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PAPER

Inkjet-printed quantum dots on paper as concept towards high-density long-term data storage

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Abstract

Handling and storing the immense amounts of data native to the information age is a major challenge in terms of technological sustainability and energy demand. To date, tape storage remains the most widespread method for data archiving, while DNA data storage appears to offer the best data density and long-term stability in the future. However, DNA data storage is still in its infancy primarily due to economic and accessibility challenges. This emphasizes the need for more practical and readily available alternatives. We present a method for data storage utilizing inkjet printable quantum dots on paper with photoluminescence (PL) readout. Our proof of principle study showcases the ability to print and stack multiple bits of data on a single spot by exploiting the unique PL properties of quantum dots. This approach utilizes easily accessible resources, including a consumer-grade printer and paper as the substrate. Additionally, we perform initial stability tests, investigate scalability by controlling emission intensity, and evaluate the potential data density achievable by our approach.

1. Introduction

Rapid advances in information technology have raised demands for high data density, cost efficiency, scalability and longevity of data storage [1]. While various technologies have proven themselves in specific areas, finding a solution that excels in all these areas is a formidable challenge. Commonly used methods include tape storage [2], hard drives [3], and printed books, each with its own set of strengths and weaknesses. Even innovative approaches such as DNA data storage or our proposed method based on quantum dots applied to paper with inkjet printers have their shortcomings. However, we anticipate that our method will prove highly competitive across multiple categories. For a more detailed comparison of different data storage technologies over the last century, we refer to the literature, e.g., [4].

The current state of the art for archiving data remains magnetic tape storage. It offers the presumably best compromise to date despite its original invention in 1928 [5] and mainstream introduction in 1953 [6]. Its comparatively good data density, reading as well as writing speeds, and decent cost efficiency have yet to be superseded by a superior solution. However, it comes with certain weaknesses, including demanding storage conditions and necessary maintenance, as data migration is typically required every 10–30 years [7]. In addition, scalability appears to be approaching its limits [8] as individual bits already reach the few-nm scale [9].

DNA data storage is one of the presumed contenders to excel in all the above criteria [8, 10]. It promises significantly higher data densities (theoretically up to exabytes per mm^3) [11, 12], and excellent long-term storage reliability [13, 14]. However, it is important to note that DNA storage is still in the developmental stage and faces challenges regarding its accessibility and retrieval [15]. Further research and development efforts are needed to transform it into a practical, cost-effective alternative suitable for widespread mass data storage.

Alternate approaches toward more efficient storage are based on optical multiplexing. Facets of this are reported for polarization [16, 17], spatial dimensions [18, 19], wavelengths [20, 21], and combinations of several of the above [22]. In particular, quantum dots have attracted attention as a viable option for data storage with their distinct emission properties and suitability for wavelength multiplexing [23–25].

The concept of using quantum dots for data security or data storage first emerged in the early 2000s, as evidenced by the patent 'Quantum dot security device and method' [26]. This patent proposed utilizing the optical properties of mixed quantum dots applied to a substrate to generate readable patterns. Quantum dots can be made water-soluble [27, 28], closely resembling standard colors such as those used in drawing or printing. This characteristic enables the creation of multiplexed patterns [29], significantly enhancing the data density per data point. A recent example of using colors to increase data density in an existing method is multi-colored QR codes [30].

Colors are traditionally applied as inks, and in the evolution of data storage methods, manual handwriting on paper was the initial approach. The solution to the increasing need for multiple copies of data and the storage of larger amounts of data was printing techniques. Inkjet printing has emerged as the most widely used form of printing and is undergoing continuous improvements in resolution [31], substrate compatibility [32], droplet control and precision [33], 3D printing [34], and overall print quality.

In our work, we demonstrate a fusion of conventional inkjet printing technology with quantum dot-based inks. By leveraging the size-dependent and consequently tunable emission wavelength of quantum dots [35], we achieve a fourfold increase in data density per dot compared to single-color data storage. Moreover, this method holds the potential for further expansion through the utilization of additional colors or parameters, such as quantum efficiency, as we will explore in our discussions.

