We demonstrate an all-optical strategy for realizing spherical three-dimensional (3D) super-resolution (~λ^3/22) spot arrays of pure longitudinal magnetization by exploiting a 4π optical microscopic setup with two high numerical aperture (NA) objective lenses, which focus and interfere two modulated vectorial beams. Multiple phase filters (MPFs) are designed via an analytical approach derived from the vectorial Debye diffraction theory to modulate the two circularly polarized beams. The system is tailored to constructively interfere the longitudinal magnetization components, while simultaneously destructively interfering the azimuthal ones. As a result, the magnetization field is not only purely longitudinal but also super-resolved in all three dimensions. Furthermore, the MPFs can be designed analytically to control the number and locations of the super-resolved magnetization spots to produce both uniform and nonuniform arrays in a 3D volume. Thus, an all-optical control of all the properties of light-induced magnetization spot arrays has been demonstrated for the first time. These results open up broad applications in magnetic-optical devices such as confocal and multifocal magnetic resonance microscopy, 3D ultrahigh-density magneto-optic memory, and light-induced magneto-lithography.

**Keywords:** inverse Faraday effect; longitudinal magnetization; magnetic-optical devices; magneto-optics; vectorial beams; vectorial Debye diffraction theory

Numerous endeavors have been devoted to the development of ingenious polarization configurations^5,10,14 in conjunction with amplitude/phase modulation^11,13,15 that can simultaneously improve the longitudinal magnetization purity and compress the dimension of the light-induced magnetization field via the inverse Faraday effect^16,17. To achieve a high spatial resolution for the light-induced magnetization, a high numerical aperture (NA) objective is required. However, the inherent depolarization effect along the axial direction^17 significantly degrades the purity of the longitudinal magnetization. Although a single spot of pure longitudinal magnetization can be achieved by using cylindrically polarized beams with a first-order vortex phase, a subwavelength spatial resolution can only be achieved along the lateral direction with a compromised resolution in the axial direction^10. As a result, the overall magnetization volume is not improved. Moreover, although some strategies, for example, a superoscillatory lens, can achieve impressive resolution improvement, further development is required to eliminate the longitudinal polarization states in the focal region to achieve a pure longitudinal magnetization spot in the future^18,19. Therefore, it is a great challenge to simultaneously realize high longitudinal magnetization purity and achieve 3D...
super-resolution along both the lateral and axial directions of the magnetization spot, which holds the key for most applications, in particular, for opto-magnetic data storage and lithography.

Here we propose and analytically demonstrate a 3D super-resolution (~\(\lambda/22\)) pure-longitudinal magnetization spot (MS) array with an individual spot size far below the diffraction limit (~\(\lambda/8\)) for the first time using a 4\(\pi\) high-NA focusing configuration\(^{20-22}\). Such a high resolution is achieved with ignorable side lobes (~10%), which is generally challenging for any super-resolution technique\(^{23,24}\). Under high resolution is achieved with ignorable side lobes (~10%), which is generally challenging for any super-resolution technique\(^{23,24}\).

By carefully manipulating the phase of the CPBG beams\(^{25}\), the illumination of tailored circularly polarized Bessel-Gaussian (CPBG) beams\(^{25}\) is achieved along both the lateral and axial directions of the MS array (Figure 1c).

### Optical scheme for achieving a pure-longitudinal MS

There are multiple ways to destructively interfere the focal electric fields of the two objectives is\(^{33}\), which can migrate freely\(^{26}\). The induced static susceptibility. Therefore, the magnetizations of the magnets in the MO material is the vector product of the electric field components being polarized along different directions, as is schematically shown in Figure 1b. One can see that the longitudinal magnetization (\(M_z\)) is induced by the electric field components polarizing parallel to the focal plane (\(E_p\) and \(E_z\)). Due to the strong electric field components along the optical axis (\(E_z\)) caused by the inherent depolarization effect of the high-NA objective, there exist significant transverse magnetization components (\(M_y\) and \(M_\phi\)). Therefore, the key to achieving pure \(M_z\) is to eliminate the \(E_z\) component by destructive interference. Since the overall area of the magnetization spot is determined by the size of the focal spot of light (Figure 1a), reducing the size of the focal spot represents a viable approach to achieving 3D super-resolved magnetization spots (Figure 1c).

