Dynamical Diffraction Theory for Wave Packet Propagation in Deformed Crystals

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We develop a theory for the trajectory of an x ray in the presence of a crystal deformation. A set of equations of motion for an x-ray wave packet including the dynamical diffraction is derived, taking into account the Berry phase as a correction to geometrical optics. The trajectory of the wave packet has a shift of the center position due to a crystal deformation. Remarkably, in the vicinity of the Bragg condition, the shift is enhanced by a factor $\frac{\Delta \omega}{\omega}$ ($\omega$: frequency of an x ray, $\Delta \omega$: gap frequency induced by the Bragg reflection). Comparison with the conventional dynamical diffraction theory is also made.

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X-ray diffraction has been used as a powerful tool for analyzing crystal structures. Conventional analysis of x-ray diffraction is based on a kinematical theory, namely the first-order Born approximation, and has played an important role in many fields beyond physics such as medical science. A theory incorporating the multiple-scattering process of x ray is called the dynamical diffraction theory, pioneered by Ewald and Darwin. In Darwin’s theory for a perfect crystal, higher-order diffraction is taken into account by superposing the diffracted waves. A perfect crystal, however, suffers from an extinction effect: the intensity of the first-order diffracted wave is reduced by the higher-order waves. In order to avoid this difficulty, mosaicness is introduced into a crystal to suppress the contributions of the higher order diffracted waves by random interferences. Thus, a perfect crystal is not preferable for the structural analysis, and applicability of the dynamical theory has been limited. However, recent technological advances for extremely intense source of x ray such as a free-electron laser will open up new fields of research associated with the dynamical theory.

On the other hand, for a deformed crystal, a dynamical theory was first developed dozens of years ago. In Refs. 4-6, eigenequations for the Bragg reflected beams are translated into differential equations. In general, it is almost impossible to analytically solve such equations, and it is not so easy to extract a clear physical picture from the numerical solutions. We note a corresponding theory for electronic systems with dislocations.

These diffraction theories assume incident waves to be plane waves or spherical waves, and concern the propagation of their wave fronts. On the other hand, one can ask what happens if the incident wave is confined in the transverse direction, i.e., a narrow beam whose trajectory is well-defined. Naively one might expect that the trajectory of such a beam is always perpendicular to the wave front, which is expected in a wave propagation from Fermat’s principle. In this Letter, we reveal an anomalous feature of such electromagnetic beams beyond this naive expectation. We derive equations of motion for an x-ray wave packet in the presence of a crystal deformation. These equations of motion incorporate the dynamical effect of a multiple scattering, and a geometrical phase associated with the wave dynamics, i.e., a Berry phase, is taken into account in a natural way. We find that such a Berry phase in deformed crystals gives rise to a shift of the center position of an x-ray wave packet, and enhances the shift by several orders of magnitude in the vicinity of the Bragg condition. Such a gigantic enhancement is physically related to a small group velocity near the Bragg condition.

A wave packet has a finite width both in real and momentum spaces due to the uncertainty principle. Such a wave packet feels “curvatures” in real and/or momentum spaces. A Berry phase describes these curvatures in real and momentum spaces, and plays a nontrivial role in the wave dynamics in constrained systems. For electromagnetic waves, such a constraint is naturally realized; electromagnetic fields are always constrained to be perpendicular to the propagation direction described by the wave vector $\vec{k}$. The Berry phase for light appears for example in a twisted optical fiber in which the trajectory of the wave vector $\vec{k}$ makes a closed loop. In this case, the polarization plane rotates during propagation, and the rotation angle is represented by a Berry phase. Another kind of constraint is realized in periodic systems, where the dispersion relation has a band structure. When a physical state is constrained on each band, the Bloch function is associated with a Berry phase in momentum ($k$) space. Namely, $k$ space is curved for each band of the Bloch wave function, which influences the wave dynamics. Onoda et al. constructed a generalized geometrical optics including a Berry phase of light.
and derived a set of equations of motion for an optical wave packet. The Berry phase in $k$ space gives rise to a transverse displacement of a wave packet, leading to the optical Hall effect. They were mainly concerned with photonic crystals for visible light, and investigated how to design the photonic band structure in $k$ space. On the other hand, this Letter focuses on the x-ray diffraction; the main interest here is how to probe a spatial deformation in $k$ space. Therefore the relevant Berry phase is distinct from that discussed in [11].