Our approach features great scaling potential, as print resolution limits are in the single-digit micron range [36] and the number of colors and intensity levels can potentially be increased by orders of magnitude. This scalability could result in data densities surpassing those of current tape storage methods, all while maintaining the inherent advantage of long-term reliability provided by acid-free paper. Furthermore, there is a possibility to develop environmentally friendly inks based on similarly safe and non-toxic quantum dots, potentially leading to a long-term data storage technique with substantially lower energy consumption for both reading and writing processes.

2. Results and discussion

The proposed method exploits the distinct PL properties and encodes data to their multiplexed emission spectra by printing combinations of quantum-dot-based inks on paper (figure 1). We use CdSe/ZnS core–shell-type quantum dots to prove the concept's viability. They are inexpensive and readily commercially available in various size distributions, each featuring spectrally narrow emission bands [38]. These properties make them highly suitable for this proof of principle study. However, it is important to note that their environmental impact is somewhat higher compared to other similarly exploitable quantum dots, such as CuInZnS.

The central emission wavelengths of quantum dots are determined by their sizes, as they confine excitations in all three dimensions [35]. Particularly relevant for their use in data storage is the size distribution of the quantum dots. It governs the spectral width of the individual emission bands as the emission from an individual single quantum dot is much narrower.

Using a solution mixture and adding the water-solvable quantum dots creates a printable liquid that can be used with an inkjet printer like commercial inks (details of the controlled quantum dot concentrations are given in the methods section).

Data are encoded by mapping single or multiple bits of information to the distinctive spectra of these quantum dots and their combinations. The data are then printed on paper in form of their corresponding quantum dot combinations, with acid-free paper being the preferred choice for long-term durability. This results in the data being stored on paper as a dot matrix featuring various 'colors' as illustrated in figure 1.

The data can be retrieved by illuminating the paper and spatially scanning the emission spectra using a standard spectrometer. Subsequently, the data are decoded from the PL spectra using the original encoding routines. Quantum dot-based inks offer a distinct advantage over standard inks, as they can be mixed within a small area without compromising their individual emission properties. The middle part of figure 1 shows the results for four different sized quantum-dot inks on paper (PL maxima at 540, 587, 629, and 657 nm). Their emission spectra exhibit minimal overlap with FWHM values of 28, 29, 26, and 27 nm for the respective maxima. As a result, these emissions can be readily distinguished. The PL spectrum obtained from the combination of all four quantum dot inks shows each of the individual features in the form of their respective emission maxima, thus providing clear proof of concept.

