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Near-field radiative heat transfer in scanning thermal microscopy computed with the boundary element method

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ABSTRACT

We compute the near-field radiative heat transfer between a hot AFM tip and a cold substrate. This contribution to the tip-sample heat transfer in Scanning Thermal Microscopy is often overlooked, despite its leading role when the tip is out of contact. For dielectrics, we provide power levels exchanged as a function of the tip-sample distance in vacuum and spatial maps of the heat flux deposited into the sample which indicate the near-contact spatial resolution. The results are compared to analytical expressions of the Proximity Flux Approximation. The numerical results are obtained by means of the Boundary Element Method (BEM) implemented in the SCUFF-EM software, and require first a thorough convergence analysis of the progressive implementation of this method to the thermal emission by a sphere, the radiative transfer between two spheres, and the radiative exchange between a sphere and a finite substrate.

1. Introduction

Radiative heat transfer in the far field [1], *i.e.* for inter-object distances larger than Wien's wavelength ($\sim 10\,\mu$m at room temperature), is well established. In contrast, near-field radiative heat transfer (NFRHT), when the gap size between objects is smaller than Wien's wavelength, is a relatively new field. It was realized few decades ago that near-field radiative effects can have an impact on the local temperature or on the heat flux transfer in scanning probe microscopy (SPM) techniques [2]. It is especially important for scanning thermal microscopy (SThM), a technique which aims at measuring local temperature or thermal properties. To understand these local measurements, a precise knowledge of all the heat transfer mechanisms between the SThM tip and the sample under study is required [3]. The contribution of near-field thermal radiation has been so far the hardest to estimate accurately due to a lack of reliable theoretical methods able to deal with complex geometries. Nevertheless, this contribution is critical when the tip is not in contact, in particular when the SThM is operated in vacuum. Tip-sample near-field heat transfer has received increasing attention during the last decade thanks to technical advances allowing experiments with improved thermal sensitivity down to the sub-nW.K^{-1} regime [4–8]. Recently, such experiments have been carried out to measure radiative heat transfer in the last few nanometers before contact [7–9]. Other SPM techniques, such as Thermal Radiation Scanning Tunneling Microscopy (TR-STM) [10,11], where the near field is scattered and detected in the far field, therefore providing sub-wavelength information on the sample [10], are also impacted by heat transfer between the probe and the sample. Obviously, various SPM techniques could also benefit from improved theoretical methods to predict NFRHT between complex objects.

NFRHT was quantified by analytical and numerical methods in various configurations. Since the seminal case of two half-spaces [12], analytical methods were developed to quantify the heat transfer between academic configurations involving planar media and dipoles, spheres, cylinders (see e.g. respectively [4,13–15]). These approaches are, however, limited to simple geometries. Numerical methods used in NFRHT are based on those for electromagnetic wave scattering and propagation. The Finite Difference Time Domain (FDTD) method [16] was used for computing the heat exchanged between arbitrary geometries in a statistical manner. It consists in applying the fluctuation-dissipation theorem to the Poynting vector averaged over many simulations by considering the random values of surface currents. The drawbacks of this method are the long computational time and the accuracy of the numerical results due to various numerical errors such as discretization of objects and numerical dispersion intrinsic to the space-time scheme. The radiative heat transfer can be also computed by implementing the Thermal Discrete Dipole Approximation (T-DDA) and the BUlk Field Formulation of ElectroMagnetism (BUFF-EM) methods. The T-DDA method was proposed initially for modelling radiative heat transfer between three-dimensional arbitrary compact objects [17] and then extended to finite-size object close to an infinite surface [18]. This method consists in discretizing

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the objects in sub-volumes, each considered as a thermally oscillating dipole. The BUFF-EM is a free, open-source software implementation of the frequency-domain volume-integral-equation (VIE) method of classical electromagnetic scattering [19,20]. It consists in modelling the bodies by volume (tetrahedral) meshes. The last approach we will discuss is based on the surface-integral-equation (SIE) formulation of classical electromagnetism that allows direct application of the boundary element method (BEM) [21]. It attracted a lot of attention because it offers considerable flexibility to handle arbitrary shapes. Contrary to the T-DDA and BUFF-EM methods, it requires to discretize the surface and not the volume, so that less mesh elements could be required. In this work, we use the implementation of this approach provided in the open-source SCUFF-EM software package developed at MIT [22]. It was already used in [7,8] for the analysis of the tip-sample radiative heat transfer, but not in a comprehensive way.

The paper is organized in two main parts devoted (i) to the SCUFF-EM computations (Sections 2 and 3) and (ii) to the AFM tip-sample radiative heat transfer (Section 4). We first perform calculations of thermal radiative emission and radiative transfer involving homogeneous spheres in Section 2 to validate the SCUFF-EM code against asymptotic formulae. Section 3 deals with thermal transfer between a sphere and a planar substrate. This configuration is of interest in scanning thermal microscopy where the probe tip can sometimes be approximated by a sphere [7,11,23]. Furthermore, spheres attached to tips have been used for experimental investigations of near-field thermal radiation [4,5]. This section explains which requirements are needed to simulate a planar surface. Sections 2 and 3 analyse numerical convergence and could be used as a guide for future computations with SCUFF-EM for other geometries. The final section introduces the numerical modelling of heat transfer between a conical probe tip and a planar substrate. The dependence of heat transfer on the gap size between a compact object (sphere or tip) and a substrate is studied in Sections 3 and 4 for highlighting the near-field characteristics. Finally we compare the flux levels computed with recent experiments and investigate the shape of the heat flux distribution on top of the sample in order to determine the spatial resolution of radiative heat transfer. This is done for two typical radii of curvature. The numerical results are compared to predictions of the Proximity Flux Approximation [24] (also called the Derjaguin approximation).

