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Direct writing-in and visualizing reading-out data storage with high capacity in low-cost plastics

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Abstract: The explosive growth of the global data volume demands new and advanced data storage methods. Here, we report that data storage with ultrahigh capacity (~1 TB per disc) can be realized in low-cost plastics, including polycarbonate (PC), precipitated calcium carbonate (PCC), polystyrene (PS), and polymethyl methacrylate (PMMA), via direct fs laser writing. The focused fs laser can modify the fluorescence of written regions on the surface and in the interior of PMMA, enabling three-dimensional (3D) information storage. Through the 3D laser processing platform, a 50-layer data record with low bit error (0.96%) is archived. Visual reading of data is empowered by the fluorescence contrast. The broad variation of fluorescence intensity assigns 8 gray levels, corresponding to 3 bits on each spot. The gray levels of each layer present high stability after long-term aging cycles, confirming the robustness of data storage. Upon single pulse control via a high-frequency electro-optic modulator (EOM), a fast writing speed (~1 kB/s) is achieved, which is limited by the repetition frequency of the fs laser.

Key words: laser modification; fluorescence; micro-nano fabrication

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1. Introduction

With the rapid development of the digital era, information including character, sound, image and video is boosting faster than ever[1–3]. People are motivated to develop information storage technology with a higher capacity, longer lifetime and lower energy consumption[2]. Offering significant advantages in terms of capability, durability and robust reliability, optical data storage has gradually replaced magnetic and other forms of data storage[3]. Generally, the writing-in of optical data storage is achieved by changing the microscale physical or chemical properties of media[4], and the read-out of stored information is realized by photoinduced fluorescence or reflectance contrast[4–9]. The writing and read-out of previous-generation optical data storage devices such as CDs, DVDs and blue-DVDs are two-dimensional (2D) optical data storage systems. Namely, they are based on the surface modification of polycarbonates[10]. Currently, such optical storage technology has encountered bottlenecks in storage capacity due to the Airy disk diffraction limit[11] and volume storage density[12, 13]. As promising approaches, near-field optical data storage[11, 14–17] and multiplexed information with multidimensional physical parameters[18–20] have been developed. However, these advanced technologies are often equipped with complicated manipulation equipment, which hinders their industrial production. Recently, fs laser-based refractive index[21] and fluorescence[22] modification technologies have attracted much attention because of their ultrahigh speed and easy implementation. Furthermore, fs-laser-matter interactions have shown the capability of 3D writing upon the effective fabrication of waveguides, microchannels and other functional devices inside the bulk of transparent materials[23–25], which could be used for 3D optical data storage[3, 24, 26, 27]. However, the data capacity is still struggling to reach the terabyte (TB) level due to the depth-dependent spherical aberration[28, 29], lower gray level assignment accuracy[22] and high bit error rate for data points[13].

Here, optical data storage with a capacity of ~1 TB is realized in PMMA by means of fs laser writing. The high data capacity arises from the 3D storage schema and multiple gray levels of fluorescent contrast. By penetrating the interior of PMMA to achieve multilayer data writing, the focused laser beam enables 3D information storage. The superior linear correlation between the single pulse energy and fluorescent contrast renders the direct writing-in of 8 gray levels. Moreover, the written data present a low bit error rate, fast writing speed, direct visual reading and high stability. This optical data storage technique is expected to offer an implementable multidimensional light disk to match the requirement of rapidly growing data volumes.

2. Experimental section/methods

2.1. Materials and methods

Transparent PMMA (CQ grade, Goodfellow, UK), polycarbonate (PC), precipitated calcium carbonate (PCC) and polystyrene (PS) samples were purchased online. The femtosecond laser used was an amplified Ti:sapphire laser with a central wavelength of 800 nm, a pulse duration of 35 fs and a

