Reconstruction of acoustic pulses for gold-cobalt bilayer structures probed with femtosecond surface plasmons

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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] and magneto-plasmonics [3,4], with most previous studies devoted to their electronic properties.

Here we extend these studies to investigate ultrafast coherent phonon pulses and report on 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, monitored with ultrafast surface plasmon interferometry [5]. This new technique is presented in Fig. 1. It allows for unambiguous characterization of arbitrary ultrafast acoustic transients.

A hybrid acousto-plasmonic 120 nm gold/35 nm cobalt/ sapphire multilayer structure was manufactured by magnetron sputtering of a (111)-oriented gold layer on top of an hcp-cobalt film deposited on a (0001) sapphire substrate. The 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 wavelength). Thermal expansion of the cobalt transducer launches a unipolar acoustic pulse in both directions. This compressional acoustic strain pulse $\eta(z,t)$ 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 inside the metal, which modulates the surface plasmon wave vector k_{sp}, when the strain pulse arrives within the surface plasmon skin depth δ_{skin}=13 nm at the gold-air interface.

We will show how to reconstruct the acoustic strain from the plasmonic pumb-probe measurements resolving the Fredholm integral equation, which describe the changing of dielectric permittivity due to action of the acoustic strain. Normally, this equation can be presented as follows:

$$\delta\varepsilon'(t) = \frac{|\varepsilon_m|}{\tau_{skin}} \int_{-\infty}^{\infty} \eta(t') \exp\left(-|t-t'|/\tau_{skin}\right) \operatorname{sgn}(t-t')\, dt' \qquad (1)$$

After taking the Fourier transform it is easy to express the equation for strain:

$$T(w) = \frac{\tau_{skin}}{|\varepsilon_m|} \frac{F[\delta\varepsilon'(t)]}{F[\exp\left(-|t-t'|/\tau_{skin}\right)\operatorname{sgn}(t-t')]} = F[\delta\varepsilon'(t)] \frac{\tau_{skin}^2}{|\varepsilon_m|} \frac{1+\tau_{skin}^2 w^2}{iw} \qquad (2)$$

Finally to get the acoustic strain profile from the measurements the thermal background should be removed and only after that the expression (2) can be used, where $\delta\varepsilon'(t)$ stands for experimental data.

This methodic was used to process the data from the set-up explained above with only difference in the thickness of gold layer which 100 nm bigger.

Simple approach to use the Fourier transform shows great results and high conformity with theoretical calculations (Fig. 1 (e)). Therefore this "in-click reconstruction" method can be used as good approach for reconstruction of acousto-plasmonics measurements without need to fit the data manually.

References

[1] S.D. Brosnon, J.G, Fujimoto and E.P. Ippen, "Femtosecond Electronic Heat-Transport Dynamics in Thin Gold Films," Phys. Rev. Lett. **59**, 1962-1965 (1987).

[2] G. Tas and H.J. Maris, “Electron diffusion in metals studied by picosecond ultrasonics,” Phys. Rev. B **49**, 15046-15054 (1994).
[3] K J. Chau, M. Johnson and A.Y. Elezzabi, “Electron-Spin-Dependent Terahertz Light Transport in Spintronic-Plasmonic Media,” Phys. Rev. Lett. **98**, 133901 (2007).
[4] A. Melnikov et al., “Ultrafast Transport of Laser-Excited Spin-Polarized Carriers in Au/Fe/MgO(001),” Phys. Rev. Lett. **107**, 076601 (2011).
[5] V.V. Temnov et al., “Active magneto-plasmonics in hybrid metal-ferromagnet multilayer structures,” Nature Photonics **4**, 107-110 (2010).
[6] V.V. Temnov, “Ultrafast acousto-magneto-plasmonics,” Nature Photonics **6**, 728-736 (2012).
[7] V.V. Temnov et al., “Femtosecond nonlinear ultrasonics in gold probed with ultrashort surface plasmons”, Nature Communications **4,** doi:10.1038/ncomms2480 (2013).
[8] K.J. Manke et al., “Detection of Shorter-Than-Skin-Depth Acoustic Pulses in a Metal Film via Transient Reflectivity’”, in *AIP Conference Proceedings* **1506**, 22-27, doi:10.1063/1.4772519 (2012).

Figures

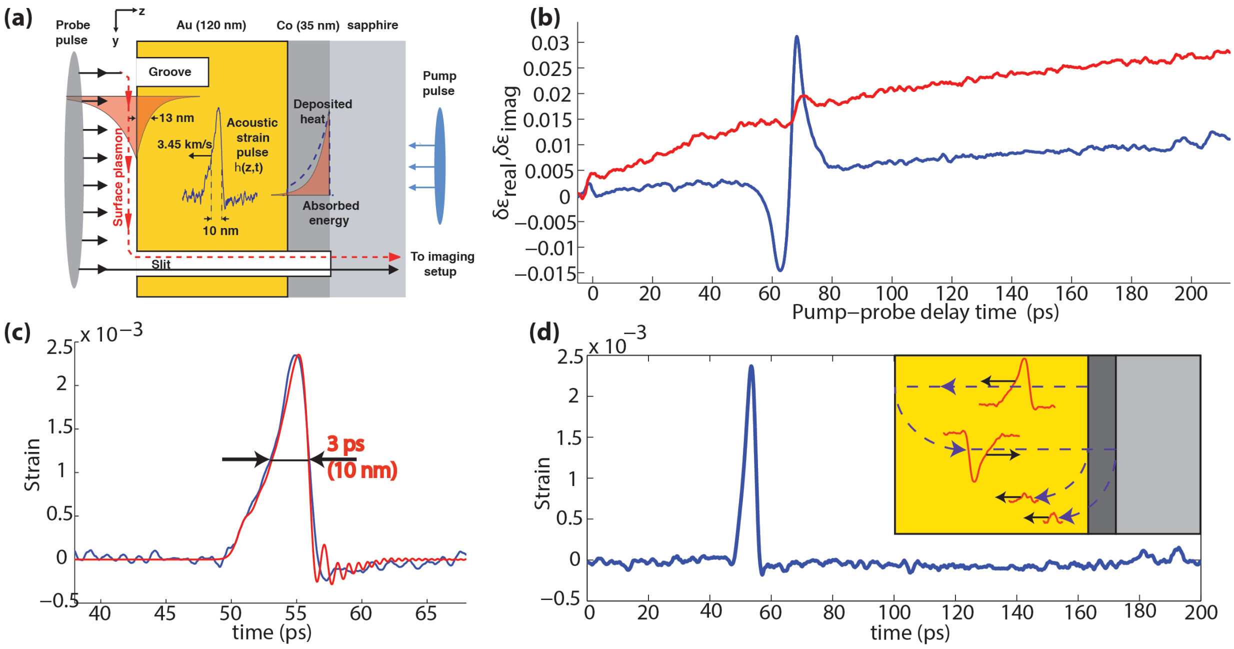

Fig. 1: (a) Schematic drawing of the acousto-plasmonic pump-probe experiment: surface plasmons propagating at the gold-air interface, probe the reflection of acoustic pulses generated in laser-heated cobalt transducer. (b) Ultrafast dynamics of the real (red line) and imaginary (blue line) parts of the surface dielectric function extracted from plasmonic interferograms with fit of thermal background (dashed lines). (c) Superposition of reconstructed (blue line) and calculated theoretically (red line) strains with pulse duration about 3 nm (FWHM). (d) Reconstructed strain profile using the Fourier approach showing all echoes delayed in time with inset explaining the obtained results.