Fourier reconstruction of picosecond acoustic pulses in femtosecond plasmonic measurements

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Abstract

Fundamental interactions induced by lattice vibrations on ultrafast time scales have become increasingly important for modern nanoscience and technology. Experimental access to the physical properties of acoustic phonons in the terahertz frequency range and over the entire Brillouin zone is crucial for understanding electric and thermal transport in solids and their compounds. Among different metal-based compounds the hybrid metal-ferromagnet multilayer structures attract particular interest because of their applications in ultrafast spintronics [1,2], magneto-plasmonics [3,4] and acousto-plasmonics [4].

Most recently, the generation and nonlinear propagation of giant (displacement in the order of 1% of the lattice constant) acoustic strain pulses in hybrid gold/cobalt bilayer structures was monitored by femtosecond plasmonic interferometry [5]. This new technique shown in Fig. 1 allows for unambiguous characterization of arbitrary ultrafast acoustic transients.

The hybrid acousto-plasmonic gold/cobalt/sapphire multilayer structures were manufactured by magnetron sputtering of a (111) - textured gold layer on top of an hcp-cobalt film deposited on a (0001) sapphire substrate. The 35 nm thin ferromagnetic cobalt layer was excited through the substrate by an ultrashort optical pump pulse and served as an efficient opto-acoustic transducer. Due to a very short electronic mean free path the diffusion of hot electrons in cobalt is particularly inefficient and the heat penetration depth only slightly exceeds the 10 nm skin depth of the pump radiation (at 400 nm optical pump wavelength). Thermal expansion of the cobalt transducer launches a unipolar acoustic pulse in both directions. This compressional acoustic strain pulse $\eta(z,t)=\eta(t+z/c_s)$ creates a layer of higher ion density which moves at sound velocity c_s=3.45 km/s in gold. As the stationary charge separation between electrons and ions in a metal is prohibited by the tiny Debye radius, the spatial profile of the electron charge density exactly follows the ionic one. Therefore, an ultrashort acoustic pulse creates a time-dependent spatial profile of the dielectric function $\delta\varepsilon'(z,t)=\varepsilon'(1+\eta(z,t))$ inside the metal, which modulates the surface plasmon wave vector k_{sp},

$$\delta k_{sp}(t)=\frac{k_0}{2|\varepsilon_m|^2}\delta\varepsilon'(t)=-\frac{k_0}{2|\varepsilon_m|\delta_{skin}}\int_0^\infty \eta(z,t)\exp\left(-|z|/\delta_{skin}\right)dz \qquad (1)$$

when the strain pulse arrives within the surface plasmon skin depth δ_{skin}=13 nm at the gold-air interface, Here $k_0=2\pi/\lambda$ denotes the vawav vector of light in vacuum the value for the complex dielectric function of gold at 1.55 eV probe photon energy reads $\varepsilon_m=\varepsilon'+i\varepsilon''$=-24.8+1.5i.

Experimental data in Fig. 1b demonstrate that the acoustic reflection indeed dramatically changes the real part of surface dielectric function $\delta\varepsilon'(t)$, whereas the imaginary part $\delta\varepsilon''(t)$ shows a feature-less thermal background.

One of the main goals of acousto-plasmonic measurements consists in the reconstruction of the unknown acoustic pulse from the strong 'acoustic echo' observed at the pump-probe delay time of about 70 ps. Taking into account the reflection of the acoustic pulse at the gold-air interface according to $\eta(z,t)=\eta(t+z/c_s)-\eta(t+z/c_s)$, equation (1) can be presented in a more convenient form

$$\delta\varepsilon'(t)=\frac{|\varepsilon_m|}{\tau_{skin}}\int_{-\infty}^{\infty}\eta(t')\exp\left(-|t-t'|/\tau_{skin}\right)\mathrm{sgn}(t-t')dt' \qquad (2)$$

Equation (2) represents a convolution of the unknown time-dependent strain $\eta(t)=\eta(z=0,t)$ with the opto-acoustic response function determined by the acoustic travel time $\tau_{ac}=\delta_{skin}/c_s$=3.8 ps through the skin depth of the surface plasmon polariton. Using convolution theorem this equation can be easily solved in the Fourier domain to obtain the strain pulse

$$\eta(\omega)=\frac{(1+\tau_{skin}^2\omega^2)}{2i\omega\tau_{skin}}\frac{\delta\varepsilon'(\omega)}{|\varepsilon_m|} \qquad (3)$$

Taking the inverse Fourier transform of $\eta(\omega)$ gives us the desired strain $\eta(t)$. The results of strain reconstruction by applying the Fourier algorithm to the experimental data in Fig. 1b are shown in Fig. 1c. Not only the main acoustic pulse but also the very weak secondary acoustic reflections at gold-cobalt and cobalt-sapphire interfaces can be accurately reconstructed using this algorithm. The only condition to the experimental data is the zero time integral under the curve $\delta\varepsilon'(t)$, which is achieved by the subtraction of the slowly varying background.

