Scattering Theory of Bardeen's Formalism for Tunneling: New Approach to Near-Field Microscopy

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(Received 19 October 1999)

We propose a new theoretical approach to near-field microscopy, which allows one to deal with scanning tunneling microscopy and scanning near-field optical microscopy with a unified formalism. Under the approximation of weak tip-sample coupling, we show that Bardeen's perturbation formula, originally derived for electron tunneling, can be derived from a scattering formalism which extends its validity to electromagnetic vector fields. This result should find broad applications in near-field imaging and spectroscopy.

PACS numbers: 61.16.Ch, 03.65.Nk, 07.79.Cz, 73.40.Gk

The development of scanning tunneling microscopy (STM) in the early eighties [1] opened the way to real-space surface study at the atomic scale. Since then, various techniques of scanning probe microscopy (SPM) have been proposed [2,3], based on local interaction between a sharp tip and the sample under study. Scanning near-field optical microscopy (SNOM) [4] is one of these techniques, which uses optical interaction in the visible or near-infrared range. SNOM has proven its ability to image optical fields and surface structure at a subwavelength scale [5]. In the field of microscopy, spectroscopy, and surface modification on the nanometer scale with visible or infrared light [6], SNOM looks complementary to other SPM techniques.

In the context of STM, some theories were developed shortly after the first experimental demonstrations, based on self-consistent methods and numerical calculations [7,8] or on analytical models [9–12]. Many of these theories [8–10] have in common the use of Bardeen's perturbation formula, originally derived for electron tunneling between two weakly coupled electrodes [13]. In particular, the approach of Tersoff and Hamann [9] remains an explicit and practical description of the STM. An important result in this approach was the direct interpretation of the STM signal as a measurement of the local electron density of state of the sample. Although this result is valid under weak tip-sample coupling, it was a breakthrough in understanding the STM images [2].

Similarly, in the context of SNOM, several theoretical methods and numerical simulations [14], as well as analytical models [15,16], have been developed, in order to improve the capability of the technique and to understand the measured signals. Although the underlying physics behind SNOM is understood to a certain extent, an overlook at the current state of SNOM leads to the two following remarks. (i) The analogy between STM and SNOM is often qualitatively put forward. In particular, some SNOM setups such as the photon scanning tunneling microscope (PSTM) were introduced by analogy between optical and electron tunneling [17]. Nevertheless, there is no unified formalism and theoretical proof of a clear and general analogy. (ii) An explicit SNOM theory was developed some years ago [15], which gave an interpretation of the signal and clarified the role of spatial filtering and polarization effects. Nevertheless, a general formalism allowing to introduce in a natural way an appropriate tip model seems to be missing [18].

In this Letter, we propose a new approach to near-field microscopy which deals with both STM and SNOM with a unified formalism. We first derive an expression of the current in the gap [19] which is valid for STM and SNOM. This expression allows an original discussion of the tunneling contribution to the SNOM signal. Then, under the approximation of weak tip-sample coupling, we derive a general expression of the signal in SNOM, which generalizes Bardeen's formula to scattering of vector electromagnetic fields. This generalization allows one to deal with SNOM using the standard formalism of STM modeling.

Let us consider the general SNOM setup depicted in Fig. 1(a), and the general STM setup in Fig. 1(b). In the SNOM situation, the tip-sample system is illuminated by a light source of arbitrary size and state of coherence, and part of the scattered energy is collected by a detector of arbitrary shape. The gap region (between the sample and the tip) is assumed to be vacuum or air. At this stage of the discussion, we concentrate on the tunneling current in both STM and SNOM, and we do not take polarization effects into account. In the STM situation, we assume that the central part (with respect to the z direction) of the gap region is of constant potential V. The state of the electromagnetic field at a given frequency ω, or a stationary state of the electron of energy E, are both represented by a scalar wave function $\Psi(\mathbf{r})$. We assume that the tip remains situated above the highest point of the surface topography (although the path followed during the scan may

