Multiple-energy x-ray holography: Polarization effect

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We present the theory for multiple-energy x-ray holography (MEXH), using a multipole expansion for the scattered field. We find that light polarization plays a crucial role in the reconstruction of the image, and we suggest how to use it in order to eliminate aberration effects. The method we propose is alternative to the scattered-wave-included Fourier transform method, but has the advantage that no theoretical calculations are required to redefine the hologram.

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I. INTRODUCTION

The idea of holography dates back to Gabor’s original works and is essentially based on the interference of optical paths. The problem has been summarized very clearly by Szöke. Consider as a simple example an emitter A at the origin and a single neighboring atom Na at a position ra. In the process known as x–ray fluorescence holography (XFH), the atom A is excited and the emitted radiation either goes directly to the detector (photofilm) at a far distance R (the so-called reference wave), or is first scattered by Na (object wave). The photofilm registers in this way the interference of the two optical paths (hologram), and gets in Fourier transform the desired information on the scatterer position ra. Equivalently, as proposed originally by Gabor, the last step can be accomplished by illuminating the photographic film with an incoming spherical wave (the decoding wave), but numerical solutions that use a digital detector and suppress the decoding wave are preferable nowadays.

A step forward in this direction has been given recently by the group of Fadley and Materlik, in using the emitter A as a detector. In this experiment (MEXH), the reference wave is furnished by a synchrotron radiation (SR) source, while the fluorescing atom A senses an electric field which is the sum of the direct wave and the scattered front (see Fig. 1) due to its neighboring atoms (object wave).

Formally, MEXH can be visualized as a time reversal situation of XFH, in the sense that the detector is substituted by the source, which obviously does not modify the hologram. XFH data can be collected at the same time by many detectors, one for each fluorescence energy of the excited atom. In spite of this, MEXH presents many hidden advantages.

First of all, the hologram χ(k) is defined in a continuous range of energies. In MEXH and in contrast to XFH, the photon energy is always above the fluorescence threshold of the emitter, thus allowing a better resolution of the image as ~k−1. Secondly, the SR photon energy in MEXH can be freely varying, and this allows one to suppress unpleasant twin-image effects, once the integration is not limited to a sphere, but is done over a three-dimensional (3D) volume in k space. A comparison between the two methods has been given recently by Len et al., and we refer to them for further details.

In this report we develop the full vector theory for MEXH. The general theory is presented in Sec. II. In Sec. III we make a multipole expansion of the previous formulas, and discuss their limit of applicability. Here is where aberration comes out, and we show in Sec. IV how to avoid it, playing with the polarization of the reference beam. Finally in Sec. V we confirm our previous findings presenting some numerical results on a real crystal structure.

II. THE BASIC EQUATIONS

Let us fix our attention on the neighboring atoms first. The external SR monochromatic field is represented by the normalized vector potential

\[ \mathbf{A}_{SR} \]

FIG. 1. The picture of MEXH process. The interference pattern is due to the different optical paths of the direct beam (dashed) and the one scattered by the neighboring atom Na (solid line).
The origin and state, and the last term in Eq. (2.5) produces density and total induced density for each transition is weighted by appropriate coefficients, which can be found by applying standard perturbation theory. Matching the density and current density operators between states, the result is the same form as in Eq. (2.1), i.e.,

\[ \rho(r, t) = \rho(r; \omega) e^{-i \omega t} + \text{c.c.} \quad (2.3a) \]

and

\[ j(r, t) = j(r; \omega) e^{-i \omega t} + \text{c.c.} \quad (2.3b) \]