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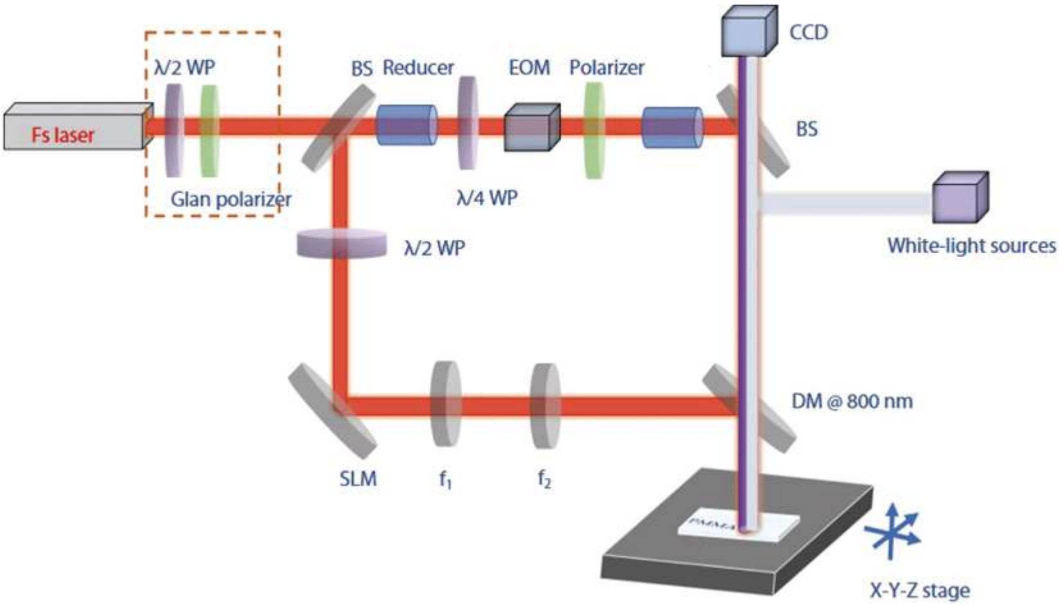

Fig. 1. (Color online) Schematic diagram of the femtosecond laser writing system.

repetition rate of 1 kHz. An array of periodic laser-induced bits was fabricated through a homemade 3D three-dimensional laser processing system. To increase the storage density, we used a high NA 100× with a 1.2 mm working distance IR objective bought from Olympus. Moreover, the environmental parameters of the aging experiment were controlled by an industrial grade constant temperature and humidity chamber.

2.2. Characterization

The UV–vis absorption spectrum, PL spectra and confocal fluorescence microscope images were measured by using a Shimadzu UV/Vis UV - 2600 spectrometer, a WITEC-R200 and K1-Fluo RT, respectively. According to the depth of the laser-modified region, an oil immersion microscope objective (Olympus, UPlanSApo, 1.2 NA, 60× magnification, 0.28 mm working distance) and an air microscope objective (Olympus, UPlanSApo, 0.75 NA, 20× magnification, 0.6 mm working distance) for confocal fluorescence microscopy images were used to focus light to the region modified by the fs laser. The mean and standard deviation of laser-induced fluorescence that was read out by commercial confocal fluorescence microscopy and were evaluated using ImageJ software.

3. Results and discussion

Fig. 1 and Fig. S1 present a sketch and actual scene of the ultrahigh-density 3D laser-based data storage platform. Here, we employed a 1 kHz, 800 nm Ti:sapphire oscillator–amplifier laser system as the light source, producing an ~3.5 mJ pulse with a laser peak width of ~35 fs. The fs laser can be controllably driven, modifying the microscale region in bulk materials or low-dimensional materials. To precisely and rapidly control the laser power, we introduce a combination of an electro-optic modulator (EOM), a quarter-wave plate and a polarizer. Moreover, we introduce a liquid crystal spatial light modulator associated with the 4f system for multibeam parallel processing[30], focal aberration correction[31] and fs-pulse shaping[32] in future exploration and research.