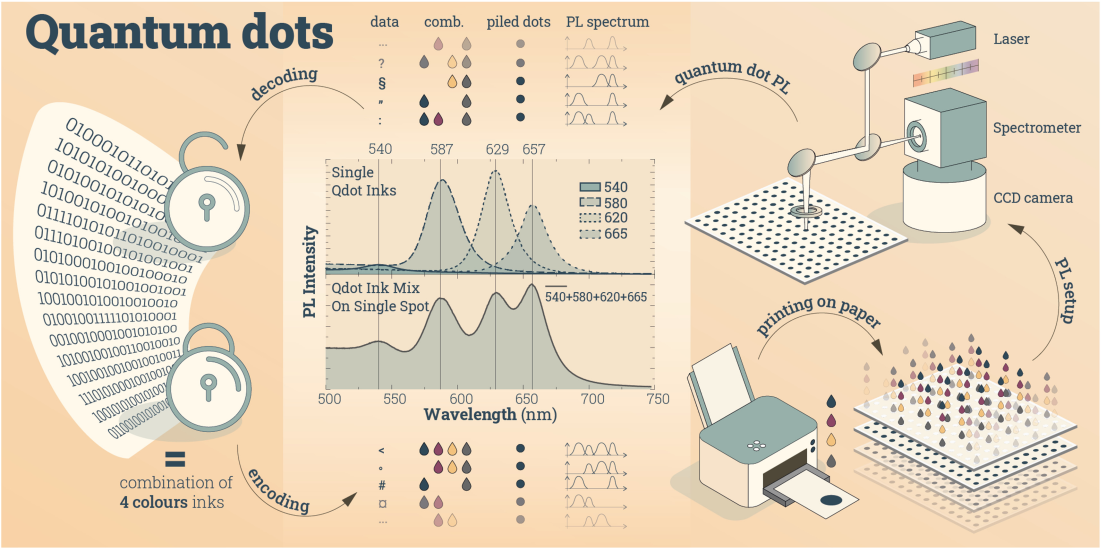


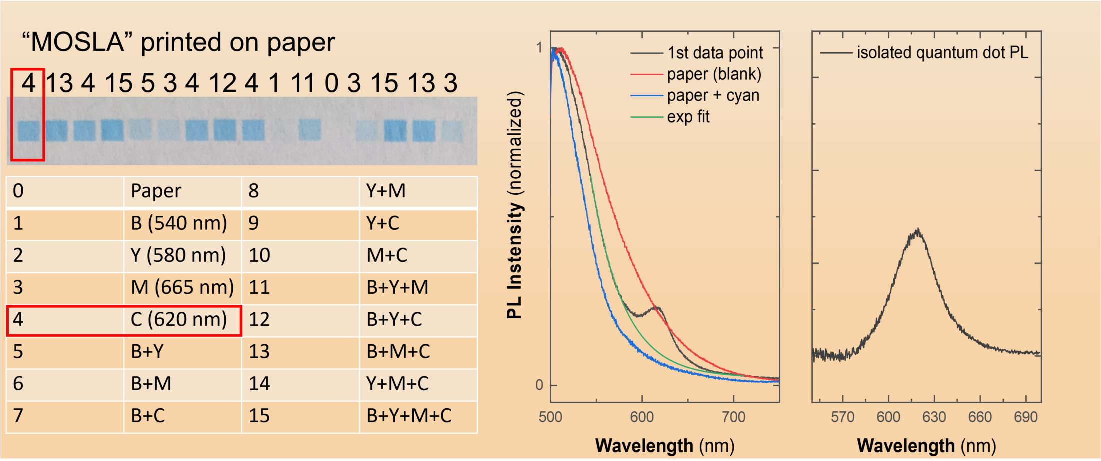
Figure 1. Quantum dot-based ink data storage process. Data are encoded to combinations of quantum dots and their associated spectra. Using an inkjet printer, the data are printed on paper in form of quantum dot combinations. The data can then be read out again by scanning the paper with a PL setup and decoding the spectra back to data. The graph in the middle shows the mixability of different quantum dot-based inks.

For our proof of principle experiment we chose four different quantum dot inks, each using one color cartridge originally intended for cyan (C), magenta (M), yellow (Y), and black (K) in standard commercial CMYK printers. The incorporation of four distinct quantum dot colors results in 15+1 unique and well distinguishable emission signatures per data point, i.e., each dot on the carrier medium. This significantly increases the data density compared to four standard inkjet colors, as explained in more detail in the data density evaluation. For the latter, it is exceedingly challenging to identify the base colors based on the emission spectra, which is crucial for data retrieval. Although it is possible to differentiate the reflectance spectra of such CMYK mixtures, the lack of clearly distinct features renders this method far less practical, necessitating advanced processing like machine learning for effective data retrieval. (cf Supplement 1, ‘Reflectance and PL spectra of standard printer inks’).

Our proof of principle system relies on a readily available, refillable consumer-grade printer (Epson eco tank 2720). While standard printer software allows for the separate printing of a dot from each color (CMYK), it does not allow us to print combinations of well-defined distributions from more than two color cartridges. To address this limitation, we have developed a software tool that directly controls the print head and works with matrices instead of RGB images. This approach solves both the challenge of mixing inks from multiple cartridges and the translation from RGB (computer) to CMYK (printer) color systems. As a conclusive demonstration of our data storage concept, we printed the acronym ‘MOSLA’ encoded with a Reed-Solomon-based code optimized for four colors. The printed data points along with the used encoding table and PL data for the first data point are shown in figure 2. To make the printed data points visible to the eye, we added cyan color to the inks. Further information can be found in Supplement 1.