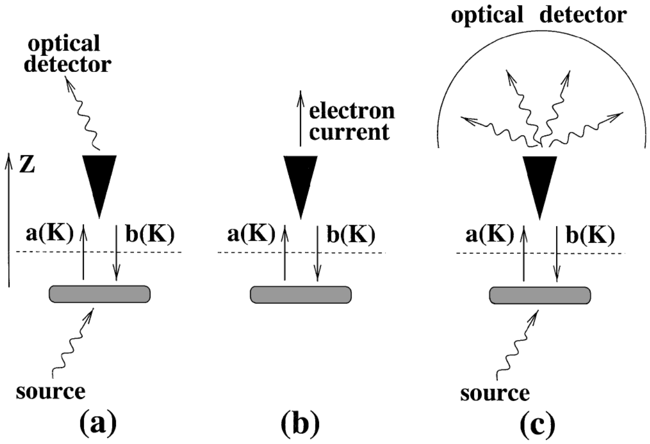

FIG. 1. (a) Scheme of a SNOM setup. Light coming from the source is scattered towards the detector through near-field coupling between the tip and the sample. (b) Scheme of a STM setup. The current is created by tunneling electrons between the tip and the sample. (c) SNOM setup with hemispherical detection.

be arbitrary). In the gap region, the wave field can be written in the form of an angular spectrum of plane waves [20]:

$$\Psi(\mathbf{r}) = \int a(\mathbf{K}) \exp(i\mathbf{K} \cdot \mathbf{R} + i\gamma z)\, d^2\mathbf{K} + \int b(\mathbf{K}) \exp(i\mathbf{K} \cdot \mathbf{R} - i\gamma z)\, d^2\mathbf{K}\,, \quad (1)$$

where $\gamma(\mathbf{K}) = \sqrt{k^2 - K^2}$ for $K^2 \leq k^2$ (homogeneous or propagating components) and $\gamma(\mathbf{K}) = i\sqrt{K^2 - k^2}$ for $K^2 > k^2$ (inhomogeneous or evanescent components). We use the notations $\mathbf{r} = (x, y, z)$, $\mathbf{R} = (x, y)$, and $K = |\mathbf{K}|$. For the electromagnetic field, $k = \omega/c$, c being the speed of light in vacuum. For the electron wave function, $k^2 = 2m/\hbar^2(E - V)$, where m is the electron mass and $\hbar$ is Planck's constant. The integrals are extended to $0 < K < +\infty$. Note that, in the case of electron tunneling in STM ($E < V$ and $k^2 < 0$), the wave function in the gap region contains evanescent waves only.

The current density associated with the wave function Ψ is $\mathbf{J}(\mathbf{r}) = A\,\mathrm{Im}[\Psi^*(\mathbf{r})\nabla\Psi(\mathbf{r})]$, where Im denotes the imaginary part and $*$ the complex conjugate. This formula represents either the momentum density of the electromagnetic field in the scalar representation or the probability current in quantum mechanics [20]. The constant A may be determined by identifying the current flux at the detector with either the energy flux of the electromagnetic field (in the case of SNOM) or the electronic current (in the case of STM). Using Eq. (1), the total current $\phi = \int J_z(\mathbf{r})\, d^2\mathbf{R}$ across a plane at a constant z in the gap region (dashed line in Fig. 1) can be cast in the following form:

$$\phi = A \int_{K^2 \leq k^2} \gamma[|a(\mathbf{K})|^2 - |b(\mathbf{K})|^2]\, d^2\mathbf{K} + A \int_{K^2 > k^2} \gamma[a(\mathbf{K})b^*(\mathbf{K}) - a^*(\mathbf{K})b(\mathbf{K})]\, d^2\mathbf{K}\,. \quad (2)$$

Although Eq. (2) simply expresses the total current flowing through the gap region, it was never used before, to our knowledge, in the context of near-field microscopy. In STM, except for a constant factor, ϕ is exactly the tunneling current which is measured in the experiment. In SNOM, ϕ is proportional to the *total* energy flux, including the flux flowing through channels that do not end up at the detector. In many SNOM experiments, only part of this flux is actually collected and contributes to the signal. ϕ would be an exact expression of the signal in situations in which a hemispherical detector is used to collect all the flux traveling in a half space, as shown in Fig. 1(c). An example of such a configuration is the tunneling near-field optical microscope [21] when complete hemispherical detection is performed, and its reciprocal setup, namely, a PSTM using hemispherical incoherent illumination [22,23].