The vectorial electric field distributions in the focal region can be calculated using the Debye diffraction theory (Supplementary Equations (1) and (2)). The interference of the focal fields of the two objectives is\(^{33}\)

\[
E_i(r, \varphi, z) = E_1(r, \varphi, z) + E_2(-r, \varphi, -z) \tag{2}
\]

where \(E_1\) and \(E_2\) stand for, respectively, the focal electric fields of the left and right objectives. The vectorial electric fields can be expressed as

\[
E_i(r, \varphi, z) = E_{i0} \left( \pm r, \varphi, \pm z \right) + E_{i1} \left( \pm r, \varphi, \pm z \right), n = 1, 2 \tag{3}
\]

where + is for the case \(n=1\) and - is for the case \(n=2\). The interference of the focal fields of the two objectives is

\[
E_{2,3,0} = A_{2,3,0} \left( r, \varphi, z \right) \exp \left\{ \pm \left( \Phi_{2,3,0} (r, \varphi, z) + \Phi_0 + k z \right) \right\} \tag{4}
\]

where \(A\) and \(\Phi\) are the amplitude and phase distributions, respectively, and \(k = 2\pi/\lambda\) is the wave vector of the incident light. The negative sign of \(r\) in \(E_2\) denotes the opposite orientations of the instantaneous polarizations of the two incident beams, which means \(\Phi(r, \varphi) - \Phi(-r, \varphi) = \pi\).

**Figure 1** Comparison of the magnetization spot produced by a single high-NA objective (a) and a 3D pure longitudinal super-resolved magnetization spot (c). (b) Schematics of the magnetization mechanism due to the inverse Faraday effect in the focal region of a high-NA objective and (d) in the focal region of a 3D super-resolved focal spot. The crosses indicate the elimination of the \(E_z\) field component, leading to pure longitudinal magnetization.
whereas the negative sign of \( z \) in \( \mathbf{E}_3 \) indicates their counter-propagation nature, which means that the \( kz \) terms cancel each other. \( \Phi_0 \) is the phase offset applied to the incident beams, and the phase difference between the two focal electric fields is \( \Delta \Phi = \Phi_{\text{f1}} - \Phi_{\text{f2}} \).

From Equations (2)–(4), one can see that it is possible to constructively or destructively interfere the electric field components from the two objectives to enhance or suppress a particular field component by manipulating the amplitude \( A \) and the phase difference \( \Delta \Phi \), which is determined by the polarization and phase of the incident beams. The \( \mathbf{E}_2 \) component can be completely removed if \( A_{\text{f1}} = A_{\text{f2}} \) and \( \Delta \Phi = 0 \). On the other hand, it can be enhanced if \( \Delta \Phi = \pi \) for incident light with any polarization state.

The previous strategy to achieve a pure longitudinal MS is to interfere two radially polarized vortex beams in the focal region of high-NA objective lenses in a 4\( \pi \) setup\(^{34} \). However, this strategy suffers from strong side lobes of up to 50\% as strong as the intensity of the central peak due to the constructive interference of the longitudinal electric field components, rendering this method less attractive for practical applications\(^{35} \). Additionally, only a single magnetization spot was achieved previously, significantly limiting the processing efficiency. In addition, to be able to achieve a strong central focal spot in the transverse focal plane, a vortex phase \( (\Phi(r,\phi) = \phi) \) is required in cylindrically polarized light beams\(^{10} \), which inevitably degrades their lateral resolutions. In comparison, circularly polarized beams have the advantage of a dominant transverse electric field in the center of the focal region to induce longitudinal magnetization. Therefore, circularly polarized incident beams have been selected in this study.

