Ultrafast Dynamics of Light Transmission Through Plasmonic Crystals

Claus Ropers [1], Roland Müller[1], Christoph Lienau[1], Gero Stibenz[1], Günter Steinmeyer[1], Doo-Jae Park[2], Yeo-Chan Yoon[2], and Dai-Sik Kim[2]

[1] Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany
E-mail: lienau@mbi-berlin.de
[2] School of Physics, Seoul National University, Seoul 151-742, Korea
E-mail: denny@phya.snu.ac.kr

Abstract. We study with 10-fs light pulses the ultrafast dynamics of light transmission through plasmonic 2D- and 1D-nanocrystals. Two interfering transmission channels are identified: ultrafast non-resonant transmission due to photon tunneling, and time-delayed resonant re-emission of surface plasmons.

1. Introduction

The linear and nonlinear optical properties of plasmonic nanostructures currently attract much attention, as such structures are promising candidates for novel applications in nano-lensing [1], perfect lensing [2], field localization, nanoscale wave-guiding [3], nano-lasing [4], and ultrafast switching. A prominent example for the unusual optical effects associated with plasmonic nanocrystals is the enhanced transmission of light through periodic nano-hole arrays in metal films [5]. It is now well understood that the optical properties of such nanostructures are governed by short-lived surface plasmon polariton (SPP) excitations with life times in the 3 – 100 fs range. Yet, the ultrafast dynamics of these excitations have largely remained elusive. Previous studies of ultrafast pulse propagation through nano-hole arrays reported a 10-fs delay in transmission [6,7]. The physical interpretation of this delay, assigned to either the finite transit time through the nanoholes [6] or to the SPP lifetime [7], however, remains controversial.

Here we report the first experimental study of ultrafast light propagation through plasmonic nano-crystals using light pulses much shorter than the SPP damping time. Phase-resolved measurements of the time structure of the transmitted light allow to clearly distinguish two different contributions to enhanced transmission: non-resonant tunneling and SPP re-radiation. We demonstrate that the optical spectra of plasmonic crystals are governed by Fano-like interferences between these channels.

2. Experimental methods

We investigate 150-300 nm thick gold films deposited onto a sapphire substrate and perforated with periodic square arrays of holes with a radius of 125 nm and

periods of 700 and 800 nm [7], or with a linear array of 50 nm wide nano-slits with periods of 600 and 700 nm. The samples are illuminated at an angle θ with weakly collimated linearly polarized 10-fs light pulses from a Ti:sapphire oscillator (Fig. 1(a)). The time structure of the transmitted light is studied using either interferometric autocorrelation (IAC) or spectral interferometry (SI).

3. Results and Discussion

In Fig. 1, autocorrelation traces of the incident light (b) and the light transmitted through a linear slit array (600 nm periodicity, 50 nm slit width) at two different angles are displayed (c,d). Linear transmission spectra are shown in the insets. In Fig. 1(c), the laser spectrum overlaps only weakly with SPP resonances. The transmitted light consists of a strong initial peak due to non-resonant transmission through the slits and a second long-lived but weak contribution due to resonant excitation and re-radiation of SPP at the sapphire/metal (SM) interface [6,7].

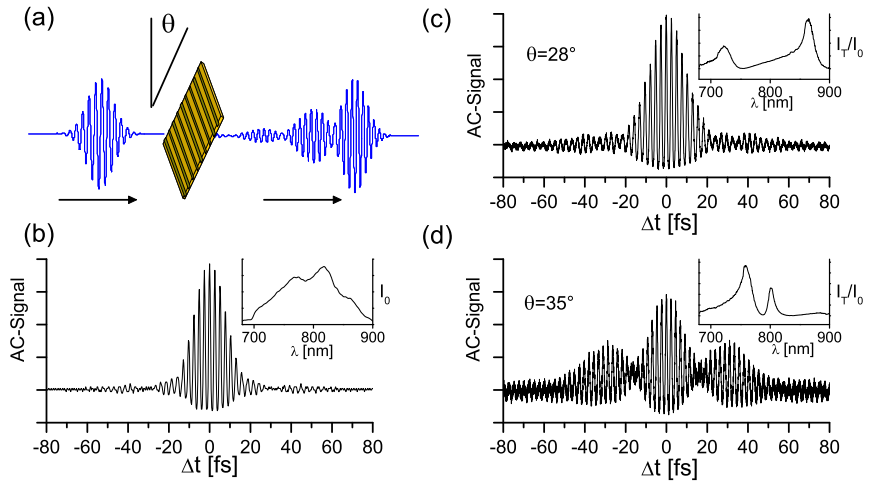

Fig. 1. (a) Schematic of the experimental setup. (b) Interferometric autocorrelation (IAC) of the incident 10-fs light pulse. (c,d) IAC of the light transmitted through a 600-nm period array of 50-nm wide nano-slits in a 150 nm thick gold film at incidence angles of θ = 28° (c) and 35° (d). The laser spectrum (b) and linear transmission spectra (c,d) are shown in the insets.

