

## Review Article

# Applications of X-Ray Holography

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X-ray holography is widely used in material, biology, and industry fields due to its potential to measure the microstructure and dynamic change of objects. In this review, the principle of X-ray holography and the development of this technology in different application fields are systematically summarized and discussed. Through analyzing the advancement of X-ray sources and recording medium, the research and development direction of X-ray holography are prospected and the overview on current strategies of novel X-ray holography is presented. It is proved that X-ray holography, as a powerful nondestructive measurement method, can be applied to a wide range of objects.

## 1. Introduction

In 1948, Gabor [1] formulated the idea of the holographic method, obtaining the first hologram and reconstructed image, for which he won the Nobel Prize in Physics. X-rays were discovered by Roentgen [2, 3] in 1895. Baez [4] combined the idea of holography with X-rays to form a new method. X-ray holography was thereafter paid increasing attention by researchers.

The development of X-ray holography was depended on the highly-bright X-ray sources, the X-ray components, and the recording medium. The idea of holography was first proposed to improve the resolution of the microscope. The first holographic experiment was performed with a visible light source. But at that time, there was no coherent light source to demonstrate all the functions of holography. To find a suitable coherent light source had become the focus of research for many years. Because of the shorter wavelength, X-rays provide a way to achieve higher resolution than visible light. The theoretical foundations of high-resolution X-ray holography were laid by Leith et al. [5–9]. By improving the resolution of the medium and the incident light source [10–15], the researchers succeeded in obtaining X-ray holograms. Especially, Tegze and Faigel [16, 17] analyzed the possibility of atomic resolution in X-ray holography and got holograms with atomic resolution, which brought X-ray

holography into a new stage of development. With the improvement of coherent X-ray sources (third-generation synchrotron radiation sources, free electron lasers, and laboratory sources) [18–26] and detector [27–29], studies have shown that the spatial resolution can be greatly improved. To solve imaging artifacts translated by high spatial frequencies, Geilhufe et al. [30] introduced three approaches. They proved that image detail smaller than the source size of the reference beam could be restored to the diffraction limit of the hologram. With the development of these technologies, the resolution of X-ray imaging has reached a higher level.

With the characteristics of short wavelength in penetrating power and high energy, X-rays are combined with holography. As a method of direct three-dimensional imaging, X-ray holography has a great deal of advantages in studying the crystal structure of objects. X-ray holography does not only have the function of optical holography but also some special properties, which makes it advantageous in the three-dimensional imaging and dynamic observation of the internal microstructure of the objects. It can be used to obtain three-dimensional images with atomic resolution, which is widely used in the research on crystals, crystal films, impurities, and lattice distortions [31]. X-rays in the ‘water window’ have a contrastingly enhancing mechanism for biological samples. The combined application of holography

and X-rays can be used to measure the three-dimensional structure of the biological molecule, the captured images of live cells, and the processing of chemical reactions [32, 33]. Some special functional materials, e.g., nanomaterial and ferroelectric material, are widely used in the production process of significant components. Although it has been a long time since the discovery of these materials, the understanding of their detailed structure and microscopic origin is still unclear. The microstructure of the film's epitaxial growth and the electrode affects the performance of the material and, ultimately, the performance of the components. X-ray holography provides a powerful tool for these studies. This paper will (1) review works of literature that address the applications of X-ray holography in various fields, (2) describe the advantages of X-ray holography, and (3) discuss the prospect of technological development and application.

## 2. Applications

**2.1. Material Field.** Material Science is the research, development, production, and application of metallic materials, inorganic nonmetal materials, polymer materials, and composites. At the atomic level, the structure and composition of elements, the spatial sequence of atoms or molecules, and the atomic motion pattern are studied in order to correctly understand and apply materials and develop new materials. Many approaches are used to measure the three-dimensional structure of element atoms and their surroundings in materials. X-ray fluorescence holography (XFH) is one of the approaches [34–37]. The use of short-pulse X-ray sources in combination with high-resolution detectors in X-ray holography can provide clear atomic images. Besides, the deformation of the crystal cannot be directly obtained by the traditional X-ray diffraction (XD) technique, while XFH can be used to solve this problem [31, 38]. Examples of applying X-ray holography in investigations of crystalline structures are discussed in the following section.

Atoms in crystals' internal structure are regularly arranged in three-dimensional space. In fact, many materials are made of a mixture of two or more elements, which affect the internal structure of the crystal elements making the sample become 'imperfect'. The first implementation of direct XFH measured strontium fluorescence (Figure 1) from a strontium titanate single crystal [17]. In addition, to measure the structure of the elementary atoms in the crystal, XFH is also used to measure the structure around the atoms in the material, which is confirmed by the research of Tegze et al. [39], who pointed out that X-ray holography could measure the three-dimensional image of the local environment of selected atoms with atomic resolution and distinguish atoms in different magnetic states. In 2006, Hosokawa et al. [40] obtained a three-dimensional atomic image around Ge atoms in the  $\text{Ge}_2\text{Sb}_2\text{Te}_5$  film with XFH. The analysis of the image showed that the  $\text{Ge}_2\text{Sb}_2\text{Te}_5$  single crystal film had no hexagonal point symmetry around Ge atoms (Figure 2). In 2009, Happo et al. [41] obtained a three-dimensional atomic image around Mn atoms in a

$\text{Cd}_{0.6}\text{Mn}_{0.4}\text{Te}$  single crystal of dilute magnetic semiconductor with XFH. In the same year, Hosokawa et al. [42] measured the three-dimensional image around the Zn atom in the  $\text{Zn}_{0.4}\text{Mn}_{0.6}\text{Te}$  crystal with XFH. In 2011, Hosokawa et al. [43] used XFH to observe the three-dimensional image of In atoms, Tl atoms in the single-crystal  $\text{TlInSe}_2$  thermoelectric material at room temperature. In 2018, Nishioka et al. [44] used XFH to measure the three-dimensional local structure of atoms in the local plane around Zn in  $\text{Mg}_{75}\text{Zn}_{10}\text{Y}_{15}$  alloy. In 2017, Stellhorn et al. [45] used XFH to measure the three-dimensional structure around the Fe and Ni atoms in  $\text{Fe}_{65}\text{Ni}_{35}$  Invar alloy. In 2019, Kimura et al. [46] used XFH to measure the three-dimensional structure around the Fe atoms in  $\text{Pb}(\text{Fe}_{1/2}\text{Nb}_{1/2})\text{O}_3$  (PFN) multiferroic material at different temperatures to study the relationship between atomic images and temperature. In 2020, Ang et al. [47] measured the original and irradiated  $\kappa$ -(BEDT-TTF) $_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$  crystals with XFH. The measurement results are used to study the effect of radiation on the local structure around Cu in the anion layer. The above experiments have proved the successful application of X-ray holography in three-dimensional imaging of atomic structures. Earlier, it was used to study the microstructure and composition of materials. In recent years, it is mainly used to study the change in materials' internal structure under different conditions, which can better evaluate the performance of materials.