Two separate contributions are clearly identified in Eq. (2). The first integral describes the contribution of waves that are homogeneous in the gap region. It simply expresses the balance between two currents flowing in opposite directions through propagating channels. The second integral describes a current flowing through evanescent (or tunneling) channels. In the case of STM, this is the only contribution to the current. This term simply reflects the net flux traveling through the tunneling channel $\mathbf{K}$, and vanishes if $a(\mathbf{K}) = b(\mathbf{K})$. Note that, if either $a(\mathbf{K})$ or $b(\mathbf{K})$ vanishes, then the contribution of this tunneling channel also vanishes. This reflects the fact that tunneling is essentially a consequence of the presence of two interfaces at close proximity (e.g., the sample and the tip). Equation (2) also demonstrates the existence of an optical tunneling contribution in any SNOM configuration. Moreover, it shows that the SNOM current travels through both propagating and tunneling channels in the gap, whereas in STM the current flows only through tunneling channels. This is a fundamental difference between SNOM and STM.

In practice, computing the SNOM or STM signal from Eq. (2) requires the knowledge of the angular spectra $a(\mathbf{K})$ and $b(\mathbf{K})$ of the wave function in the gap region. These are solutions of a difficult scattering problem in a confined geometry, which can, in general, be solved only numerically. Nevertheless, under the approximation of weak tip-sample coupling, it is known in STM modeling that Bardeen's formula can be used to describe the tunneling current [2,8–10]. We shall now give a new derivation of this formula, based on a scattering formalism. This approach generalizes Bardeen's original formula, by showing that (i) it describes both the tunneling current and the current flowing through propagating channels, and that (ii) it also applies to vector electromagnetic fields.

Let us consider the general SNOM setup depicted in Fig. 2(a). The illumination is done by a plane wave with a wave vector $\mathbf{K}_s$, a unit amplitude, and a state of polarization described by the complex unit vector $\mathbf{a}_s$. The signal is recorded by a detector placed in the far field, in a direction defined by the wave vector $\mathbf{K}_d$. We assume that the detection is performed with a polarizer (analyzer) whose polarization direction is defined by the unit vector $\mathbf{a}_d$. Note that this represents the most general configuration, because an extended and/or unpolarized source or detector can be described by adding the contributions of a set of incoming or outgoing plane waves. Depending on the experimental setup, the summation should be done with a properly defined degree of coherence and/or polarization [23]. Without loss of generality, we have chosen the transmission geometry shown in Fig. 2(a), but the argument can be easily extended to any SNOM setup. Using a scattering formalism, we describe the sample, the tip, and the entire setup by their generalized transmission coefficients $\overleftrightarrow{t}_s(\mathbf{K},\mathbf{K}_s)$, $\overleftrightarrow{t}_d(\mathbf{K}_d,\mathbf{K})$, and $\overleftrightarrow{T}(\mathbf{K}_d,\mathbf{K}_s)$. These coefficients are elements of the scattering matrix of each system in a plane-wave basis [24]. The signal S is the flux of the Poynting vector (current density) at the detector position (i.e., in the far field). The far-field asymptotic expression of the electromagnetic field in the direction $\mathbf{K}_d$ can be obtained by the stationary-phase technique [20]. In this condition, the expression of the signal is

$$S = 2\pi^2 \epsilon_0 c \gamma^2(\mathbf{K}_d)\,|\mathbf{a}_d \cdot \overleftrightarrow{T}(\mathbf{K}_d,\mathbf{K}_s) \cdot \mathbf{a}_s|^2. \qquad (3)$$

