Project No: 254996

Project Acronym: OPTONANOMECH

Project Full Name: Operation of Cavity Optomechanics in Fluids for Ultrasensitive Mass Detection

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AGENCIA ESTATAL CONSEJO SUPERIOR DE INVESTIGACIONES CIENTIFICAS

Marie Curie Actions

Final Report

Nanomechanical resonators, especially nanocantilevers, have become the basis of a wide variety of applications from scanning probe microscopy to mass sensing. The concept is the measurement of the resonance frequency variations that in the case of scanning probe microscopy arise from the intermolecular forces between the microcantilever and the probed surface; and in the case of mass sensing are induced by the mass added on the surface of the resonator. The ever increasing control in nano-fabrication techniques give as result the size shrinking of the resonators to the nanoscale, which has allowed the attainment of unprecedented sensitivities of zeptogram-scale mass sensing and zeptonewton-scale force detection. These achievements have enabled milestones such as single electron-spin paramagnetic resonance and protein spectrometry.

In parallel, last years have witnessed a crescent interest in the interplay of light and matter. In that sense, the optomechanics field has rapidly grown for exploiting the forces exerted by the electromagnetic radiation on mechanical structures. The canonical experiment consists of positioning a freestanding mechanical element with a high-reflectivity coating near a mirror to establish an optical cavity with high finesse. This conceptually simple experiment was born as the basis of the current gravitational wave detectors, in which a moving mirror substitutes one of the reference mirrors of an optical Michelson interferometer. More complex embodiments include coupling the mechanical element to microtoroidal cavities, to high-finesse Fabry−Peŕot cavities formed between two macroscopic rigid mirrors and the use of external phononic bandgap shields. A primary goal of optomechanics is to exploit the optical forces that emerge in the cavity, usually referred as radiation pressure, to cool down the fundamental vibration mode of the mechanical system. This goal has inspired analogue methodologies such as the electromechanical coupling between a mechanical resonator and a quantum bit. We believe that the advances in optomechanics can also be exploited for enhancing the detection limits of nanomechanical sensors. However, when the mechanical system dimensions are shrunk beyond the optical wavelength, strong diffraction effects emerge and the optical back-action is largely reduced.

The goal of the project OPTONANOMECH is to circumvent the problems derived from the diffraction limit by developing a nano-optomechanical platform to study the dynamics of nanomechanical systems. Following an approach orthogonal to current MEMS/NEMS, integrated photonic circuits are employed for device actuation and detection. The nanomechanical resonator forms part of the photonic circuit; therefore, the actuation and detection of the resonator motion can be achieved all-optically. The integrated approach circumvents the diffraction limit native to MEMS/NEMS systems and allows for unprecedented sensitivity. This all-photonic approach eliminates the need for establishing electrical connections to the nanomechanical resonator. Therefore geometrical limitations imposed by the required isolation for electrical interconnects are eliminated. In addition, optical cavities can be employed to obtain enhanced sensitivity and to provide amplified dynamic feedback. Our concept allows us to transfer the non-linear potential imposed by the light to the mechanical resonance. Optical cavities are also used as sensors themselves; therefore, this is a good opportunity to combine the optical spectroscopy with mass sensing using NEMS. As it was described in the original proposal, the main objective of the project is to reach a mass sensitivity of 10^{-20}g in physiological environments.

During the development of this project, the researcher has demonstrated that the optical resonances associated to the light confinement in subwavelength semiconductor structures allow to efficiently extend optomechanics to nanoscale objects such as silicon nanowires. This discovery opens the door to novel cavity designs to efficiently cool tiny mechanical elements with the prospect of achieving the quantum limit more quickly than with micromechanical structures. On the other hand, this phenomenon enables the development of self-sustained nanowire oscillators in which the optical energy is converted into mechanical vibration at the natural frequency with unprecedented stability. These devices offer significant advantages with respect to previous frequency detection methods, as there is no need of external excitation, which allows a high degree of simplification in the design of sensors based on nanowires.