Material preparation technology and conditions or special application requirements make crystals contain impurities, and the atomic structure around the impurities will change, resulting in local lattice distortion. XFH can achieve high-resolution imaging of the three-dimensional distribution of atomic structures, and it is very sensitive to the positional fluctuations of atoms from the ideal position. It can also obtain information about local lattice distortion in the process of generating three-dimensional atomic images. Therefore, XFH is very suitable for the analysis of the atomic structure around the impure atoms in the crystal. The research of Hayashi et al. [48] also proved this, for he pointed out that XFH can be used to observe three-dimensional atomic images around specific elements within a radius of nm order and described the local lattice distortions around specific elements. In 2006, Kopecký et al. [49] used X-ray diffuse scattering holography to determine the position of Mn atoms in GaMnAs doped with Mn and obtained new information about the local atomic structure (Figure 3). In 2011, Hayashi et al. [50] used XFH to measure the hologram of  $\text{ZnSnAs}_2:\text{Mn}$  thin film and observed the local structure around Mn. In 2018, Kimura et al. [51] used XFH to obtain the in-plane atomic image near In on an In-doped  $\text{Bi}_2\text{Se}_3$  topological insulator. He found the local lattice distortion and discussed the reason. The measurement results obtained by combining XFH with other experiments or algorithms sometimes are better than using XFH only. In 2017, Hosokawa et al. [52] pointed out the impurity sites of Mn atoms, the distance between Mn-Te atoms, the local lattice distortion and the positional fluctuations around Mn atoms that can only be measured with a combination of the XFH and X-ray absorption fine-structure (XAFS) measurements

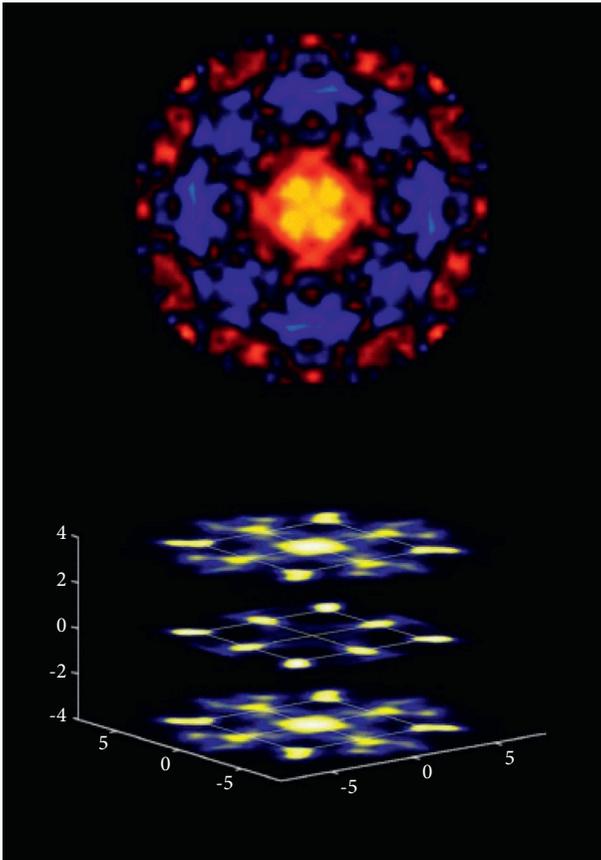


FIGURE 1: Three-dimensional holographic image of  $\text{SrTiO}_3$ , showing only the Sr atoms [17].

on a single crystal of a  $\text{Bi}_2\text{Te}_3\text{Mn}_{0.1}$  topological insulator. In 2020, Stellhorn et al. [53] used XFH with a sparse modeling algorithm to analyze the local structure around the doped atoms in  $\text{Nd}:\text{LaF}_3$  single-crystal scintillator. He determined that the atoms in the crystal were replaced by impurities and caused very small lattice distortions.

Although XFH has great advantages in measuring the internal structure of materials, it still has shortcomings, and that is why Multi-energy X-ray holography (MEXH) (Figure 4) is used as a new approach to holography. In 1995, Gog et al. [55] used MEXH to image the local atomic environment of Fe atoms in a hematite crystal, which showed that image aberrations caused by single energy were effectively suppressed. Gog et al. [56] pointed out that the accuracy of using MEXH to determine the position of atoms and the degree of the suppression of repetitive image twinings will be affected by the total amount of imaging under single energy measured within the valid time, the integration and processing approaches in different energy measurements. In 1998, Novikov et al. [54] used multiple energy recording X-ray holograms to image three-dimensional images of  $\text{Cu}_2\text{O}$  crystal structure for the first time and clearly displayed the first Cu–Cu coordination shells. In 2000, Adams et al. [57] used MEXH to image a reciprocal holographic experiment on  $\text{Cu}_3\text{Au}$  single crystal and obtained a three-dimensional image of the  $\text{Cu}_3\text{Au}$  atomic structure. The reconstruction of the measured hologram showed the

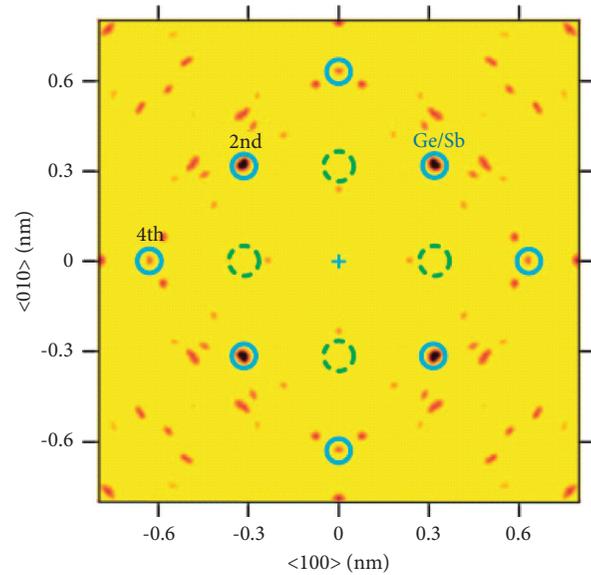


FIGURE 2: (Color) Atomic images around the central Ge atom [40].

positions of the nearest and the nearest neighbors of Cu atoms, which are identical to the actual ones. In 2001, Hayashi et al. [58] used MEXH to image the local atomic environment of Zn atoms doped in GaAs crystal. Studies have shown that MEXH can be successfully used in the measurement of atomic structure, the surrounding structure of crystals, and the atomic structure of impurities in crystals. Besides, multiple scattering [59] and accidental image cancellations [60] can be suppressed effectively by MEXH, which has an advantage in the improvement of image quality.

