Quantum Sensing with Scanning Near-Field Optical Photons Scattered by an Atomic-Force Microscope Tip

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(Dated: December 12, 2022)

Scattering scanning near-field optical microscopy (s-SNOM) is known as a promising technique for overcoming Abbe diffraction limit and substantially enhancing the spatial resolution in spectroscopic imaging. The s-SNOM works by exposing an atomic force microscope (AFM) tip to an optical electromagnetic (EM) field, while the tip is so close to a sample that the incident beam lies within the near-field regime and displays nonlinear behaviour. We suggest replacing the incident field by quantized EM fields, i.e. photons, and propose a quantum model for the suggested system, by employing electric-dipole approximation, image theory, and perturbation theory. Quantum state of scattered photons from the AFM tip is extracted from the proposed model, which contain information about electrical permittivity of the dielectric material beneath the tip. The permittivity of the sample can be extracted through spectroscopic setups. Our proposed scheme can be used for quantum imaging or quantum spectroscopy with high resolution.

For the past few decades, various techniques have been proposed for overcoming the $\lambda/2$ diffraction limit, also known as the Abbe diffraction limit \cite{1-4}. These techniques are based on apertures \cite{5} and scattering probes \cite{6}. Scattering scanning near-field optical microscopy (s-SNOM) is one of the reliable techniques that has attracted more attention recently, due to the independence of resolution to the wavelength. In this technique, the resolution only depends on the size of the setup.

The s-SNOM technique is leveraged on placing an atomic force microscope (AFM) tip close to the surface of a desired sample; light interacts with the AFM tip instead of directly interacting with the dielectric sample. In this approach, the resolution is limited to the size of the AFM tip. This technique provides imaging information with high nanoscale resolution, which has been recorded to be within the order of 1 nm \cite{7}.

Many research groups have studied the interaction of a classical light with an AFM tip in spectroscopic usage, mainly through the image theory \cite{8-10}. Also, interaction of a classical electromagnetic (EM) light with an AFM tip doped with 3-level quantum particles has been studied recently \cite{11}. To the best of our knowledge, the interaction between a quantized EM wave (i.e. a photon) and an AFM tip, in the presence of a dielectric material, has not yet been studied.

We investigate the interaction of near-field photons incident to an AFM tip, close to the surface of a dielectric sample. The goal is to study the effect of the dielectric material on the scattered photons from the AFM tip. As shown in Fig. 1, we consider an AFM tip, comprised of atomic-size quantum dots, such as phosphorous (P) atoms doped in silicon (Si) \cite{12,13}, Si-based dangling-bonds (DB) \cite{14,16}, diamond-based nitrogen-vacancy (NV$^-$) centers \cite{17}. Incident photons interact with the tip and excite the quantum dots (QDs); as the QDs return to ground state, photons are generated and scattered out of the tip. Since the AFM tip is placed close to the dielectric half-plane surface, spontaneous emission of the quantized EM fields from the AFM tip is modified in the presence of the dielectric surface, resulting in changes in the Fock state of the re-emitted photons. To model the interaction between the AFM tip and the sample, we consider using the image theory. The AFM tip exposed by incident photons is modeled by an electric dipole. The sample is replaced by an effective image dipole containing the dielectric properties of the material, including the permittivity. The AFM tip is so close to the surface ($\approx$ 30 nm, the rim zone) that the near-
field electrodynamics applies in this scale, and Van der Waals force is induced between the tip dipole and its image dipole in the sample. The effect of the sample on the scattered photons from the AFM tip, is modelled by employing modified second-order perturbation theory. Fock state of the scattered photons from the AFM-tip is then calculated. The scattered photons contain the electrical properties of the sample and can be derived through spectroscopy setups.

Now we elaborate on our proposed system and the suggested quantum model for describing it. For the AFM tip, a sphere with a radius of \( \leq 10 \) nm is considered, filled with a certain amount of QDs, such as DBs \([14-16]\), NV\(^-\) centers \([17]\), or Si-doped P atoms \([12, 13]\). If the size of the QD is smaller than the spatial extension of the photon’s wavepacket, the electromagnetic field can be considered constant on the QD; hence, with long-wave approximation, the QD is considered as an electric dipole. Conditioned to have matching between photons’ energy and the energy difference of the two quantum states of the QDs, we use the electric dipole approximation to model the AFM tip, exposed to a stream of photons. Transition between the two states of the quantum dot is then given by \( d_{aa'} = \langle a' | q r | a \rangle \) for \( r \ll \lambda \), where \( d_{aa'} \) is the QD effective electric-dipole moment, and \( |a\rangle \) and \( |a'\rangle \) are the ground state and the excited state of the QD, respectively. Parameter \( r \) is the spatial dispersion of the QD, which is assumed to be smaller than the wavelength of the incident photon \([18]\). Assuming that the AFM-tip dipole is at distance \( R \) from the surface of a nearby dielectric sample, it results in an image dipole in the sample that is located at the same distance below the surface, as illustrated in Fig. 2. According to the image theory, the whole effect of the half-space sample on the tip’s dipole can be modeled by considering its corresponding image dipole. As shown in Fig. 3, it is assumed that the charges of the image dipole are separated by the same distance as the main dipole, but with different values \([19, 20]\). The image of a single charge in a dielectric material is given by

