

Complex γ -Ray Hologram: Solution to Twin Images Problem in Atomic Resolution Imaging

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A new technique for high fidelity three-dimensional imaging of atomic structure with γ -ray holography is demonstrated. A complex hologram was constructed from holograms recorded for different values of the nuclear scattering amplitude on both sides of the ^{57}Fe Mössbauer resonance. The holographic reconstruction was applied to this complex hologram resulting in a twin-image-free image of the bcc Fe local structure. The proposed procedure allows the removal of the twin images for all real space, making γ -ray holography an unambiguous tool for atomic and magnetic structure imaging.

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Internal source holography (ISH) [1] was proposed as a tool for three-dimensional model-free imaging of atomic structure. ISH employs the fact that the interference pattern formed by radiation emitted from a localized source inside a sample can be interpreted as a hologram of the local atomic structure. ISH was introduced in electron diffraction [2] and subsequently applied in x-ray fluorescence [3] and bremsstrahlung [4] experiments. For electromagnetic radiation the time-reversed version of ISH is more promising as it was experimentally demonstrated for x rays [5] and γ rays [6]. Holographic methods are not limited to systems with a long-range order but can also be applied to clusters, surface adsorbates, and impurities [7].

In the first step of such holographic procedure, an interference pattern,

$$I = \left| R + \sum_i O_i \right|^2 = R^2 + 2 \operatorname{Re} \sum_i R^* O_i + \dots, \quad (1)$$

of waves O_i scattered by the atoms and a coherent reference wave R is recorded. If the object's waves are small, a three-dimensional image of the object can be numerically reconstructed from the measured pattern in the second step [8].

As seen from Eq. (1), only the real part of the complex interference term (hologram) may be recorded, which results in the presence of twin images in real-space reconstruction. For ISH real and twin images occupy the same region in space distorting the reconstructed image.

In the time-reversed γ -ray holography [6], a sample containing a Mössbauer isotope (^{57}Fe) is illuminated by radiation from a radioactive source (^{57}Co). The incident photons can be directly absorbed in a recoilless, resonant process (reference wave) or they can be additionally resonantly scattered on other nuclei (object waves). The intensity of the resulting interference field is probed at nuclear sites by measuring the conversion electron yield as the illumination direction is varied. The nuclear scattering cross

section value is high and the observed holographic modulation is several percent of the total signal which results in reasonable acquisition times even for a tabletop experiment with a natural radioactive source. The most promising feature of γ -ray holography is the imaging possibility of a three-dimensional magnetic structure, similarly as in theoretically predicted method of spin electron holography [9] but without any of the drawbacks connected with the sophisticated nature of low-energy electron scattering. It could be performed by tuning to specific Mössbauer transition between the sublevels split in the internal magnetic field specific for a crystal site. It has been suggested previously [10] that a model experiment demonstrating the magnetic resolution of γ -ray holography could be performed for magnetite in which Fe ions occupy two different crystallographic positions and the magnetic hyperfine fields at both sites differ enough to resolve site specific holograms.

Unfortunately, this possibility has not been realized yet. The main reason is the presence of holographic twin images. For specific systems, the real and twin images overlap and for certain combinations of interatomic distances and incident radiation energy they can add up to zero, completely removing the information about particular scatterers from the reconstructed real-space image [11]. This difficulty may be overcome in electron holography and multiple energy x-ray holography by collecting holograms for many wavelengths [12]. This method cannot be applied in the γ -ray holographic Mössbauer experiment where only a single wavelength is accessible. However, very recently, for optical holography, it was demonstrated that twin images can be eliminated by reconstructing a complex hologram which contains information about both the real and imaginary parts of the scattered waves [13]. In the present Letter, a similar method is applied for γ -ray holography. The proposed method takes advantage of the possibility of changing the phase of the nuclear scattering amplitude by detuning from the resonance [10], in a way similar to the x-ray anomalous diffraction techniques [14].

We show a high fidelity 3D real-space reconstruction of the bcc-Fe structure, completely free of twin images, obtained from a complex hologram, which had been constructed from the experimental holograms. In addition, presented results link the anomalous x-ray diffraction methods with the idea of complex holography.