The extremely strong and ultrafast X-ray pulses from free-electron lasers make it possible to study the fundamental aspects of complex transient phenomena in materials. In 2007, based on femtosecond time-delay X-ray holography, Chapman et al. [61] monitored the dynamics of the polystyrene sphere and observed the explosion after the initial pulse. They pointed out that the three-dimensional dynamics of materials can be studied on the timescale of atomic motion (Figure 5) with the help of ultrafast X-ray sources. In 2012, Wang et al. [62] used resonant X-ray holography to perform femtosecond single-shot imaging of nanoscale ferromagnetic spin sequences and proved the feasibility of highly efficient single-shot imaging of spin-resolved electronic structures. In 2021, Keskinbora et al. [63] pointed out that the single reconstruction capability of Structured Illumination X-ray Holography (StIXH) is expected to be used in measuring the unreproducible dynamics in ultrahigh time resolution with highly repeating rate X-ray source. The scope of microscopic research can be extended to more samples, such as biological cells or electromagnetic devices.

It is worth mentioning that Complex X-ray Holography (CXH) [64] was proposed to record the phase of scattered X-rays when using resonant X-ray scattering. In 2004, Takahashi et al. [65] succeeded in using the CXH to reconstruct the isolated As atomic images in GaAs crystal. The

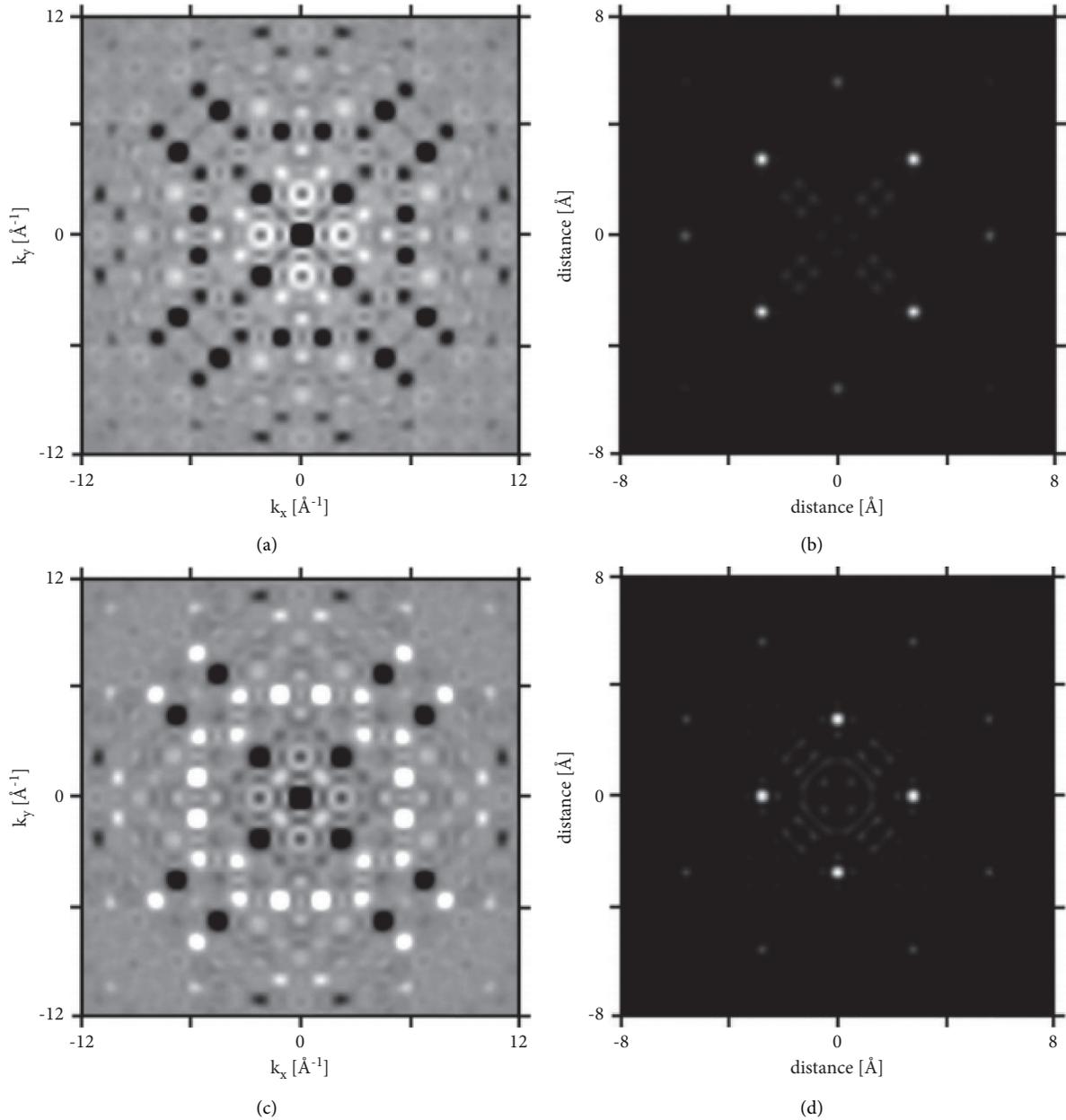


FIGURE 3: Simulated holograms of clusters of atoms corresponding to the volume of eight unit cells of single-crystal GaAs doped by Mn in (a) substitutional and (c) interstitial positions. (b, d) Real-space images of planes parallel to the (001) crystallographic plane containing reference Mn atoms reconstructed from holograms (a) and (c), respectively [49].

results indicated that the accuracy of the reproduced images can be improved with the CXH. Obviously, its application in measuring material structure is not as wide as that of XFH.