The technical limitations of our consumer-grade printer prevent the automation of positioning each dot and thus do not allow us to present high-density data. However, a custom printer with a print head that can be moved in two dimensions, rather than just one, should solve this issue [37].

The printer is controlled using the printer-specific command language to navigate the print head and access the print head nozzles directly. As there is no programming guide for the ESC/P (Epson Standard Code for Printers) language [38] used by the default Epson drivers, we use the Gutenprint 5.3.3 drivers on a Notebook with the Arch Linux operating system. Gutenprint generates print commands in the ESC/P2 language (Epson Standard Code for Printers, version 2), for which a programming guide is available. As a basis for our quantum-dot printing application, we use the escp2-client [38]. It provides a python wrapper around ESC/P2 commands and information regarding their usage. The printer-specific header and footer commands are obtained by adding a virtual Epson eco tank 2720 to the CUPS (Common Unix Printing System) and printing an empty page using this virtual printer to generate commands for the Epson eco tank 2720. The resulting binary *.prn file is then parsed using the Gutenprint parse-escp2 script to identify the header and footer in the*.prn file. The technical reference sheet on the Epson homepage provides information on the number of nozzles per color.



0	Paper	8	Y+M
1	B (540 nm)	9	Y+C
2	Y (580 nm)	10	M+C
3	M (665 nm)	11	B+Y+M
4	C (620 nm)	12	B+Y+C
5	B+Y	13	B+M+C
6	B+M	14	Y+M+C
7	B+C	15	B+Y+M+C

Figure 2. Data reading of the first data point in ‘MOSLA’. The top left shows the word ‘MOSLA’ printed as code on paper. Each data point contains information which can be compared to the encoding table below. The right side displays the measured PL spectra of the marked data point on paper (black), the PL of paper without a data point (red), the PL of cyan ink printed on paper (blue), and an exponential fit for the paper PL of the measured data point (green). The PL spectrum on the right side demonstrates the isolated PL of the data point, with background PL from the paper subtracted using the green line’s exponential fit.

A python script is used to generate the printer command that utilizes the escp2-client commands relevant to quantum-dot printing, i.e., dot and dot-matrix generation, together with an interactive jupyter notebook and a docker container for portability. The script employs nested lists to represent nozzles, with the outer list representing all nozzles of one color on the print head, each inner list representing one row of nozzles, and each element in a list a singular nozzle. Each element can take the values between 0 and 3, with zero leading to an inactive nozzle during printing and 1, 2, 3 for droplets of increasing size.

The data is protected using a Reed-Solomon code, using the Galois Field (2^8), with the irreducible polynomial $x^8 + x^4 + x^3 + x^2 + 1$ and two error correction symbols. Following that, the encoded data is transcoded from binary to a quaternary system, where each quaternary symbol corresponds to one printer color. Using two error correction symbols allows for reliable correction of one error when the position of the erroneous dot is unknown, with the potential to correct up to four errors if all erroneous dots are part of the same encoded byte. If the positions of the erroneous dots are known, the error correction capabilities double, with two errors that can be reliably corrected and up to eight correctable errors if all erroneous dots are part of exactly two encoded bytes.

Following the successful proof of principle, we conduct an initial assessment of the method’s long-term data storage viability by exposing it to external stress in the form of temperature. For this assessment, we created printed areas measuring 0.5×0.5 cm^2 on paper, utilizing ink with a PL maximum at 657 nm. The samples are then tempered in an ambient atmosphere for 3 to 12 h. Figure 3(a) shows examples of the corresponding emission spectra recorded at room temperature under ambient conditions after heat exposure. The samples show great thermal stability when exposed to temperatures within the range of -4 °C and $+35$ °C. However, exposure to temperatures of 40 °C and above resulted in discernible degradation in quantum dot PL intensity. This result is in line with previous tests using a thermal-based inkjet printer instead of a piezo-based one. Here, employing the thermal-based printer led to lower PL intensities of the printed quantum dots. However, with a piezo-based inkjet printer we obtained similar results to those obtained with a pipette, making them the logical choice. Additionally, it is noteworthy that the ink solutions added for printing did not cause changes in the PL, except for the anticipated decrease in intensity.

