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Scattering of twisted light from a crystal

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Abstract
Recent years have seen significant progress in the generation and application of twisted beams carrying orbital angular momentum. Here we study the elastic scattering of twisted Bessel light from a crystal and compare our predictions with the results for incident plane-wave radiation. Based on form-factor approximation our numerical calculations of the differential scattering cross sections have been carried out for a crystal of lithium at x-ray energies. It is shown that the use of twisted light can lead to a measurable change in the scattering cross section for the nanocrystals approaching a few nm in size.

Keywords: angular momentum of light, optical vortex, nanocrystal, x-ray scattering

(Some figures may appear in colour only in the online journal)

1. Introduction

The elastic scattering of x-rays by bound atomic electrons has become an excellent probe of the structure of matter [1]. One of the most intriguing example of this is small angle x-ray scattering enabling high-throughput analysis of protein structure [2]. Another interesting example is conventional x-ray imaging used to produce an image of an object, which would otherwise be invisible to the human eye [3]. For instance, medical imaging with x-rays enables us to reconstruct the full three-dimensional structure of a patient's head [4]. Moreover, x-ray diffraction experiments provided decisive structural parameters of the DNA molecule [5].

The essential physics of these processes has been known and understood for many years now when considering the scattering of plane-wave radiation. However, this is not the case for twisted light beams that carry a nonzero projection of the orbital angular momentum (OAM) onto their propagation direction and whose intensity pattern has an annular character [6]. During recent years intense studies have been performed to explore how such twisted beams interact with matter. In particular, it was shown experimentally that the selection rules can be modified in the excitation of atoms by twisted light [7, 8], while the possibility of the production of beams with OAM in the radiative recombination or high-order harmonic generation was emphasized in [9, 10]. A detailed theoretical analysis involving twisted beams was carried out also for electromagnetically induced transparency and for Rayleigh scattering from atomic ensembles [11, 12].

In order to understand how the scattering from crystalline materials depends on the 'twistedness' of incident x-rays, we present here a theoretical analysis of the elastic scattering of twisted Bessel beams from a single crystal of lithium. In section 2 we express the scattering amplitudes for twisted light in terms of their plane-wave analogs. By making use of these amplitudes, we then derive the differential scattering cross section. Section 3 describes our numerical calculations, which show that the scattering cross section is sensitive to the projection of the total angular momentum (TAM) of twisted beams and differs from the standard plane-wave case when the size of the crystal is reduced to the nanometer scale. Finally, a summary of our results is given in section 4. Atomic units ($\hbar = e = m_e = 1$, $c = 1/\alpha$) are used throughout the paper unless otherwise indicated.

2. Theory

2.1. Scattering of plane waves from a crystal

We begin by considering the scattering of plane-wave photons from a crystalline material comprised of atoms in the form-factor approximation that is valid for low-Z atoms and photon energies large compared with binding [13]. In this approximation, the scattering amplitude for a crystal can be written in general as [1]

$$\mathcal{M}_{fi}^{\mathrm{pl}} = \boldsymbol{\epsilon}_i \cdot \boldsymbol{\epsilon}_f^* \sum_n^{\text{all atoms}} \mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i - \boldsymbol{k}_f)\cdot \boldsymbol{b}_n} f_n(\boldsymbol{k}_i, \boldsymbol{k}_f), \tag{1}$$

where $\boldsymbol{k}_{i,f}$ and $\boldsymbol{\epsilon}_{i,f}$ are the wave and polarization vectors of the circularly polarized incident and outgoing photons, respectively, and where $f_n(\boldsymbol{k}_i, \boldsymbol{k}_f)$ is the atomic form factor of the atom situated at position $\boldsymbol{b}_n$, which is defined as [13, 14]

$$f(\boldsymbol{k}_i, \boldsymbol{k}_f) = \int \rho(r)\, \mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i - \boldsymbol{k}_f)\cdot \boldsymbol{r}}\, \mathrm{d}^3\boldsymbol{r}. \tag{2}$$