When the overlap between SPP resonances and laser spectrum is optimized by angle tuning (Fig. 1(d)), the SPP contribution is strongly enhanced, and the time profile of the transmitted light is dominated by the pronounced polarization interference between two SPP resonances persisting for more than 80 fs [8]. The IAC measurements show directly that the time structure of the transmitted field

$E_t = E_{nr} + E_{SPP}$ is given as a superposition between the short burst due to non-resonant transmission through the nanoslits $\boldsymbol{E_{nr}}$ and the weakly damped emission from different SPP resonances $\boldsymbol{E_{SPP}} = \sum_n \boldsymbol{E_n} \cdot \exp\left(-\boldsymbol{i}\omega_n \boldsymbol{t} - \gamma_n \boldsymbol{t}\right)$. The SPP dispersion relation for a flat metal film predicts SPP resonance frequencies $\omega_{n,AM}$ and $\omega_{n,SM}$ for the air-metal and sapphire-metal interface, respectively. In the spectral domain this gives rise to asymmetric Fano-like lineshapes

$$I(\omega) = \left| a_{nr}(\omega) + \sum_n \frac{b_n}{\omega - \omega_n + i\gamma_n} \right|^2 \tag{2}$$

as recently proposed [8,9]. Similar experimental results as those shown in Fig. 1 are also obtained for two-dimensional nanohole arrays.

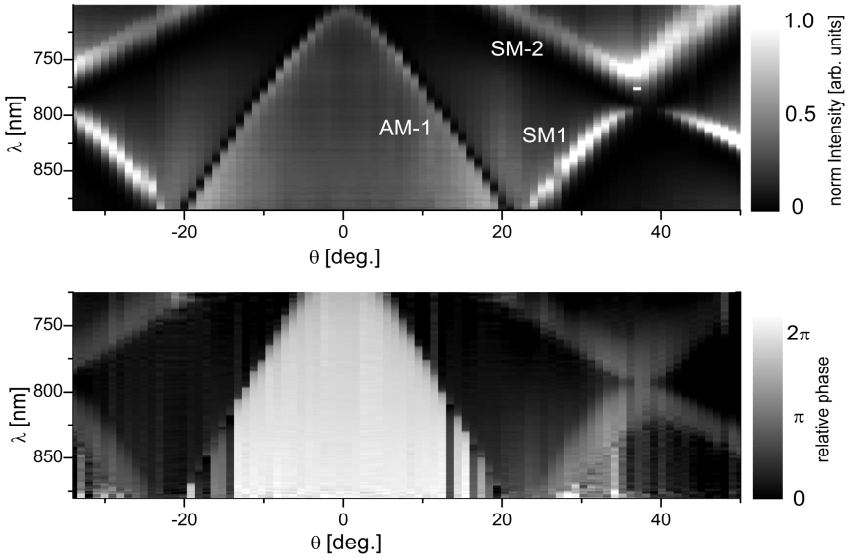

Fig. 2. Spectral intensity (a) and spectral phase (b) of the light transmitted through a 600-nm period array of 50-nm wide nano-slits in a gold film. The phase data are obtained by spectral interferometry with 10-fs light pulses as a function of incidence angle θ. Different SPP transmission resonances on the air/metal (AM) and the sapphire/metal (SM) interfaces are resolved, showing distinctly different phase signatures.

The interferometric autocorrelation measurements alone do not allow for a quantitative analysis of the different interfering contributions or for a study of the effects of coherent coupling between SPP resonances and the formation of bandgaps in these plasmonic nanocrystals. We thus use spectral interferometry to directly measure the phase of the transmitted field by interfering it with a replica of the 10-fs incident laser. Results of the angle-dependent measurements are summarized in Fig. 2 for the 600-nm period slit array. Different SPP resonances at either the air/metal (AM) or sapphire/metal (SM) interface are clearly resolved. The resonances are each characterized by an asymmetric line shape (Fig. 3(a)) and

a pronounced phase variation near the resonance (Fig. 3(b)). The phase signatures for AM and SM resonances are distinctly different. The lineshape model introduced above allows for a quantitative description of the amplitude and phase variations.

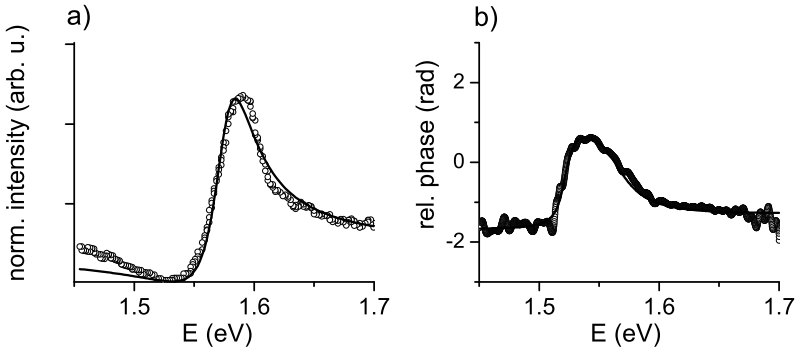

Fig. 3. Experimental transmission spectrum (a) and spectral phase (b) of the light transmitted through a 700-nm period array of 50-nm wide nano-slits in a gold film (open circles) and comparison to the lineshape model introduced in the text (solid lines). The asymmetric shape of the transmission spectrum arises from the Fano interference between non-resonant transmission and SPP re-radiation.