To further assess the potential data stability across numerous readout cycles, we subject the samples to continuous, long-term laser exposure to check for signs of photodegradation. Figure 3(b) presents the corresponding emission spectra. Notably, during the entire investigated time period of 10 h, we observe virtually no decay in the quantum dot PL intensity. This result implies that the method can endure more than 360,000 readout cycles, assuming a conservative readout time of 100 ms.

While these findings do not replace the need for future in-depth long-term testing with optimized inks, printers, and paper substrates, these initial stability tests undeniably point to great potential for long-term data storage under ambient conditions.

It should be noted that the paper substrate exhibits a broad emission band spanning from below 500 nm to well above 600 nm, as depicted in figure 3(b). The emission profile is primarily dictated by the paper’s intrinsic characteristics and lacks distinct maxima. However, it may pose a challenge in discerning quantum dot maxima,

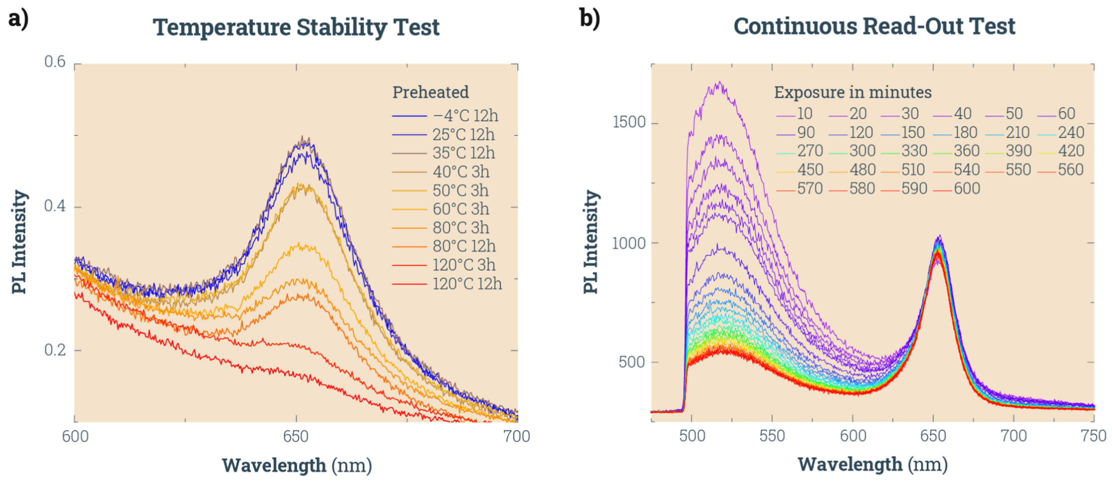


Figure 3. (a) Temperature stability tests. The samples undergo controlled heating in an oven under ambient conditions for specific durations, after which their photoluminescence (PL) is measured at room temperature. The graph demonstrates excellent thermal stability within the temperature range of −4 to +35 °C, with a noticeable decline in PL intensity observed at temperatures exceeding +35 °C. (b) Continuous read-out tests. The graph displays PL intensities recorded at different time intervals during continuous exposure (100 ms snapshots) spanning over 10 h.

particularly within the 500–550 nm range. Fortunately, potential issues arising from this optical background can be effectively addressed. Solutions include using specialized paper with reduced optical background, employing quantum dots with maxima at higher wavelengths, or increasing their concentration to enhance the intensity of their PL relative to the paper luminescence.

In summary, the background luminescence of the paper should not impact the data storage performance, as its influence can be mitigated, and the PL intensity of the quantum dots remains remarkably stable, as shown in figure 3(b).