To find the two quantities on the right-hand side, we must pay attention to (1) the atoms are centered at \( r_s \), and not at the origin and (2) the wave functions \( \psi \) depend on the relative position with respect to the nucleus

\[ x = r - r_s \quad (2.4) \]

and are otherwise independent of \( s \). This means that the induced density (or current) for each atom, factorizes as a prefactor \( \exp(i \mathbf{k} \cdot \mathbf{r}_s) \) times a term which is function of \( x \) in Eq. (2.4) and is formally independent of the atomic position. The total induced density (or current) is the sum over atoms. With this in mind the result is

\[ \rho(r; \omega) = \sum_s e^{i \mathbf{k} \cdot r_s} \rho(x), \quad j(r; \omega) = \sum_s e^{i \mathbf{k} \cdot r_s} j(x) \quad (2.5) \]

where

\[ \rho(x) = -\frac{e^2}{mc^2} \sum_{\beta=1}^3 \sum_m \frac{\langle \psi_m^* (x) \psi_m (x) | m | e^{i \mathbf{k} \cdot x'} p_{\beta} | n \rangle}{\hbar (\omega_{mn} - \omega - i \delta)} \]

\[ - \langle n | e^{i \mathbf{k} \cdot x'} p_{\beta} | m \rangle \psi_m^* (x) \psi_n (x) \frac{\hbar (\omega_{mn} + \omega + i \delta)}{\hbar (\omega_{mn} - \omega - i \delta)} \epsilon_{\beta} \quad (2.6) \]

and

\[ j_{\alpha} (x) = -\frac{e^2}{mc} \sum_{\beta=1}^3 \left\{ \delta_{\alpha \beta} e^{i \mathbf{k} \cdot x} | \psi_n (x) \rangle^2 \right\} \]

\[ - \frac{1}{m} \sum_m \left\{ \psi_m^* (x) \frac{p_{\alpha} \psi_m (x)}{\hbar (\omega_{mn} - \omega - i \delta)} \right\} \left\{ \langle n | e^{i \mathbf{k} \cdot x'} p_{\beta} | m \rangle \psi_m^* (x) \psi_n (x) \right\} \epsilon_{\beta} \quad (2.7) \]

In Eq. (2.7) the dyadic over the momentum operator \( \mathbf{p} = -i \hbar \nabla \) means that it operates symmetrically both on the right and on the left, according to the square bracket in Eq. (2.2b). In the first row in Eq. (2.7) one recognizes the diamagnetic contribution which does not depend on the virtual states \( m \), while in the denominators we have defined

\[ \omega_{mn} = (E_m - E_n) / \hbar, \]

and \( E_{m,n} \) are energies of the excited and core electron level. It can be verified from Eqs. (2.6) and (2.7) that Eqs. (2.3a) and (2.3b) satisfy the charge conservation \( \nabla \cdot j + \rho = 0 \). Once the charges and currents are given, the electric field at the emitter is calculated from the Maxwell equations. Outside the sources, this can be found from the vector potential only as

\[ \mathbf{E} = \frac{i}{k} \nabla \times \mathbf{B}, \quad \mathbf{B} = \nabla \times \mathbf{A}, \quad (2.8) \]

where

\[ \mathbf{A}(r; \omega) = \frac{1}{i} \int \frac{e^{i |r - r'|}}{|r - r'|} j(r'; \omega) d^3 r'. \]

Changing the integration variable as in Eq. (2.4), then using the definition in Eq. (2.5), one gets

\[ \mathbf{E}_{\text{obj}} (r; \omega) = \frac{i}{\omega} \sum_s e^{i \mathbf{k} \cdot r_s} \]

\[ \times \left\{ \nabla \times \int \left[ \frac{e^{i |r - r_s - x'|}}{|r - r_s - x'|} \times j(x') \right] d^3 x' \right\}. \quad (2.9) \]

The last equation is the object wave due to the scatterers. A similar result has been obtained by Fonda\(^{10}\) by computing the field through the vector and scalar potentials, but erroneously the latter was neglected from the start. Fonda’s result, in the present notation, is \( \mathbf{E}_{\text{obj}} = i k \mathbf{A} \), which is equivalent to dropping one term of the double vector product in Eq. (2.9). Further on, we show in Sec. IV how this correction becomes crucial for the polarization dependence of the reconstructed image, which is the main point of this paper.