Based on the self-developed laser writing system, various kinds of low-cost plastics, such as PC, PCC, PS and PMMA, which do not fluoresce in the visible spectrum before laser modification, can be considered candidates for optical storage research. Once the chosen regions of PC, PCC, PS and PMMA are irradiated by the laser, significant fluorescence is observed from the laser irradiated regions by confocal microscopy equipped with a reading laser wavelength of 405 nm and with a highly sensitive photomultiplier with a filter ranging from 420–750 nm (Fig. S2). The PL may originate from fs-laser-induced bond scission of plastics, which can serve as the fluorescence emission center[27]. The PMMA PL emission wavelength shown in Fig. 2(a) varies with the excitation laser wavelength. This may be ascribed to light-matter interactions producing active clusters[33, 34] or to red-edge excitation effects[33]. Regarding storage capacity, the diffraction-limited resolution, $r = \frac{0.61\lambda}{\mathrm{NA}}$ (where λ is the wavelength of the incident laser), is critical for optical writing ability[35]. Judging from the scanning electron microscopy (SEM) image in Fig. 2(b), the size of the modified region on the PMMA surface was ~200 nm, indicating that the diffraction limit can be exceeded during irradiation. However, even though cold knifes, such as ultrashort pulse interaction-based writing processes[36], can break the diffraction limit, the size of the memory bit is still limited by the resolution of the reading device. Based on current common lenses (e.g., NA ~ 1.2, 60×), a typical reading resolution of 600 nm is shown in Figs. 2(c) and 2(d).

For effective use of their interior space, modification should be attempted by focusing the beam into the interior of the low-cost plastics. PMMA was purchased from the Goodfellow company, the absorption spectrum of which is shown in Fig. 3(a). The high transmittance of transparent PMMA in the visible wavelength indicates that it is feasible to write information into the interior volume using a fs laser so that a slight pattern can be recorded at different depths of the low-cost transparent material by adjusting the *z* position of the laser focal plane. Via a fixed excitation energy, a uniform intensity of fluorescence can be written into each data layer. The fluorescence intensity can be assigned to the “on” or “off” state as the “0” and “1” states in traditional CDs or DVDs. The 3D confocal fluorescence microscope image shown in