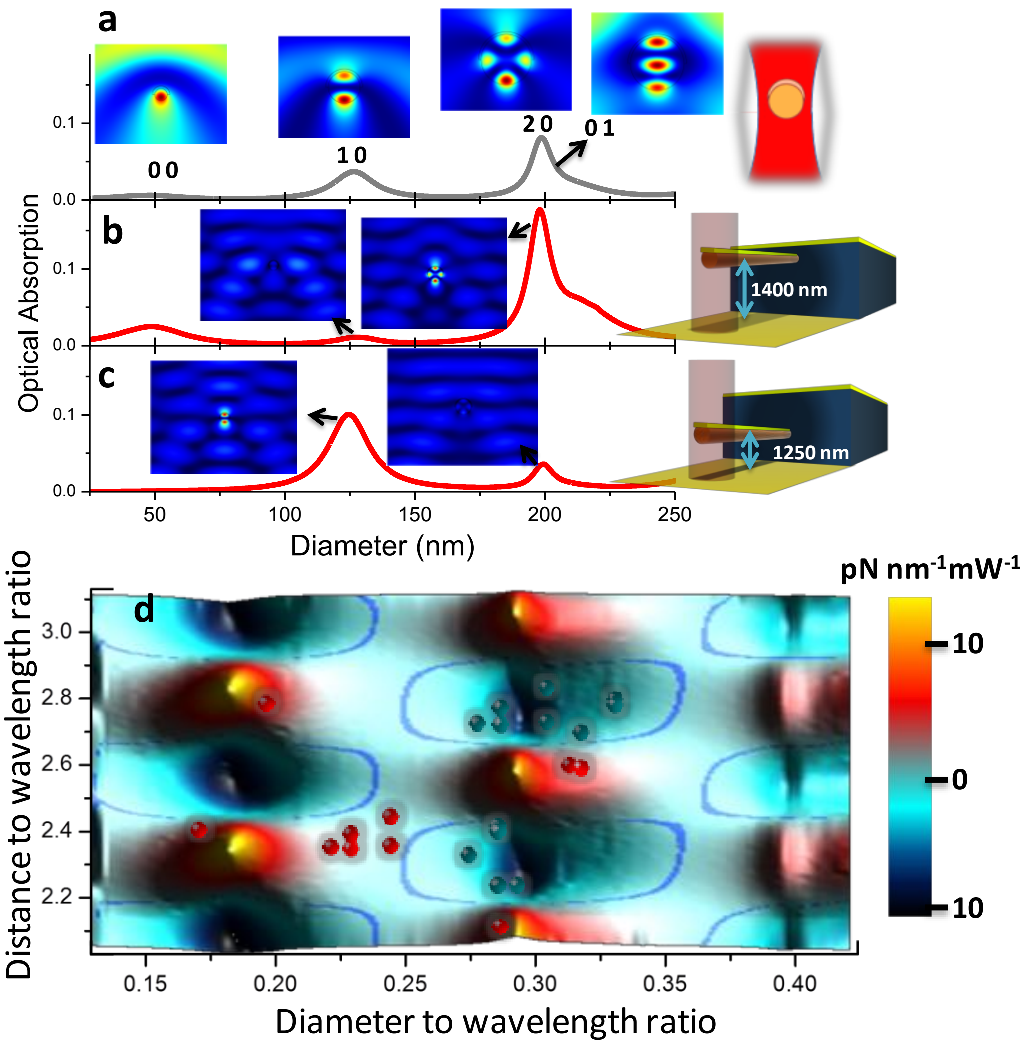

Figure 1. Effect of light confinement within the nanowire on the optomechanical coupling. a, Theoretical calculation of the power absorption of the gold coated nanowires free in space as a function of the nanowire diameter. The series of peaks featuring the spectrum can be associated, to a very good approximation, with the morphology-dependent optical resonances, also known as whispery gallery modes or just Mie resonances. The distributions of the square of the electric field norm for the first four optical resonances are also included. For the diameters and wavelengths used in this work, the nanowires support approximately three optical resonances that correspond with the modes (1,0), (2,0) and (0,1). **b, and c,** Power absorption of the gold coated nanowires when are situated at 1400 and 1250 nm from the substrate, respectively. The interference field

modulates the power absorbed by the electromagnetic modes of the nanowire. This effect is significant in the absorption of the modes (1 0) and (2 0) whose electrical intensity distribution can be observed in the insets **d,** Calculated color intensity map of the feedback gain of the cavity per light power unit at a wavelength of 633 nm, respectively, as a function of the distance between the nanowire and the substrate and the diameter of the nanowire, both normalized to the incidence wavelength. The calculations were performed for an incidence power of 100 μW and a beam spot size of 2 μm. The contour lines separate the regions of cooling and amplification. The experimental diameter and cavity distance coordinates of the nanowires in which amplification and cooling was observed are included as red and cyan circles, showing a good agreement with the theoretical predictions.