This result shows that the basic quantity to compute is $M_{ds} = \mathbf{a}_d \cdot \overleftrightarrow{T}(\mathbf{K}_d,\mathbf{K}_s) \cdot \mathbf{a}_s$, which is analogous to the elastic tunneling matrix element in Bardeen's formalism [13]. We now assume that the coupling between the tip and the sample is weak. In the scattering picture, this means that the current in the gap results from fields that have been scattered once at the tip or at the sample. In this case, the transmission coefficient of the system is

$$\overleftrightarrow{T}(\mathbf{K}_d,\mathbf{K}_s) = \int \overleftrightarrow{t}_d(\mathbf{K}_d,\mathbf{K}) \cdot \overleftrightarrow{t}_s(\mathbf{K},\mathbf{K}_s)\, d^2\mathbf{K}\,, \qquad (4)$$

where the integral is extended to both propagating and tunneling channels. We see that, in the case of weak coupling,

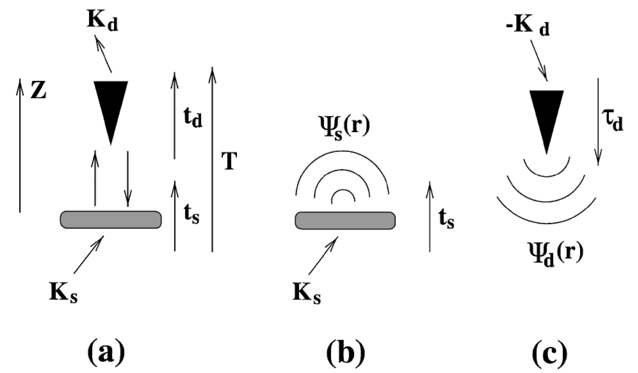

FIG. 2. (a) General SNOM setup with directional illumination and detection. (b) Illustration of the meaning of the sample wave function $\mathbf{\Psi}_s$. (c) Illustration of the meaning of the tip wave function $\mathbf{\Psi}_d$.

the signal can be calculated from the transmission coefficients of the sample and the tip, considered as independent systems. We will now transform Eq. (4) into an expression in direct space, involving two wave fields that are solutions of the two scattering problems in Figs. 2(b) and 2(c). This will lead to a generalization of Bardeen's formula to scattering of electromagnetic vector fields. Let $\mathbf{\Psi}_s(\mathbf{r})$ be the (vector) electric field, in the gap region, that results from scattering of the illuminating plane wave (wave vector $\mathbf{K}_s$, polarization state $\mathbf{a}_s$) by the sample, in the absence of the tip. Let $\mathbf{\Psi}_d(\mathbf{r})$ be the (vector) electric field, in the gap region, that results from scattering by the tip of a plane wave of amplitude unity coming from the direction of the detector (wave vector $-\mathbf{K}_d$, polarization state $\mathbf{a}_d$). The explicit expressions of these wave fields are

$$\mathbf{\Psi}_s(\mathbf{r}) = \int \overleftrightarrow{t}_s(\mathbf{K},\mathbf{K}_s) \cdot \mathbf{a}_s \exp(i\mathbf{K} \cdot \mathbf{R} + i\gamma z)\, d^2\mathbf{K}\,, \qquad (5)$$

$$\mathbf{\Psi}_d(\mathbf{r}) = \int \overleftrightarrow{\tau}_d(\mathbf{K},-\mathbf{K}_d) \cdot \mathbf{a}_d \exp(i\mathbf{K} \cdot \mathbf{R} - i\gamma z)\, d^2\mathbf{K}\,, \qquad (6)$$

