



All-Optical Phase-Change Memory in a Single Gallium Nanoparticle

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We report on the first demonstration of a quaternary-logical resonatorless optical memory element with information encoded in the structural phase of a single 80 nm gallium nanoparticle. The size of the memory element is comparable with bits in next-generation hard disks, and radically smaller than previously suggested memories exploiting optical resonators. Furthermore, the energy required for switching the nanoparticle is an order of magnitude less than needed in DVD, DVR, or hard disk technologies.

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Phase-change memories [1–3] are strong candidates for addressing challenges in size and power consumption of electronic memories [4], with data recording done by switching the material between amorphous and crystalline phases, much in line with today's DVD or DVR technology. However, polymorphic systems exist in which crystalline-to-crystalline transitions can provide for higher-base logics.

In this Letter we demonstrate the principle of a quaternary-logical [5] optical memory element with information encoded in the structural phase of a single 80 nm gallium nanoparticle, with its four logical states written by optical pulses of a few pico-Joules energy. This memory element is comparable in size with bits in next-generation hard disks, and radically smaller than previously suggested memories exploiting optical resonators. Furthermore, the energy required for switching the nanoparticle is an order of magnitude less than needed in DVD, DVR, or hard disk technologies. This novel principle of operation equally well applies to methods for achieving plasmonic switching [6,7].

Phase transitions in nanoparticles can be continuous and reversible in their nature, occurring through a dynamic coexistence of their different structural forms. However, polymorphic nanoparticles can in certain conditions be locked into metastable states, in which they remain regardless of changes in temperature [Fig. 1(a)]. As the different crystalline phases of a single nanoparticle possess different optical cross sections of absorption and scattering, one may obtain a logical element by coding each distinct optical characteristic by a unique label. This does not necessarily apply only to binary systems, as employed in the vast majority of today's devices, but can just as well be a ternary, quaternary, or even higher-base logics [5] [Fig. 1(b)]. The transitions between the different phases, or equivalent logical states, may be activated by increasing or decreasing the temperature of the medium. In the case of optical data storage, this could be done by external excitation using optical pulses. The idea of achieving binary or even higher-base logics in optical resonators was proposed

and studied extensively in the 1980s, in optically bi- and multistable configurations where optical nonlinearities were employed together with a feedback to lock devices into stable states. However, these solutions all had in common that their minimum spatial size was limited to one optical wavelength as they involve feedback from interference. Phase memories, however, store information