The experimental measurements of the tapered NWs resonance frequencies were performed using a homemade hybrid interferometric optical system[1-3]. The nanowire size actively selects the amount of light confined within the nanostructure, depending on the incident laser wavelength. The researcher also reported the use of similar confined electromagnetic modes in sub-wavelength structures such as diamond triangular nanobeams[4,5]. Evanescent fields emanating from these confined modes overlap with the standing wave generated by reflections from the silicon surface. Therefore, the phase contrast caused by nanowire vibrations produces a detectable signal, fig1.a-c. Depending on the sign of the optomechanical feedback force[1,2], the effective damping acting on the resonator can be positive (cooling, blue regions in fig.1d) or negative (amplification, red areas in fig.1d).

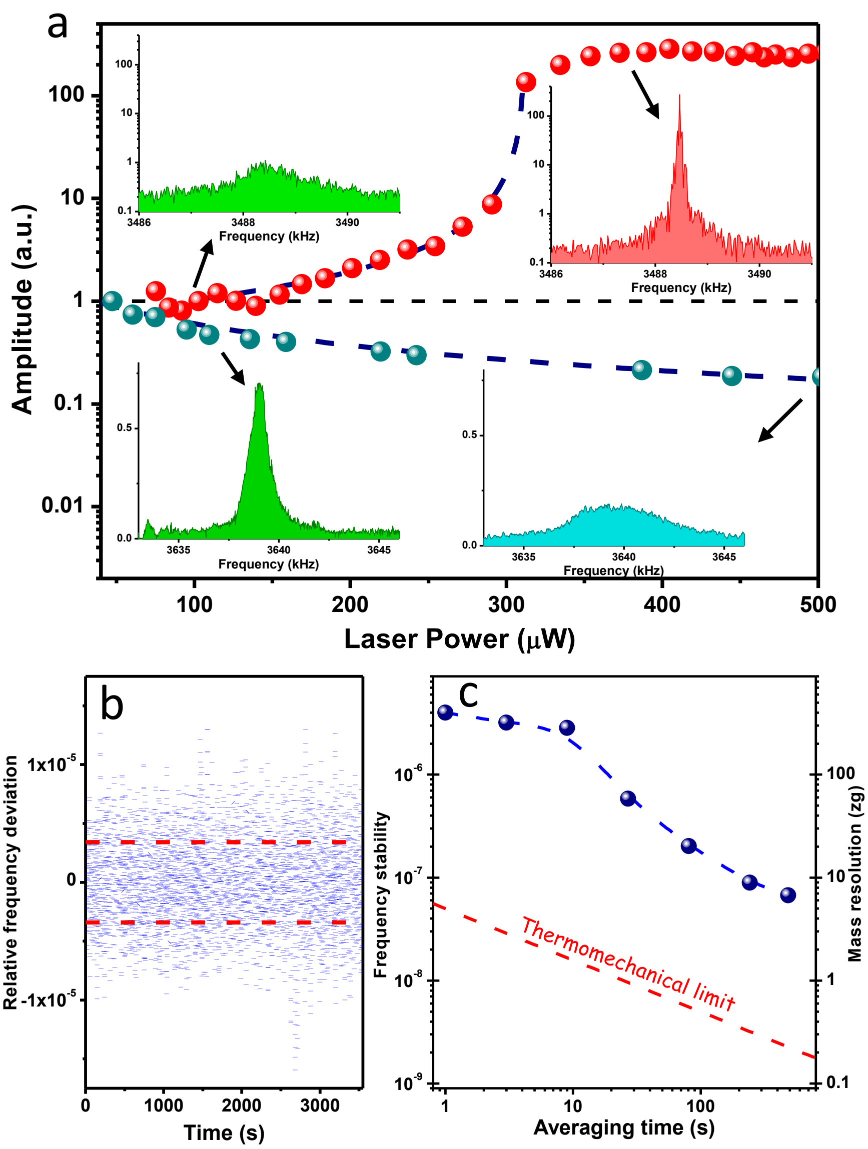

Figure 2. Effect of the laser intensity on the optomechanical coupling and frequency stability of self-sustained nanowire oscillators. a, Amplitude at resonance of two nanowires as a function of the laser power intensity (symbols). The fundamental vibration mode of one nanowire is optically amplified (red symbols) whereas is cooled in the other case. The effective temperature achieved for the cooling case is of about 60 K for a laser power of 500 μW. In the case of optical amplification, the amplitude moderately grows with the laser intensity until it undergoes an abrupt increase of about 20 times when the laser power is 310 μW. Above this critical laser power, the nanowire exhibits self-sustained oscillation. The insets show the amplitude spectral density of the thermomechanical fluctuation for the two nanowires for a laser power of 50 and 350 μW. In the case of optical amplification, the achieved quality factor is of 263 000 that corresponds to an extremely small linewidth of 23 Hz. **b,** One hour recording of the relative frequency deviations of the self-sustained nanowire oscillator. The integration time is of one second and the frequency is of 3505 kHz. The red dashed lines represent the standard deviation of the frequency. **c,** Standard deviation of the relative frequency shifts and corresponding mass resolution as a function of the

integration time. The mass resolution is approximately given by the product of two times the effective mass of the nanowires (≈50 fg). The red dashed line represents the ultimate frequency stability imposed by the thermomechanical limit at room temperature.