Here $\rho(r)$ denotes a spherically symmetric charge distribution. Because we are describing the atom in a single-electron model, it is possible to decompose the charge distribution into a sum of terms corresponding to individual electrons. To proceed further, we use the expansion of a photon plane wave in terms of spherical Bessel functions [15]

$$\mathrm{e}^{\mathrm{i}\boldsymbol{k}_i\cdot \boldsymbol{r}} = 4\pi \sum_{l,m} i^l j_l(kr)\, Y_{lm}^*(\theta_{k_i}, \phi_{k_i})\, Y_{lm}(\theta, \phi) \tag{3}$$

with the magnitude of the photon wave vector $k = \omega/c$. By using the above expansion in the form factor (2) and integrating over the angles, we find that

$$f(\boldsymbol{k}_i, \boldsymbol{k}_f) = \sum_{l,m} \mathrm{e}^{-\mathrm{i}m\phi_{k_i}} \tilde{f}_{lm}(\theta_{k_i}, \boldsymbol{k}_f), \tag{4}$$

where the function $\tilde{f}_{lm}(\theta_{k_i}, \boldsymbol{k}_f)$ reads

$$\begin{aligned} \tilde{f}_{lm}(\theta_{k_i}, \boldsymbol{k}_f) &= 4\pi\, Y_{lm}(\theta_{k_i}, 0)\, Y_{lm}(\theta_{k_f}, \phi_{k_f}) \\ &\times \int_0^\infty j_l^2(kr) \sum_e [f_e^2(r) + g_e^2(r)]\, r^2 \mathrm{d}r. \end{aligned} \tag{5}$$

The symbol $\sum_e$ denotes the sum over all bound electrons, while $f(r)$ and $g(r)$ are, respectively, the large and small radial components of the Dirac wave function. A pair of these bound-state radial wave functions is obtained here by solving a pair of radial Dirac equations with the GRASP2K package [16].

We are particularly interested in the case of body centred cubic (bcc) structure adopted, for example, by lithium at room temperature. The bcc structure is a cubic lattice with two atoms per unit cell, or two interpenetrating simple cubic sublattices specified by a set of vectors of the form [17]

$$\begin{aligned} \boldsymbol{b} &= n_1\boldsymbol{a}_1 + n_2\boldsymbol{a}_2 + n_3\boldsymbol{a}_3, \\ \boldsymbol{b}' &= \left(n_1 + \frac{1}{2}\right)\boldsymbol{a}_1 + \left(n_2 + \frac{1}{2}\right)\boldsymbol{a}_2 + \left(n_3 + \frac{1}{2}\right)\boldsymbol{a}_3, \end{aligned} \tag{6}$$

where $\boldsymbol{a}_1$, $\boldsymbol{a}_2$, and $\boldsymbol{a}_3$ are the lattice vectors, whereas n_1, n_2, and n_3 are integers. Then, on assuming that all of the atoms in the unit cell are identical, the scattering amplitude for plane waves becomes

$$\begin{aligned} \mathcal{M}_{fi}^{\mathrm{pl}} &= \boldsymbol{\epsilon}_i \cdot \boldsymbol{\epsilon}_f^* \sum_{l,m} \mathrm{e}^{-\mathrm{i}m\phi_{k_i}} \tilde{f}_{lm}(\theta_{k_i}, \boldsymbol{k}_f) \\ &\times \sum_{n_1,n_2,n_3} [\mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i - \boldsymbol{k}_f)\cdot \boldsymbol{b}} + \mathrm{e}^{\mathrm{i}(\boldsymbol{k}_i - \boldsymbol{k}_f)\cdot \boldsymbol{b}'}]. \end{aligned} \tag{7}$$

If we take the direction of the incident plane wave to be along the z axis, the differential scattering cross section averaged over final polarizations may be written in terms of the scattering amplitude as

$$\frac{\mathrm{d}\sigma^{\mathrm{pl}}}{\mathrm{d}\Omega} = r_0^2 \sum_{\epsilon_f} |\mathcal{M}_{fi}^{\mathrm{pl}}(\theta_{k_i} = 0)|^2, \tag{8}$$

where r_0 is the classical electron radius, while the relation $\sum_{\epsilon_f} |\boldsymbol{\epsilon}_i \cdot \boldsymbol{\epsilon}_f^*|^2 = (1 + \cos^2\theta_{k_f})/2$ for the polarization vectors can be used herein. From these expressions, we see that the differential cross section (8) can be utilized to successfully analyze the angular properties of the scattered light.