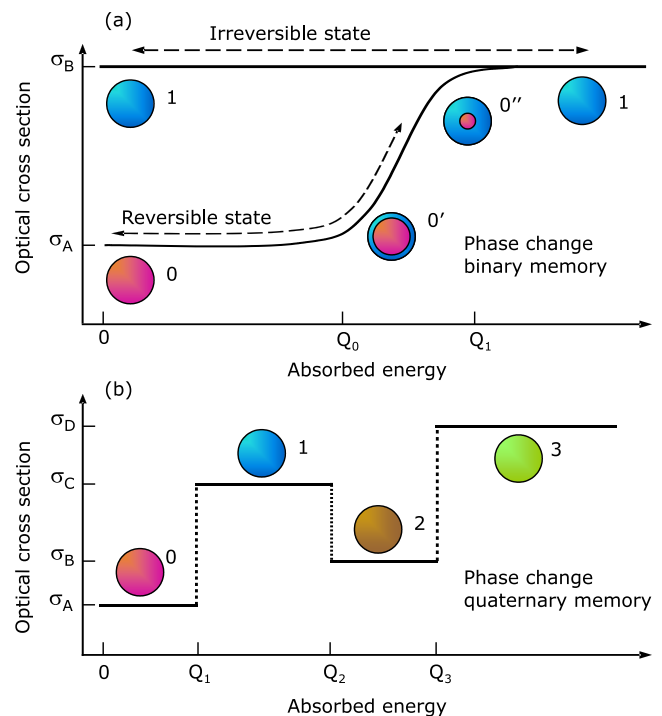


FIG. 1 (color online). (a) Binary memory functionality in a nanoparticle. Once the excitation passes the upper level Q_1 for full transformation from phase 0 into phase 1, the particle remains in this metastable phase even as the excitation is withdrawn. Shell structures indicate transitional mixed-phase states occurring in the range between Q_0 and Q_1 . (b) Simplified generic diagram illustrating quaternary memory functionality in a nanoparticle, employing four different phases, each labeled as a unique logical state. Mixed-phase states occurring between different phases are not shown.

in the phase of the material and are not subject to this constraint. In this respect, nanoparticles for data storage are particularly promising as they provide intrinsic mechanisms of phase metastability, require a very small amount of energy per logical state to write, and offer an outstanding information storage density.

With the starting point in today's data storage density of 0.015 Tb/in^2 in Blu-ray DVD or DVR and recent records of 0.23 Tb/in^2 in hard disks employing perpendicular recording technology [8], the general interest in phase-change materials has intensified, and only recently a density as high as 3.3 Tb/in^2 was reported in a binary phase-change material [3] written by means of a heated atomic force microscopy (AFM) tip. In recent experiments on reversible light- and electron beam-induced structural transformations in gallium nanoparticles [9] and, in particular, in the observation of solid-to-solid light-induced phase transitions in a single nanoparticle [10], it was demonstrated that a nanoparticle with a diameter of the order of 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.

In our experiment, the switching between the nanoparticle's four different structural states, corresponding to a *quaternary-logical memory element*, is achieved by excitation with single laser pulses, thus writing information into the nanoparticle, while the reading of this information is done using a pump-probe arrangement, sampling changes in the reflectivity of the nanoparticle. We find that the energy required for switching between the logic states can be as low as 1.5 pJ , an order of magnitude less than the energy required in today's state-of-the-art hard disk technology. This value should be compared with the thermodynamic lower limit obtained from the latent heat necessary for a particle of this size to undergo a β -to-liquid phase transition [11], which is of the order of 80 fJ . In fact only a fraction of the pulse energy is actually absorbed by the particle, with the rest scattered by and transmitted through it. The absorbed part of the pulse energy is spent to heat the particle to the phase transition temperature and to provide for the latent heat of transition. A fraction of the absorbed energy also dissipates radiatively, and as heat in the fiber tip.

By using the atomic beam from an effusion cell [Fig. 2(a)], the gallium nanoparticle was grown at the 30 nm aperture of the tip of a gold-coated tapered fiber [Fig. 2(b)] maintained at 80 K during growth. Using the light-assisted deposition technique [12], gallium was deposited at a surrounding pressure of 10^{-6} mbar at a rate of 3.0 nm/min for 30 min , forming a nanoparticle in the center of the aperture of the fiber tip [Fig. 2(c)]. Having grown the nanoparticle in this position, an external fiber-optical setup was used for its excitation and monitoring, as illustrated in Fig. 3.