We apply the discovered optomechanical phenomena to induce self-sustained oscillations in nanowires with very high spectral purity[1]. This state is reached when the gain of the feedback loop reaches a critical value in which the damping rate becomes negative. The transient oscillation then grows exponentially until the nonlinear saturation mechanisms of the system come into play and the oscillation reaches a steady state with virtually no noise in the amplitude and small phase fluctuations of diffusive nature, fig.2. Here the saturation comes from the nonlinear behavior of the light intensity modulation. The transition from optical amplification to self-oscillation is seen as an abrupt increase of the amplitude of several orders of magnitude when the laser intensity reaches a critical value that is of 310 μW in the case shown in Figure 2a. We characterize the frequency stability of the self-sustained oscillation of the nanowire for a laser power of 350 μW. The frequency fluctuations for an averaging time of 1 s were of 3×10^{-6} during one hour of experiment. Figure 2c shows the standard deviation of the frequency as a function of the integration time. For integration times of about one minute, the frequency fluctuations are reduced to 10^{-7}. The ultimate limit in the frequency noise of the self-sustained oscillation is imposed by the phase diffusion that approximately follows the Einstein equation for diffusion processes. A compelling feature of resonant nanowires is their capability for ultrasensitive mass sensing and for discerning subtle variations in the Young's modulus, which point out these devices as promising tools for biological nanomechanical spectrometry. In this sense, we have plotted the mass resolution based on the frequency stability values in Figure 2c. Our current mass sensitivity is of 100 zg for integration times of few seconds. The sensitivity can be largely enhanced by using smaller nanowires, higher vibration modes and by optimization of the readout setup. The achievement of a mass resolution of 0.1 zg (10^{-22} g) would open the door for weighing proteins with sensitivity down to a single aminoacid.

In order to characterize the ultimate limits of our optical detection method, we synthetized structures in the CVD (chemical vapor deposition) chamber grown under particular conditions to obtain that the nanowire diameter linearly decreases from the clamped to the free end[3]. Tapering during growth occurs via two possible mechanisms: on the one hand, gradual size reduction of the metal catalyst gold nanoparticle during growth by either diffusion, evaporation or chemical reaction results in a time-varying nanowire growth diameter; on the other hand, dissociative adsorption on the gas/solid interface produces a progressive thickening in the radial direction. Although tapered Si NWs have been usually considered undesirable for most applications, and many efforts regarding growth optimization have focused on obtaining uniform NWs with negligible diameter variation, this effect offers exceptional opportunities for obtaining devices with matchless performance characteristics that were also studied during the development of this project[3].