2.2. Scattering of twisted light from a crystal

Having studied the scattering of plane waves, we now move on to consider the scattering of twisted light from crystalline materials. Let us briefly review the twisted Bessel beam with a well-defined longitudinal momentum k_{z_i}, modulus of the transverse momentum $\varkappa$, photon energy $\omega = ck = c\sqrt{k_{z_i}^2 + \varkappa^2}$, as well as the projection m_γ of the TAM upon its propagation (z) direction. Such a Bessel beam is uniquely characterized by its vector potential defined by [18]

$$\boldsymbol{A}^{\mathrm{tw}}(\boldsymbol{r}) = \int a_{\varkappa m_\gamma}(\boldsymbol{k}_{\perp_i})\, \boldsymbol{\epsilon}_i\, \mathrm{e}^{\mathrm{i}\boldsymbol{k}_i\cdot \boldsymbol{r}} \frac{\mathrm{d}^2\boldsymbol{k}_{\perp_i}}{(2\pi)^2}, \tag{9}$$

where the amplitude $a_{\varkappa m_\gamma}(\boldsymbol{k}_{\perp_i})$ is given by

$$a_{\varkappa m_\gamma}(\boldsymbol{k}_{\perp_i}) = (-\mathrm{i})^{m_\gamma}\, \mathrm{e}^{\mathrm{i}m_\gamma\phi_{k_i}} \sqrt{\frac{2\pi}{k_{\perp_i}}}\, \delta(k_{\perp_i} - \varkappa). \tag{10}$$

As seen from these expressions, Bessel beam can be viewed as a superposition of plane waves with wave vectors $\boldsymbol{k}_i$, lying on a cone with an opening angle $\theta_{k_i} = \arctan(\varkappa/k_{z_i})$. For the sake of simplicity, we consider only the transverse momentum of the photon smaller than its longitudinal momentum, $\varkappa \ll k_{z_i}$. Within this paraxial approximation, the opening angle θ_{k_i} is also very small, and the TAM projection m_γ of the Bessel beam is just a sum of the OAM and the helicity [18].

We have seen above that a Bessel beam can be written as an integral of the standard plane-wave components with the function $a_{\varkappa m_\gamma}(\boldsymbol{k}_{\perp_i})$. We therefore conclude that the scattering amplitude for twisted light can be found by integrating the plane-wave amplitude (7), namely

$$\mathcal{M}_{fi}^{\mathrm{tw}} = \int a_{\varkappa m_\gamma}(\boldsymbol{k}_{\perp_i})\, \mathcal{M}_{fi}^{\mathrm{pl}}(\boldsymbol{k}_{\perp_i}) \frac{\mathrm{d}^2\boldsymbol{k}_{\perp_i}}{(2\pi)^2}. \tag{11}$$

Upon integration over the transverse momentum with the help of the delta function, we see that $k_{\perp_i} = \varkappa$. The scattering amplitude can be simplified further by noticing that $\boldsymbol{\epsilon}_i \cdot \boldsymbol{\epsilon}_f^* \approx 0.5\,(1 + \lambda_i\lambda_f\cos\theta_{k_f})\, \mathrm{e}^{\mathrm{i}\lambda_i(\phi_{k_f} - \phi_{k_i})}$ for sufficiently small opening angles θ_{k_i}, with the helicities $\lambda_{i,f}$ of the incident and scattered photons [18]. Then, the integration over the azimuthal angle ϕ_{k_i} shows that the main contribution to the amplitude (11) comes from the terms in equation (4) with $m = m_\gamma - \lambda_i$, assuming the crystal to be centered on the beam axis (see figure 1). Finally, we can write down the

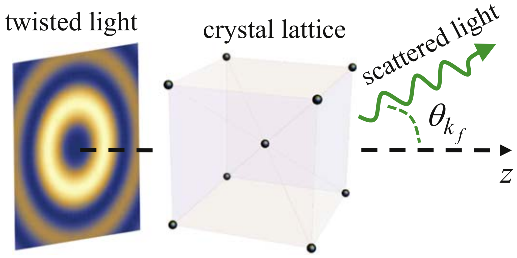

Figure 1. Geometry of the scattering of twisted light from a crystal lattice centered on the beam axis. The emission direction of the outgoing photons is characterized by the angle θ_{k_f}, while the quantization (z) axis is chosen along the propagation direction of the incoming Bessel photons.

scattering amplitude for twisted light in the following form

$$\begin{aligned} M_{fi}^{\mathrm{tw}} &= \frac{(-\mathrm{i})^{m_\gamma}}{2}(1 + \lambda_i\lambda_f\cos\theta_{k_f})\,\mathrm{e}^{\mathrm{i}\lambda_i\phi_{k_f}} \\ &\times\sum_l \tilde{f}_{lm_\gamma-\lambda_i}(\theta_{k_i}, \boldsymbol{k}_f) \\ &\times\sum_{n_1,n_2,n_3}[\mathrm{e}^{\mathrm{i}b_z k_{z_i}-\mathrm{i}\boldsymbol{b}\cdot\boldsymbol{k}_f} + \mathrm{e}^{\mathrm{i}b_z' k_{z_i}-\mathrm{i}\boldsymbol{b}'\cdot\boldsymbol{k}_f}]. \end{aligned} \tag{12}$$