The conceptual design of the 4\( \pi \) microscopic system based setup is shown in Figure 2, where the incident beams are two counter-propagating CPBG beams with the same handedness of polarization, which illuminate on the spatial light modulators (SLMs)\(^{36,37} \) and are focused by the high-NA objective lenses (NA = 1). Without additional phase modulation from the SLMs \( (A_{\text{f1}} = A_{\text{f2}}, \text{ and } \Delta \Phi = 0) \), only \( M_z \) shows a non-zero value in the center of the focal region due to the removal of \( E_z \). In addition, the transverse electric field components \( (E_r \text{ and } E_\phi) \) constructively interfere in the vicinity of the focus along both the \( z \) and \( r \) axes, which is favorable to sharpening the MS. Therefore, a single 3D super-resolution pure-longitudinal MS can be created by the two CPBG beams in the focal region. By modulating the wavefronts of the CPBG beams with the designed MPFs encoded by the SLMs, which consist of the superposition of the desired multiple off-focus and off-axis phase patterns, arrays of identical 3D super-resolution pure-longitudinal MSs can be created in an isotropic MO medium placed in the focal region perpendicular to the optical axis.

**RESULTS AND DISCUSSION**

**Generation of 3D super-resolution MS via amplitude modulation**

The shape of the focal spot can be controlled by varying the effective NA of the focusing objective through adjusting the amplitude distribution of the incident beam. Here we design the amplitude of the incident beam based on the Bessel–Gaussian (BG) function\(^{25} \), which can be tuned by manipulating the order of the Bessel function \( m \) as

\[
l_0(\theta) = \text{exp} \left[ -\left( \frac{\sin \theta}{\sin \alpha} \right)^2 \right] J_m \left( \frac{2 \sin \theta}{\sin \alpha} \right)
\]

where \( l_0 \) is the amplitude distribution at the back aperture and \( \theta \) and \( \alpha \) are the convergence angle and the maximum, respectively. \( J_m \) is the \( m \)th Bessel function of the first kind. When \( m = 0 \), it is a Gaussian beam (Figure 3a). When \( m \) is a non-zero integer, a doughnut beam with a zero amplitude in the center is formed (Figure 3b and 3c). By increasing the value of \( m \), the radius of the central zero amplitude is increased, which results in a higher effective NA, so that only the focal region is illuminated.
lobes of cylindrically polarized vortex beams with previously reported side radial magnetization component (Figure 3m), and the FWHM in the axial direction (axial direction are enormously suppressed by the $4\pi$ addition, the full width at half maximum (FWHM) values along the leading to pure longitudinal magnetization (Figure 2f, 2i and 2l). In addition, due to the focal spot elongation in the axial direction, the side lobe of the MS is minimized.

The resulting magnetization distributions of BG beams of different orders ($m=0$, 1 and 2) are shown in Figure 3. Since the nonzero radial magnetization component ($M_r$) only appears in the out-of-focus region, it is not shown here. Compared to the case of one CPBG beam focused by a single high-NA objective (Supplementary Figs. S1 and S2), the azimuthal magnetization component ($M_\phi$) vanishes completely regardless of the beam order (Figure 3e, 3h and 3k), leading to pure longitudinal magnetization (Figure 2f, 2i and 2l). In addition, the full width at half maximum (FWHM) values along the axial direction are enormously suppressed by the $4\pi$ system (Figure 3m), and the FWHM in the axial direction ($W_z$), which in the transverse direction ($W_x$) and the strength of the side lobes are tunable by the order of the Bessel function (Figure 3n and 3o). It is found that a remarkable almost spherical (aspect ratio of 97%) and 3D super-resolution longitudinal magnetization spot arrays can be achieved at the optimized position when $m=2$ (Figure 3o). In the meantime, the maximal side lobe strength is only 9.6% of the central peak along the axial direction (black line in Figure 3o). Such exceptionally low side lobes significantly distinguish the current approach from that of the $4\pi$ strongly focused cylindrically polarized vortex beams with previously reported side lobes of >50%34. The calculated $W_x$ are 0.592\lambda, 0.470\lambda and 0.435\lambda for $m=0$, 1 and 2 (Figure 3m–3o), respectively, while the $W_z$ are 0.319\lambda, 0.395\lambda and 0.449\lambda, respectively, which are significantly smaller than the wavelength. It is obvious that $W_x$ and $W_z$ follow an opposite trend as predicted. To further verify these trends, we select the higher beam order of $m=4$ to portray the light-induced magnetization pattern (Supplementary Fig. S3). It is obvious that the value of $W_z$ has increased, leading to a much larger MS compared to the optimized case at $m=2$.