\[
q' = -\alpha q, \quad \alpha = \frac{\epsilon - 1}{\epsilon + 1}
\]

where \( \alpha \) is the dielectric-air coefficient, which is a function of \( \epsilon \), i.e. the electric permittivity of the dielectric material. Many groups have studied the coulomb energy of particle-surface interactions \([21, 22]\). Subsequently, the energies associated with the states of the image dipole (i.e. \( |b\rangle \) and \( |b'\rangle \)) are given by,

\[
E_b = \alpha^2 E_a, \quad E'_b = \alpha^2 E'_a,
\]

where \( E_a \) and \( E'_a \) are the energies associated with the states of the AFM tip dipole. The same modification applies for the energy band gap of the image dipole, i.e. \( \Omega' = \alpha^2 \Omega \).

To model photons incident to the AFM tip, we consider the near-field interaction. Study of electromagnetic waves and photons in an area very close to a material lies in the near-field electrodynamics, which is a great field of interest \([23-26]\). As incident photons are reflected from the AFM tip, there are a few of them with imaginary \( k \)-vectors, called the evanescent photons, that decay as they pass through the very small area between the AFM tip and the nearby sample. As a consequence, spontaneous emission of the photons scattered from the AFM tip is modified due to the nearby dielectric sample. In other words, the presence of a dielectric material near to the AFM tip affects the incident-photon AFM-tip interaction, resulting in a signature of the electric permittivity of the dielectric surface, in the quantum state of the photons scattered from the AFM tip.

Due to the near-field electrodynamics, interaction between the electric dipole of the tip and its image dipole is studied by Van der Waals force. This approach has many interest in scanning near-field optical microscopy \([9]\).
To guarantee for being in the near-field region, hence ensuring Van der Waals interaction between the two dipoles, the distance between the AFM dipole and the corresponding image dipole should be small enough to satisfy,

\[ |r_a - r_b| < < c_0 \omega_{ab}^{-1} \omega_{bb}^{-1}, \]

where \( \omega_{ab}^{-1} \) and \( \omega_{bb}^{-1} \) are the Bohr frequencies of the dipoles [13], \( r_a \) and \( r_b \) are the position vectors of the dipoles, and \( c_0 \) is the speed of light in vacuum, see Fig. 2.

To model the tip-sample Van der Waals interaction, we propose a modified perturbed Hamiltonian representing the AFM tip dipole and the image dipole interaction [27]. The effective perturbation Hamiltonian corresponding to the instantaneous dipole–dipole interaction is obtained by [13],

\[ \delta \hat{H} = -\frac{1}{|r_a - r_b|^3} \frac{1}{c_0} \hat{d}^a \cdot \delta_T (r_a - r_b) \hat{d}^b, \]  

where \( \hat{d}^a \) and \( \hat{d}^b \) are the electric-dipole moment operators of the main dipole and the image dipole respectively [13]. The parameter \( \delta_T \) is the transverse delta function, representing the direction along the line passing through the dipoles. If the dipoles are polarized in the z direction (see Fig. 3), the inner product between the operators vanishes. The electric dipole operators are,

\[ \hat{d}^a = d_{a\sigma} (\hat{\sigma}^a_+ + \hat{\sigma}^a_-) \quad \text{and} \quad \hat{d}^b = \alpha d_{b\sigma} (\hat{\sigma}^b_+ + \hat{\sigma}^b_-) \]  

where \( \alpha \) is given by Eq. 4 and contains information about the permittivity of the sample, \( d_{i\sigma} \) for \( i = \{a, b\} \) is the electric dipole moment, and \( \sigma^a_\pm \) are the ladder operators defined by \( \sigma^a_\pm = |i\rangle\langle i'| \) and \( \sigma^a_\pm = |i\rangle\langle i'| \). Eventually, the perturbed Hamiltonian turns to

\[ \delta \hat{H} = \frac{1}{|r_a - r_b|^3} \frac{1}{c_0} \alpha (\sigma^a_+ \sigma^b_+ + \sigma^a_- \sigma^b_- + \sigma^a_+ \sigma^a_- + \sigma^a_- \sigma^b_+). \]  