Adding from Eq. (2.1) the direct contribution of the reference wave, i.e.,

\[ \mathbf{E}_{\text{ref}} (r; \omega) = i k e^{i \mathbf{k} \cdot r}, \quad (2.10) \]
we get the total field
\[ E_{\text{tot}} = E_{\text{ref}} + E_{\text{obj}} \] (2.11)
that acts as a perturbation on the fluorescing atom A.

**III. THE MULTIPOLAR EXPANSION**

The states of A involved in the transition are again core electron levels. This means that one needs Eq. (2.11) for \( r = x \) and \(|x| \leq d\), with \( d \) of the order of the Bohr radius. In the same way, the integral in Eq. (2.9) is confined to \(|x'| \leq d\), i.e., in the volume where the current in Eq. (2.2b) is appreciably different from zero. Since \( r_s \) is of the order of the lattice parameter, it follows that
\[ r_s \gg x, x'. \]
This allows us to use for the propagator in Eq. (2.9) the so-called plane wave approximation (PWA)\(^{10}\)
\[ e^{ik|\mathbf{r} - \mathbf{r}_s - x'|} \left| \frac{\mathbf{r} - \mathbf{r}_s - \mathbf{x}'}{|\mathbf{r} - \mathbf{r}_s|} \right| e^{-ik\cdot \mathbf{x}'} \] (3.1)
with \( \mathbf{k}_s = -\mathbf{k} \cdot \mathbf{r}_s \) the scattered momentum. One gets
\[ E_{\text{obj}}(\mathbf{x}, \omega) = \sum_s k^2 e^{ik\cdot \mathbf{x}'} \left[ (\mathbf{M}_s - (\mathbf{M}_s \cdot \hat{\mathbf{k}}) \hat{\mathbf{k}}) \right] \]
\[ + \frac{1}{ikr_s} \left[ 3(\mathbf{M}_s \cdot \hat{\mathbf{k}}) \hat{\mathbf{k}} \cdot \mathbf{r}_s - \mathbf{M}_s \right] \]
\[ \times \left[ 1 - \frac{1}{ikr_s} \right] e^{-ik\cdot \mathbf{r}_s} e^{ik\cdot \mathbf{r}_s} \] (3.2)
with
\[ \mathbf{M}_s = \frac{i}{\omega} \int e^{-ik\cdot \mathbf{x}'} \mathbf{j} \left( \mathbf{x}' \right) d\mathbf{x}' \cdot d^3x'. \] (3.3)
Equation (3.2) has been written for completeness. For \( kd \ll 1 \), \( \exp(-ik\cdot x') \simeq 1 \) in Eq. (3.3) and \( \mathbf{M}_s = \mathbf{p} \) with
\[ \mathbf{p} = \int \mathbf{x}' \rho(\mathbf{x}') d^3 \mathbf{x}' \] (3.4)
the electric dipole moment and \( \rho \) the density in Eq. (2.6). In this case the object field in Eq. (3.2) is constant over the atom. In the two extreme limits \( kr_s \gg 1 \) and \( kr_s \ll 1 \) one discovers in it the familiar result (see Ref. 11) of the radiative and the near static field in the dipole approximation.