With this result in mind, when the scattered photons are again observed with a polarization-insensitive x-ray detector, the differential scattering cross section from a crystalline material for twisted light is given by

$$\frac{\mathrm{d}\sigma^{\mathrm{tw}}}{\mathrm{d}\Omega} = r_0^2\sum_{\epsilon_f}|\mathcal{M}_{fi}^{\mathrm{tw}}|^2. \tag{13}$$

Below we shall discuss in detail how this cross section depends on the 'twistedness' of incident x-rays.

3. Results and discussion

Let us first work out the scattering of plane waves. It is a well-known result for any particular crystalline material that the lattice sum in the scattering amplitude (7) is a maximum when the momentum transfer $\boldsymbol{k}_i - \boldsymbol{k}_f$ coincides with a reciprocal lattice vector, a fact which is sometimes known as the Laue condition for the observation of x-ray diffraction [1]. In this case all phases in the lattice sum $\sum_n \exp[\mathrm{i}(\boldsymbol{k}_i - \boldsymbol{k}_f)\cdot\boldsymbol{b}]$ are 2π or a multiple thereof, making the sum equal to the huge number of terms. As an example, we consider here a crystal of lithium with a lattice parameter of 3.5 Å and a beam of light of photon energy 8.04 keV corresponding to the wavelength 1.54 Å of the K_α line in Cu, which is frequently used in the laboratory. Such twisted x-rays have been produced experimentally at storage rings either with a circular phase plate [19] or with a rectangular aperture [20]. The Laue condition for the given system is satisfied when the crystal is oriented, for instance, at Euler angles of $\{74°, 50°, 69°\}$ with regard to the scattering plane spanned by the vectors $\boldsymbol{k}_i$ and $\boldsymbol{k}_f$. Within this geometry, the diffraction conditions dictate that the plane-wave differential cross section (8) is sharply peaked about the scattering angle $\theta_{k_f} = 36.2°$ for the (110) reflection, as illustrated by the tabulated data [21]. Our numerical calculations confirm fairly well such a behavior, as shown in the top panel of figure 2.

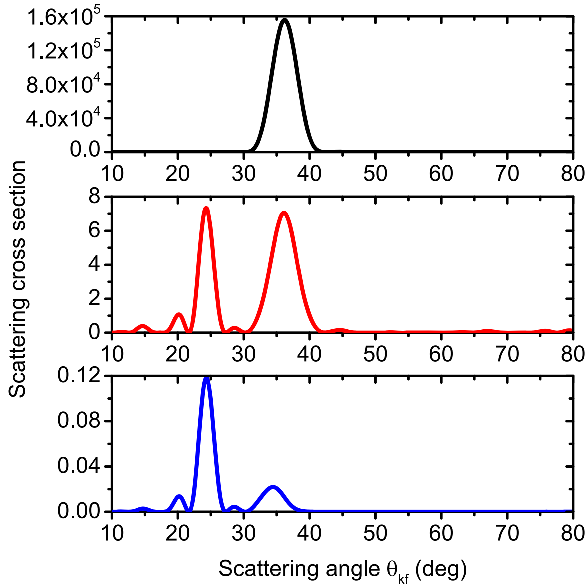

Figure 2. Differential scattering cross section (in r_0^2/sr) for the Li crystal of about 1.5 × 1.5 × 1.5 nm in size oriented at Euler angles of $\{74°, 50°, 69°\}$ with respect to the scattering plane spanned by the incident and scattered direction vectors. Cross section for incident plane waves (top panel) is compared with those for twisted beams with TAM projections $m_\gamma = 2$ (middle panel) and $m_\gamma = 3$ (bottom panel). Calculations were performed for the photon energy $\hbar\omega = 8.04$ keV, helicity $\lambda_i = +1$, and opening angle $\theta_{k_i} = 4°$.

