ULTRAFAST ELLIPSOMETRY OF LASER-INDUCED PHASE TRANSITIONS IN MATTER

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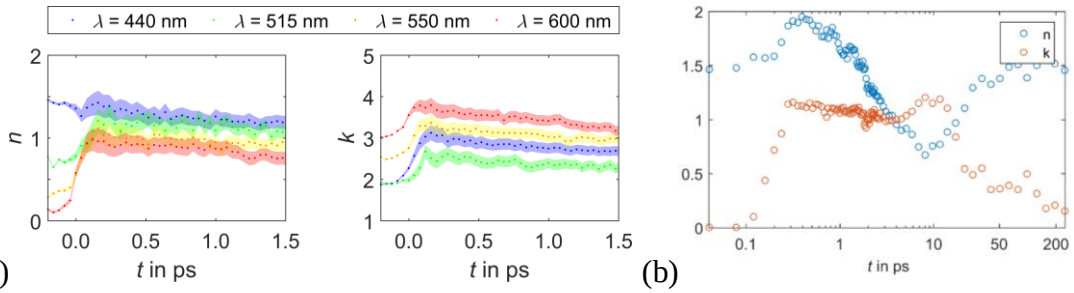
Irradiating matter with ultrafast laser radiation induces transient states with physical and chemical properties far away from any equilibrium. Thin gold film on a glass substrate (thickness $d_{\mathrm{Au}} = 200$ nm, roughness R_a =3 nm) and bulk PMMA have been excited by single pulsed femtosecond laser radiation ($\tau_{\mathrm{H}} = 40$ fs) and the temporal evolution of the complex refractive index was detected spatially- and spectroscopically-resolved by a self-developed pump-probe ellipsometer. The gold film excited at the pump wavelength $\lambda = 800$ nm exhibits for a fluence of 1 J/cm² (below gentle ablation [1]) and a probe wavelength $\lambda = 440$, 515, 550, and 600 nm a complex dynamics of the refractive index n and the extinction coefficient k (Fig.1 a). Apparently, the refractive index converges for all investigated wavelengths about 100 fs after excitation to a value close to 1 describing the heating of the electron system, in which the refractive index for $\lambda = 440$ nm demonstrates a slight decrease and for $\lambda = 515$, 550 and 600 nm the refractive index increases strongly compared to the value at rest. Thereafter, a slight decrease of the refractive index is obtained, depicting the beginning of the cooling of the electrons [2].

PMMA, an organic dielectric material, is transparent for radiation at small intensities in the VIS and IR spectral range, but at large intensities (I =3·10^{13} W/cm²) the radiation induces field ionization generating free electrons [3, 4]. Exciting above ablation threshold with a single laser pulse ($\lambda = 800$ nm) a strong increase in the refractive index and extinction coefficient is induced within 100 fs probing at the wavelength $\lambda = 440$ nm. The extinction coefficient lasts about 20 ps, then decreasing again, depicting the lifetime of the laser-induced free electrons.

Keywords: ultrafast; transient state; ablation



(a) (b)

Fig. 1. Refractive index and extinction coefficient as function of time after excitation with single pulsed laser radiation ($\tau_{\mathrm{H}} = 40$ fs, $\lambda = 800$ nm) (a) of gold for different probe wavelengths, and (b) of PMMA for the probe wavelength $\lambda = 440$ nm.

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