Giant, ultrafast optical switching based on an insulator-to-metal transition in VO_2 nano-particles: photo-activation of shape-controlled plasmons at 1.55 μm.

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A new generation of devices where the electronic, optical or magnetic state of a system can be controlled optically on the ultrafast timescale[1] is one of the most compelling technological ramifications of the rapidly advancing field of strongly correlated electrons[2]. However, for real-world applications it is also necessary to incorporate these compounds in appropriate environments (e.g. optical fibers or silicon-based electronics), to ensure compatibility with existing technologies (e.g. telecom wavelengths), room temperature operation and limited power densities. Here, we report on the study of the photo-activated optical switching in nano-rods of strongly correlated VO_2. The particles are grown by ion-implantation and self-assembly within a Silica matrix or an optical fiber, operate at room temperature and can be switched between the insulating and metallic phase within less than 100 fs. The energy threshold to achieve switching corresponds to approximately 500 pJ within the core of a single mode fiber and is compatible with current diode technologies. Tailoring of the spherical/cylindrical geometry results in control of the spectral response of the system, which is dominated by the impulsive formation of a surface plasmon upon the insulator-to-metal transition. The response at the technologically important 1.55 μm wavelength is in this way maximized.

The ultrafast response of strongly correlated compounds has important ramifications in both fundamental science and technology. The many phase transitions exhibited by these systems are in fact associated with large changes in their electrical, optical and magnetic properties. Such multi-stable behavior is also extremely sensitive to external stimuli, due to the many interacting degrees of freedom and competing states of the system. Recently, a number of studies have reported large optical nonlinearities in such strongly correlated solids[3]. However, compatibility with existing technologies, operating temperatures and wavelengths are often critical limitations for practical applications.

Symmetrically, fabrication of nanometer-scale structures allows for the design of specially tailored materials that adapt the property of extended solids to the specific application[4]. Recently, the design of VO_2 nanoparticles in silica and in optical fibers has been demonstrated, opening attractive new avenues for optical-device applications[5].

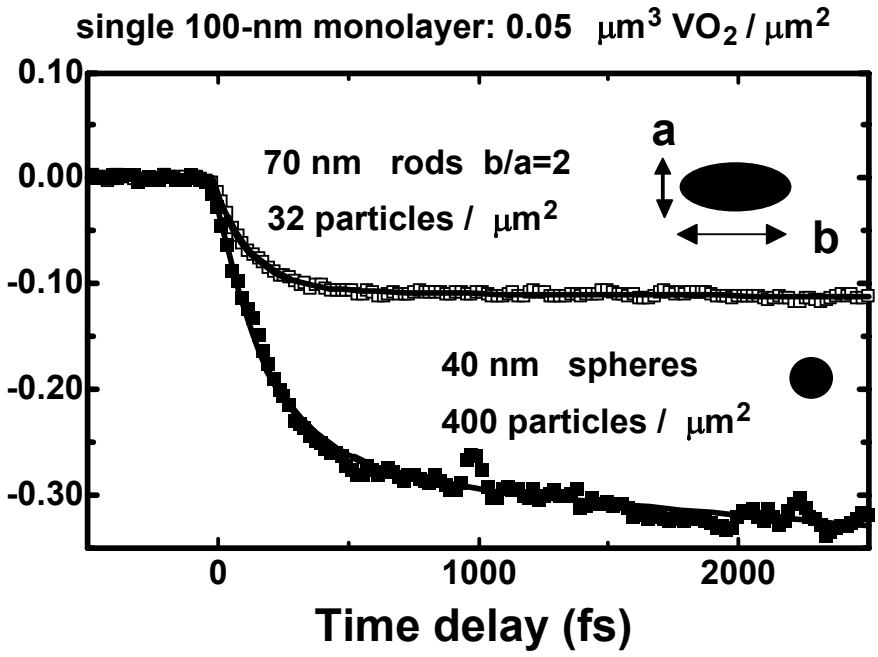

Figure 1. Ultrafast response at 1.55 micron wavelength for nanospheres and nanorods. The excitation wavelength is 800 nm. The particles are grown by ion implanting Vanadium and Oxygen in stoichiometric quantity and by annealing and self assembly. The total amount of material is nearly identical in the two cases, distributed over a layer that is approximately 200-nm thick.

Important features of the VO_2 phase transition are: (1) the possibility of photo-inducing it on the ultrafast timescale[6,7], (2) the large optical changes in the near IR, (3) the near-room temperature transition point (340 K). The availability of these crystallites makes it possible to exploit the change in optical properties associated with its insulator-to-metal transition in a fiber-optic environment.