The small size of the nucleus as compared to the radiation with a x-ray wavelength and a relatively simple polarization dependence of nuclear scattering allow us to write the holographic formula for a single scattering nucleus as [10]

$$\chi_1(\mathbf{k}, \Delta E) \approx 2 \operatorname{Re} \left[\frac{f(\Delta E)}{r} e^{i(kr + \mathbf{k} \cdot \mathbf{r})} \right]. \quad (2)$$

Equation (2) describes only the interference term between the reference wave, traveling from the γ -ray source placed far from the sample to an absorbing nucleus (the reference nucleus) located inside the sample at the origin, and an object wave additionally scattered by a single nucleus placed at position \mathbf{r} . In Eq. (2) \mathbf{k} is the wave vector of the incident plane wave and the exponential factor results from the optical path differences. In the nuclear resonant scattering amplitude $f(\Delta E) = |f(\Delta E)|e^{i\phi(\Delta E)}$ only the dependence on the energetic shift ΔE from exact nuclear resonance, which has the width of the order of 10^{-9} eV, is shown explicitly. This parameter may be easily tuned by changing the constant value of the Doppler velocity of the γ -ray source moved by a Mössbauer transducer. In Eq. (2), the angular dependence of the scattering amplitude on the wave vector and the magnetic field directions, characteristic for the M1 transitions, has been omitted for simplicity. A more accurate expression for the γ -ray hologram, including the vectorial nature of the scattered waves, may be found in Ref. [10] or in Ref. [15], where it was derived in a rigorous quantum-mechanical approach. For monochromatic radiation or radiation with a Lorentz-like energy spectrum $f(\Delta E)$ has the following properties characteristic for resonance scattering [16]: $\operatorname{Re}f(\Delta E) = -\operatorname{Re}f(-\Delta E)$, $\operatorname{Im}f(\Delta E) = \operatorname{Im}f(-\Delta E)$, and $\phi(\Delta E = 0) = -\pi/2$.

The twin-real image problem may be demonstrated considering the simplest centrosymmetric system consisting of a reference nucleus placed at the origin and two scattering nuclei at positions \mathbf{r} and $-\mathbf{r}$. Then, the corresponding hologram function may be written as

$$\chi_2(\mathbf{k}, \Delta E) = 4|f(\Delta E)| \cos[kr + \phi(\Delta E)] \cos(\mathbf{k} \cdot \mathbf{r})/r. \quad (3)$$

The real-space image function obtained from such a hologram by applying the Fourier-like holographic transform [8] may be expressed as

$$U(\mathbf{R}) = \iint_S \chi_2(\mathbf{k}, \Delta E) e^{i\mathbf{k} \cdot \mathbf{R}} d\sigma_k \propto \cos[kr + \phi(\Delta E)] \times [j_0(k|\mathbf{R} - \mathbf{r}|) + j_0(k|\mathbf{R} + \mathbf{r}|)]/r, \quad (4)$$

where the spherical Bessel functions j_0 are simply the real or twin image of a point scatterer. It is apparent that the hologram and the reconstruction image are blind with respect to nuclear pairs for some values of the kr product, for which the cosine value reaches zero. It is also visible that this condition varies for different values of the scattering phase shift.

Consider first a situation of scattering with pure imaginary or pure real scattering amplitude ($\phi = -\pi/2$ or $\phi = 0$). Hereafter, the corresponding holograms will be called real and imaginary holograms, respectively. In Fig. 1(a) calculated holograms $\chi_1(\mathbf{k})$ of a single nucleus are shown for both cases. The real-space image functions reconstructed from $\chi_2(\mathbf{k})$ holograms are shown in Fig. 1(b) as a function of the distance between the imaged nuclear pair and the reference nucleus. It is visible that both dependencies are exactly in antiphase, which means that the twin images can be removed by measuring both a real and an imaginary hologram. Imaginary holograms have already been recorded experimentally [6]. Unfortunately, it is not possible to directly record a real γ -ray hologram because the scattering amplitude is real only far from the resonance where the nuclear scattering cross section is negligible.

A hologram is a linear diffraction pattern. It is a linear function of the scattering amplitude unlike a conventional diffraction pattern recorded in the far field containing its squared values. Furthermore, γ -ray holograms recorded close to the Mössbauer resonance correspond to the same

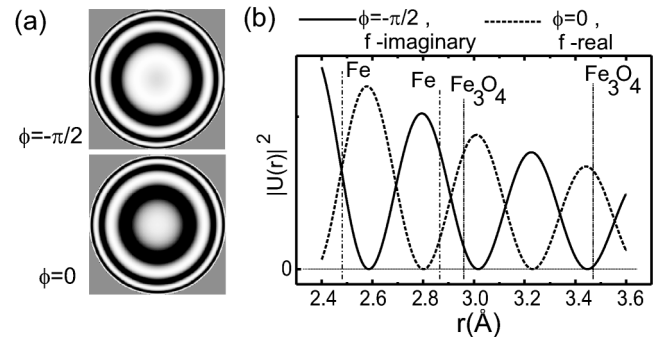


FIG. 1. (a) Calculated real and imaginary γ -ray holograms of a single nucleus. (b) Illustration of the real-twin images cancellation. Real-space image function reconstructed from holograms of two nuclei placed symmetrically with respect to the absorbing nucleus shown as a function of the distance between the absorbing and scattering nuclei. The solid line represents the real-space image function obtained from an imaginary hologram and the dashed line an image from a real hologram. The vertical lines mark the position of nearest Fe nuclei in bcc Fe and in magnetite, showing that for the latter the real-twin images cancellation is severe.

