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# Nanoscale Photo-Orientation of Azo-Dyes in Glassy Environments Using Polarized Optical Near-Field

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**Abstract :** Recent advances in improving information storage performance are inseparably linked with circumvention of fundamental constraints such as the supermagnetic limit in heat assisted magnetic recording, charge loss tolerance in solid-state memory and the Abbe's diffraction limit in optical storage. A substantial breakthrough in the development of nonvolatile storage devices with dimensional scaling has been achieved due to phase-change chalcogenide memory, which nowadays, meets the market needs to the greatest advantage. A further progress is aimed at the development of versatile nonvolatile high-speed memory combining potentials of random access memory and archive storage. The well-established properties of light at the nanoscale empower us to use them for recording optical information with ultrahigh density scaled down to a single molecule, which is the size of a pit. Indeed, diffraction-limited optics is able to record as much information as ~1 Gb/in2. Nonlinear optical effects, for example, two-photon fluorescence recording, allows one to decrease the extent of the pit even more, which results in the recording density up to ~100 Gb/in2. Going beyond the diffraction limit, due to the sub-wavelength confinement of light, pushes the pit size down to a single chromophore, which is, on average, of ~1 nm in length. Thus, the memory capacity can be increased up to the theoretical limit of 1 Pb/in2. Moreover, the field confinement provides faster recording and readout operations due to the enhanced light-matter interaction. This, in turn, leads to the miniaturization of optical devices and the decrease of energy supply down to ~1 μW/cm$^2$. Intrinsic features of light such as multimode, mixed polarization and angular momentum in addition to the underlying optical and holographic tools for writing/reading, enriches the storage and encryption of optical information. In particular, the finite extent of the near-field penetration, falling into a range of 50-100 nm, gives the possibility to perform 3D volume (layer-to-layer) recording/readout of optical information. In this study, we demonstrate a comprehensive evidence of isotropic-to-homeotropic phase transition of the azobenzene-functionalized polymer thin film exposed to light and dc electric field using near-field optical microscopy and scanning capacitance microscopy. We unravel a near-field Raman dichroism of a sub-10 nm thick epoxy-based side-chain azo-polymer films with polarization-controlled tip-enhanced Raman scattering. In our study, orientation of azo-chromophores is controlled with a bias voltage gold tip rather than light polarization. Isotropic in-plane and homeotropic out-of-plane arrangement of azo-chromophores in glassy environment can be distinguished with transverse and longitudinal optical near-fields. We demonstrate that both phases are unambiguously visualized by 2D mapping their local dielectric properties with scanning capacity microscopy. The stability of the polar homeotropic phase is strongly sensitive to the thickness of the thin film. We make an analysis of α-transition of the azo-polymer by detecting a temperature-dependent phase jump of an AFM cantilever when passing through the glass temperature. Overall, we anticipate further improvements in optical storage performance, which approaches to a single molecule level.

**Keywords :** optical memory, azo-dye, near-field, tip-enhanced Raman scattering
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