It is shown that X-ray holography is successfully applied in the three-dimensional measurement of atomic structure and the structure around atoms with a high resolution. In particular, XFH has shown significant advantages in three-dimensional imaging and has been widely used. The most applied cases of XFH are to measure changes in the local atomic structure caused by impurities in different materials, including single crystal, amorphous phase change dielectric film, ferromagnetic semiconductor film, and dilute magnetic semiconductor single crystal, thermoelectric material,

multiferroic material, topological insulator, etc. MEXH can simultaneously record a variety of different energy and summarize the data of several incident energies, which can thereby effectively suppress the aberration of the holographic image, the twin images, interobject multiple scattering, self-interference and to improve the quality of reconstructed images. XFH can measure crystal deformation, which cannot be done by traditional XD technology. The advantage in measuring the lattice distortion of atoms and impurity atoms in the crystal lattice, observing the degree of distortion, and analyzing the causes of the distortion is conducive to a better understanding of the properties of materials. With the development of a new

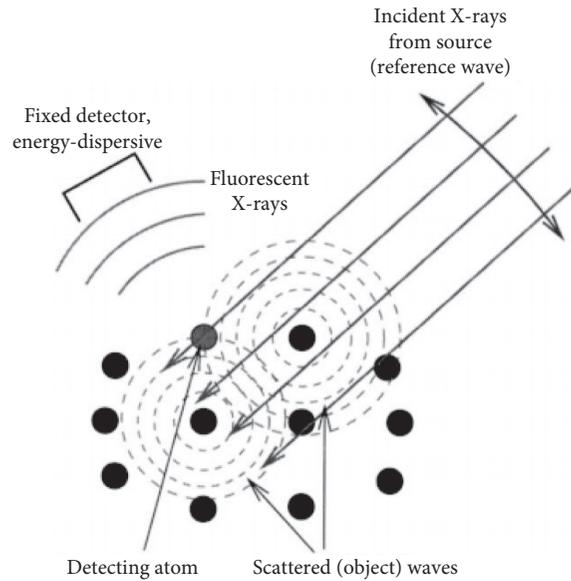


FIGURE 4: Formation of holograms in MEXH approaches [54].

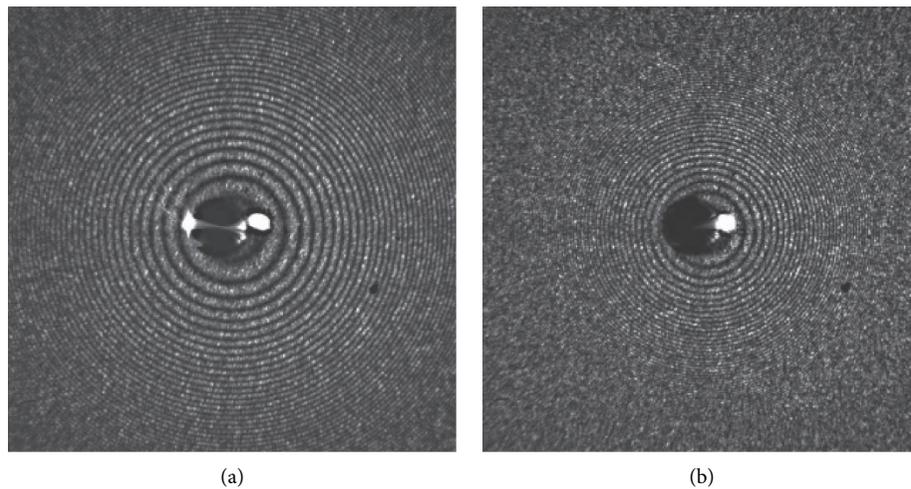


FIGURE 5: Time-delay X-ray holograms of 140 nm-diameter polystyrene spheres. The time delays were  $348 \pm 1$  fs (a) and  $733 \pm 2$  fs (b). The pulses were 32 nm wavelength and 25 fs duration with intensities  $(0.5 \pm 0.2) \times 10^{14} \text{ W cm}^{-2}$ . The intensities of the holograms are shown on a linear greyscale, to a half-width of  $4.5 \mu\text{m}^{-1}$ . Deriving the time delays and the change in the optical path through the exploding particles from the fringe pattern, we have found that the particle sizes are determined from the envelope of the intensity [61].

generation of highly coherent X-ray sources, the X-ray holography that uses the generation of synchrotron radiation sources or free-electron X-ray lasers is used to obtain the nanoresolution imaging on the timescale of atomic motions. The development of X-ray holography is closely related to the development of X-ray sources and detectors. It is believed that with the development of science and technology, X-ray holography will see greater improvements in imaging quality, measurement time, and application range.

**2.2. Biological Field.** Biological imaging is one way to obtain microstructure images of biological cells and tissues and further to understand various physiological processes of

biological cells through image analysis. In 1980, Kirz and Sayre [66] proposed the use of X-ray microscopy to achieve three-dimensional images of microscopic organisms. In 1987, Howells et al. [67] used X-ray technology to measure pancreatic zymogen granules. Limited by the poor coherence of the X-ray source and the low resolution of the detector, the resolution obtained in the experiment has not exceeded the resolution of the optical microscope.

With the development of modern biology and genetics, determining the structural information of the measured objects is becoming the key to solving the problem. There is an urgent need for tools to study the structure and function of biological macromolecules. The advantages of X-ray holography in life-science research are as follows: (1) high

resolution, (2) direct measurement and processing on live samples with its ability to distinguish atoms, (3) dynamic observation of the processing. Biological cells can be analyzed by X-rays with the latest development in X-ray focusing, diffraction data analysis, and coherent imaging reconstruction algorithms [68]. The development of new technology makes it possible to study biological cells in different preparation states (freeze-drying, low-temperature vitrification, chemical fixation, and living cells).

Ultrafast X-ray imaging can achieve high-resolution images, which traditional imaging techniques cannot obtain when measuring living samples [69]. In 1991, Nugent et al. [70] pointed out that coherent soft X-ray holography is a technology that can achieve high-resolution imaging. Because the technology and components required for the experiments had not been developed to an advanced enough state, the advantages of coherent soft X-ray holography were not displayed at that time. Based on the need to produce three-dimensional images of comparably bigger life-science samples with a resolution of about 10 nm, Howells et al. [71] proposed a new form of Fourier-transform X-ray holography, which has higher resolution and can determine the phase and amplitude of the diffracted wave field. In 2000, Murray et al. [72] used X-ray holography to reconstruct the crystal structure of the enzyme-product complex of the hammerhead ribozyme and observed the interaction between residues and functional groups. In 2011, Gorniak et al. [33] demonstrated the first digital X-ray hologram of a biological sample recorded in the water window (Figure 6), which greatly promoted the development of imaging hydrated biological material with photons. In 2016, Tomita et al. [73] observed X-ray fluorescence holograms from protein crystals for the first time with minimal radiation damage and started a promising approach for investigating the metal active-sites in biomacromolecules. In the same year, Nicolas et al. [74] used a combination of scanning small-angle X-ray Scattering (SAXS) and full-field holography to test actomyosin in freeze-dried neonatal rat cardiomyocytes. It is shown that X-ray holography is ideal in completing missing scattered data at low momentum transfer by the structure factor, extending the covering range of spatial frequencies by two orders of magnitude. In 2017, Tomita et al. [75] used XFH to measure hemoglobin (Hb) and obtained the atomic image of the hemoglobin environment in a single subunit of Hb. Tomography is a well-established X-ray technique for imaging a medical object in three dimensions by generating images of certain layers [76]. In 2020, Kuan et al. [77] used X-ray holographic nanotomography (XNH) to reconstruct the main dendrites and axon branches of neurons of *Drosophila melanogaster* and mice with a resolution of sub-100 nm.