For x rays the limit \( kr_s \gg 1 \) is appropriate, but \( k \sim d^{-1} \) is required for good resolution. This means that only the first term contributes to the sum in Eq. (3.2), while in Eq. (3.3) one is led to attempt an expansion as
\[ e^{-ik\cdot \mathbf{x}'} \approx 1 - ik\cdot \mathbf{x}' + \ldots. \]
The result is
\[ E_{\text{obj}}(\mathbf{x}, \omega) = \sum_s k^2 e^{ik\cdot \mathbf{x}'} \left[ [\mathbf{r}_s \times (\hat{\mathbf{r}} \times \mathbf{p})] + [\hat{\mathbf{r}} \times \mathbf{m}] \right] \]
\[ - \frac{i}{3!} [\mathbf{r}_s \times (\hat{\mathbf{r}} \times \mathbf{Q}_s)] e^{ik\cdot \mathbf{r}_s}, \] (3.5)
where \( \mathbf{m} \) is the magnetic dipole moment,
\[ \mathbf{m} = \frac{1}{2c} \int \mathbf{x}' \times j(\mathbf{x}') d^3 \mathbf{x}', \] (3.6a)
\[ (\mathbf{Q}_s)_a = \sum_{\beta=1}^{3} Q_{a\beta}(\mathbf{k}_s)_\beta, \] (3.6b)
and \( Q_{a\beta} \) is the electric quadrupole moment tensor
\[ Q_{a\beta} = \int (3x'_x x'_y r^2 - 2x'_x x'_y r^2 \delta_{a\beta}) p(\mathbf{x}') d^3 \mathbf{x}'. \] (3.6c)
To make the expansion consistent, we shall suppose in the following that the last two terms in Eq. (3.5) are small. Equally well we shall suppose that the same happens for the fluorescing atom A, such that the limit \( x = 0 \) in Eq. (3.5) is appropriate. The yield is simply \( |\mathbf{E}(\mathbf{x} = 0)|^2 \) and function of the direction of the laser beam only. The hologram follows as
\[ \chi(\mathbf{k}) = \left( |\mathbf{E}_{\text{tot}}|^2 - |\mathbf{E}_{\text{ref}}|^2 \right) / |\mathbf{E}_{\text{ref}}|^2 \] (3.7a)
\[ \approx \frac{1}{k^2} (|\mathbf{E}_{\text{ref}}|^2 \cdot \mathbf{E}_{\text{obj}})^c.c., \] (3.7b)
and in the last line the self-hologram \( -\mathbf{E}_{\text{obj}}^2 \) has been neglected. Using Eqs. (2.10) and (3.2) the result for Eq. (3.7b) is
\[ \chi(\mathbf{k}) = k \sum_s f_s e^{ik\cdot \mathbf{r}_s} e^{-ik\cdot \mathbf{r}_s} + c.c., \] (3.8)
where the scattering amplitude \( f_s \) is given by
\[ if_s = (\mathbf{e}^* \times \hat{\mathbf{r}}_s) \cdot (\mathbf{p} \times \hat{\mathbf{r}}_s) + \mathbf{e}^* \times \hat{\mathbf{r}}_s \cdot \mathbf{m} - \frac{i}{3!} \mathbf{e}^* \times \hat{\mathbf{r}}_s \cdot (\mathbf{Q}_s \times \hat{\mathbf{r}}_s). \] (3.9)
This equation shows that \( f_s \) is complex and is polarization dependent. These properties of \( f_s \) can cause difficulties with the imaging of atom positions. It turns out that the quadrupole correction causes a shift,\(^{8}\) while the dipole terms give rise to a distortion of the image (aberration effects). In practical cases (see Ref. 8 for details) the shift is \( \sim 1/10 \) of the resolution and thus negligible. On the contrary the distortion is significant and is polarization dependent, as discussed in the next section.

**IV. ANGULAR ANISOTROPIES**

For a constant \( f_s \) in Eq. (3.8), the Fourier transform
\[ U(\mathbf{r}, k) = \frac{1}{4\pi} \int \chi(\mathbf{k}) e^{-ik \cdot \mathbf{r}} d\mathbf{k}, \] (4.1)
with $d\hat{k}$ denoting the solid angle $d\Omega_{\hat{k}}$, solves completely the problem of the atomic position. In this case\(^5\)
\[ U(r; k) = k \sum_{s} f_s e^{ikr_s} \frac{\sin(k|r-r_s|)}{r_s} + k \sum_{s} f_s^* e^{-ikr_s} \frac{\sin(k|r+r_s|)}{k|r+r_s|} \] (4.2)
and shows spherical illuminated spots with resolution $\sim k^{-1}$ centered at the atoms and their twins. The twins do not represent a problem in MEXH. In fact, as suggested by Barton,\(^7\) they can be mostly eliminated by integrating Eq. (4.1) over the energy as
\[ U(r) = \int U(r; k) e^{-ikr} k^2 dk \] (4.3)
and we shall not discuss it. Instead we shall use Eq. (4.1) and concentrate on how to render $f_s$ constant.