where $\overleftrightarrow{t}_d$ is related to $\overleftrightarrow{\tau}_d$ by the reciprocity theorem $\gamma(\mathbf{K}_d)\overleftrightarrow{t}_d(\mathbf{K}_d,\mathbf{K}) = \gamma(\mathbf{K})\overleftrightarrow{\tau}_d^{\,T}(-\mathbf{K},-\mathbf{K}_d)$, the superscript T denoting the transposed tensor [24]. From Eqs. (4)–(6), one obtains the following expression for the matrix element M_{ds}:

$$M_{ds} = \frac{1}{8\pi^2 i\gamma(\mathbf{K}_d)} \int \left[\mathbf{\Psi}_d(\mathbf{r}) \cdot \frac{\partial \mathbf{\Psi}_s}{\partial z}(\mathbf{r}) - \mathbf{\Psi}_s(\mathbf{r}) \cdot \frac{\partial \mathbf{\Psi}_d}{\partial z}(\mathbf{r})\right] d^2\mathbf{R}\,, \qquad (7)$$

where the integral is performed along a plane at a constant z in the gap region.

Equation (7) is the main result of this Letter. It is similar to Bardeen's formula for the elastic tunneling matrix element $M_{\mu\nu}$ between a state Ψ_μ of the probe and a state Ψ_ν of the sample [see, e.g., Eq. (3) in Ref. [9]]. Note that the complex conjugation of the tip wave function $\mathbf{\Psi}_d$ does not appear in Eq. (7). This point is not fundamental. Bardeen's formula is exactly retrieved when using a tip wave function $\mathbf{\Psi}_d' = \mathbf{\Psi}_d^*$, namely, the *time reversed* of the wave function $\mathbf{\Psi}_d$ introduced in Eq. (6). When the tip is lossless, this difficulty can be overcome by choosing a real solution for $\mathbf{\Psi}_d$. This is the usual choice that is made in STM modeling based on Bardeen's formula [9,12]. Our formalism clearly demonstrates that the tip wave function must be chosen as a solution of the scattering problem in Fig. 2(c), followed by time reversal, for the integrand in Eq. (7) to take the form of a current operator, as in Bardeen's original paper [13]. The general derivation of Eq. (7) given in this Letter shows that Bardeen's formula applies (i) to the general situation where the current in the gap flows through both propagating and tunneling channels, and (ii) to electromagnetic vector fields. From a fundamental point of view, this constitutes an important generalization of Bardeen's original formula. From a practical side, Eqs. (3) and (7) provide an expression of the SNOM signal using the same formalism as in standard STM modeling [9,10,12]. This unification could greatly improved the understanding of the SNOM signal.

Finally, let us comment on the choice of the tip wave field $\mathbf{\Psi}_d$ in SNOM modeling. The simplest model could consider the tip as a small sphere. In this case, it can be shown [25] that one retrieves the same result as that obtained in Ref. [15]. Except for some polarization effects, the signal in this model is proportional to $|\mathbf{\Psi}_s|^2$ at the location of the probe. This is also the result of the Tersoff and Hamann theory developed for STM [9]. Nevertheless, recent experimental studies of the apertureless setup have shown that this model is not sufficient to describe polarization and spectroscopic effects [18]. A theory including an appropriate tip model is needed to account for these effects [26]. The generalized Bardeen formula derived in this Letter provides a natural way to include realistic tip shapes in SNOM modeling.

In summary, we have presented a new formalism for near-field microscopy which unifies SNOM and STM. We have given an explicit expression of the total current in the gap region, which demonstrates the role of optical tunneling in SNOM. Under the approximation of weak tip-sample coupling, we have given a new and general derivation of Bardeen's perturbation formula, which applies to currents flowing through both propagating and tunneling channels, and to electromagnetic fields. The results in this Letter should find broad applications in near-field imaging and spectroscopy using visible or infrared light.

This work was supported by the French-Spanish Integrated Program PICASSO. We acknowledge helpful discussions with P. Chaumet, J.-J. Greffet, and M. Nieto-Vesperinas.

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