Quantum states of the system are given by

\[ \Sigma_{1, n} |a, b, n\rangle, \Sigma_{2, n} |a, b', n\rangle, \Sigma_{3, n} |a', b, n\rangle, \Sigma_{4, n} |a', b', n\rangle, \]

which are comprised of the energy states of the tip dipole and its image in the dielectric sample, and the fock state \( |n\rangle \) representing the photons incident on the AFM tip. Since perturbation in the system results from the effect of the image dipole on the tip dipole, as presented in Eq. 4, there is no direct effect on the fock state of the incident photons in the perturbation Hamiltonian. Furthermore, the first order perturbation applies for charged particles, which is not the case for the dielectric sample, in our desired system. Therefore, modified second order perturbation theory is implemented to calculate the perturbed states,

\[ \sum_{n=1}^{\infty} \beta_{1n} |a, b, n\rangle^{(2)} = \sum_{n=1}^{\infty} \frac{1}{2(2R)^3} \frac{a_1^2 (\frac{1}{c_0} \alpha)^2}{(\Omega(1 + \alpha^2))^2} |a, b, n\rangle, \]

where \( \beta_{1n} \) is the coefficient of the perturbed state \( |a, b, n\rangle \). Similar results can be obtained for other three states which will be shown later. Presuming to have a condition under which the incident photons interact one-by-one with the AFM tip (for instance, having a single-photon emitter as a source of generating incident photons), the state of such photons would be represented by [1]. Then, the density matrix of our desired system would be a \( 4 \times 4 \) matrix given by,

\[ \rho_{ab} = |\Psi\rangle \langle \Psi| = \begin{bmatrix} \beta_{11}^2 & \beta_{12}^2 & \beta_{13} & \beta_{14} \\ \beta_{21}^2 & \beta_{22}^2 & \beta_{23} & \beta_{24} \\ \beta_{31}^2 & \beta_{32}^2 & \beta_{33} & \beta_{34} \\ \beta_{41}^2 & \beta_{42}^2 & \beta_{43} & \beta_{44} \end{bmatrix} \]  

where \( \beta \) parameters are the coefficients of every possible state in our system and are given by,

\[ \beta_{11} = \beta_{33} = a_{1, 3} = \frac{1}{2(2R)^3} \frac{a_1^2 (\frac{1}{c_0} \alpha)^2}{(\Omega(1 + \alpha^2))^2}, \]

\[ \beta_{22} = \beta_{44} = a_{2, 4} = \frac{1}{2(2R)^3} \frac{a_2^2 (\frac{1}{c_0} \alpha)^2}{(\Omega(1 - \alpha^2))^2}. \]  

Suppose that using a proper mechanism, single photons scattered from the AFM tip, are detected by a single photon detector. These photons contain information about the electric properties of the dielectric sample. State of the scattered photons is obtained by reducing the state of the whole system, by tracing out the states of the subsystems, associated with the AFM dipole and its image dipole interaction. Consequently, the state associated with the scattered photon is

\[ |\Psi_1\rangle = \sqrt{\beta_{11}^2 + \beta_{22}^2 + \beta_{33}^2 + \beta_{44}^2} |1\rangle. \]

Assuming that both dipoles are initially in their ground states, then only \( \beta_{11}^2 \) remains nonzero. Substituting its value from Eq. 10, the state of the scattered photon is obtained to be,

\[ |\Psi_1\rangle = (a_1 - \frac{1}{2(2R)^3} \frac{a_1^2 (\frac{1}{c_0} \alpha)^2}{(\Omega(1 + \alpha^2))^2}) |1\rangle, \]

where \( a_1 \) is the coefficient of the unperturbed state \( |a, b, 1\rangle \). The result clearly shows that the state of a single scattered photon is bearing information about the properties of the dielectric sample, which is hidden in \( \alpha \). It also depends on the distance \( R \) of the tip from the sample, and the energy difference between the two states of the tip dipole \( \Omega \). Furthermore, the effective perturbed Hamiltonian, given by Eq. 4, leads to an energy shift, \( \Delta E \), of the states \( |a, b, 1\rangle \) given by

\[ \Delta E = -\frac{1}{2(2R)^3} \frac{a_1^2 (\frac{1}{c_0} \alpha)^2}{(\Omega(1 + \alpha^2))}. \]
This energy shift is negative, indicating that Van der Waals interaction lowers the ground state energy of the tip dipole. Also, it varies with $R^{-3}$, which means the tip-sample force varies as $R^{-4}$.

To conclude, we showed that the quantum behavior of an AFM tip exposed to incident photons is modified by existence of a dielectric material nearby. This modification is resulted by the Van der Waals interaction between the tip’s dipole and an its image dipole, representing the properties of the dielectric. Finally, the fock state of a scattered single photon is obtained through the perturbation theory and can be used to specify the permittivity of the dielectric material. The proposed quantum s-SNOM provides the opportunity of imaging and spectroscopy with higher spatial resolution.

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