One way is to use the scattered-wave–included Fourier transform (SWIFT) method proposed by Saldin et al.\(^12\) In it, the Fourier transform of Eq. (4.1) is done not on the bare hologram $\chi(k)$ as one gets from the data, but on the redefined quantity
\[ \chi_{\text{SWIFT}}(k; r) = \frac{\chi(k)}{f_r} \]
where
\[ f_r = f_s(\hat{r}_s - \hat{r}) \]
and $f_s$ is defined in Eq. (3.9). Using Eq. (3.8) one notes that as $r$ in Eq. (4.1) approaches $r_s$, $f_s/f_s \approx 1$, which guarantees that both shifts and distortions are eliminated in Eq. (4.1).

The method applies for any polarization, but problems may arise when the ratio $f_s/f_s$ is $0/0$. In addition the dipole moments and the quadrupole term are assumed to be known.

Another way to render $f_s$ constant, would be instead to use the bare experimental quantity $\chi(k)$, but to play with the polarization. Neglect for the moment the corrections in Eq. (3.9) and concentrate on the first term only. To make life easier, here and in the following we shall suppose that $p$ is in the direction of the field, i.e., $p = a \hat{e}_{\text{ref}}$ with $a$ real. Then
\[ f_s \sim |\epsilon \times \hat{r}_s|^2 = 1 - |\epsilon \cdot \hat{r}_s|^2 \] (4.4)
and $f_s = 1$ if
\[ \epsilon = \epsilon_s = \frac{\hat{r}_s \times \hat{k}}{|\hat{r}_s \times \hat{k}|}. \] (4.5)
The choice made in Eq. (4.5) seems to require the prior knowledge of the position $r_s$. However, we can proceed and come back to this point after Eq. (4.7), because it is not necessary to know $r_s$ before taking measurements. The method we propose is the following.

Let $\epsilon_1$ be an arbitrary polarization, let $\epsilon_2 = \hat{k} \times \epsilon_1$ be obtained by a rotation of $\pi/2$ over the direction of the reference beam, and take
\[ \epsilon_3 = \frac{1}{\sqrt{2}} (\epsilon_1 + \epsilon_2). \]
Measure the yields for $\epsilon_1$, $\epsilon_2$, $\epsilon_3$ and call $\chi_1$, $\chi_2$, and $\chi_3$ the respective holograms. Then the hologram $\chi_{(\alpha, \beta)}$ for any polarization
\[ \epsilon = \alpha \epsilon_1 + \beta \epsilon_2, \quad \alpha^2 + \beta^2 = 1, \] (4.6a)
with real $\alpha$ and $\beta$, is given by
\[ \chi_{(\alpha, \beta)} = (\alpha - \beta) (\alpha \chi_1 - \beta \chi_2) + 2 \alpha \beta \chi_3. \] (4.6b)
This equation follows from the analogous relation between scattering amplitudes in the approximation (4.4). The proof is below.

For the three polarizations $\epsilon_1$, $\epsilon_2$ and $\epsilon_3$ have from Eq. (4.4) $f_s^{(1)} = 1 - (\epsilon_1 \cdot \hat{r}_s)^2$, $f_s^{(2)} = 1 - (\epsilon_2 \cdot \hat{r}_s)^2$, and
\[ f_s^{(3)} = 1 - \frac{1}{2} (\epsilon_1 \cdot \hat{r}_s)^2 - \frac{1}{2} (\epsilon_2 \cdot \hat{r}_s)^2 - (\epsilon_1 \cdot \hat{r}_s)(\epsilon_2 \cdot \hat{r}_s). \]
For the general $\epsilon$ of Eq. (4.6a), and $\alpha$ and $\beta$ real, we have instead
\[ f_s = 1 - \alpha^2 (\epsilon_1 \cdot \hat{r}_s)^2 - \beta^2 (\epsilon_2 \cdot \hat{r}_s)^2 - 2 \alpha \beta (\epsilon_1 \cdot \hat{r}_s)(\epsilon_2 \cdot \hat{r}_s). \]
Using the equation for $f_s^{(3)}$ to eliminate $(\epsilon_1 \cdot \hat{r}_s)(\epsilon_2 \cdot \hat{r}_s)$, then using the equations for $f_s^{(1)}$ and $f_s^{(2)}$, as well as $\alpha^2 + \beta^2 = 1$, one has
\[ f_s = (\alpha - \beta) (\alpha f_s^{(1)} - \beta f_s^{(2)}) + 2 \alpha \beta f_s^{(3)}. \]
From Eq. (3.8), the same relation holds for the three holograms, thus Eq. (4.6b) follows.

