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ULTRAFAST DYNAMICS OF HEAT GENERATION IN PLASMONIC NANOSTRUCTURES

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14. ABSTRACT

To study heat generation in plasmonic nanostructures, Zhang group has developed analytic and numerical methods to study the dynamics of plasmonic structures. The group has also studied the substrate effect on the plasmonic structures. To demonstrate the effect of the substrate on the phonon modes, the mechanical motion has been simulated as a function of frequency for different coupling strengths between the nanostructure and the substrate.

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1 Summary

To study heat generation in plasmonic nanostructures [1], Zhang group has developed analytic and numerical methods to study the dynamics of plasmonic structures. The group has also studied the substrate effect on the plasmonic structures [2]. To demonstrate the effect of the substrate on the phonon modes, the mechanical motion has been simulated as a function of frequency for different coupling strengths between the nanostructure and the substrate. Such nanostructures can form resonances for the enhancement of electromagnetic intensity, resulting in numerous applications including quantum electrodynamics, optical nonlinearities, optical forces, surface enhanced Raman spectroscopy and biosensing.

2 Introduction

The objective of this project is to investigate the ultrafast dynamics of heat generation in plasmonic nanostructures with designed geometries [1]. The dynamic processes are studied both analytically and numerically. The understanding of such a dynamic process and the development of such a methodology enable our capability in designing and optimizing nanoscopy plasmonic heat sources [3] for customized applications, which is of critical important for civilian and defense applications, in particular to sensor system thermal management relevant to Air Force and Department of Defense.

3 Methods, Assumptions, and Procedures

We have developed analytic and numerical methods to study the dynamics of a gold sphere in a glass matrix. The radial displacement at the surface shows a resonance corresponding to the phonon mode. The analytic and numerical solutions both give a resonant frequency and a quality factor in reasonable agreement with the approximate values. We have also studied the effects on the nanoscale cross structures. To demonstrate the effect of the substrate on the phonon modes, we simulate the mechanical motion as a function of frequency for different coupling strengths between the nanostructure and the substrate.

4 Results and Discussion

We have studied plasmonic nanostructures that exhibit localized phonon modes. The phonon modes of a metal nanostructure are high frequency mechanical distortions of the metallic lattice, and are the solutions of the Navier equation [4] with continuity of displacement and stress at the nanostructure boundaries. The phonons modify the optical properties of a nanostructure through mechanical distortion of the geometry and strain-induced modifications of the refractive index of the metal. The two contributions to the optical response are of the same order of magnitude. Through their sensitivity to geometry and material properties, the surface plasmon serves as a local probe of coherent acoustic phonons. In time domain spectroscopy (pump-probe), the pump excites a plasmon which rapidly decays to non-thermalized electrons [5]. The electrons thermalize through scattering processes on a sub picosecond timescale raising the electronic temperature and changing the interband transition rates. The electrons then rapidly heat the lattice through electron-phonon scattering which generates coherent acoustic phonons, the sinusoidal oscillations with a period of ~100 ps.

5 Conclusion

Using the finite element solver, we have calculated, in the frequency domain, the response of a nanoparticle to an impulse strain in the metal due to the pulsed laser excitation (Figure 1). The thermal strain generates a force which sets the particle in motion. Given the mechanical motion, we take a simple model for the detection by the plasmon and assume the transient transmission change will be proportional to the change in length.

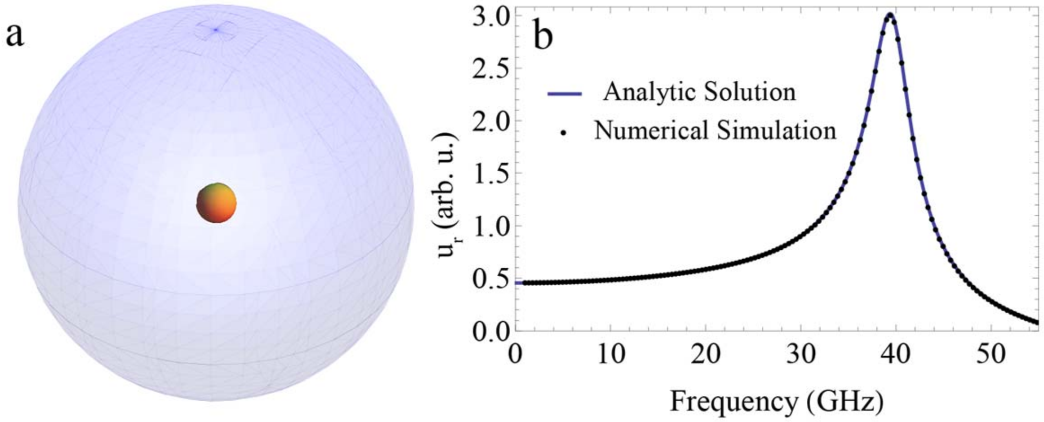

Figure 1. Analytic and numerical solutions for a gold sphere in a glass matrix.