It is shown that X-ray holography has been successfully applied in the imaging of biomolecular structures and achieved high-resolution images of the crystal structure of samples, e.g., enzyme-product complexes, protein crystals, hemoglobin, hydrated biological samples, and nerve tissue. The correct understanding of biological structure is conducive to understanding its change process, being helpful to further understand its impact on the realization of specific

functions. The nervous system is mainly composed of nerve tissues, which can regulate and control the physiological activities of the human body. The reconstruction of the main part of neurons is beneficial to the research of neural circuits.

X-ray holography can not only be used in the measurement of biological cells but also in the inspection and treatment of diseases. The disadvantage of cancer radiotherapy is that the normal tissues adjacent to the tumor cells will be affected during the treatment. In 1999, Madjidi-Zolbin and Jafari [78] pointed out that X-ray holography can accurately focus radiation on tumor cells (Figure 7) and reduce the damage to the healthy tissues surrounding the tumor cells during treatments. In 2008, Nesterets et al. [79] used X-ray phase-contrast in-line holography with ultrafast laser-based X-ray source to study the distribution of proliferating cell density with stationary cell cores and shells. The contrasting reconstructed images by phase was enhanced. The result proved the feasibility of this approach for microimaging of small soft tissue avascular tumors. In 2020, Dahlin et al. [80] used X-ray phase-contrast holographic nanotomography to perform three-dimensional scanning of neural tissues on the subcellular scale and achieved high-resolution neural three-dimensional imaging. The understanding of peripheral nerve structure helps to explain the pathology of neuropathy and to analyze the cause of illness. At the same time, the success of the experiment proved the effectiveness of this approach in peripheral nerve biopsies. In 2020, Samber et al. [81] used synchrotron radiation-based nanoscale X-ray fluorescence and X-ray online holography to study the distribution of nanoscale iron in a single fibroblast from Friedreich's ataxia (FRDA) patients. In this research, various micrometre-sized iron-rich organelles were revealed for the first time, which provided an innovative way to understand FRDA.

Besides, XFH can be used to study the local atomic structure of inorganic crystals and soft material samples, thus providing atomic resolution structural information and distinguishing experiments of different valence states of the same element in the sample [82]. Tegze et al. [83] pointed out that X-ray holography may be able to measure single molecules and viruses, and this will require ultrafast X-ray sources.

Studies have proved that X-ray holography has successfully achieved the three-dimensional measurement of biological microstructures with high resolution. In addition to the need to improve the resolution of images, the development of biological imaging technology also needs to enhance the real-time and continuous research of imaging. The goal of imaging is fully revealing the biological function by achieving continuous tracking of a single biologically functional molecule and recording its physiological process in detail. An important prerequisite for X-ray holography to be widely used in disease diagnosis is noninvasive. It can accurately focus on the lesion to achieve fixed-point detection and treatment, reducing damage to other parts. The application of bioimaging technology in clinical medical diagnosis has been paid more and more attention. In the past few decades, in macromolecular crystallography, the overall damage caused by X-ray irradiation has always been a