Taking for $\alpha$ and $\beta$ the values
\[ \alpha_s = \frac{\hat{r}_s \cdot \epsilon_2}{|\hat{r}_s \times \hat{k}|}, \quad \beta_s = - \frac{\hat{r}_s \cdot \epsilon_1}{|\hat{r}_s \times \hat{k}|}, \] (4.7)
and noting that $\epsilon_2 = \hat{k} \times \epsilon_1$ and $\epsilon_1 = \epsilon_2 \times \hat{k}$, one obtains from Eq. (4.6a)
\[ |\hat{r}_s \times \hat{k}| \epsilon = (\hat{r}_s \times \hat{k} \cdot \epsilon_1) \epsilon_1 - (\hat{r}_s \times \hat{k} \cdot \epsilon_2) \epsilon_2 = (\hat{r}_s \times \hat{k} \cdot \epsilon_1) \epsilon_1 + (\hat{r}_s \times \hat{k} \cdot \epsilon_2) \epsilon_2 = \hat{r}_s \times \hat{k}. \] (4.8)
Thus $\epsilon = \epsilon_s$ and $f_s = 1$ as wanted.

To implement Eq. (4.7) we need a preliminary estimate of $r_s$. Such an estimate will usually be provided by an examination of the holograms obtained from $\chi_1$, $\chi_2$, and $\chi_3$. Any bright spot seen in these holograms, if it really corresponds to an atom, will give a better image when use is made of the optimal polarization for that spot, constructed through Eq. (4.7). The procedure can be iterated by reading off a new $r_s$ from the improved image. The point, of course, is that all this can be done in software, without new measurements. Alter-
natively, one can optimize the entire hologram at once by replacing \( \mathbf{r}_1 \) with \( \mathbf{r} \) in Eq. (4.7).

We show now with an example how, without corrections, distortion is effectively present. Suppose that the atom \( N_a \) is placed at \( \mathbf{r}_a \) on the \( z \) axis and take

\[
\epsilon_1 \approx \frac{\mathbf{z} \cdot \hat{k}}{\left| \mathbf{z} \times \hat{k} \right|} \tag{4.9}
\]

in the \((x,y)\) plane. Then \( \left| \epsilon_1 \times \hat{r}_a \right|^2 = 1 \), but

\[
\left| \epsilon_2 \times \hat{r}_a \right|^2 = \cos^2 \psi',
\]

(4.10a)

where

\[
\hat{k} = (\sin \theta' \cos \varphi', \sin \theta' \sin \varphi', \cos \theta').
\]

(4.10b)

Keeping only the contribution of \( N_a \) to the hologram, a spherical spot is found for \( \epsilon_1 \), but not for \( \epsilon_2 \) as we now show.

According to Eq. (4.1), the atom’s image in this case is given by

\[
\frac{1}{4 \pi} \int \cos^2 \theta' \exp\{-i \mathbf{k} \cdot (\mathbf{r} - \mathbf{r}_a)\} d\Omega'.
\]

(4.11)

The exponential can be expanded as

\[
4 \pi \sum_{lm} (-i)^l Y_{lm}(\theta, \varphi) Y^*_{lm}(\theta', \varphi') f_l(\xi),
\]

(4.12)

where \( \theta, \varphi \) are the angles of the vector \( \mathbf{r} - \mathbf{r}_a \) and

\[
\xi = k |\mathbf{r} - \mathbf{r}_a|.
\]

(4.13)