The overall magnetization distribution of the spot is plotted in Figure 4a, which shows pure longitudinal magnetization in the focal region. In comparison, the single-objective case shows a strong azimuthal magnetization in the focal region (Figure 4b), resulting in a 3D polarized magnetization (marked by the two green ellipses) due to the existence of the $E_z$ light field component. To further show the magnetization profile, corresponding 3D iso-surface plots of the magnetization are shown in Figure 4c and 4d, where the 3D super-resolved quasi-spherical magnetization field can be clearly identified. To evaluate the best achievable 3D magnetization resolution, the voxel size of the MS can be calculated as $4\pi/3 \times (W_x/2)^2 \times W_z/2$, where the MS is approximated as an ellipsoid. In the optimized case of $m=2$, a 3D minimum MS with a super-resolved voxel of $\sim 2^3/22$ (Figure 4c) can be achieved, far smaller than the diffraction limit of $2^3/8$. This 3D super-resolved quasi-spherical MS can find enormous potential applications in ultra-compact opto-magnetic devices36,39 and confocal and magnetic resonance microscopy4. For example, based on the best-achieved 3D resolution, the storage density in the MO material is estimated to be 45 Tbits cm$^{-2}$, which is 2.2 Tbits inch$^{-2}$ (at an 800-nm illumination wavelength) in terms of areal density, components with a large convergence angle are focused by the objectives. Therefore, two effects are evident in the focal region: (i) the lateral resolution of the MS becomes higher and (ii) the axial resolution of the MS becomes lower. In this way, by properly searching for the balance point, it is possible to achieve the MS with the best resolution along all the three directions. In addition, due to the fact that the wavelength of the light is not shown here. Compared to the case of one CPBG beam focused by a single high-NA objective (Supplementary Figs. S1 and S2), the azimuthal magnetization component ($M_\phi$) vanishes completely regardless of the beam order (Figure 3e, 3h and 3k), leading to pure longitudinal magnetization (Figure 2f, 2i and 2l). In addition, the full width at half maximum (FWHM) values along the axial direction are enormously suppressed by the $4\pi$ system (Figure 3m), and the FWHM in the axial direction ($W_z$), which in the transverse direction ($W_x$) and the strength of the side lobes are tunable by the order of the Bessel function (Figure 3n and 3o). It is found that a remarkable almost spherical (aspect ratio of 97%) and 3D super-resolution longitudinal magnetization spot arrays can be achieved at the optimized position when $m=2$ (Figure 3o). In the meantime, the maximal side lobe strength is only 9.6% of the central peak along the axial direction (black line in Figure 3o). Such exceptionally low side lobes significantly distinguish the current approach from that of the $4\pi$ strongly focused cylindrically polarized vortex beams with previously reported side lobes of >50%34. The calculated $W_x$ are 0.592\lambda, 0.470\lambda and 0.435\lambda for $m=0$, 1 and 2 (Figure 3m–3o), respectively, while the $W_z$ are 0.319\lambda, 0.395\lambda and 0.449\lambda, respectively, which are significantly smaller than the wavelength. It is obvious that $W_x$ and $W_z$ follow an opposite trend as predicted. To further verify these trends, we select the higher beam order of $m=4$ to portray the light-induced magnetization pattern (Supplementary Fig. S3). It is obvious that the value of $W_z$ has increased, leading to a much larger MS compared to the optimized case at $m=2$.

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Creation of 3D super-resolution longitudinal magnetization arrays via MPF

For real-life applications, improving the processing efficiency by using the 3D super-resolution longitudinal MSs requires creating MS arrays with arbitrarily controlled spot locations and numbers. The current approach to generate a multifocal array is to iteratively optimize the phase modulation at the back aperture of the objective to achieve the desired intensity distribution of arrays in the focal region. This iterative approach is not only time-consuming but also lacks physical insight. Therefore, here we propose and demonstrate an analytical method to generate the desired MS arrays by using MPFs (Figure 5a).