Interestingly, the spurious emission related to the paper substrate at around 520 nm quenches, presumably due to photobleaching of the paper substrates. A direct comparison with the quantum dot PL emphasizes how stable the latter is against photodegradation [39–41].

While paper is susceptible to damage when handled roughly or exposed to moisture, it has proven its reliability in data archiving for centuries. It shares this vulnerability with other data archiving methods like tape storage and DNA. Given that standard archiving conditions already address these concerns, the development potential of this approach is likely to remain untouched.

3. Data density evaluation and future challenges

In addition to longevity, data density stands out as a critical metric when evaluating potential long-term data storage methods. Traditionally, data have been encoded in binary, represented by sequences of zeros and ones, corresponding to the standard on or off machine code. To encode any character, such as a letter, the American Standard Code for Information Interchange (ASCII) system requires eight bits or one byte of binary information. This implies the necessity to account for all 256 possibilities of the ASCII code in a bit sequence before moving on to the next character.

Such binary encoding can be readily replaced with colored dots. Using a single color is sufficient to represent the binary code by the presence or absence of a color. Colors based on quantum dots are mixable, allowing multiple colors to coexist on a single spot, as demonstrated in figure 1. This expands the number of possible states in a single spot according to the formula 2^n, where n is the number of colors utilized. Consequently, two blendable colors offer four possibilities per data point. This means that two individual bits can be accommodated within a single data point, reducing[the number of data points or dots required for the same information. A standard inkjet printer typically supports four colors, translating to 16 possibilities per data point, which increases the data density by a factor of four compared to traditional binary code. A detailed explanation on data density increase can be found in Supplement 1, 'Further explanations for the data density calculations'.

Several factors influence the maximum data density in this approach:

(1) Spot Size: The size of an individual spot (data point) is a critical factor. It can be limited either by the print resolution during data writing or by the optical resolution during data reading. Optical readout is often diffraction limited, providing considerably higher resolution than the printer.

(2) Number of Colors: The number of distinguishable colors in a spot is crucial, as it strongly influences the number of possible states.

(3) Intensity Levels: The ability to generate various PL intensities of each color through different quantum dot densities, and to reliably distinguish them, is another critical factor. The minimum intensity is then set by the desired signal-to-noise ratio.

(4) Encoding Technique: The chosen encoding method plays an important role as it determines the amount of information needed to store a piece of data.

Building upon our successful proof of principle demonstration, we aim to provide a realistic estimate of the attainable data density for storage utilizing printable quantum dots on paper.

Increasing the print resolution is presumably the most straightforward approach to enhance the data density. An alteration in print resolution from 300 to 2400 dpi amplifies the data density by a factor of 64, as eight times more dots can be printed in the x and y directions. Using 4 colors and no intensity levels, this results in an approximate storage capacity of 253 MB per DIN A4 page or 44.16 KB mm^{-3} considering the 0.1 mm thickness typical for 80 g mm^{-2} paper. Pushing to current technological limits, a theoretical spot size of 25 $\mu\mathrm{m}^2$ [31] would still be within the diffraction limit and yield a data storage capacity of 1.148 GB per DIN A4 page, translating to 0.2 MB mm^{-3}.

Optical crosstalk becomes a relevant concern when approaching diffraction-limited spot sizes. However, if the diffraction limit is still better than the print resolution, the impact of optical crosstalk should theoretically be negligible.

The data density can be further enhanced by using more colors. This increases the data density to 8/n data points or dots per byte, with n representing the number of colors. This is calculated by the following equation:

$$256^x = 2^n \tag{1}$$

Here x corresponds to the number of bytes per data point, 256 is the number of possibilities for ASCII code, and 2 is the number of possible read states for each color.

This scalability demands an expansion beyond commercial four-color printing. However, with specialized printing devices, this should not pose a significant challenge. Besides the printer, n is then determined by the available number of distinguishable quantum dots. The quantum dots we purchased would allow for up to 8 different colors in the spectral range from 540 to 680 nm, resulting in an increase in data density by a factor of 2. Further possibilities for enhancement include extending the spectral range or utilizing quantum dots with even narrower emission lines to introduce additional colors.