2. Validation of SCUFF-EM

In this section, the accuracy of the SCUFF-EM code as a function of object and mesh size will be compared with known analytical results. SCUFF-EM is a free, open-source software implementation of the boundary-element method (BEM) for electromagnetic scattering. SCUFF-NEQ is an application code in the SCUFF-EM suite for studying non-equilibrium (NEQ) electromagnetic-fluctuation-induced phenomena. It gives the radiative heat transfer or emission rates for bodies of arbitrary shapes (spheres, cylinders, interlocked rings, conical shapes [7,21,25], etc.) and arbitrary (linear, isotropic, piecewise homogeneous) frequency-dependent permittivity and permeability. The numerical calculations were performed on the P2CHPD cluster (high-performance computing facility at Université Claude-Bernard Lyon 1). Many nodes may be used for parallel or sequential computing. Each node has 64 GB RAM and two processors with 16 cores Intel(R) Xeon(R) CPU E5-2670@2.6 GHz.

We first consider thermal radiative emission and radiative transfer involving homogeneous spheres, the simplest compact objects. Thermal emission by a homogeneous sphere is known analytically, in a framework related to Mie theory [26,27].

2.1. Thermal emission of a sphere

The thermal radiation power emitted by a sphere is given by

$$Q_{\text{rad}}(T) = \int_0^{\infty} \Theta(\omega, T)\tau_{\text{rad}}(\omega, R)d\omega \tag{1}$$

where R is the sphere radius, $\Theta(\omega, T) = \frac{\hbar\omega}{e^{\frac{\hbar\omega}{k_B T}} - 1}$ is the mean energy of the Planck oscillator at temperature T and τ_{rad} denotes a temperature-independent dimensionless transmittivity that can be computed using the Mie coefficients [26,27].

In this work, only isothermal objects are studied. Two materials are considered: SiO_2 and SiC. The optical properties of SiO_2 are taken from Ref. [28]. The dielectric function of SiC is given by

$$\varepsilon(\omega) = \varepsilon_\infty\left(1 + \frac{\omega_L^2 - \omega_T^2}{\omega_T^2 - \omega^2 - i\Gamma\omega}\right)$$

where $\varepsilon_\infty = 6.7$, $\omega_L = 1.825 \times 10^{14}$ rad.s^{-1}, $\omega_T = 1.494 \times 10^{14}$ rad.s^{-1} et $\Gamma = 8.966 \times 10^{11}$ rad.s^{-1}.

We first consider a single SiC sphere. Fig. 1a compares the spectrum of τ_{rad} of the SiC sphere of radius 0.1 µm, computed for two different meshes, and the results obtained by the analytical model (black curve). Blue crosses correspond to the numerical results obtained for a coarse mesh which consists of 172 nodes and 507 edges with a mesh element size of 0.035 µm. Red circles are the numerical results for the finer mesh with 494 nodes and 1473 edges with a mesh element size of 0.017 µm. We note that τ_{rad} shows two maxima close the SiC resonance ($\varepsilon(\omega) = -2$ for $\omega \simeq 1.5 \times 10^{14}$ rad.s^{-1} and $\omega \simeq 1.75 \times 10^{14}$ rad.s^{-1}). For each frequency, the computation takes 12 s for the coarse mesh and 25 min for the fine mesh. From Fig. 1a, the numerical results are in good agreement with analytical results. As expected, Fig. 1b represents the relative error of numerical results compared to analytical ones. The numerical results converge as the mesh gets finer. However, the relative error stays around 20% close to the resonance and does not decrease when the mesh size is decreased.

Fig. 2a represents τ_{rad} of a smaller SiC sphere of radius 0.0125 µm. Similarly to the previous test case, two meshes are considered: one containing 172 nodes and 507 edges with a mesh element size of 0.004 µm, and another one consisting of 492 nodes and 1467 edges with a mesh element size of 0.002 µm. The relative error is shown in Fig. 2b. Numerical results for the coarse mesh (blue crosses) are consistent with analytical results except in the low-frequency range. These results become worse for the fine mesh because there is an inherent numerical difficulty for the computation at low frequencies or for very small objects, typically for the case where the ratio between the element size and the wavelength is lower than 2×10^{-4}. We will therefore need to pay attention to this point in what follows.

The refractive index $n = \text{Re}(n) + i\text{Im}(n)$ sets two characteristic lengths $\lambda/\text{Re}(n)$ and $\lambda/\text{Im}(n)$ to which the mesh size should be compared. As a result, we now study the effect of the refractive index n on the convergence by analyzing the variation of τ_{rad} of a sphere of 10 µm radius for different meshes at a given frequency 1.88×10^{14} rad.s^{-1} (the wavelength in vacuum is 10 µm). Two meshes are considered, with mesh element sizes $\Delta x = 1$ µm and $\Delta x = 2$ µm. Fig. 3a represents the relative error as a function of the ratio between Δx and the wavelength in the sphere $\lambda/\text{Re}(n)$, for $\text{Im}(n) = 0.01$ with $\text{Re}(n)$ varying from 10^{-3} to 10^3. Fig. 3b shows the relative error as a function of the ratio between Δx and the penetration depth $\lambda/(2\pi\text{Im}(n))$, for $\text{Re}(n) = 1$ with $\text{Im}(n)$ varying from 10^{-2} to 10^4. Red curves correspond to the results for the coarse mesh and blue curves are results obtained for the fine mesh. We observe that when the mesh size is reduced twice, relative errors decrease three times while the computational