Let us introduce a six-dimensional electromagnetic field vector;

$$\vec{\Gamma}_k(\vec{r}) = \frac{1}{\sqrt{2}} \left( \sqrt{\varepsilon(\vec{r})} \vec{E}_k(\vec{r}) \right) = \frac{1}{\sqrt{2}} \left( \frac{\vec{F}_E}{\vec{F}_H}(\vec{r}) \right),$$

where $\varepsilon$ and $\mu$ are the dielectric constant and the magnetic permeability, respectively. Hereafter we put $\mu = h = 1$. The wave equations for $\vec{F}_E, H$ are

$$\nabla \times \left[ \nabla \times \frac{1}{\sqrt{\varepsilon(\vec{r})}} \vec{F}_E(\vec{r}) \right] = \frac{\omega^2}{c^2} \vec{F}_E(\vec{r}),$$

$$\nabla \times \left[ \nabla \frac{1}{\sqrt{\varepsilon(\vec{r})}} \nabla \times \vec{F}_H(\vec{r}) \right] = \frac{\omega^2}{c^2} \vec{F}_H(\vec{r}),$$

which are Hermitian. In these wave equations, we assume a periodic dielectric function in a crystal with respect to a primitive vector $\vec{a}$: $\varepsilon(\vec{r} + \vec{a}) = \varepsilon(\vec{r})$. In the dynamical theory, the eigenmodes of an electromagnetic wave are Bloch functions under the periodic condition. They are written in the six-dimensional representation as $\vec{\Gamma}_{kz}(\vec{r}) = e^{i\vec{k} \cdot \vec{r}} \vec{U}_{kz}(\vec{r})$, where $\vec{U}_{kz}(\vec{r})$ is a periodic function satisfying $\vec{U}_{kz}(\vec{r}) = \vec{U}_{kz}(\vec{r} + \vec{a})$, and $k_z$ represents the polarization state. Figure 1(a) shows the constant-frequency contour in $k$ space, where the wave vector has a gap at the Bragg condition. Such a gap in $k$ corresponds to an energy gap at the Bragg condition $|\vec{k}| = |\vec{k} - \vec{G}|$, as shown in Fig. 1(b). In the energy gap, the wave number becomes pure imaginary and no x ray can propagate through a crystal. Due to this gap, the dispersion relation for the Bloch function has a band structure, and the Bloch functions are specified by a band index $n$; $\Gamma_{nkz}(\vec{r})$. Using the Bloch functions, we construct a wave packet whose distribution function in $k$ space has a sharp peak around the center of the momentum $\vec{k}$. The wave packet has a peak around the center position $\vec{r} = \vec{r}_c$ in real $r$ space. We consider slowly varying perturbations which do not induce the interband transitions, and keep only the diagonal matrix elements with respect to the band index $n$. The distribution in $k$ space gives an interference between components of different wave vectors, which can be expressed in terms of the Berry curvature in $k$ space associated with each band. In addition, we introduce a crystal deformation whose spatial variation is small within the width of the wave packet. Such a slowly varying deformation leads to the Berry connection in $r$ space, as we will show later.

![Figure 1](image)

**FIG. 1:** (a) Constant-frequency surface and corresponding wave vector and reciprocal vector $\vec{k}$ and $\vec{G}$, respectively. (b) Dispersion relation. At the Bragg condition, $\vec{k}$ and $\omega$ have gaps $\Delta k$ and $\Delta \omega$.

Let us derive equations of motion for such a wave packet. An effective Lagrangian for a wave packet is calculated in a similar manner as in [12, 13] to be

$$L(\vec{k}, \vec{k}_c, \vec{r}_c, \vec{k}_z, \dot{\vec{k}}_c) = -\omega_n + \vec{k}_c \cdot \vec{r}_c + \frac{i\omega}{\Delta k} \cdot (\vec{U}_{kz} | \nabla_{\vec{r}_c} \vec{U}_{kz}) + i\vec{k}_c \cdot (\vec{U}_{kz} | \nabla_{\vec{r}_c} \vec{U}_{kz}) + i\vec{U}_{kz} \cdot \partial_t \vec{U}_{kz}.$$  