We measured the profiles of the reflectivity amplitude at the fundamental mechanical resonance[6] of the nanowire for transversal magnetic (TM) and transversal electric (TE) laser polarizations (Fig. 3a, circles). We have also included the theoretical profile of the nanowire oscillation amplitude (in spectral density units) calculated by combining finite element method (FEM) simulations and the fluctuation-dissipation theorem for comparison[6] (Fig. 3a, line). The measured reflectivity amplitude due to the thermomechanical displacement exhibits maxima and minima along the nanowire axis, which is unexpected. In other words, the reflectivity amplitude is not proportional to the vibration amplitude as it occurs in the linear regime of Fabry-Perot cavities. For TE polarization, reflectivity amplitude maximum is located near the fixed end (x_0≈2.5 μm), where the vibration amplitude is smaller. In Fig. 3b, we plot the vibration amplitude versus the reflectivity amplitude for the nanowire region near the clamping (x_0<2.5 μm). Displacement sensitivity on the verge of 1 fm/Hz$^{1/2}$ is achieved for our reflectivity noise, 10^{-7}. The obtained sensitivity is even one order of magnitude better than that obtained with state of the art interferometry on reflective flat surfaces with areas that largely exceed the wavelength; and it is three orders of magnitude higher than the previously reported sensitivities in nanowires. On the contrary, the nanowire is almost undetectable for TM polarization near the free end (at x_0≈9 μm) where the vibration amplitude is higher. The displacement sensitivity strongly depends on the light polarization as shown in Fig. 3c for different laser beam positions along the nanowire. In all cases, the maximum light confinement is achieved at either, TM or TE polarizations. The displacement sensitivity when the laser beam is focused near the middle of the nanowire (position (a)) enhances for TM polarization. Conversely, the displacement sensitivity at the illumination position (b) closer to the free end enhances for TE polarization. At position (c), near the free end, the displacement sensitivity is almost insensitive to the light polarization.

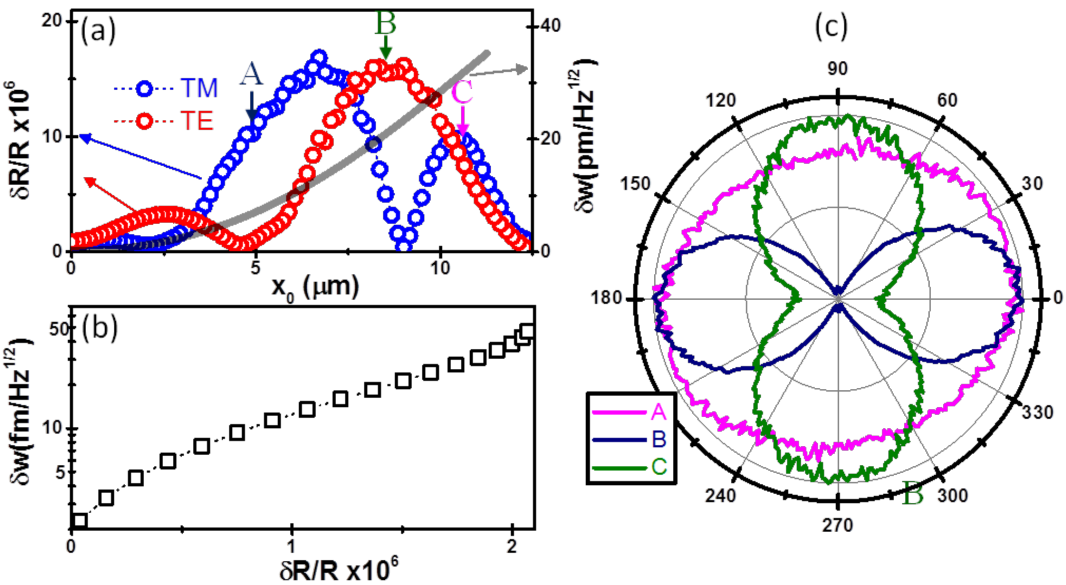

Figure 3. Measuring the reflectivity fluctuations along a tapered nanowire. (A) Experimental profiles of the reflectivity amplitude (symbols) at the fundamental mechanical resonance of the nanowire, and vibration amplitude profile (line) calculated by the finite element method and the fluctuation-dissipation theorem. The reflectivity amplitude is non monotonic along the nanowire, exhibiting maxima and minima, indicating that the reflectivity and vibration amplitudes are not linearly correlated as it occurs in Fabry-Perot cavities. **(B)** Vibration amplitude versus the reflectivity amplitude for the nanowire region near the clamping (x_0<2.5 μm). Displacement sensitivity on the verge of 1 fm/Hz$^{1/2}$ is achieved. **(C)** Experimental reflectivity amplitude as a function of the angle between the electric field polarization vector and the nanowire axis for the laser beam positions, x_0, marked in **(A)** as (a), (b) and (c).