To calculate the phase distribution at the back aperture analytically, we convert the vectorial diffraction integral (Supplementary Equation (1)) into a Fourier transform of the weighted field (Supplementary Equation (8)). In this way, the electric field distribution in the focal region is the convolution of the Fourier transform of the induced magnetization of the 4π focusing system (Figure 5a) and the single-objective focusing system (Figure 5b). Creation of 3D super-resolution longitudinal MSs requires creating MS arrays: (i) the uniformity of the magnetization amplitude, which is important parameters are determined to evaluate the performance of the MS arrays; (ii) the uniformity of the magnetization amplitude, which is defined as 1 – D, where D represents the maximum difference among

\[
t(x, y) = \sum_{g=1}^{G} \sum_{i=1}^{S} \exp \left[-2\pi \left( \pm a_g x \pm b_i y \right) \right] \times \sum_{t=1}^{T} \exp(\pm ikc_t \cos \theta)
\]

where G, S, and T are integers, giving the entire numbers of spots along the x, y, and z directions, respectively; g, s, and t, in sequence, ranging from 1 to G, 1 to S and 1 to T; and \(a_g\), \(b_i\) and \(c_t\) represent the lateral and axial displacements, respectively. Therefore, the phase function is a superposition of the electric fields of each spot at the back aperture.

Here we take the MPFs with an equidistant interval of \(a_g = b_i = c_t = 8\lambda\) and fixed unit number of \(G = S = T = 4\) as an example. The resulting phase pattern (Figure 5a) arranges periodically in the entire pupil plane to shift the single MS to the appointed positions along both the transverse and axial directions. This behaves as a distinct and versatile phase-only 3D grating. Figure 5b represents the 3D iso-magnetization surface of the induced magnetization distribution at \(M = M_{\text{max}}/2\) when the optimized phase pattern (Figure 5a) is imposed on the incoming beams at the pupil plane. It is shown that a 4 × 4 × 4 MS array can be produced, and the spacing between adjacent layers is 8\(\lambda\). Cross-section maps of the 3D MS array in the xy and rz planes are shown in Figure 5c and 5d, respectively.

More importantly, the 3D resolution of any MS (white dashed lines in Figure 5c and 5d) is identical to the case without modulation from the MPFs (Figure 5e). Furthermore, the magnetization direction of each spot inside the array is completely longitudinal within the main lobe, which is identical to the case for the single MS (Figure 5f). The same principle can be used to create multifocal spot arrays with an arbitrary number of spots and configuration, which is analogous to the corresponding optical arrays. The cases of 2 × 2 × 2 and 3 × 3 × 3 MS arrays are demonstrated in Supplementary Figs. S4 and S5. Here we compare the absolute values of a single magnetization spot and a magnetization spot in an array. The energy conversion efficiency is also studied by summing up the overall energy in the array and comparing the value to that of a single spot. It has been found that more than 90% of the overall energy distributes uniformly to the spots in the array.

In addition to the spatial resolution and polarization, two other important parameters are defined to evaluate the performance of the MS arrays: (i) the uniformity of the magnetization amplitude, which is defined as 1 – D, where D represents the maximum difference among

\[
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all MSs in the normalized magnetization distribution and (ii) a spherically symmetrical distribution for each MS. The optimized uniformities are 100%, 98.4% and 96.4% for two-layer, three-layer and four-layer 3D super-resolution longitudinal MS arrays, respectively (Figure 5e, also see Supplementary Figs. S4e and S5e). Such a high uniformity can offer an excellent tolerance in multilayer high uniformity can offer an excellent tolerance in multilayer magnetically recording and maintain high accuracy in multifocal transversely super-resolved.

Figure 5 The $4 \times 4 \times 4$ 3D super-resolution longitudinal MS array induced by the optimized MPFs acting on the left-handed circularly polarized BG beams in the $4\pi$ focusing geometry. (a) Phase pattern of the MPFs with $G=S=7=4$ and $a_0=b_0=c_0=8\lambda$. (b) 3D iso-magnetization surface of the MS array with $M=M_{\text{max}}/2$. (c and d) Contour plots of the total magnetization field in the $xy$ and $rz$ planes. (e) Relevant magnetization cross-section profiles in c and d, respectively (black and red lines are along the $x$ and $z$ axes, respectively. The green line represents the $x$-axis cross-section of the magnetization pattern of a single focal spot without the use of MPFs). (f) Enlarged individual spot inside the MS array and its polarization distribution in the $rz$ plane.