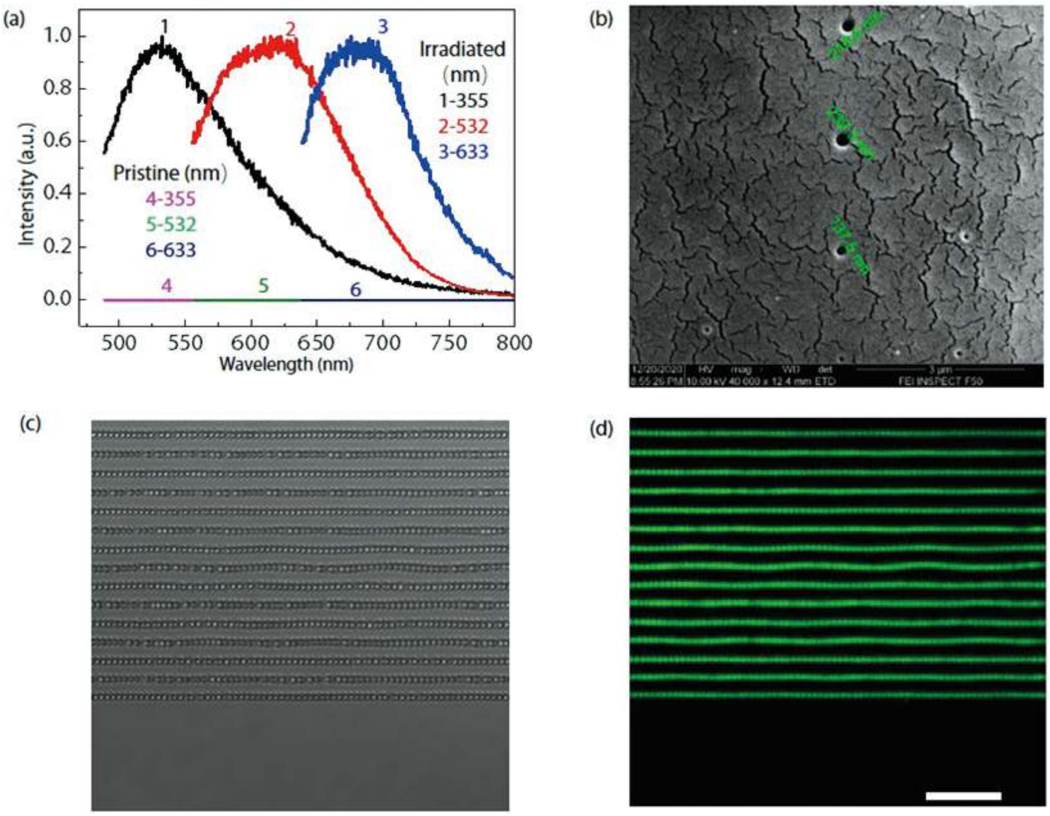

Fig. 2. (Color online) (a) PL spectra of pristine and laser-irradiated PMMA (single fs). The PL data were acquired using 355, 532 and 633 nm lasers. (b) SEM image of dots modified by threshold fs laser power at the surface. (c, d) Optical and confocal fluorescence microscopy images of the dots beneath the 30 μm surface. The scale bar is 8 μm. Here, 14 dots can be observed when the measurement length is 8 μm, suggesting that the dot spacing is ~600 nm.

Fig. 2(b) further proved that 3D optical storage was successfully released based on the different layer writing. Based on photoinduced fluorescence, bit data or designed patterns, such as the Emblem of Nanjing Normal University and Southeast University, the Great Wall and Albert Einstein can be stored inside the bulk sample, as shown in Fig. 3(c) and Fig. S3.

By adjusting the power density of the focused laser, multiple gray levels can be assigned, further improving the data storage capacity. Modulating the pulse duration or pulse energy, the writing parameters could be used to achieve controllable fluorescence intensity[22]. To expand the storage capacity of each single pulse recording bit, pulse energy-related fluorescence intensity variation was examined, as shown in Fig. 4(a). A regular array of modified regions (1st layer −10 μm beneath the surface) was retrieved under excitation, the center distance between two dots was 2 μm, and the rows were separated by 2 μm[37]. The corresponding PL intensity values acquired using the fluorescence reading software show distinct linearity, as shown in Fig. 4(b)[3, 24, 38]. According to the variability and lower bit error rate, at least 8 gray levels can be calibrated, corresponding to 3 bits, improving the storage capacity of each single pulse. PL intensity values in deeper layers were also acquired, as shown in Figs. 4(c), 4(d) and S4. Even though the required pulse energy is different from that of the 1st layer, distinct linearity of the PL intensity can also be obtained in each layer, offering equivalent gray levels. The higher pulse energy needed for writing deeper layers is because the objective was without compensating spherical, and absorption changes were inevitable with increasing depth[6, 39, 40]. It should be noted that the low threshold of pulse energy, high signal-to-noise ratio and the absence of cross-talk between layers suggested that the storage capacity limitation could be further improved. Nevertheless, stacking 50 layers with a spacing of 10 μm (Fig. S4), a resolution of 600 nm and a data bit of 3 can afford a storage capacity of 1 TB on both sides of a standard disc. Even though the capacity used here should be defined as equivalent capacity, higher storage can be expected if advanced reading techniques are adopted, and higher storage can be expected if advanced reading techniques are adopted.

The bit error rate in the optical disk is extremely important to realize industrial application[41]. In this work, the freshly modified PMMA with an 8 gray level only exhibited a 0.96% bit-error rate, as shown in Fig. 5(a). Bit-error-rate stability is also crucial for practical implementation[42]. To confirm whether the fs laser-written optical disk can be used as a long-term data storage medium, the modified sample was placed in an environment with 85% relative humidity and 85 °C for 72 h. As shown in Fig. S5, the gray level of aged samples maintained high visible stability without any crosstalk. The 8-gray level presented a 2.2% bit-error rate, suggesting that the stored data were considerably stable, as shown in Fig. 5(b). The improved fluorescence intensity may be induced by the formation of more reactive groups through thermal treatment[24]. Hence, our PMMA-based optical disk can be viewed as a reliable and stable optical storage medium that can be preserved under ambient conditions for a green future.