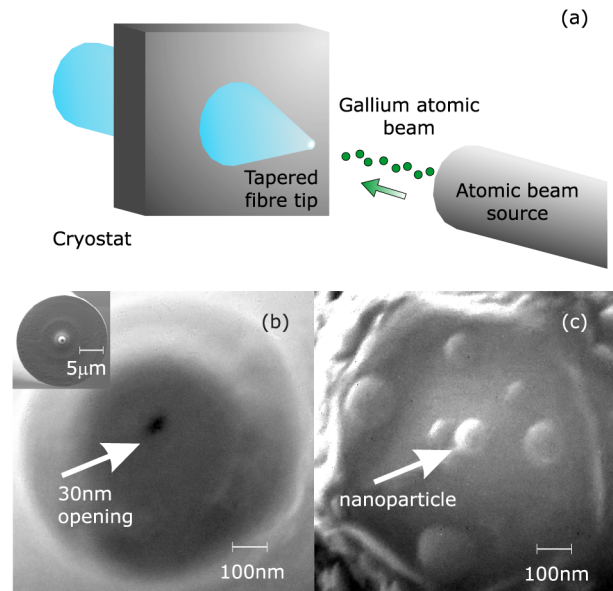


FIG. 2 (color online). (a) Schematic picture of the deposition process to form a gallium nanoparticle at the fiber tip. Below, scanning electron microscope images of the aperture of the fiber tip are shown (b) before and (c) after growth of the gallium nanoparticle. The size of the nanoparticle is 80 nm .

In order to read out the reflectivity of the nanoparticle and thus its structural phase, a pump-probe technique was employed. A continuous-wave diode laser of 1310 nm wavelength probed the particle reflectivity, while another diode laser of 1550 nm wavelength modulated at a frequency of $f = 2.5 \text{ kHz}$ acted as the pump. The reflected probe signal was measured by a photodetector, while a wavelength-division multiplexer (WDM) combined with a bandpass filter blocked the reflected pump light. The

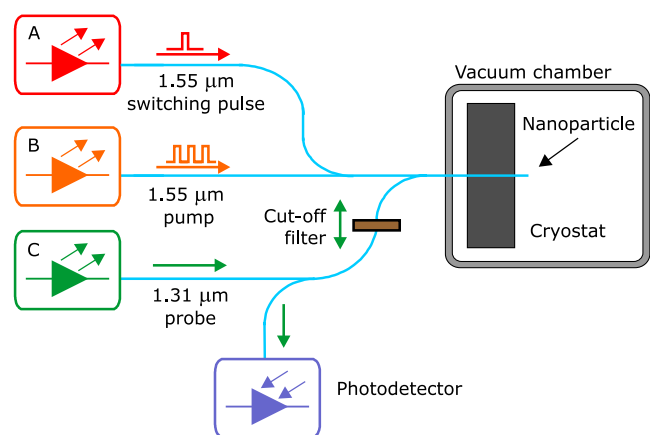


FIG. 3 (color online). The switching and diagnostics setup. Laser A delivers on-demand single pulses, used for switching the nanoparticle into the four different phases corresponding to quaternary-logical memory states. The low-power modulated pump laser B and continuous-wave laser C are used for monitoring the particle's logical state.

pump-probe technique provides a reliable way of monitoring transitions between phases of the particle. As the modulated pump gives small periodic increases in the temperature of the particle, the modulated component of the reflected probe signal is proportional to the temperature derivative of the particle's reflectivity. Its detection using a lock-in amplifier offers a substantially higher signal-to-noise ratio in monitoring the structural phase of the nanoparticle than a direct measurement of small changes in the reflectivity of the particle.

As the temperature is increased from $T = 100$ to $T = 160$ K, a sequence of narrow peaks in the induced reflectivity change is recorded, as shown in Fig. 4(a). Each peak corresponds to a structural transformation between two phases of the nanoparticle, or equivalently two logical states, with positive and negative peaks corresponding to pump-induced increases and decreases in the nanoparticle reflectivity. Starting from a temperature of $T = 100$ K, we label the consecutive phases between the transition peaks as 0, 1, 2, and 3, to represent the logical states of the memory. As the nanoparticle is cooled back to $T = 100$ K, it remains in state 3 until the pump and probe lasers are switched off, allowing the effective nanoparticle temperature to decrease enough for the system to relax to the initial state.

In order to demonstrate the memory functionality of a single gallium nanoparticle, the switching between different logical states must be performed in a controlled way. In addition, in order for the nanoparticle to function as a higher-order logical element, the possibility to switch the ground state into any targeted higher state by direct excitation is essential. In our experiment this control was achieved by means of a third, manually triggered 1550 nm pulsed laser, providing the optical writing pulses (laser A in Fig. 3). While the pump laser operates at an intensity such that the energy supplied is insufficient to induce a complete phase transition in the nanoparticle, the switching laser operates at a higher power, at which a complete transition with a single optical pulse is possible. The total energy in the pulse is changed according to which state is targeted in the transition.