Aperiodic and asymmetric multilayer MS arrays can also be induced by designing more tailored MPFs. Here we take an Archimedean spiral 3D super-resolution longitudinal MS array as an example. To this end, the phase function of the MPFs in Equation (4) should be replaced by the formula:

$$t(x,y) = \sum_{g=1}^{G} \exp[-i2\pi(a_g x + b_g y)] \exp(ikc_g \cos \theta)$$  (7)

with $a_g = t_g \cos(t_g)$, $b_g = t_g \sin(t_g)$, $t_g \in (0, 2\pi) \times g/G$, $c_g = (0, 4\pi) \times g/G$, and $G$ represents the total number of MSs in the curved array. Under such a circumstance, Figure 6 depicts the Archimedean spiral 3D super-resolution longitudinal MS array with $G=18$. It is observed from Figure 6a that the phase diagram of the renewed MPFs presents a spiral and helical pattern. Figure 6b shows the 3D iso-magnetization surface ($M=M_{\text{max}}/2$) of the MS array when the incident fields are modulated by the spiral phase revealed in Figure 6a. As expected, an MS array is created with 18 spots in an Archimedean spiral pattern, where each spot locates at its predetermined spatial position in a robust way. To visualize the demonstrated magnetization feature more intuitively, we produce cross-section maps of the 3D Archimedean spiral-shaped MS array in the $xy$ and $rz$ planes, respectively, as shown in Figure 6c and 6d. Further, it is revealed that the resolution of any MS (the blue rectangle regime in Figure 6f) is almost the same as the value without being subjected to the spiral MPFs (Figure 6e). We also find that the magnetization direction of the selected MS inside the array is purely longitudinal (Figure 6f). It should be noted that the aspect ratio of the MS in the Archimedean spiral array is $\sim 97\%$, and the relevant uniformity is 90% (Figure 6c and 6d).

From the experimental perspective, multiple phase filters can be encoded by a dynamic and phase-only spatial light modulator (SLM)
to realize three-dimensional super-resolution longitudinal magnetization spot arrays. The current commercially available SLMs (for example, Holoeye Pluto) are able to produce 256 levels of phase modulation in the range of 0–2π. The errors in the phase modulation can be minimized by phase calibration to tune the gamma curve. In this way, we expect less than 5% error in the phase modulation. It should be noted that the phase-only SLM must have at least 8 modulation levels to achieve a highly uniform (90%) spot array \(35,37\). To consider the effect of errors in the phase modulation, we implement 10% randomness in the phase modulation in our simulation, resulting in only minor decreases (~3%) in the efficiency and uniformity of the array.

CONCLUSIONS
In summary, we have theoretically demonstrated light-induced magnetization by tightly focusing two modulated circularly polarized BG beams in the \(4\pi\) configuration. Due to the destructive interference of the axial electric field component, a pure longitudinal MS is achieved. Through amplitude modulation based on the BG function, a quasi-spherical 3D super-resolution (~\(\lambda/22\)) MS with ignorable side lobes has been demonstrated for the first time. More significantly, arbitrary MS arrays, for example, Archimedean spiral 3D super-resolution longitudinal MS arrays, can be generated by MPFs, designed analytically according to the location and number of the MSs. The generated MS arrays exhibit excellent uniformity and almost spherical symmetry, making them an appealing platform for developing light-induced magneto-lithography devices. Most importantly, using the proposed method, all three properties of a MS array, namely, the magnetization direction, the 3D resolution and the number and arrangement of the MSs, can be simultaneously and independently controlled, which makes this method extremely flexible. One can arbitrarily adjust any of the properties depending on the application requirements without compromising the others. The demonstrated light-induced magnetization might also shed light on the spin–spin interaction, which ignites a new research field in magnonics\(^{39}\). From an experimental perspective, the possibility of achieving powerful MS arrays using circularly polarized beams with robust MPFs is particularly fascinating due to the application of dynamic SLMs\(^{34,35}\). The MS arrays can be applied broadly in multiple atom trapping and transport, confocal and multifocal magnetic resonance imaging, and multilayer magneto-optical recording and storage. The uniform longitudinal MS arrays can also be applied to light-induced magneto-lithography devices with high efficiency\(^{51,52}\).
CONFLICT OF INTEREST

The authors declare no conflict of interest.

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