Here, we study the near and mid IR photo-response of VO_2 nanoparticles in silica, showing that a dramatic increase of absorption can be photo-induced on ultrafast time scales. In brief, the transition from the insulating to the metallic state is responsible for the large, ultrafast changes in the optical properties, with a combined contribution from the rearrangement in the density of electronic states (bandgap collapse) and from the formation of a dielectrically confined surface plasmon in the metallic phase. The semiconducting gap, around 0.7 eV, disappears upon the phase transition to the metal state. Furthermore, since the features of the surface-plasmon resonance depend on the size and shape of the VO_2 nanoparticles, the optical switching characteristics of nanorods of different geometry appear to exhibit substantial differences. Proper design can thus tailor optimal response at telecom wavelengths.

In figure 1 we report the ultrafast response in the near IR, induced by 100-fs excitation at 800 nm with 1 mJ/cm^2. We observe an initial drop in the optical transmission of single monolayers of spheres and rods,

respectively. The reduced transmission is associated with the formation of the metallic phase and it is found to be pulse-width limited, i.e. faster than 120 fs. This is consistent with the fundamental timescale of 75 fs found in the bulk[8]. In the nano-spheres we also find a slower decay constant of 3 ps, possibly related to coherent acoustic phenomena[9]. Importantly, we find that the response can be strongly affected by the shape of the nanoparticles, resulting in a factor of 2.5 more effective optical switching for the spheres, despite the fact that in both cases the two samples contained the same amount of VO_2.

To further substantiate our assignment of the physical origin of this optical switching, we note that the dynamics exhibit a well-defined fluence threshold and saturation, indicative of a photoinduced phase-transition. We also report the spectral response of the nanorods at 1-ps pump-probe time delay for different wavelengths in the IR. A clear resonance in the $\Delta T/T$ signal between 1 and 2 microns is observed, in very good agreement with static spectroscopic studies of the thermally induced phase transition[3].

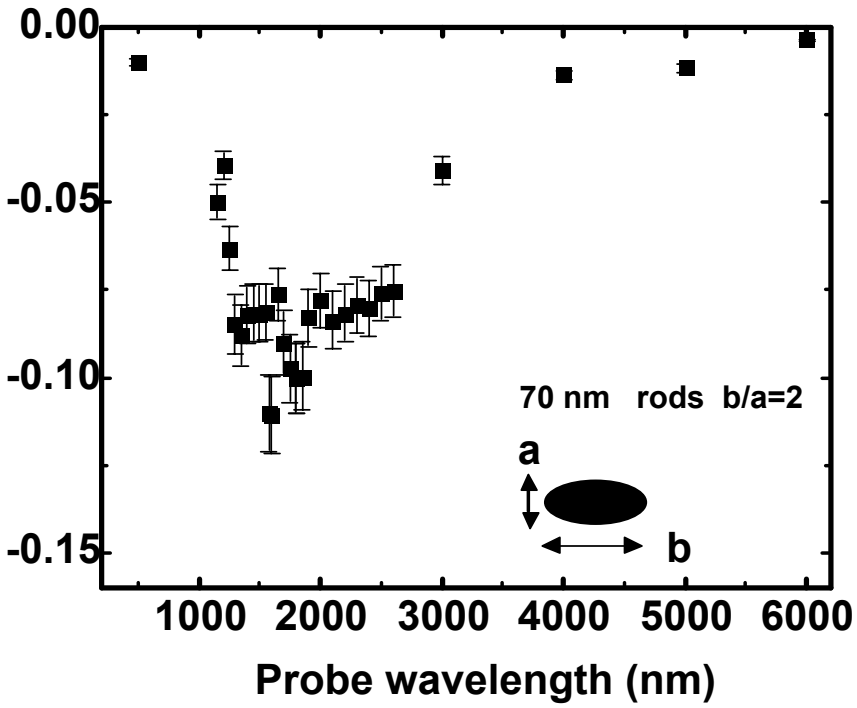

Figure 2. Wavelength dependence of the time-resolved signal. The conditions are identical to those reported in figure 1, whereby every spectral point is recorded at 1 ps time delay.

In summary, we have studied the near-IR photo-induced response of VO_2 nanoparticles in a silica matrix. We find that the photo-induced insulator-to-metal transition is responsible for very large changes of the optical properties in the infrared, in particular at the technologically relevant wavelength of 1.55 microns. Furthermore, we find that the magnitude of the response can be tailored by means of engineering size and shape of the particles during the self-assembly step of the growth procedure. Shape and size act on the optical properties through the surface plasmon resonance. Higher modulation depths can presumably be obtained by increasing the density and implantation depth of the VO_2 nanoparticles, whereby a true transparent-to-opaque transition could be obtained. It is also important to point out that the 1 mJ/cm^2 threshold for the photo-induced phase transition is equivalent to 500 pJ pulse for a typical 50-μm^2 mode size in a single-mode fiber, making this and similar schemes attractive for real-world applications.

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[1] Ultrafast switching

[2] strongly correlated electrons

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