Another challenging way to achieve enhanced data densities involves increasing the number of states associated with each color, such as various intensity levels. This leads to an expansion of the possibilities per data point. While the number of colors alters the exponent in equation (1), which calculates the number of possibilities, the number of states per color changes the base. The general formula describing the bytes written per data point is $256^x = y^n$, where y represents the number of states or intensity levels per color. Clearly, the increase in data density is logarithmic and not linear in this scenario. Distinguishable emission intensity levels can be realized using different quantum dot concentrations in the inks [29]. This expected result is also shown in our own measurements (figure 4).

Drawing from our experience, it is worth noting that incorporating numerous intensity levels with many colors simultaneously can present challenges. As multiple quantum dots superimpose, it can become increasingly difficult to make fine distinctions in intensity. This complexity arises from factors like diminishing optical excitation and potential light scattering, which can pose challenges in achieving precise intensity differentiations. In our current assessment, employing up to 4 distinct intensity levels appears to be a feasible option, particularly when using a wide array of different colors.

To assess what a well optimized data density for this approach might look like, we combine all these ideas and calculate with a reasonable 112.89 $\mu\mathrm{m}^2$ spot size (2400 dpi resolution), 0.1 mm paper thickness, 32 colors, and 4 intensity levels. With these assumptions, the possible data density grows to 0.7 $\mathrm{MB/mm}^3$ or 4.07 GB per DIN A4 page. While not surpassing the latest LTO-9 standard, which offers up to 80 $\mathrm{MB/mm}^3$, the expected data density falls in a range where possible benefits over magnetic storage in terms of energy efficiency and longevity could become decisive. By exceeding current technological print resolutions limits and respecting the diffraction limit, i.e., spot sizes below 1 $\mu\mathrm{m}^2$, data capacity would exceed 459 GB for similar parameters and a DIN A4 page,

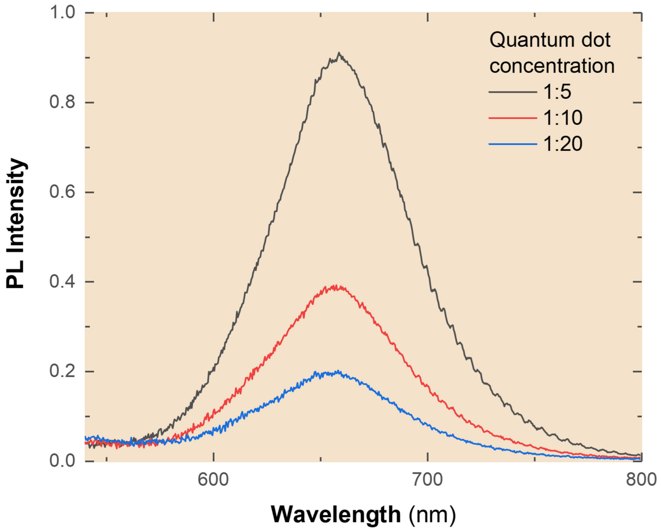


Figure 4. The graph shows distinguishable PL intensity levels as another potential increase in storage density. These distinct levels are attained by using varying quantum dot concentrations in the inks. Here, concentrations of 1:5, 1:10 and 1:20 were used for the measurement.

achieving the same data densities as LTO-9 with 80 MB/mm^3 (cf Supplement 1, 'Further explanations for the data density calculations').

While data density is the most straightforward use for the mixed states compared to binary on or off states, specialized code could potentially unlock additional benefits, particularly when employing a novel and specialized coding approach. These advantages may extend beyond increased data density and encompass improved error correction and enhanced security protocols.

It is important to note that the slower readout time of a PL setup, when compared to fast data storage options such as Flash, is not a significant concern in the context of long-term data archiving.

In the future, overcoming several key challenges will be essential to advance this method beyond its current proof of principle stage. Among these challenges, the following are likely to be the most crucial ones.