This sensitivity value provides enough room to detect shorter nanowires that exhibit higher resonance frequencies and smaller masses. This directly results in higher sensitivity in mass sensing or force gradient detection. In Fig. 4a we plot the theoretical thermal amplitude at resonance as a function of the nanowire length in vacuum (quality factor≈3000) and in aqueous solution (quality factor≈3). The optical detection paradigm demonstrated in this project would enable the measurement of the Brownian fluctuations at resonance of nanowires with lengths of ≈200 nm and 700 nm in vacuum and liquid, respectively. The mass sensitivity in vacuum without excitation would be of few hundreds of yoctogram, and near one yoctogram with external excitation (Fig. 4b). This outstanding sensitivity can potentially be applied for obtaining new insights into diffusion of atoms and molecules along nanowires as well as for chemical identification of molecules and proteins. Aqueous solutions are the natural environment where biological molecules adopt their native structure and interact with other molecules to perform complex functions in cells. Also, optical detection is best suited for liquid environments. The viscous damping highly reduces the quality factor, but this price is cheap compared with the fascinating applications that tiny nanowires can find in bionsensing and intracellular sensing. Few hundreds of zeptograms can be detected with short nanowires in liquid, which corresponds to the main objective of the project OPTONANOMECH. This sensitivity would enable the detection of single ligand-receptor events. In addition, nanowires can safely penetrate the plasma membrane and enter biological cells for drug and gene delivery, electrophysiology and endoscopy. We envision that our findings will also place at reach intracellular sensing based on nanowires.

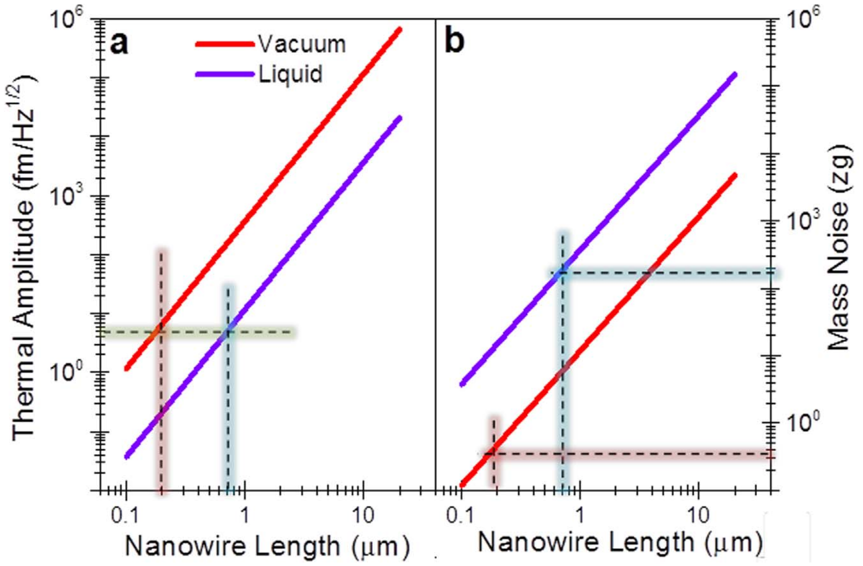

Figure 4. Detection limits of a mass sensor based on a 50 nm wide nanowire. (A) Theoretical thermal amplitude at resonance and **(B)** mass detection limit with no external excitation as a function of the nanowire length for quality factors of 3000 (that mimics the vacuum situation) and 3 (that mimics the situation when the nanowire is in liquid). The theoretical displacement detection limit is of about 5 fm/$Hz^{1/2}$ for laser powers of ≈400 μW that enable the measurement of the thermal fluctuations at resonance of nanowires with lengths of ≈200 nm and 700 nm in vacuum and liquid, respectively. The resulting mass sensitivity for these nanowires is ~100 yoctograms in vacuum and ~100 zeptograms in liquids.

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