The first two terms on the right-hand side describe the wave front propagation, which exist even for plane waves or spherical waves. The last three terms represent the additional physics which appears only in the wave packet dynamics, leading to Berry curvatures. This effective Lagrangian is analogous to that for electronic systems [12, 13]. Variational derivative of the Lagrangian gives the following equations of motion;

$$\dot{\vec{k}}_c = -\frac{\partial \omega_n}{\partial \vec{k}_c} + \Omega_{\vec{k}} \cdot \vec{k}_c + \Omega_{\vec{r}_c} \cdot \vec{r}_c - \Omega_{it},$$

$$\dot{\vec{r}}_c = \frac{\partial \omega_n}{\partial \vec{k}_c} - \Omega_{\vec{k}} \cdot \vec{k}_c - \Omega_{\vec{r}_c} \cdot \vec{r}_c + \Omega_{it},$$

$$|\dot{\vec{k}}_c| = i\vec{r}_c \cdot \vec{A}_l |\vec{k}_c| + i\vec{k}_c \cdot \vec{A}_l |\vec{k}_z|,$$

where

$$(\Omega_{\vec{k}})_{\alpha \beta} \equiv \Omega_{\alpha \beta} k_{\beta} \equiv \langle \partial_{\alpha} \vec{U}_{kz} | i\partial_{\beta} \vec{U}_{kz} \rangle - \langle \partial_{\beta} \vec{U}_{kz} | i\partial_{\alpha} \vec{U}_{kz} \rangle,$$

$$(\vec{A}_l)_{\alpha \lambda \nu} \equiv i(\vec{U}_{n\lambda \nu \vec{k}_z} | \nabla_{\vec{r}_c} \vec{U}_{n\lambda \nu \vec{k}_z} ),$$

$$(\vec{A}_l)_{\nu \lambda \alpha} \equiv i(\vec{U}_{n\lambda \nu \vec{k}_z} | \nabla_{\vec{r}_c} \vec{U}_{n\lambda \nu \vec{k}_z} ).$$

$|\vec{k}_z|$ has two components representing the polarization states, and $\lambda$ is a polarization index. The vectors $\vec{A}_l \vec{k}_z$, called Berry connections in mathematics, are “vector potentials” in such a sense that their rotation gives the Berry curvatures. Each term in the above equations of motion can be interpreted as follows. The first terms on the right-hand side of Eqs. (1) and (2) reproduce geometrical optics. The second term in Eq. (1) represents effects of the crystal deformation, while the third one corresponds to a Lorentz force which is zero in a non-magnetic system. The second term in Eq. (2) represents the anomalous velocity of light [13, 14], giving rise to an optical Hall effect [11]. The third term represents
the displacement of a ray due to a crystal deformation. Equation (3) describes the time evolution of the polarizations. Since we consider a nonmagnetic system, $\Omega_{\Gamma_0} = 0$. We also assume that the perturbations are independent of time; $\Omega_{\Gamma_0} = \Omega_{\Gamma k} = 0$.

Consider a system with a deformation, as shown in Fig. 2, where the spatial variation of the deformation is small within the width of the wave packet. We assume that the wave packet has a sharp peak in $k$ space around the center $\vec{k}_c$, with its width much smaller than $|\vec{G}|$. This implies that the peak in $r$ space is much wider than the lattice constant. In this situation, the eigenfunction of an atomic displacement by several orders of magnitude in the vicinity of the Bragg condition. We assume that the wave vector $\vec{k} \parallel \vec{G}$, and the amount of the shift becomes maximum when $\vec{k} \parallel \vec{G}$. This factor differs for polarizations, as shown in Fig. 2.

This formula is the central result of this Letter. The direction for the shift is parallel to $\vec{G}$, and the amount of the shift becomes maximum when $\vec{G} \parallel \vec{u}$. Such a shift is enhanced by a factor $\frac{\omega}{2|\vec{k}|^2 \Delta \omega}$. This factor differs for polarization states, s- and p-polarizations, which are defined relative to the plane spanned by $\vec{k}$ and $\vec{G}$. The different size of the gap for each polarization state gives the different amount of the shift. When we inject an unpolarized beam, it splits into two beams with s- and p-polarizations, as shown in Fig. 2.