Dye system selection should prioritize optimization over availability. Environmentally friendly alternatives, such as quantum dots based on CuIn/ZnS, can replace CdSe/ZnS quantum dots. Commercially available printer solutions favor water-soluble quantum dots over oil-soluble ones due to their similar viscous properties to standard inks, ensuring printability. To enhance the variety of possible inks, printer technology needs to be optimized in this regard. While maximizing print resolution is challenging, it offers the highest benefits regarding data density. The maximum resolution achievable in inkjet printing primarily depends on factors like nozzle size, drop volume, the interaction of drops with the substrate, and ink spreading on the surface [33].

Another limiting factor will be the physical dimensions of quantum dots once such printing resolutions in the μm range are achieved, *e.g.*, by nano-scale nozzles, specialized printers, and fully refined inks. Even when the size of the quantum dots is one or two orders of magnitude below the printing resolution, the smallest possible data points will still expand in size as multiple colors and intensity levels are employed. This expansion, attributed to 3D stacking resulting from the increased number of quantum dots per data point required to represent the data accurately, will ultimately limit the print resolution for a given number of colors and intensity levels.

4. Conclusion

In conclusion, this study presents a compelling proof of principle for a data archiving method utilizing readily available resources such as an inkjet printer, water-soluble quantum dots and paper together with a PL setup for optical readout. Printable inks based on quantum dots, such as the CdSe/ZnS core–shell type, offer the advantage of mixability without the loss of information and thus allow multiple bits to be stored in a single data point. This has the potential to significantly increase the data density, as the number of data points required per byte depends on the number colors and intensity states within the system.

Our initial stability assessments offer promising prospects for long-term viability. The temperature stability tests revealed resilience within the range of −4 °C to +35 °C. Additionally, the continuous read-out examinations demonstrated no discernible decay in PL intensity over a ten-hour excitation period. This equates to a minimum of 360,000 read accesses, assuming exposure times of 100 ms each.

Future developments hold immense potential for advances in data density compared to conventional methods such as standard offset printing, microfiche, multi-colored QR codes or tape storage. Combined with further optimized, environmentally friendly inks, this approach has the potential to become an energy-efficient, long-term stable alternative to the current state of the art in data storage by tape.

5. Methods

5.1. Sample preparation

The commercial CdSe/ZnS quantum dots (NanoOptical Materials Inc.) are delivered in a borate buffer and have acid ligands to make them water solvable. When added to the ink mixture consisting of 75% demineralized water, 12.5% triethylene glycol monobutyl ether (Sigma Aldrich), and 12.5% ethylenglycol (Carl Roth), they yield working concentrations at ratios of 1:1 to 1:5 for ink solution to quantum dots. The data presented correspond to ratios of 1:1.5, 1:3, and 1:5 for quantum dots emitting at 540 nm, 580 nm and 665 nm, and 620 nm, respectively.

5.2. Optical readout

The data are read using one of two PL setups. One setup consists of a 405 nm laser excitation source, a spectrometer, and a Si based charge-coupled device (CCD) camera. The emission is detected in reflection geometry. For the long-term stability tests, we utilize a Ti:Sapphire laser excitation source emitting approximately 100 fs pulses at 810 nm with a 78 MHz repetition rate. A frequency-doubling crystal (β-barium-borate (BBO)) converts the laser fundamental to a wavelength of 405 nm for pulsed excitation. The emission is detected confocally in backscattering geometry using an aluminum-coated Schwarzschild objective and a spectrometer equipped with a Si based CCD camera. The samples are mounted on a 3D stage to facilitate movement and control the readout position.

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Data availability statement

All data that support the findings of this study are available from the authors on request (and any supplementary files).

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Disclosures

The authors declare no conflicts of interest.

Supplemental document

See Supplement 1 for supporting content.

Code availability

The modified software we used to control the printer and to encode or decode data can be found here:

https://github.com/MW55/escp2-quantum-dot-client
https://github.com/MW55/quantum-dot-codec

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