The high-resolution zoom of the acoustic pulse presented in Fig. 1d is in excellent agreement with the theoretically predicted pulse shape [5]. We have tested this fairly simple Fourier algorithm with many acousto-plasmonic data sets and obtained surprisingly good results. Therefore we believe that the described Fourier-based "one-click" reconstruction algorithm for ultrashort acoustic pulses represents a robust alternative to the iterative method proposed earlier [5]. We expect that the developed Fourier method can be also successfully applied to extract the acoustic pulse shapes from much simpler time-resolved reflectivity measurements [6].

References

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Figures

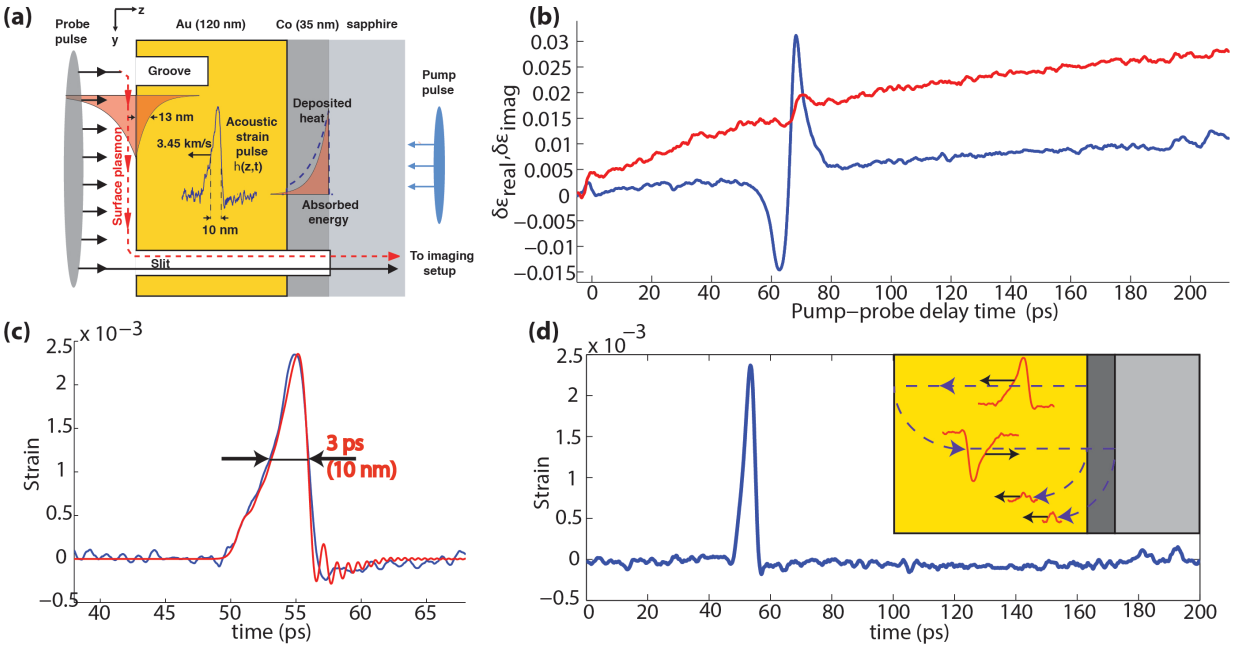

Fig. 1: (a) Schematic drawing of the acousto-plasmonic pump-probe experiment: surface plasmons propagating at the gold-air interface, probe the reflection of acoustic pulses generated in laser-heated cobalt transducer. (b) Ultrafast dynamics of the real (blue line) and imaginary (red line) parts of the gold dielectric function measured with femtosecond plasmonic interferometry. (c) The Fourier reconstructed strain shows the main acoustic pulse as well as two secondary reflections from the gold-cobalt and cobalt-sapphire interfaces. (d) The reconstructed acoustic pulse (blue line) is in excellent agreement and with a 3 ps long theoretical pulse (red line), which resembles the exponential profile of deposited heat in cobalt. See Ref. [4,5] for more details.