Now we give a realistic estimate for the enhancement factor $\frac{\omega}{2|\vec{k}|^2 \Delta \omega}$ and the shift $\Delta \vec{r}_c$ shown in Fig. 2. For an x ray, the dielectric constant can be approximated as $\epsilon(\vec{r}) = 1 - \omega_p^2/\omega^2$, where $\omega_p$ is a plasma frequency, and typically $\omega_p^2/\omega^2 \sim 10^{-6}$. Hence the value of the enhancement factor for an x ray can be $\frac{\omega}{2|\vec{k}|^2 \Delta \omega} \sim 10^{-6}$, which yields $\frac{\Delta \omega}{\omega} \sim 10^{-6}$. For a deformation of $|\vec{u}| \sim 0.1$ mm, the displacement can be $\Delta \vec{r}_c \sim 0.1$ mm. It means that the deformation in an atomic scale gives rise to a macroscopic shift of the wave packet. Therefore, the factor
plays the role of a “lens”, which gigantically magnifies the geometrical effect on the wave packet dynamics. Such a gigantic enhancement of the shift is a consequence of an energy gap and can be physically interpreted as follows. In the vicinity of the Bragg condition, we have a very small group velocity $\vec{v}_k$. Such a slow wave packet has enough time to interact with electrons, resulting in a remarkably large optical effect. This kind of an enhancement at the edge of the energy gap is often studied in optical waves in photonic crystals [17]. Photonic crystals, however, have a large energy gap, and hence the enhancement is much smaller.

We note that the distribution of the wave packet in $k$ space is desired to be narrower than $\Delta k$. The components of wave numbers outside of this $\Delta k$ width do not show the enhanced shift. To make the whole wave packet shift, an x ray with a wider spot size $l_\perp \sim (\Delta k)^{-1} \sim 0.1$ mm is desirable, because its peak width in $k$-space is as small as $\Delta k \sim 10^{-6}|\vec{G}|$. The amount of the shift for such a beam is $\Delta \vec{r}_c \sim l_\perp$. For a typical beam whose spot size is $l_\perp \sim 100$ nm, only a small portion of the wave packet is shifted, and rest propagates without shifts. It is because the width in $k$ space of this wave packet is larger than the gap $\Delta k$. It is then difficult to measure the shift.

Let us compare the present theory with the conventional diffraction theory. In the conventional kinematical theory, only an equation of motion for $\vec{k}_c$ is concerned. The equation for $\vec{k}_c$ describes the propagation of the wave front. The term $\frac{\partial \vec{\omega}}{\partial \vec{v}}$ can deflect the beam; its amount, however, is not enhanced by the dynamical effect. The conventional dynamical theory deals with a part of the wave front which propagates through a deformation, and calculate the multiple scatters which result in refraction. Although a small part of the wave front has often been regarded as a wave packet, our theory reveals a novel physics of a wave packet, apart from wave front physics. The anomalous enhancement of the shift of the wave packet in this Letter occurs because the wave packet is not a monochromatic wave, but a mixture of waves with slightly different wave numbers. An interference between these waves gives rise to the enhancement discussed in this Letter. This physics of a wave packet is absent in a plane wave or a spherical wave.

It should be warned that such a displacement is completely different from refraction. In refraction, the spot position on the screen is dependent on the distance between a sample and a screen, because the refraction is caused by the shift of $\vec{k}_c$. On the other hand, the shift of $\vec{r}_c$ obtained in this Letter is independent of the distance. Hence the shifts of $\vec{k}_c$ and $\vec{r}_c$ have different physical origins and are experimentally distinguishable.

In summary, we have developed a dynamical diffraction theory for a wave packet propagation in a deformed crystal. Equations of motion for a wave packet are derived. The central result is the shift of the trajectory given in Eq. (4); the center of position of the wave packet is displaced by a crystal deformation parallel to the reciprocal lattice vector $\vec{G}$, and the amount of the shift has maximum when $\vec{G} \parallel \vec{u}(\vec{r})$. Remarkably, such a shift of the wave packet is giganticly enhanced by the factor $\Delta k$, which is $\sim 10^6$ for an x ray. Since the factor is different for each polarization state, the wave packets with no-polarization splits into two beams with independent polarizations. In contrast with the conventional dynamical theory which focuses on the wave front and requires numerically solving the Maxwell equations, our theory treats the narrow beam and provides an easy way to evaluate a displacement of the wave packet for a given deformation. An x-ray wave packet with a good coherency is useful to measure a deformation or dislocation in a crystal.

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