Important insight into the physics of enhanced light transmission through metal films perforated with periodic nanohole arrays is gained from this analysis. First, our results give clear evidence for two distinct transmission channels. A fraction of the incident light directly tunnels through the nanoholes and is rescattered into the far-field. Second, SPP modes can be excited at both the AM and the SM interface by the grating coupling effect at specific resonance energies. The lifetime of these SPP modes is much longer than the non-resonant tunneling time and can exceed 100 fs. It is limited by a Rayleigh-like scattering of these evanescent modes into progating far-field radiation, with the nanoholes acting as scattering centers [7]. The linear far-field transmission spectra result from the interference of these two transmission channels, and thus SPP excitation may both enhance and decrease the far-field transmission, depending on the phase delay between the two contributions. A generalized Fano model quantitatively describes the far-field transmission spectra. The model allows to derive the dispersion relation of this plasmonic crystal with the energy positions ω_n and the damping rates γ_n of the SPP resonances. Direct evidence for the formation of plasmonic bandgaps comes from the observation of a 40-meV splitting at the crossing of the SM(1) and SM(-2) resonances. An even more striking consequence of the coherent coupling between different SPP eigenmodes is the pronounced variation of the damping rates γ_n near the crossing region. The coupling leads to the formation of new SPP eigenmodes having different spatial overlap with the nano-slits. As the nano-slits

serve as local scattering centers converting evanescent SPP fields into far-field radiation [7], a variation of the spatial mode structure gives rise to a change in the radiative damping of the SPP excitation and thus of γ_n. This effect, which is analogous to the Dicke sub-/superradiance in coupled atomic systems, is directly observed from the reduction in line width and amplitude of the SM(1) resonance near the crossing region (Fig. 2). At certain angles, we observe ultranarrow transmission resonances with linewidths corresponding to SPP lifetimes of over 300 fs, which is more than 30 times longer than reported in previous time-resolved transmission experiments [6,7]. Spatially resolved near-field SPP spectra, reported elsewhere, give evidence for the different symmetry of the sub- and superradiant SPP modes.

In summary, we have presented the first experimental study of ultrafast light propagation through plasmonic nanocrystals with a time resolution beyond the damping times of the relevant surface plasmon polaritons. The dynamics of two distinct transmission channels – non-resonant tunneling and SPP re-radiation – are directly resolved, and it is shown that the linear transmission spectra of such nanostructures are governed by interferences between both contributions. Coherent couplings between different SPP resonances, *i.e.*, formation of plasmonic bandgaps and sub-/superradiant damping are quantitatively probed. We believe that similar studies are highly relevant for a detailed microscopic understanding of the linear and nonlinear optical properties of plasmonic and photonic crystals.

Acknowledgements. We gratefully acknowledge financial support of the work in Germany by the DFG (SFB296) and the European Union (SQID) and that in Korea by MOST (NRL and Nano-Photonics program) and KOSEF (SRC program).

References

1 H. J. Lezec, A. Degion, E. Devaux, R.A. Linke, L. Martin-Moreno,F. J. Garcia-Vidal, T. W. Ebbesen, in *Science*, Vol. 297, 820, 2002.
2 J. B. Pendry, in *Physical Review Letters*, Vol. 85, 3966, 2000.
3 S. A. Maier, P. G. Kik, H. A. Atwater, S. Meltzer, E. Habel, B. E. Koel, A. A. G. Requicha, in *Nature Material*, Vol. 2, 229, 2003.
4 D. J. Bergman and M. I. Stockman, in *Physical Review Letters*, Vol. 90, 027402 2003.
5 T. W. Ebbesen, H. J. Lezec, H. F. Ghaemi, T. Thio, P. A. Wolff, in *Nature*, Vol. 391, 667, 1998.
6 A. Dogariu, T. Thio, L. J. Wang, T. W. Ebbesen, H. J. Lezec, in *Optics Letters*, Vol. 26, 450, 2001.
7 D. S. Kim, S. C. Hohng, V. Malyarchuk, Y. C. Yoon, Y. H. Ahn, K. J. Yee, J. W. Park, J. Kim, Q. H. Park, and C. Lienau, in *Physical Review Letters*, Vol. 91, 143901, 2003.
8 R. Mueller, V. Malyarchuk, and C. Lienau, in *Physical Review B*, Vol. 68, 205413, 2003.
9 C. Genet, M. P. van Exeter, and J. P. Woerdman, in *Optics Communications*, Vol. 225, 331, 2003.