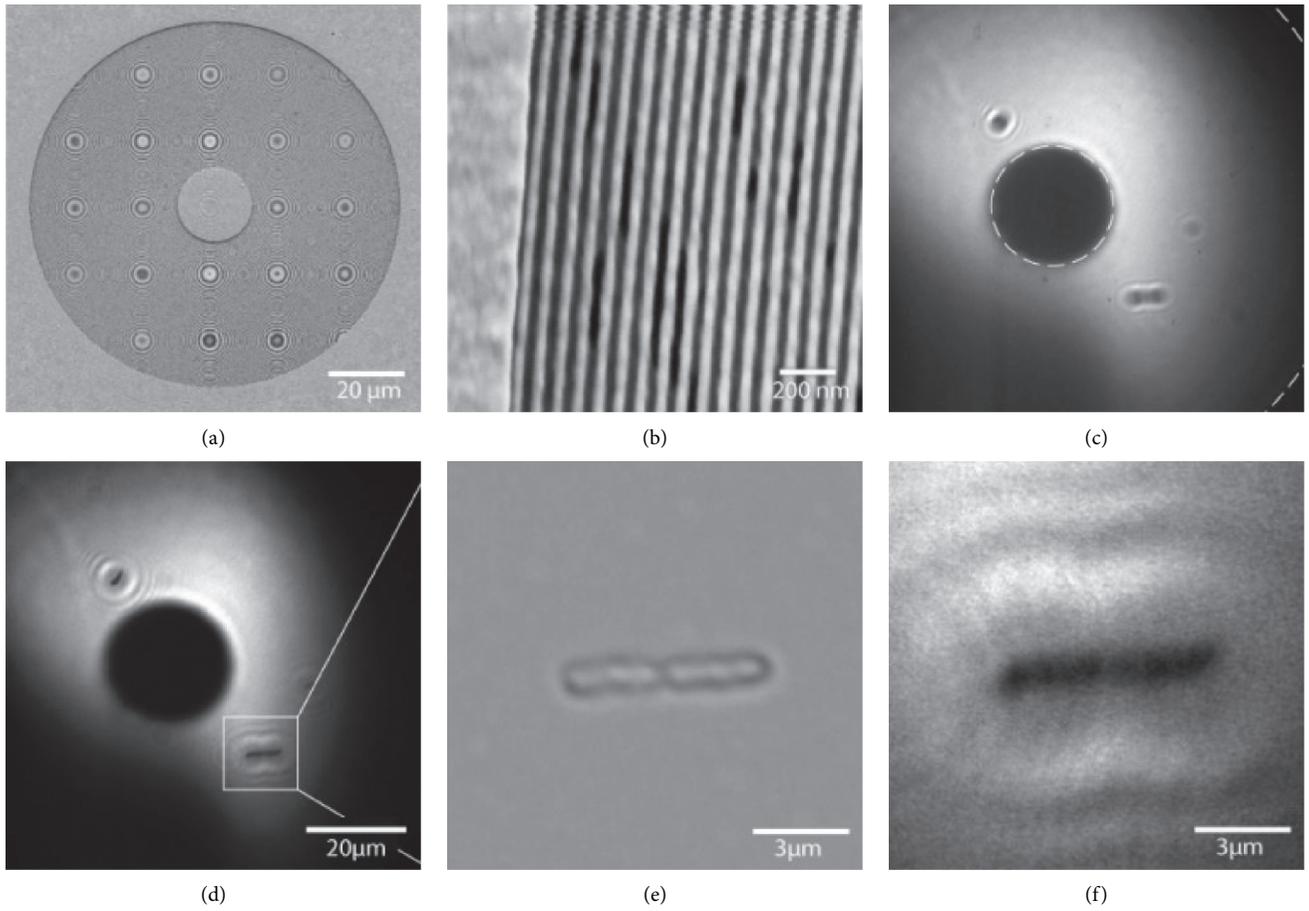


FIGURE 6: Imaging the marine bacteria *Cobetia marina* in the water window at  $\lambda_3 = 2.68$  nm using digital in-line holography. (a) SEM image of the used zone plate with a clearly visible central beam stop. (b) Close-up of the zone plate's outermost zones. (c) X-ray hologram with *Cobetia marina* in the lower right corner. (d) Reconstruction of hologram (c). (e) Sample *Cobetia marina* under an optical microscope in bright field illumination (100x, NA = 0.9). (f) Magnified ROI of the reconstructed image as indicated by the rectangle in (d) [33].

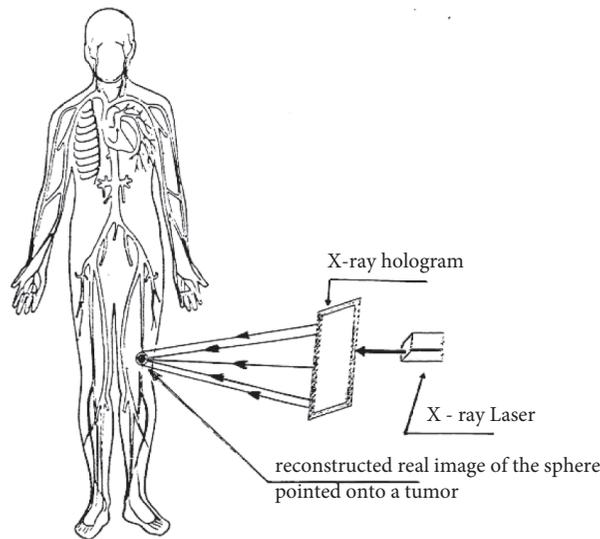


FIGURE 7: A schematic illustration of cancer treatment by X-ray holography [78].

problem faced by scholars, but the best solution currently proposed is cryogenic cooling. The highly coherent light source enables X-ray holography to achieve dynamic measurement in the femtosecond range. With the improvement of new technologies and approaches, the measurement time can be shortened and it is practical to complete the measurement before the sample becomes invalid. The new generation of free electron laser type X-ray source combined holography may be able to measure the three-dimensional structure of single molecules, viruses, and other tiny systems that cannot be crystallized, which will greatly promote the development of this process.

**2.3. Industrial Field.** With the rapid development of industry, the miniaturization, integration, and intelligence of components have made functional materials with special properties widely used, which demands higher requirements for material performance.

The research on nanomaterials is indispensable with special physical properties, chemical properties, and application value. The experiments show the successful measurement of nanostructured materials, which can help to understand the type, quantity, and internal structure of substances and to obtain products with special functional requirements. One of the significant applications of X-ray holography is to measure nanostructures. There are several ways that can be used to measure tiny structures with high resolution, e.g., high-brightness synchrotron radiation accelerator, optimized testing technology, and improved experimental equipment combined with X-ray holography. In 2005, Hellwig et al. [84] used soft X-ray spectroscopy holography to measure the magnetic nanostructures, whose reconstructed image had a spatial resolution below 50 nm. In 2007, Scherz et al. [85] demonstrated phase imaging of magnetic nanostructures by resonant soft X-ray holography. It is shown that the use of quantitative and spectral phase approaches allows high-contrast imaging of nanoscale electronic and magnetic levels while increasing the depth of detection and reducing the radiation dose by an order of magnitude. In 2009, Streit-Nierobisch et al. [86] proposed that resonant soft X-ray holography could be used to measure the magnetic domain structure of different Co/Pt multilayer films (Figure 8). By studying focused ion beam (FIB)-induced anisotropic modulation and vertical domain structure in nanostructure samples, the size and configuration of domains can be controlled by adjusting the amount of ionic agent applied in a single point. In 2010, Chamard et al. [87] demonstrated the three-dimensional imaging of SiGe nanocrystal with Bragg Fourier transform holography and obtained the shape, the internal views of the density, and displacement field. The simplicity and stable inversion have opened up a new way of in-situ study of the inhomogeneous strain field in the nanocrystal. In 2011, Kim et al. [88] obtained the reconstructed image of nanomaterials with the resolution of 87 nm with single-shot Fourier-transform X-ray holography and X-ray laser. In 2020, Zhang et al. [89] reconstructed the evolved nanostructures in ultrathin films with X-ray waveguide fluorescence holography. The analysis

results showed that the controllable synthesis of nanostructure ultrathin films may become a reality.

It is obvious that researchers have increased their research on nanomaterials and have gradually deepened it in recent years due to the development of a new generation of synchrotron radiation sources and detectors. The approaches of measurement with efficient image processing programs have improved experimental devices and optimized reconstruction algorithm. With other technologies, they have greatly promoted the precise measurement of nanostructures.