We further analyze the scattering of twisted x-rays. When assuming a small crystal size of about 1.5 nm or 2.3 nm on each side (i.e. a crystal built up from 250 or 686 atoms, respectively), the differential scattering cross sections for Bessel beams (13) with the TAM projections $m_\gamma = 2$ (middle panel) and $m_\gamma = 3$ (bottom panel) differ considerably from the plane-wave results (8) and exhibit several peaks, whose position depends strongly on the TAM of light (see figures 2 and 3). Moreover, the magnitude of these peaks rapidly decreases with increasing values of the photon's TAM m_γ, because the intensity of twisted light at the beam center, where the crystal is located, quickly diminishes as well. The scattering pattern exhibits almost no dependence on the dispersion of the opening angle θ_{k_i}. However, if we take a sufficiently large crystal size (about 14 nm or larger), then there is only one sharp maximum for twisted beams occurred at the same scattering angle as for the plane waves. This point is illustrated in figure 4, which also indicates a general insensitivity of the angular distribution of the emitted radiation to the phase structure of a single twisted beam when using macroscopic atomic ensembles, as has been noted in our previous scattering study [12]. A similar emission pattern for paraxial Bessel and plane-wave incident radiation is observed because in both cases the lattice sum is the dominant factor in the amplitude (12) for a rather large number N of atoms in the crystal, thus suppressing ($\sim 1/N$) all peaks in the scattering cross section except the one dictated by the Laue condition [1]. It is worth stressing that the crystal displacement of about $3/\varkappa$ ($\sim$1.1 nm for the opening angle and energy considered here) for $m_\gamma - \lambda_i = 2$ or $2/\varkappa$ ($\sim$0.7 nm) for $m_\gamma - \lambda_i = 1$ from the central dark spot of twisted beam to its bright ring

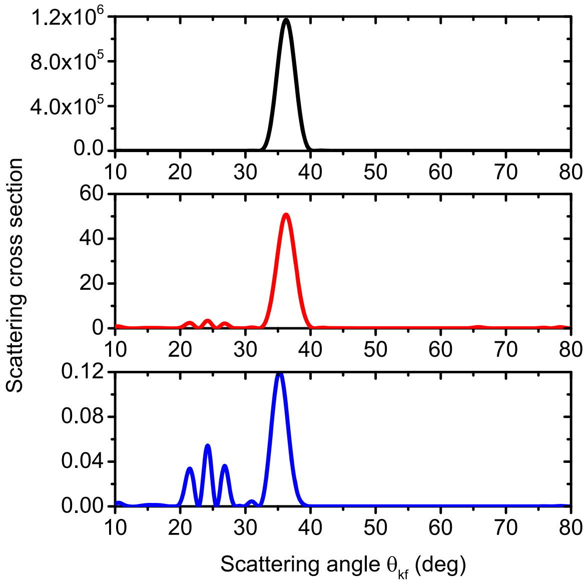

Figure 3. The same as figure 2, but for the Li crystal of about $2.3 \times 2.3 \times 2.3$ nm in size.

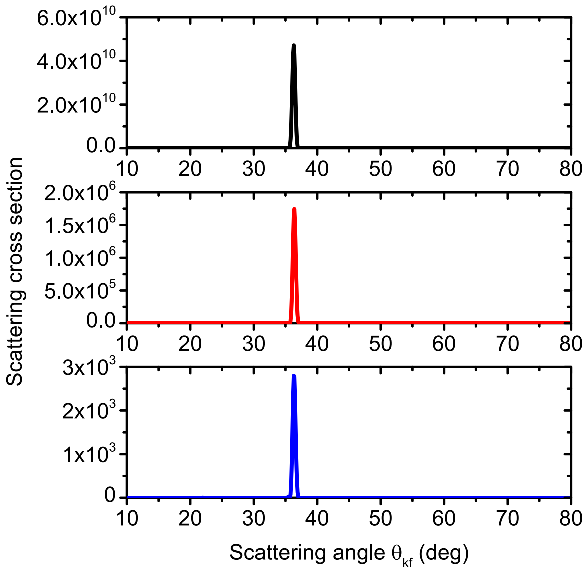

Figure 4. The same as figure 2, but for the Li crystal of about $14 \times 14 \times 14$ nm in size.

arising from the imperfections in positioning of the crystal, which has a typical value of 20 nm in current x-ray diffraction experiments using optical trapping techniques [22], can also lead to plane-wave results because then there is no influence of the OAM on the atomic electrons.

4. Summary

In summary, the scattering of twisted light from a single crystal at x-ray energies has been investigated. Special emphasis was placed on the collision of the Bessel photons with a crystal of lithium. We have demonstrated how the angular momentum properties of incoming radiation can be traced back to the differential scattering cross section. Indeed, the scattering pattern was shown to depend strongly on the projection of the TAM of light for the nanocrystals approximately a few nm in size. Although our present computations were carried out for Li crystals only, a similar behavior can also be observed for other crystalline materials.

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