wavelength since the relative energy change has a value of only 10^{-13} . Consider therefore two superpositions of holograms recorded symmetrically for both sides of the Mössbauer resonance:

$$\chi_{\pm}(\mathbf{k}, |\Delta E|) = \chi_1(\mathbf{k}, +|\Delta E|) \pm \chi_1(\mathbf{k}, -|\Delta E|). \quad (5)$$

Using the properties of the nuclear scattering amplitude described above, it can be shown that

$$\chi_{\pm}(\mathbf{k}, |\Delta E|) = \begin{cases} -4 \operatorname{Im}[f(|\Delta E|)] \sin(kr + \mathbf{k} \cdot \mathbf{r})/r, \\ 4 \operatorname{Re}[f(|\Delta E|)] \cos(kr + \mathbf{k} \cdot \mathbf{r})/r, \end{cases} \quad (6)$$

which is an important result for the purpose of this study. The linear superpositions from Eq. (5) of the holograms recorded symmetrically on both sides of the resonance are also holograms. Moreover, the resulting holograms are equivalent to real and imaginary holograms independent of the ΔE parameter. The relative intensity of the holograms is given by a factor $c = \operatorname{Im}f(\Delta E)/\operatorname{Re}f(\Delta E)$ for which the value of the order of 2 can be obtained even close to the resonance.

To show that the proposed procedure can be realized experimentally we performed a holographic experiment in a setup similar to the one described in Ref. [6], based on a conversion electron Mössbauer spectrometer. For practical experimental reasons, two γ -ray holograms of a $^{57}\text{Fe}(001)/\text{MgO}(001)$ epitaxial film were recorded with a ^{57}Co source on the left side of the $\frac{1}{2} \rightarrow \frac{3}{2}$ Mössbauer transition and on the right side of the $-\frac{1}{2} \rightarrow -\frac{3}{2}$ transition. Both lines give identical holograms in the resonance. In addition, these are the only transitions, where the scattered radiation has exactly the same energy as the incident one. For other transitions the spin-flip effect allows the emission of two radiation components and decreases the contrast of the hologram. The holograms were recorded symmetrically with respect to resonance with their effective energetic separation ($2\Delta E$) being equal to the experimental FWHM of the Mössbauer line, i.e., only a double decrease of the total number of counts with respect to the resonance detection occurred. The corresponding asymmetry factor had a value of $c = 2$ which allowed the construction of the linear superposition from Eq. (6) with a good signal-to-noise ratio.

Figure 2(a) shows linear superpositions of the recorded holograms. A slowly varying background coming from the reference wave was subtracted and the holograms were symmetrized by operations corresponding to the fourfold symmetry characteristic for the bcc structure of α -Fe. To enhance the signal coming from the nearest nuclei a low-pass Fourier filter was applied to the images. For the following real-space transformation this filter was omitted. For comparison, real and imaginary holograms calculated according to Eq. (2) for two coordination shells around the absorbing nucleus are shown in Fig. 2(b). The agreement

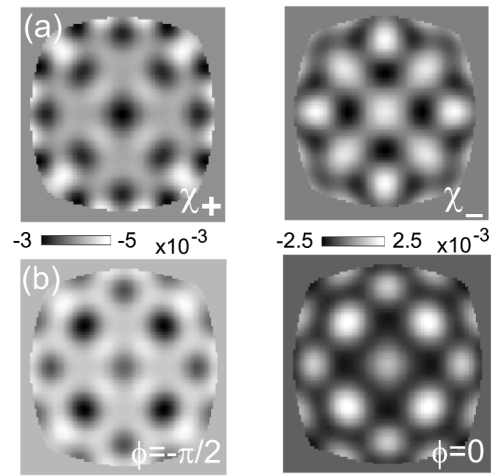


FIG. 2. (a) Holograms of bcc Fe local structure in an epitaxial Fe(001) film obtained by linear superpositions (sum and difference) of two holograms recorded in experiment on opposite sides of the Mössbauer resonance. The gray intensity scale refers to the total signal and is of the order of 5×10^{-3} . (b) Calculated real and imaginary holograms of nuclei in two coordination shells around the absorbing nuclei.

is good, confirming the possibility of obtaining holograms differing in the phase by $\pi/2$ in an experiment.

Figure 3 shows real-space images obtained from the constructed experimental real and imaginary holograms expanded to a full reciprocal-space sphere in order to obtain isotropic spatial resolution [17]. The intensities of the nuclear images of the nearest and the next-nearest neighbors are different for both real-space images as predicted by Eq. (4) and shown in Fig. 1(b) what is caused by the twin-images effect.