The integral can be done by using the orthogonality relations of spherical harmonics. The result, which does not depend on \( \varphi \), can be written as

\[
U_\theta(\xi) = \frac{1}{3} \left[ j_0(\xi) - 2 j_2(\xi) P_2(\cos \theta) \right],
\]

(4.14)

where \( P_2 \) is a Legendre polynomial and \( j_0 \), \( j_2 \) are spherical Bessel functions. Note that \( U_\theta(0) = 1/3 \). For \( \theta = 0 \)

\[
U_\parallel(\xi) = \frac{1}{\xi} \left[ 1 - \frac{2}{\xi^2} \sin \xi + \frac{2}{\xi} \cos \xi \right],
\]

(4.15a)

while for \( \theta = \pi/2 \)

\[
U_\perp(\xi) = \frac{1}{\xi^2} \left[ \sin \xi - \cos \xi \right].
\]

(4.15b)

More in general for any \( \theta \)

\[
U_\theta(\xi) = U_\parallel(\xi) \cos^2 \theta + U_\perp(\xi) \sin^2 \theta.
\]

(4.16)

The contours of constant \( U \) resemble ellipses. In particular, we can examine the contour \( \xi = \xi(\theta) \), solution of \( U_\theta(\xi) = 0 \). We see that \( U_\parallel \) vanishes for \( \tan \xi = \xi/(1 - \xi^2/2) \), thus \( \xi(\theta = 0) \approx \pi/1.5 \). On the other hand, \( U_\perp \) vanishes for \( \tan \xi = \xi \), i.e., \( \xi(\pi/2) \approx \pi/0.7 \). Thus the two axes of the constant-\( U \) ellipses are approximately in the ratio \( 1.5/0.7 > 2 \).

V. NUMERICAL RESULTS AND CONCLUSIONS

Numerical calculations have been carried out for a Fe bcc structure, to see how the method works. With the emitter at the origin, we focus our attention on the nearest neighbor spot \( \mathbf{r}_a = (\frac{1}{2} \frac{1}{2} \frac{1}{2} + \frac{1}{2})a \), where \( a = 2.87 \) Å is the lattice spacing for iron. The plane is the \((1 1 0)\) one and the directions \([110]\) and \([001]\) as indicated in the figures.

The reference beam energy has been chosen to minimize out of phase overlap between an atom and its twin, a well known problem in single-energy holography. In fact the sum in Eq. (3.8) is invariant for \( \mathbf{r}_a \rightarrow - \mathbf{r}_a \), thus one gets (apart from a \( 2k^2 \) factor) the dimensionless quantity

\[
\chi(k) = \sum_s f_s \frac{\cos(kr_s)}{kr_s} \sin(k \cdot \mathbf{r}_s).
\]

(5.1)

with \( f_s \approx 1 \) for both polarizations given in Eq. (4.4). The prefactor \( P = \cos(kr_0)/kr_0 \) selects the possible energies for the image reconstruction as \( \cos(kr_0) = 1, -1 \) which correspond to local maxima (minima) in the \( |E_{\text{ed}}|^2 \) intensity in Eq. (3.7a). We concentrate here on maxima, and take as the most suited energies for the \( r_a \) scatterer the values \( E_1 = 5 \) keV and \( E_2 = 10 \) keV. The calculations are multiple energy in the sense that \( E_1 \) and \( E_2 \) are peak energies of Gaussian beams with energy spreads indicated in the figure captions and discussed more fully below. We do not perform the further average of Eq. (4.3) because it can introduce distortions of its own and we want to focus on the polarization effect.

Figures 2(a) and 2(b) are for energy \( E_1 \). Figure 2(a) is for polarization \( \epsilon_1 \), here both orthogonal to \( \hat{r}_a \) and \( \hat{k} \) as in Eq. (4.5) for \( \hat{r}_a = \hat{r}_a \). A spherical spot is clearly shown, in spite of the possible disturbances induced by other scatterers, and neglected in the analysis of the previous section. Figure 2(b) refers to \( \epsilon_2 \) polarization, gotten by rotating \( \epsilon_1 \) by \( \pi/2 \) around \( \hat{k} \) as indicated previously. Here the central spot is distorted and very weak, due to the interference of other scatterers which we discuss below. The main intensity appears to be concentrated at the emitter only (lower-left corner). Actually, the intensity at the emitter is the same in Figs. 2(a) and 2(b), but we use a rescaling in Fig. 2(b), to make it clearer.

