Nanoparticle optical memory function

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ABSTRACT

Light and electron-beam induced structural transformations in nano-particles of polymorphic metals provide a paradigm for achieving resonator-less optical memory functionality with picojoule energy requirements. We demonstrate optical memory function with gallium nanoparticles grown by light-assisted self-assembly at the end face of an optical fiber. The memory is written by single optical control pulses, with the state of the bistable nanoparticle film monitored by a continuous wave low intensity reading laser, measuring the reflectivity of the particles.

Keywords: Metal nanoparticles, Optical memory, Data storage techniques.

1 INTRODUCTION

The demand for controlling light with light and plasmonic signals in highly integrated circuits is one of the driving forces behind the significant effort and progress that lately has been put into research in nanoscale-sized particles and thin films. In this respect they promise to provide the functionality of optical switching and signal conditioning between elements of three-dimensional, highly integrated photonic-band-gap optical signal routers and two-dimensional plasmonic waveguides, ultimately in combination with two- or three-dimensional arrays of miniature semiconductor lasers. While most applications of optical memory functionality rely on the bulk wave propagation properties of a nonlinear medium together with an optical feedback, such as for example in a cavity or ring resonator, the nanoparticles instead rely on the significant hysteresis of their thermo-physical and optical properties. The advantage of using this configuration is that the physical size and power consumption are significantly reduced.

In the terminology of light-by-light signal control, it is implied that either the intensity or phase of the light wave is changed substantially by the presence of a control signal. In an all-optical device this is achieved by using a control lightwave to change the optical characteristics of the medium as seen by a signal lightwave.

In recent experiments with a single nanoparticle [1], we demonstrated that a nanoparticle with a diameter of a few tens of nanometers exhibits equilibrium coexistences between a number of solid and disordered structural phases with very different dielectric properties, which can be controlled by optical excitation in a highly reversible and reproducible fashion. The energy required to exercise such control is only a few picojoules. In this paper we report on light induced structural transformations which in the confined conditions of a nanoparticle provide a paradigm for achieving resonator-less optical memory functionality with picojoule energy requirements.

2 MEMORY FUNCTIONALITY

In contrast to bulk solids where phase transitions are abrupt and characterized by discontinuous structural transformations, phase transitions in nanoparticles proceed through a dynamic coexistence of structural forms, making them continuous and reversible [2,3]. As a nanoparticle is brought to the verge of what would be a first order structural phase transition in the corresponding bulk medium, its structural composition can become significantly more sensitive to external stimulation. If the structural phases involved have different dielectric properties, a continuous structural transformation will be accompanied by a continuous change in the optical properties of the nanoparticle, thus effectively providing an optical nonlinearity.

![Figure 1: Schematic picture of a nanoparticle undergoing a structural transformation.](image)

The transition may be activated by increasing the ambient temperature or by external optical excitation at a fixed ambient temperature. In the later case the transformation is driven by the laser-induced heating or excitation of the material's electronic structure. By increasing the level of excitation, the nanoparticles are sequentially converted from stage I, through II and III, to stage IV, as illustrated in Fig. 1. Reversible changes occur in the excitation range between II and III. If the excitation is withdrawn before stage IV is achieved, the nanoparticle...
will return to its initial state, i.e. stage I. As the particles are fully converted into phase IV, they will not return to their low-temperature phase without substantial overcooling.

For example, a gallium nanoparticle probed using low intensity continuous wave (CW) laser light needs to be overcooled by more than 90 K before it returns to the low-temperature phase. Such a nanoparticle is therefore an inherently bistable object, and this presents an opportunity for creating an optical memory element.

**Figure 2:** Experimental arrangement for demonstrating optical memory function in gallium nanoparticles: (a) components inside the deposition chamber and AFM image of a gallium nanoparticle film on the core of the cleaved optical fiber; (b) optical setup for memory reading and writing, outside the chamber.

By employing the hysteresis, we have demonstrated optical memory functionality with gallium nanoparticles grown by light-assisted self-assembly at the end face of an optical fiber, using the setup illustrated in Fig. 2a. A pulsed semiconductor laser operating at a wavelength of 1.5 μm was used to generate single control pulses with 1 μs duration (Fig. 2b). These pulses were used to switch the gallium nanoparticles from a low reflectivity state, labeled as logical state ‘0’, into a high reflectivity state, labeled as logical state ‘1’. The state of the bistable nanoparticle film was monitored by a second CW low intensity reading laser, used for measuring the reflectivity of the particles.

**Figure 3:** Nanoparticle undergoing light-induced structural transformations as an inherently bistable element. Hollow arrows indicate temperature set points, solid lines show the action of the control optical pulse, and dashed lines show the evolution of the system on termination of the control pulse. The dotted line shows system reset.