The twin images will disappear for a hologram containing the whole information about the scattered waves, i.e.,

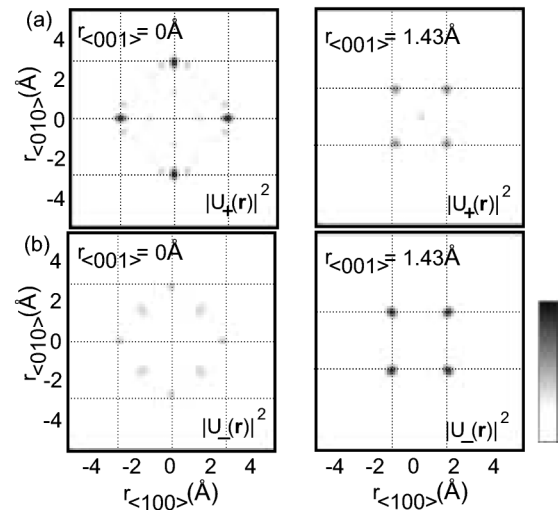


FIG. 3. Real-space images reconstructed from a sum (a) and a difference (b) of experimental holograms recorded for opposite sides of the Mössbauer resonance. Cuts are presented for the plane containing the absorbing nucleus and 1.43 Å above it.

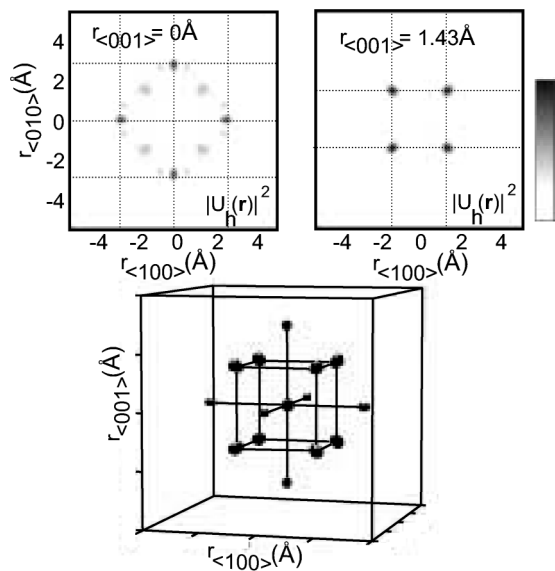


FIG. 4. Real-space images reconstructed from a constructed complex hologram. Cuts are presented for the plane containing the absorbing nucleus and 1.43 Å above it. The 3D image is presented in a form of an isosurface at 40% of the maximum value of the whole real-space image. The sphere in origin represents the absorbing reference nucleus. The sticks have a length equal to the lattice constant of bcc Fe.

about the real and imaginary part of the holographic interference term. The real and imaginary holograms from Eq. (6) can thus be combined to form a complex hologram,

$$h(\mathbf{k}, |\Delta E|) = \chi_+(\mathbf{k}, |\Delta E|) + ic\chi_-(\mathbf{k}, |\Delta E|), \quad (7)$$

which contains such information.

The real-space image of the bcc-Fe epitaxial film obtained from the complex hologram defined in Eq. (7) is presented in Fig. 4. The nuclei in the two coordination spheres are well imaged with an isotropic resolution of less than 0.5 Å and with an experimental error in positions of about 0.1 Å. Further nuclei are not imaged because of the divergence of the γ -ray beam and corresponding experimental angular resolution, which is a simple consequence of the sampling theorem. The intensities of the nuclear images in two coordination shells of 3D real space are almost equal, proving that the cancellation of real and twin images is eliminated.

The presented procedure is very much different from the multiple-energy holographic algorithm [12]. Only two holograms are needed for the complete removal of the twin images, whereas in the multiple-energy scheme a high number of holograms is needed. This result indicates a practical possibility of using γ -ray holography to study the geometrical and especially the magnetic atomic structure of thin films and multilayers containing ^{57}Fe . Holographic

diffraction using γ rays seems to be well suited for the investigation of those kinds of systems, since the high divergence of the beam from radioactive sources fits very well with the slow angular variation of the holographic signals. Systems of interest do not need to have either long-range or chemical order. In addition, for symmetrical detuning from the resonance, the mass absorption as well as the nuclear and electronic scattering cross sections are identical for $\pm\Delta E$ and the related background will vanish for the difference hologram. Therefore, for a particular system, if the geometrical structure is known (and a pure real hologram is not blind to it) it will be possible to construct only the difference of the recorded diffraction patterns and the troublesome procedure of the background removal will be redundant. This will enable experiments on complex magnetic systems with a nonperfect structure and on magnetic nanostructures with low concentration of the Mössbauer isotope (effectively only about 10–20 monolayers of ^{57}Fe), for which electronic scattering in the substrate is dominant and the time instabilities of the detection system cannot be avoided due to long acquisition times.

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