Photoinduced surface dynamics of V_3O_5: visualization of autocorrelation function from light scattering data

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Abstract: Ultrafast transition in V_3O_5 shows complex dynamics of the surface autocorrelation function. Phase-retrieval algorithms with filtering of stochastic scattering component provide reconstruction of specific/hidden features of photoexcited surface.

Optical visualization of ultrafast surface dynamics is one of the perspective directions in elastic light scattering metrology. Time- and angle-resolved hemispherical light scattering (*tr*-ARHELS) technique is one of the most powerful methods to obtain accurate information about transient properties of phase-change materials with femtosecond resolution in time and with submicron resolution in space [1]. While most angle-resolved light scattering measurements usually provide the bidirectional scatter distribution function (BSDF) and, in some cases, the power spectral density (PSD) of the surface [2], these data can also be used to obtain the surface autocorrelation function (ACF). The numerical computation of the ACF from PSD or from BSDF data by inverse Fourier transform usually suffers from significant error owing to the absence of experimental data in the low and high range of spatial frequencies. This problem can be resolved by applying phase-retrieval algorithms with reconstruction of absent experimental data along with data filtering procedure [3].

In this work, we report on the visualization of ACF dynamics upon photoinduced phase transition in the correlated oxide V_3O_5. This material shows reversible insulator-to-metal transition IMT at temperature T_c ~425 K [4,5]. The transition can be also induced by intense light pulse on an ultrafast time scale. The main attention in the present work was paid to time-resolved ACF of self-organized submicron domain structure superimposed onto stochastic surface relief of a V_3O_5 120-nm-thick film. The ACF for all components of surface roughness shows rapid growth during the IMT, indicating increase of optical homogeneity of surface roughness in lateral direction.

The sample was prepared by reactive DC magnetron sputtering from a vanadium target of 99.95% purity [6]. The substrate roughness was the main factor which defined the surface morphology of V_3O_5 film. Since the substrate was amorphous quartz, the grains orientation of V_3O_5 film was random. Nevertheless, some surface areas showed a local ordering of V_3O_5 domains on the mesoscale.

Ultrafast pump-probe laser spectroscopy measurements were performed with a Spectra-Physics femtosecond laser system. The system generates 1 mJ 130 fs laser pulses with central wavelength λ=800 nm at 1 kHz repetition rate. After passing a set of mirrors, polarizers, beamsplitters and nonlinear crystal, the laser pulse was divided to pump (λ=800 nm) and probe (λ=400 nm) pulses. These pulses in collinear geometry were used in an ultrafast scatterometer described elsewhere [6]. A motorized computer-controlled optomechanical delay line provided time-delay between pump and probe pulses. The sample was installed inside the scatterometer, and the pump pulse was focused within a 0.6 mm diameter spot, providing excitation fluence of 10 mJ/cm^2. The probe pulse was overlapped with the pump pulse at normal incidence and focused into a 60 μm spot, while its intensity was reduced by several orders of magnitude to exclude the possibility of any nonlinear interaction with the sample. Scattered light of the probe pulse was collected within the front hemisphere by an elliptical mirror and projected to a CCD camera. Obtained scattering indicatrices were recalculated into BSDF and PSD data arrays.

For time-resolved measurements of light scattering dynamics we choose an area on the sample surface with high crystallinity, where V_3O_5 grains are organized in spatially-ordered/twinned slab domains. The scattering data were analyzed in terms of Elson's vector theory of light scattering by surface roughness [7]. For this analysis the surface PSD function was calculated from BSDF data. Optical constants used in the calculations were obtained from additional angle-resolved reflection coefficient measurements for *s*- and *p*-polarized incident light. The contribution to the PSD of slab domains highly oriented in lateral direction was obtained by filtering of stochastic PSD component. This was achieved by using different window functions for the Fourier transform in a multistep computation algorithm [3].