Figures 3(a) and 3(b) refer to the second energy \( E_2 \). The shorter wavelength allows here a better resolution of the central spot, and at the same time a better positioning at the center of the figure. Figure 3(a) is for \( \epsilon_1 \) polarization and shows a perfect spherical image. Figure 3(b) instead refers to \( \epsilon_2 \) polarization. Now the image has become half as intense as before and is distorted as well. The polarizability choice seems to be crucial again for the atom observability.

We make now further comments on the figures and give details of the calculations. With energy \( E_2 \), the atoms \((001)\) and \((110)\) cannot be observed since in both cases \( \cos(kr_0) \) \( \approx 0 \). Surprisingly also the \((111)\) atom (upper-right corner) is missed, although at a distance \( 2r_a \) from the emitter. On the contrary two spurious spots appear in the figure: the first along the diagonal; the second on the abscissa. These artifacts are not new, and in no way related to the polarization...
In all cases we find that the angular averaging is irrelevant as far as $U(r)$ is concerned. The primary role is played by the integration over the modulus, i.e., over the energy. Here we use a 1D Gaussian convolution $g \approx \exp(-k^2/\Delta k^2)$ on the grid data. The effect of using the Gaussian is twofold, and well discussed in Ref. 3. On the one hand it is nothing but the energy spread of the primary beam. On the other, it acts as a low-pass filter to wash out all the distant scatterers, and allows a rapid convergence as the number of the bulk crystal atoms under the sum in Eq. (3.8) is increased. In particular using a sphere of radius $R = a \times N$ we find for $\Delta k \approx \pi/(aM)$ and $N > M$, the result to be independent of the sphere’s radius.

We mention also another role played by the averaging, which is connected with the ill-defined value of bare FT $U$ at $r = 0$. This can be simply shown in the scalar case taking $f_s = 1$ constant. Inserting Eq. (5.1) in Eq. (4.1) and performing the angular integration, get

$$U(0) \approx -2 \sum \frac{\sin(2kr_s)}{(2kr)^2},$$

which is clearly undetermined. On the other hand the same expression can be found from Eq. (5.1) by treating the sum over $s$ as a continuum variable over the angle. This is the contribution of distant scatterers to the hologram and the indeterminacy regards now a physical quantity. It is removed once the integration over the energy window $\Delta k$ is performed.

In Figs. 3(a) and 3(b) we fix $\sigma = \Delta k/k = 7.2 \times 10^{-2}$, close to the Tegze and Faigel value. For Figs. 2(a) and 2(b) we use instead twice the above value since the energy is now half as before. All the $\sigma$ values indicated in the captions ensure a good convergence for $R = 4a$ (536 scatterers) we use in all the figures.

The numerical calculation is performed with a FORTRAN program. Once the averaged holograms $\chi_1$, $\chi_2$, and $\chi_3$ are
calculated on the $5^\circ \times 5^\circ$ grid, the FT requires 2' and 30'' of computer time for each reconstruction at $E = 10$ keV. For Figs. 2(a), 2(b) the requirement reduces to $\sim 1'$. To get a better contrast in the figures, we follow the standard procedure used in atomic holography, namely, plotting $U^2$ rather than $|U|$. After this, $U^2$ has been redefined such that $0 \leq U^2 \leq 1$ in the whole $0 \leq x \leq \sqrt{2}a$, $0 \leq y \leq a \ (1\bar{1},0)$ plane, or in the relevant part of it.

In conclusion, we can assert that the images obtained for optimal polarization are significantly better than those for a polarization orthogonal to it for several energies, and thus also when a further averaging over energies is performed.

The reconstruction of the image with the optimal choice of polarization can be gotten with a reasonable effort from the experimental holograms for three polarizations.

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