Epitaxial growth refers to the growth of a single crystal layer on a single crystal substrate under certain requirements. It has the same crystal orientation as the substrate. Many electronic devices are fabricated with the technology of epitaxial growth on single-crystal substrates. Studies have shown that the local atomic structure of thin film samples can be obtained by XFH. In 2004, Sekioka et al. [90] found that whether the material accepts radiation has a significant difference in the local atomic structure hologram of the  $\text{EuBa}_2\text{Cu}_3\text{O}_{7-x}$  (EBCO) superconductor films measured by XFH. In 2014, Happo et al. [91] used XFH to observe the local atomic structure around Ge and Mn atoms in  $\text{Ge}_{0.6}\text{Mn}_{0.4}\text{Te}$  thin films and found the local lattice distortions around Ge atoms (Figure 9). In 2015, Hayashi et al. [92] used XFH to evaluate the crystal structure of  $\text{ZnSnAs}_2$  thin films.

The ferroelectric material is a kind of functional material with ferroelectric effects as ferroelectricity and piezoelectricity. It has very significant applications in microelectronics, photovoltaics, and sensors to make ferroelectric memories, pyroelectric infrared detectors, spatial light modulators, optical waveguides, and so on. Scott [93] researched ferroelectric materials and found that their applications are still in the development stage. Hayashi et al. [94] used XFH to analyze the local structure around Ti in the ferroelectric material  $\text{Pb}(\text{Zr}_{0.7}\text{Ti}_{0.3})\text{O}_3$  crystal (Figure 10). Essential for the design of new and improved ferroelectric materials, the result can be used to understand the local structure of lead zirconate titanate (PZT) and reveal the microscopic origin of high dielectric and piezoelectric response.

The demand and requirements for energy to be green and sustainable are becoming a trend, in which lithium batteries have become a mainstream energy source. Widely used as a high-efficient and environmentally friendly high-temperature fuel cell, the performance of battery electrodes is affected by their microstructure and electrochemical performance. X-ray nanoholography can also be used to study the microstructure of the electrode of solid oxide fuel cells. Based on the requirements of brightness and hard X-ray, this technology is nondestructive. To obtain three-dimensional images requires the use of a third-generation synchrotron. Villanova et al. [95] performed quantitative phase-contrast X-ray nanoholographic imaging on a large number of solid oxide fuel cells (SOFC) anodes composed of Ni/YSZ cermet. Then Grindler et al. [96–99] studied the microstructure of electrodes based on different objectives. For comparison between different electrodes and electrode

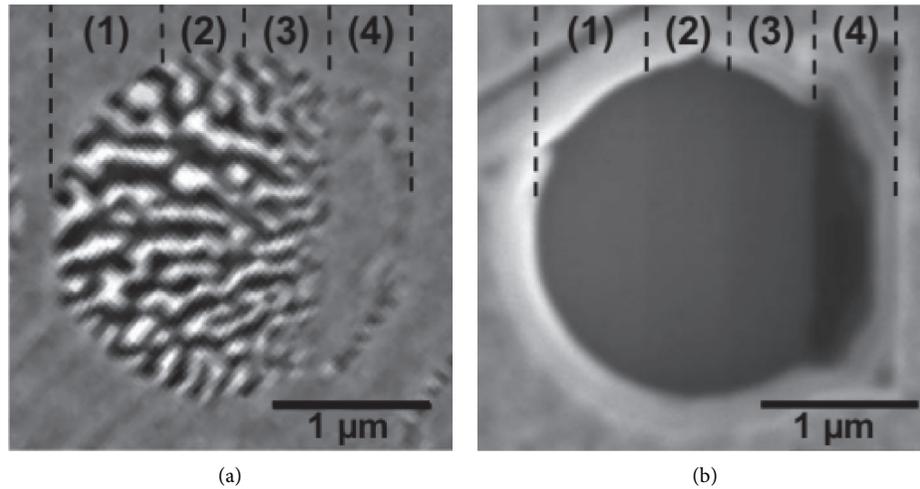


FIGURE 8: (a) Domain image of a Co/Pt sample structured with stripes of increasing ion dose, labeled (1–4), at remanence. (b) SEM image of the sample [86].

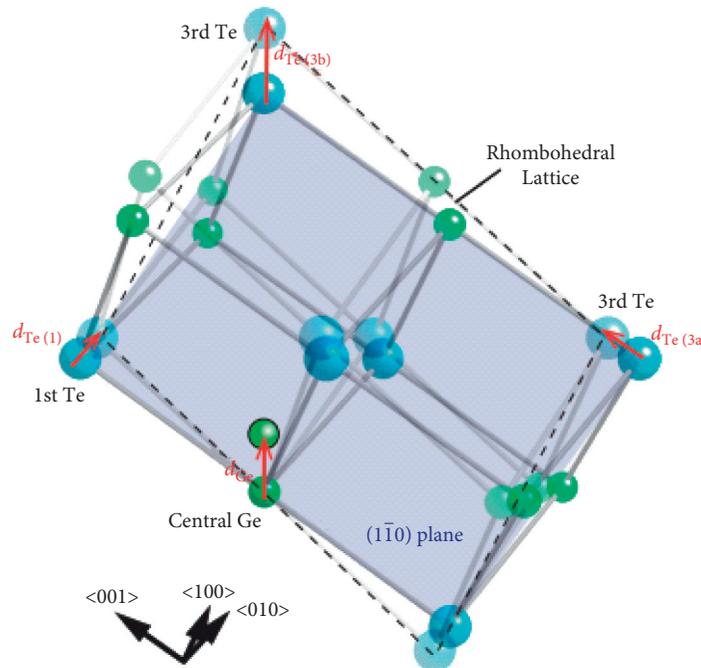


FIGURE 9: (Color) Distortion model around the central Ge atom in  $\text{Ge}_{0.6}\text{Mn}_{0.4}\text{Te}$ . For clarity, the distortions in the illustration are much larger than the real ones (the distortion of the lattice angle is about 5 times as large as the true value) [91].

optimization, Nguyen et al. [100] used quantitative phase-contrast X-ray nanoholography to analyze the microstructure of  $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$  high-energy density electrodes and quantified them.

Recently, a new application of X-ray holography has been discovered. Turnbull et al. [101] researched nanoscopic lamellae of centrosymmetric ferromagnetic alloy and showed tilted holographic images at  $30^\circ$  incidence. They have proved the successful application of X-ray holography in identifying the topology of localized structures in nanoscale magnetism. Blukis et al. [102] used XFH to image the magnetic structure of the Tazewell IIICD meteorite cloud

area with a spatial resolution of 40 nm. This is the first case of applying XFH to measure the magnetization of a single magnetic particle. Its response to a magnetic field can directly measure its magnetic stability and the strength of particle interaction. It provides a new approach for studying the magnetic field of the early Solar System and further for studying the formation of the Solar System and the process of early planetary evolution.