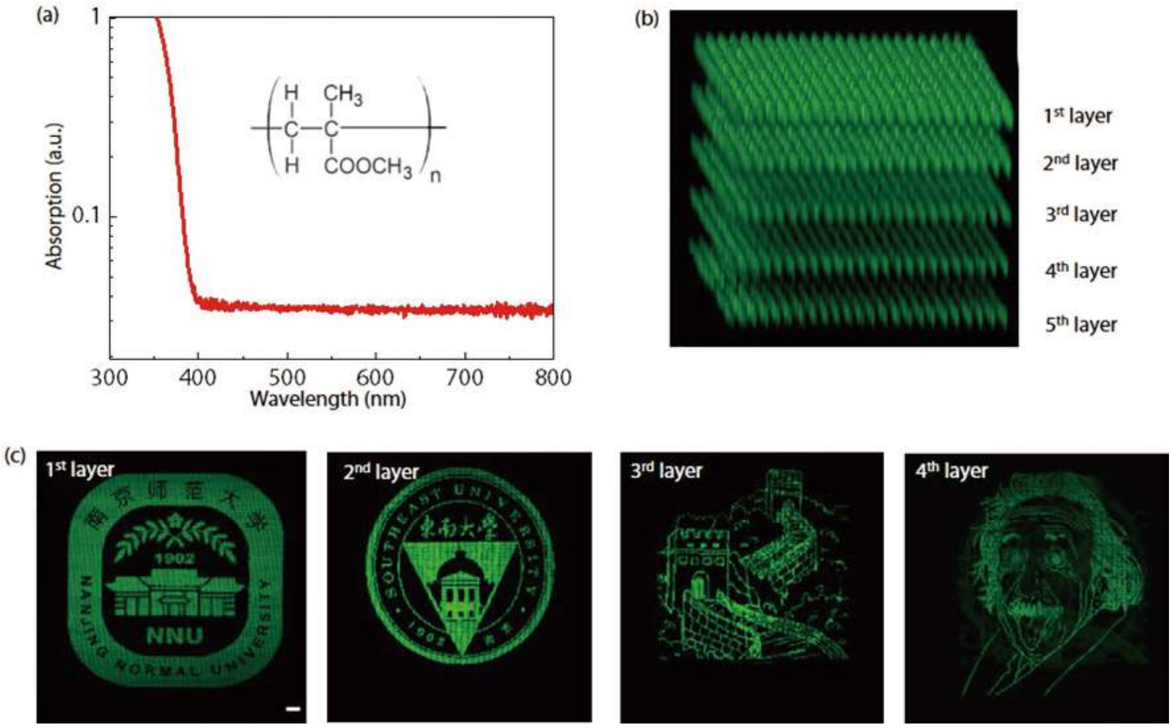

Fig. 3. (Color online) (a) Absorption spectrum of pristine PMMA, the chemical structure of which is shown in the inset. (b) The 3D fluorescence structure of the array with fixed writing power (40 nJ) inside PMMA separated by 10 μm, which was read out by confocal fluorescence microscopy. (c) Confocal fluorescence microscopy image of the fs laser-modified Emblem of Nanjing Normal University and Southeast University, The Great Wall and Albert Einstein in the 1st, 2nd, 3rd, and 4th layers, respectively, scale bar of which is 30 μm. Specific patterns can be stored in PMMA with a layer separated by 10 μm.