Figure 1 shows the filtered autocorrelation function ACF* of V_3O_5 slab domains for unexcited surface [Fig. 1(a)], and the evolution of ACF* within 50 ps time scale [Fig. 1(b)]. ACF* is shown in arbitrary units with respect to the normalized nonfiltered ACF of stochastic surface roughness. For these calculations we used Gerchberg-Saxton error reduction (ER) and Hybrid Input-Output (HIO) algorithms, which produced an accurate reconstruction of the ACF* and PSD pair [6]. The structural ordering of slab domains results in the oscillation of ACF*. The amplitude of the oscillation changes noticeably upon photoexcitation, as the V_3O_5 switches into metallic state.

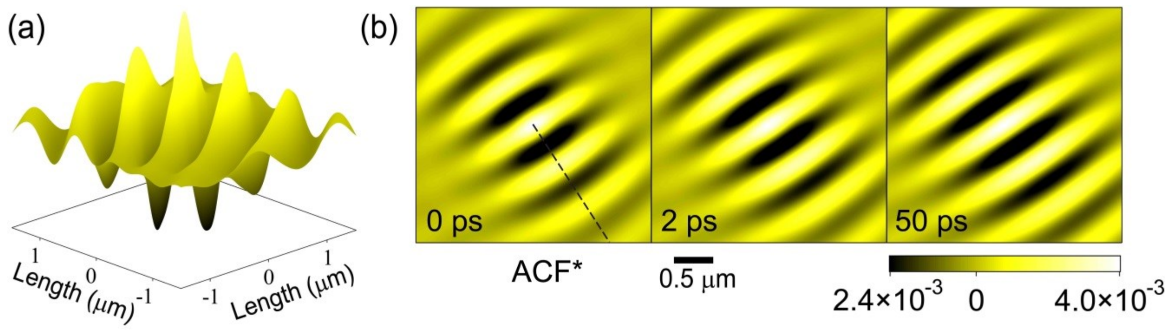

Fig. 1. Autocorrelation function of periodic domain structure: (a) ACF* of unexcited V_3O_5; (b) evolution of ACF* upon photoexcitation.

The cross-section of the ACF* (Fig. 2) along the direction specified by the dashed line in Fig. 1(b) at time t= 0 ps shows the growth of the ACF* amplitude due to the evolution of surface optical properties with time. Inset in Fig. 2 shows that the amplitude of the oscillatory maximum at 0.6 μm lag length builds up within less than 5 ps, indicating that the main structural surface dynamics associated with light-induced IMT completes within this time scale. Increase of the ACF*(0.6 μm) amplitude on 20% is associated with noticeable increase of optical homogeneity of each surface domain. After 5 ps the optical roughness remains nearly unchanged.

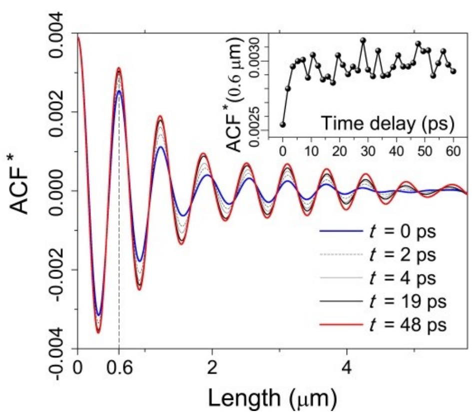

Fig. 2. Cross-section of ACF*.

In summary, the V_3O_5 surface dynamics was visualized during light-induced IMT by computing the transient ACF from *tr*-ARHELS experimental data. The *tr*-ARHELS technique provides powerful tool to monitor specific features of the surface roughness or dielectric constant inhomogeneity with metrological precision. BSDF and PSD data filtering can be performed as a multistep process by using different window functions for the Fourier transform as a part of ER and HIO algorithms. This can be used for the detection and visualization of ACF and transient dynamics of specific/hidden features of surface optical roughness.

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