In this section, X-ray holography is mainly reviewed for three-dimensional imaging of nanomaterials, thin film crystals, ferroelectric materials, and electrode structures. Fundamentally, these research objects are on functional

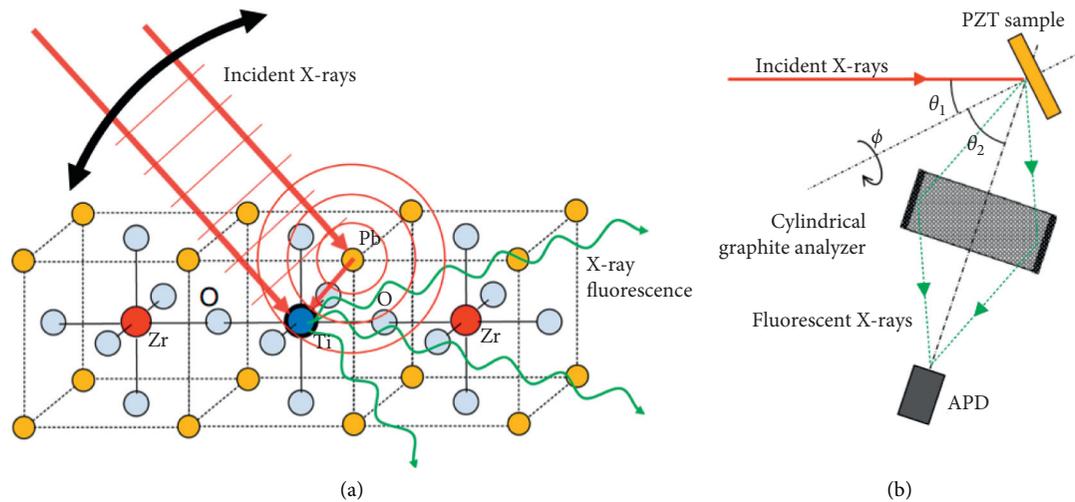


FIGURE 10: Principle and experimental setup of XFH. (a) Principle of inverse XFH. In this study, Ti in PZT was the target element. (b) Experimental setup for inverse XFH [94].

materials with special purposes but are different from general materials. X-ray holography can realize the measurement of the three-dimensional atomic structure of nanostructured materials, which is the basis for understanding nanomaterials. In reality, most nanomaterials are manufactured artificially with the premise of understanding. The epitaxial growth technology improves the flexibility of device design and the performance of the device. X-ray holography successfully realized the three-dimensional imaging of the extrinsic long film structure. Through the measurement of structural changes among different elements, the influence of change in external conditions on the internal element structure can be obtained. This is helpful for the study of the working process of that material. As a special functional material, ferroelectric materials have significant applications in many aspects. The three-dimensional imaging of the internal microstructure of ferroelectric materials is mainly used to study the composition and structure of materials, which helps to understand the microscopic origin of special properties and make better use of ferroelectric materials. The measurement of the microstructure of the electrode helps to study the influence of the internal substance of the electrode, its shape on the performance of the electrode, and the optimization of the electrode. Through the measurement of structural changes, the influence of changes in external conditions on the internal element structure of the material and among different elements can be obtained, which are helpful for the study of the working process of the material. Research on materials is the key to the progress and development of industry.

Through X-ray holography, the understanding of these special materials has been further improved. The rapid development of science and technology puts forward higher requirements for materials. So to discover more properties of materials becomes the focus of the following research. However, research on composite nanomaterials, high-performance ferroelectric materials, low-temperature solid

oxide battery materials, and high-performance epitaxial growth technology are still in their infancy. With the improvement of technology, X-ray holography, we believe, will become an important tool for more in-depth material research.

### 3. Conclusions

As a mature approach of three-dimensional imaging, X-ray holography has many advantages and a wide range of applications in material science, biomedical science, and industry, which attracts many researchers to invest increasing energy in the study and its development. The research and applications of X-ray holography have been more than a century. They have drawn extensive attention from scholars from the beginning. As an important tool, X-ray holography has shown many strong advantages, such as high spatial resolution, short measurement time, and more advanced image reconstruction approaches. It is not difficult to find that the application range and measurement results of X-ray holography are related to the improvement of test equipment and technological progress. These technologies include many aspects. The first of them is the X-ray source. The development of synchrotron radiation accelerators and free electron lasers can provide brighter and better coherent light sources, which can excite shorter pulses to achieve femtosecond imaging. In this case, the holographic images like 'continuous' to achieve dynamic measurement of the microstructure. The second is the improvement of measurement approaches. The higher measurement requirements have led to the emergence of many measurement approaches, including improvements of the existing approach and the combination with traditional approaches. The third is the upgrade of the image reconstruction algorithm. The second step of holography is an image reconstruction based on the principle of diffraction, providing the phase information of the image. Combined with more optimized algorithms, high-resolution images and more information can be obtained.

Although X-ray holography is currently in full swing, its technology still faces many challenges. For example, X-ray holography can measure the structure of materials with atomic resolution and also their microstructures on prepared biological samples. But there are great challenges in measuring the microstructure of living cells. As is known to all that X-rays have radiation, which can damage the structure of the tested sample and make the measurement results inaccurate. This is a huge challenge to measure the microstructure of sensitive materials and live biological samples with X-ray holography. Imaging neuron networks provide a basis for understanding the nervous system. Nanoscale X-ray holography based on synchrotron radiation can perform three-dimensional imaging of nerve tissue on a subcellular scale, which is tremendously helpful to understand the pathological mechanism of neurological diseases. The next focus of research will be about how to perform three-dimensional imaging on a smaller scale while obtaining more information. Radiation therapy is a useful means to treat some diseases. X-ray holography can accurately focus the radiation on the diseased part and reduce the damage to other parts. In order to obtain a smaller damage range, an urgent problem to be solved is how to achieve the precise positioning of the target. The development and application of high-performance functional materials are related to the development of the industry. However, the current technical conditions are limiting a comprehensive understanding of the properties of these materials and the mastery of the preparation process. With the advancement of X-ray holography, it is believed that these problems will finally be solved. The key research is on X-ray sources as for the progress and development of X-ray holography. The fourth-generation synchrotron radiation emitter can produce high-brightness, short-pulse X-ray sources. Combined with high-performance detectors and optimized algorithms, it is expected to achieve dynamic and accurate measurements of smaller structures on the femtosecond scale while expanding the range of measurement. In the future, further theoretical and practical research will promote the application of X-ray holography to the three-dimensional measurement of more diverse microstructures.

### Conflicts of Interest

The authors declare no conflicts of interest.

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