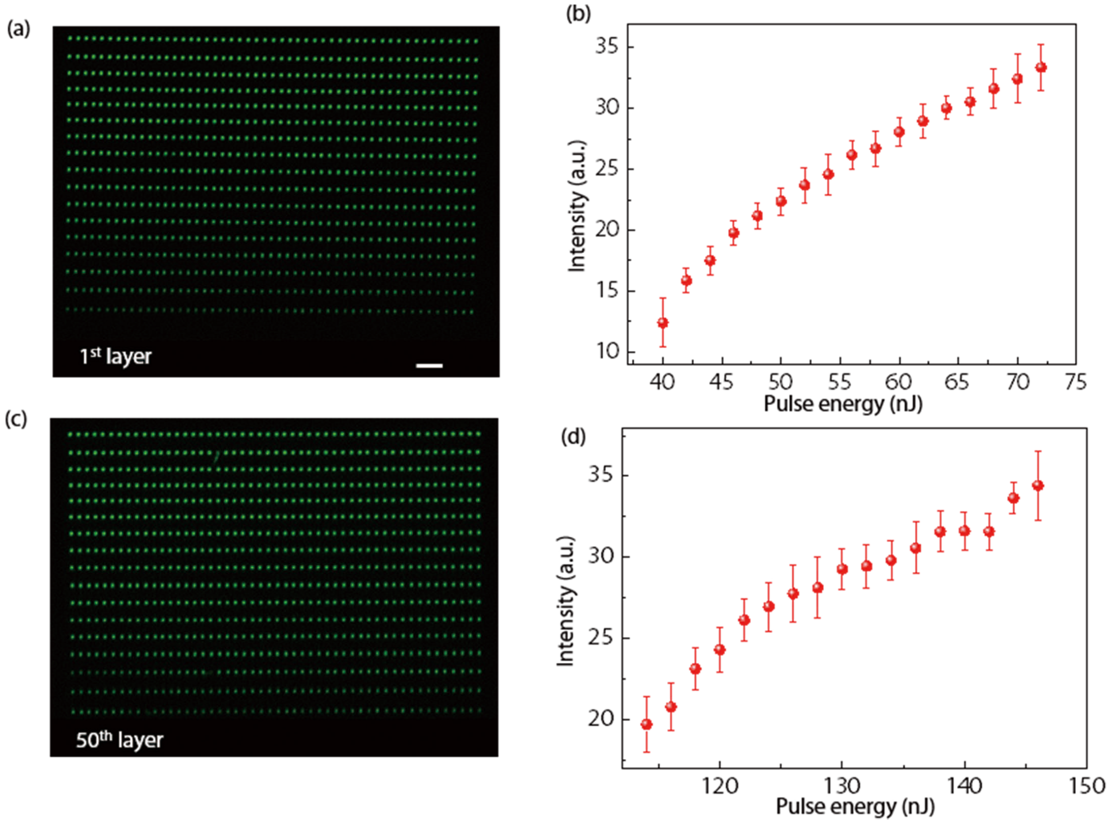

Fig. 4. (Color online) (a, c) Confocal fluorescence microscopy images of the dots (1st and 50th layers) written by a fs laser with a single pulse, scale bar: 20 μm. (b, d) show the fluorescence intensity change according to pulse energy evolution, which are acquired from the 1st and 50th layers, respectively. The threshold power is 40 nJ for the 1st layer and 114 nJ for the 50th layer.

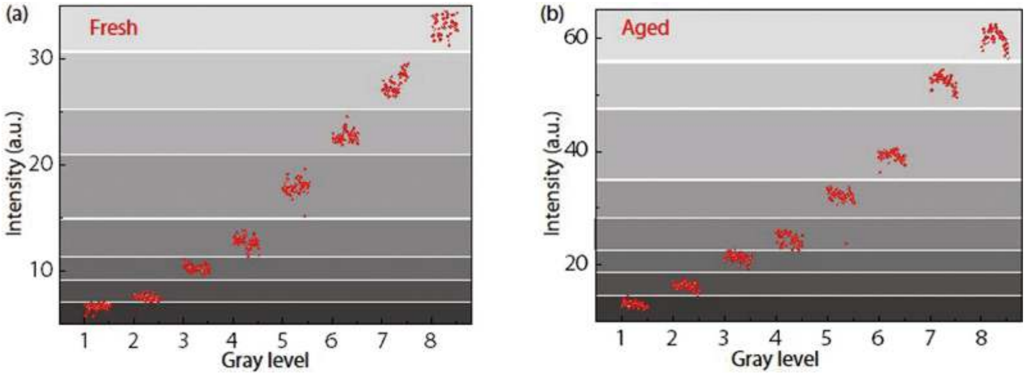

Fig. 5. (Color online) Gray level assignment based on fluorescence distinguish. It should be noted that both (a) the fresh sample and (b) the aged sample can be of capability of 8 gray level assignment.

4. Conclusion

In conclusion, we reported 3D optical memory in PMMA using the femtosecond pulsed laser processing platform. Multilayered bit patterns (bit resolution = 600 nm, layer separation =10 μm, gray levels = 8) can be recorded into the volume and can be conveniently read out through commercial confocal microscopes. The change in fluorescence intensity corresponding to the gray level of memory bits remained stable in extreme aging circumstances, which was essential for long-term preservation. This high storage optical disk can provide an innovative perception to advance optical data storage technology for information explosions.

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Appendix A. Supplementary material

Supplementary materials to this article can be found online at https://doi.org/10.1088/1674-4926/43/6/062301.

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