Hz compared with bare NR. When adding 30 DNA molecules, the changed frequency $\delta\omega_2 = 19.14$ Hz. The right inset of Fig. 4 shows the linear relationship of the frequency shift and the number of added DNA molecules. We can conclude that the smaller effective mass and higher frequency of NR enhance the sensitivity of mass measurement from the above relationship of the frequency-shift and mass-variation.

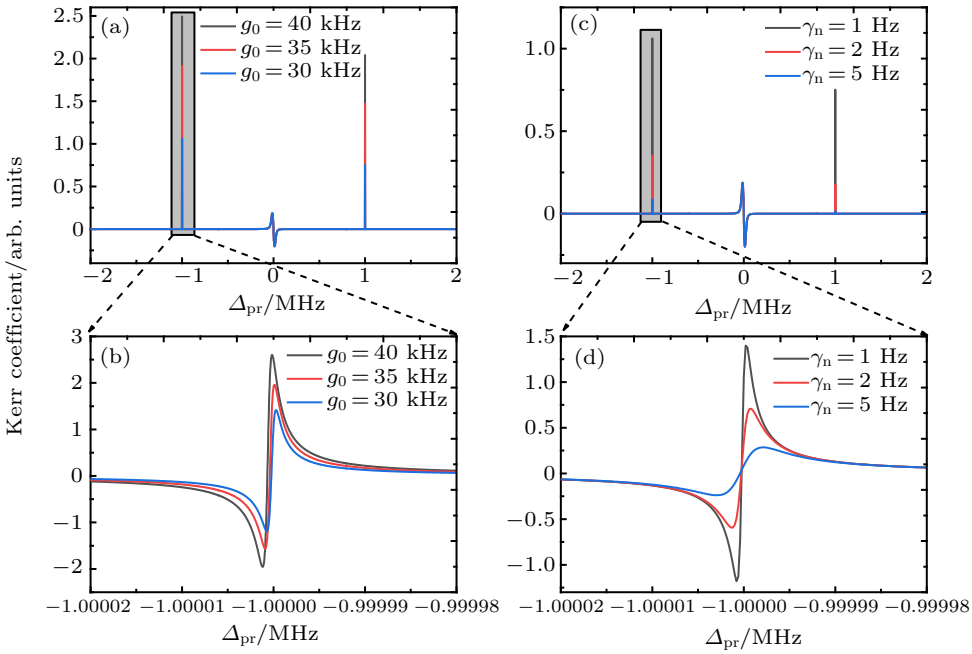


Fig. 3. (a) The optical Kerr coefficient as functions of the probe field from the exciton for different coupling strengths. (b) The detailed parts of the left peaks in panel (a). (c) The optical Kerr coefficient as functions of the probe field from the exciton with several different decay rates. (d) The detailed parts of the left peaks in panel (c).

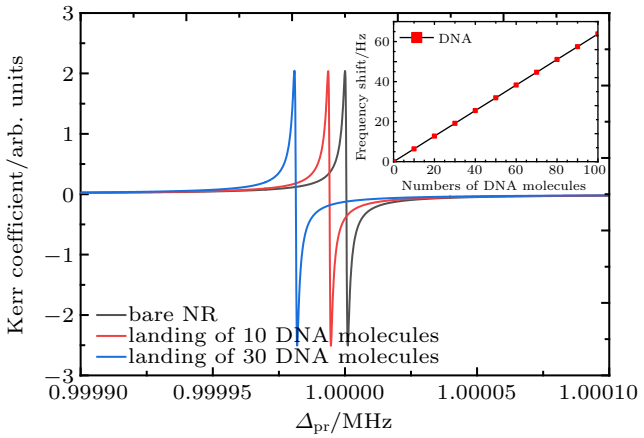


Fig. 4. The optical Kerr coefficient without anything else and with 10 or 30 DNA molecules on the surface of NR. The inset shows the relationship between the frequency shift of NR and the number of added DNA molecules.

As mentioned in the previous section, the traditional mass spectrometer is difficult to detect the mass of some particles which are hardly ionized. Fortunately, the scheme of mass measurements proposed here does not require ionization. The mass of measured particles can be easily obtained by the nonlinear Kerr spectrum. Compared with the electric measurement method which will lead to the heating effect and energy loss, our proposed scheme can avoid the shortcoming effectively. Although optical mass sensors using a single laser detection has been demonstrated in cavity optomechanical systems,[59] the pump–probe technique is based on the coherent effect of microwave distinguishing from the single light detection method, which not only overcome the problem that the single light detection can only be targeted at low-frequency oscillators but also improve the sensitivity and accuracy at the same time on account of not including any electrical parameters. Last but not least, the mass measurement method through the nonlinear Kerr spectrum may be immune on the detection noise compared with the linear optical spectrum.[60]

4. Conclusion

We proposed a mass measurement scheme based on the hybrid spin–nanoresonator system with the microwave pump–probe techniques. By detecting the nonlinear Kerr spectrum of the probe fields, the mass of the attached particles at the free vibrating end of the NR can be obtained accurately. The mass sensing based on the spin–oscillator hybrid system with the pump–probe method has obvious advantages in comparison to the traditional electrical measurement method and single-light measurement method. Compared with the previous mass sensing scheme in both theory and experiment, the hybrid system presented here has a remarkable property, that is, the system can be carried out at room temperature. Finally, we hope our proposal can be realized in the future.

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