With increasing ambient temperature for a nanoparticle initially in the ground state 0, we investigated phase transitions obtained by sending a single optical pulse at $T_0 = 120$ K. For a pulse of energy 4.8 pJ (as reaching the particle) and duration 1 μ s, the nanoparticle was found to fully transform its phase from the 0 into the 3 state. As the ambient temperature was further increased to $T = 160$ K, no other peaks were found, as shown in Fig. 4(b). The lack of peaks in the remaining part of the scan is explained by the fact that the energy deposited was large enough to fully transform the particle directly from state 0 to state 3. After exposure to the optical pulse, the nanoparticle remained in this higher state even as the ambient temperature was reduced to the initial $T = 100$ K, due to

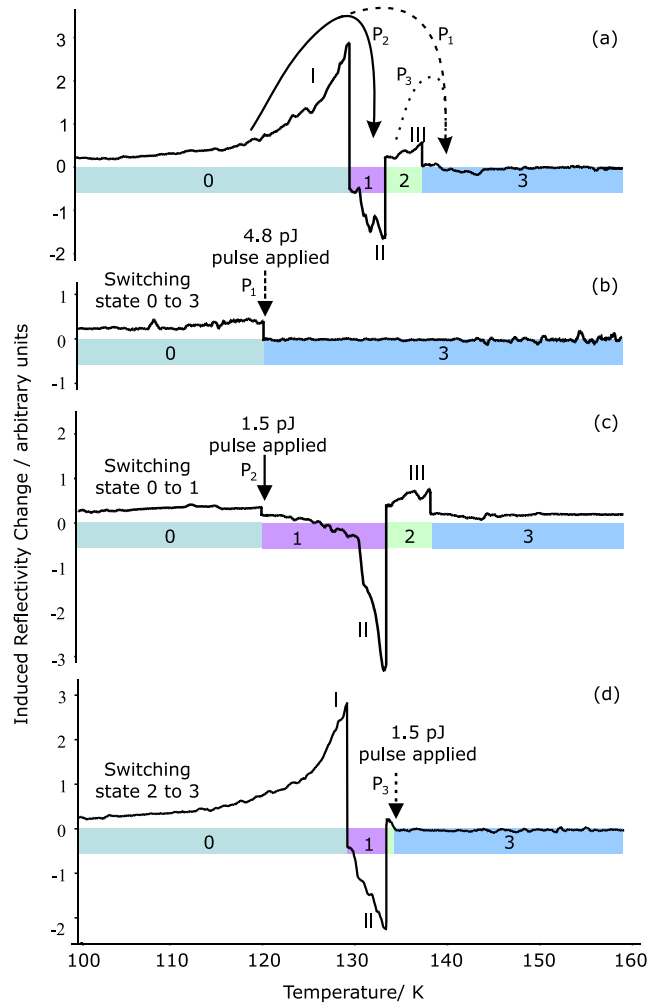


FIG. 4 (color online). Quaternary memory functionality of the nanoparticle, with the particle state monitored using the pump-probe technique. In (a), the temperature dependence is recorded in the absence of writing pulses, while in (b) and (c) switching optical pulses of 1 μ s duration and energies 4.8 and 1.5 pJ were applied at $T = 120$ K. In (a), the particle undergoes a series of phase transitions between four states with different optical properties, while in (b) and (c) the particle is directly switched from state 0 into states 3 and 1 (indicated by labels P_1 and P_2), without passing through intermediate stages. Finally, in (d) a 1.5 pJ pulse was applied at $T = 135$ K, switching the nanoparticle from state 2 to state 3 (indicated by label P_3).

the hysteresis present. Thus we have used a single laser pulse to write the logical state 3 into the nanoparticle memory element. In order to erase the memory of the nanoparticle, the temperature was brought down to $T = 100$ K and the lasers were switched off. Once this was done, the nanoparticle returned to the ground state 0, as was also verified by an additional scan of the ambient temperature which reproduced the graph shown in Fig. 4(a).