In the absence of the control pulse the reflectivity shows a well-defined hysteresis on cycling the temperature. However, as the temperature of the nanoparticles is set below the lower transition temperature $T_{0*}$ the control pulse causes only a reversible change in the state of the nanoparticles (Fig. 3a). As the temperature $T_0$ is set to a temperature higher than $T_{0*}$, within the region of the hysteresis curve, the control pulse energy becomes sufficient to fully convert the nanoparticles into state ‘1’ (Fig. 3b), where they remain even after the pulse has passed. The system then stays in state ‘1’ regardless of the presence of additional control pulses (Fig. 3c). In order to restore state ‘0’, one can decrease the temperature of the nanoparticles below the hysteresis boundary $T_{0*}$ (Fig. 3d), completing the cycle of writing and erasing information to give EPROM functionality.

As the temperature set point can be controlled optically by the intensity of the reading beam, in addition to the temperature $T_0$, we have also experimentally shown that one can in fact achieve all-optical erase functionality as well, simply by adjusting the reading signal power. Thus, a nanoparticle can constitute an optical memory bit which can be read and erased by a weak probe beam, and written by a single optical control pulse.

3 **EXPERIMENTAL SETUP**

In demonstrating the memory functionality, the film of gallium nanoparticles was fabricated by deposition on the end face of an optical fiber, using an atomic beam from an effusion cell and the light assisted deposition technique as
The nanoparticles were grown to a median diameter of 50 nm. By growing the film of nanoparticles at the end face of an optical fiber, in the setup shown in Fig. 2, it is ideally positioned for its optical probing and excitation. The fiber with the film of nanoparticles was attached to a liquid nitrogen cooled cryostat inside a vacuum chamber, allowing the temperature of the film to be varied in the range from 80 K to 300 K.

Two lasers were used as light sources in the optical characterization of the film of nanoparticles. The first one was a 1 mW CW diode laser operating at a vacuum wavelength of 1310 nm, which together with an InGaAs photo detector probed the film reflectivity. The second one was a pulsed diode laser operating at a vacuum wavelength of 1550 nm, which was used to optically excite the nanoparticles by generating single optical pulses on demand.

4 EXPERIMENTAL RESULTS

The film reflectivity in the absence of optical excitation was recorded as function of temperature in a sweep from 80 K to 300 K at a rate of 2 K/min. In this sweep the film reflectivity changed from 7.2 to 7.6 percent, undergoing a sharp transition at 200 K. The intermediate region between states II and III as shown in Fig. 1 was found to correspond to the temperature interval between 195 K and 220 K. The corresponding reflectivity curve for scanning the temperature in the reverse direction was then recorded, and was found to stay in the high reflectivity level all the way down to 80 K. In order to switch back into the low reflectivity state ‘0’, the probe laser needed to be switched off, so that the effective film temperature was further reduced. Once the probe laser was switched on again, the initial level ‘0’ was recovered.

In order to demonstrate the memory functionality the gallium nanoparticle film one needs to be able to lock it into the high reflectivity state at a constant temperature. In the experiment as here reported, this was achieved by using the manually triggered 1550 nm pulsed laser, with the sample kept at a temperature close to the phase transition $T_{\text{set}} = 170$ K. In order to switch the film from the logic ‘0’ state to the ‘1’ state, a single optical pulse of peak power 13.1 mW and pulse duration 1 µs was used to excite the system. After exposure to the optical pulse, the film was found to be locked into the high reflectivity state ‘1’ in which it remained when keeping the temperature constant. This provides proof that the gallium nanoparticles can be switched by a single optical pulse from the low reflectivity state ‘0’ to the high reflectivity state ‘1’ at a constant temperature, hence showing memory write functionality.

The written state is maintained until the nanoparticle film is substantially cooled, by reducing the background temperature to a level below $T_m$ at which the particles return to the ‘0’ state, hence also providing memory erase functionality.

5 CONCLUSIONS

In conclusion, we have experimentally demonstrated that gallium nanoparticles undergoing light-induced structural transformations possess all elements necessary to act as a rewritable all-optical memory. Although for this first proof of concept a film of nanoparticles was used, each particle can potentially act as a single memory bit. By taking the total energy supplied to the gallium film with the writing pulse and dividing it by the estimated number of nanoparticles present in the effective area of the wave guided mode of the fiber, an upper limit of the total energy required to switch a single nanoparticle from state ‘0’ to state ‘1’ is obtained as 400 fJ. This estimate clearly shows the potential for single nanoparticles to provide memory functionality in future nanophotonic devices, operating at a very low power requirement.

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REFERENCES