We calculate the dynamics of a 40 nm gold sphere in response to an impulsive thermal strain. (a) A schematic of the system consisting of a sphere in the center of an infinite glass medium. (b) The radial displacement at the surface shows a resonance corresponding to the phonon mode. The analytic and numerical solutions both give a resonant frequency of 39.59 GHz and a quality factor of 9.4. These values are also in reasonable agreement with the approximate resonance frequency and quality factor given by the simple relations:

$$f \approx v_p / (2R) \approx 40.5\ \text{GHz} \ \text{ and } \ Q \approx \pi \rho_{Au} v_{p,Au} / (2 \rho_{SiO_2} v_{p,SiO_2}) \approx 7.49\,. \qquad (1)$$

We have also studied another model plasmonic structure – the cross nanostructures that exhibit complex localized phonon modes (Figure 2). The mode in which the two arms oscillate in phase is the symmetric mode and the mode where the arms oscillate out of phase is the anti-symmetric mode. [2] These modes can be viewed as the hybridization of the extensional modes of the two arms. The modes are symmetric and anti-symmetric in the context of coupled mode theory and not in terms of spatial symmetry. The phonon excitation is simulated by calculating the thermal expansion of the nanoparticle due to a raised temperature, relative to that of the substrate. The temperature increase causes a strain proportional the temperature change and the coefficient of thermal expansion of the gold. The distorted geometry is shown in Figure 1(a) with a color scale to indicate the magnitude of the displacement from the original geometry, where red indicates a larger displacement. The displacement is then calculated for each frequency given the thermal stress. We use periodic boundary conditions on the outer boundaries and a perfectly matched layer on the bottom to absorb propagating acoustic waves. The detection of the phonon by the plasmon is simulated by assuming the shift in the plasmon resonance, and thus the amplitude of the differential transmission, is proportional to the change in length along the axis parallel to the probe polarization. The length change is calculated by integrating the displacement parallel to the probe polarization across that face of the nanostructure.

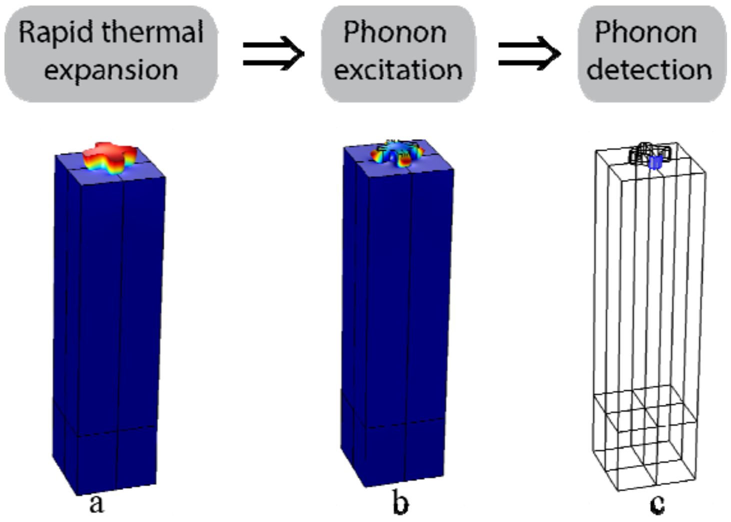

Figure 2. Simulation procedure of cross nanostructures.

Figure 3 shows the results of nanomechanical simulations of the phononic response of isolated metallic nanostructures and nanostructures in contact with a fused silica substrate. An isolated bar (a,b, blue) has one resonance (extensional mode) and two when coupled to a substrate (a,b, green). An isolated cross (c,d, blue) with non-equal arms has two modes, the symmetric and anti-symmetric modes. When coupled to the substrate (c,d, green), more modes are excited. In the isolated symmetric cross (e,f, blue), only the symmetric mode is excited due to the symmetry of the excitation process, despite the anti-symmetric mode being an eigenmode of the system. The largest amplitude peaks are not affected by the lattice period and thus represent localized acoustic vibrations rather than surface acoustic waves.

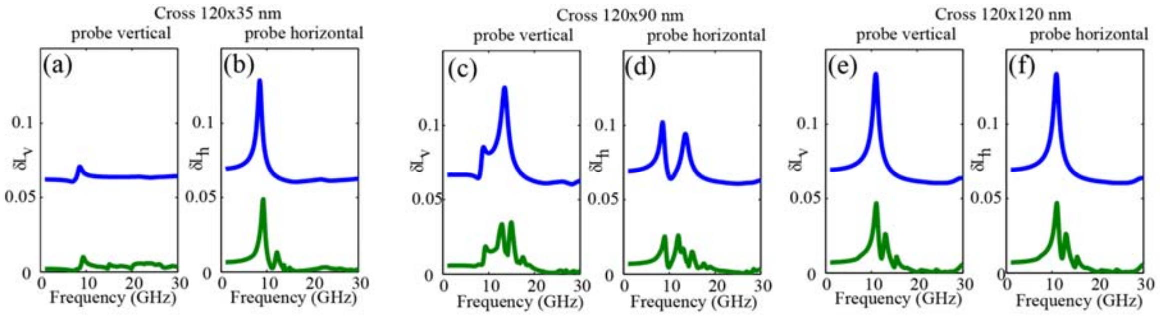

Figure 3. Nanomechanical simulations of the phononic response of isolated metallic nanostructures (blue) and nanostructures in contact with a fused silica substrate (green).

The polarization manipulation of the probe surface plasmon allows selective detection; manipulating the pump polarization, however, does not allow selective excitation due energy transport by super diffusive electrons. The amplitudes of the phonon eigenmodes can be controlled through design of the nanostructures and understanding of the hot electron dynamics. In particular, we use the fact that the phonons are excited due to an isotropic heating to prevent the excitation of anti-symmetric modes by designing the structure to be symmetric. The understanding of the ultrafast dynamics of plasmonic heat sources at nanoscale will open the door for many applications in physics, biology, and materials, such as plasmonics enhanced catalysis, optofluidics, photothermal cancer therapy.

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