The entire cycle can also be performed all-optically by keeping the cryostage temperature at a fixed value located

below the lower limit of the hysteresis, and by using the heating effect of the probe laser to obtain an effective set point within the hysteresis loop. In this case the erasing operation is executed simply by switching off the probe laser. Hence the writing, reading, and erasing cycle of the nanoparticle memory can be repeated time and time again without need to adjust the background temperature, with the nanoparticle thus operating more like a typical memory element. For the sake of simplicity, however, the former format was chosen.

One can set the nanoparticle into one of the intermediate memory states by using an optical pulse with less total energy than for the 0 to 3 state transformation. For example, Fig. 4(c) shows the result of increasing the temperature as before to $T_0 = 120$ K but instead using a single optical pulse of energy 1.5 pJ and pulse duration 1 μ s for the excitation of the nanoparticle. In this case the absence of peak I and the presence of all the remaining peaks shows that the phase of the nanoparticle was transformed from state 0 into state 1. Finally, it is not necessary for the nanoparticle to initially be in state 0, as switching between other states is obviously also possible. Figure 4(d) shows precisely this for the transition between the states 2 and 3. In this case the temperature was increased to $T_1 = 135$ K, placing the nanoparticle in state 2, at which point a 1.5 pJ pulse again was applied. This time, the absence of peak III proves that the particle was switched to the intended state. Following the phase diagram of gallium [11], the states 0, 1, 2, and 3 can be attributed to the γ , ε , β , and liquid phases.

This first demonstration of an optical quaternary-logical element has several important implications. For example, in arithmetics it allows for entirely new algorithms in computation where complex number arithmetic is considerably simplified and error accumulation can be reduced [5]. The switching properties of the single nanoparticle are not limited to purely optical waves, but can also be applied to surface plasmon polariton waves, which are electromagnetic waves traveling at the interface between a metal and a dielectric. In this application, the switching of the state of a nanoparticle situated in a gap in a plasmonic waveguide or nanoparticle array can radically alter the transmission across the gap [6,7].

Historically, the addressing of optical memory elements has been performed by diffraction-limited means, with the exception of magneto-optical recording in which the writing process is performed using an electromagnetic read-write head. In order to provide for a read-write functionality not suffering from the diffraction limitation, a mechanical setup similar to that of today's hard disks could be employed, but using a tapered fiber tip for the read-write operation rather than an electronic coil.

In conclusion, we have demonstrated that a single gallium nanoparticle undergoing light-induced structural transformations possesses all the necessary characteristics

to act as a rewritable all-optical quaternary memory element. The data density for a hexagonal close-packed lattice of 80 nm particles is 0.1 Tb/in², if only one binary bit of information is stored in each nanoparticle. However, for the quaternary memory functionality, each nanoparticle is capable of storing twice as much information, and hence this estimate is doubled to yield 0.2 Tb/in². Although this assumes a convenient way to address individual particles packed together, it is a well-founded indication of what may be achieved using this technique, and should be compared with today's density of 0.015 Tb/in² in Blu-ray DVD and recent records of 0.23 Tb/in² in hard disks employing perpendicular recording technology, going beyond the superparamagnetic limit [8]. In terms of energy requirements, we have achieved writing functionality with as little as 1.5 pJ of total pulse energy. This is to be compared with state-of-the-art figures of 15 pJ for the energy required to write single bits of information [1]. Once again, this figure should be used as a mere indication as the value will depend on the details of the structure being used. However, both estimates clearly show the potential for single nanoparticles to provide memory functionality in future highly integrated nanophotonic devices, operating at very low power levels.

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