

Tailored Complex Potentials and Friedel's Law in Atom Optics

Claudia Keller, Markus K. Oberthaler, Roland Abfalterer, Stefan Bernet,
Jörg Schmiedmayer, and Anton Zeilinger

*Institut für Experimentalphysik, Universität Innsbruck,
Technikerstrasse 25, A-6020 Innsbruck, Austria*

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Tailored complex potentials for atoms can be made of two overlapping standing light waves, one on resonance and one far detuned. The observed diffraction asymmetry of Bragg diffraction of such light structures is due to a corresponding asymmetry of the Fourier components of the potential. In crystal physics this is known as a violation of Friedel's law. [S0031-9007(97)04304-4]

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It is often found that concepts of photon optics can be adapted to matter-wave optics. In our article we choose the conjugate approach. We use the simplicity of the interaction between light and matter waves to design complex periodic potentials for the matter waves and reveal optical concepts. As an example, we investigate a violation of Friedel's law due to fundamental optical principles in a very controlled system.

Typically, diffraction phenomena are invariant under an inversion of the crystal, even when the elementary cell of the crystal possesses no symmetry. This empirical rule is generally referred to as Friedel's law [1]. However, violations of this rule are known from diffraction experiments of x rays or electrons at solid state crystals [2], for example, due to the presence of "anomalous" (absorptive) scatterers. In this Letter we present a violation of Friedel's law in a very different system, where atomic matter waves are diffracted at specially designed "crystals" of light [3,4]. The diffraction asymmetry is due to the interaction of both "normal" and anomalous scattering at superposed refractive and absorptive subcrystals, respectively [5]. This mechanism even works although our light crystal obviously cannot be really absorptive for the atoms, but only changes their internal state.

In our experiment (Fig. 1), we detect atom intensities depending on the atom's incidence angle at the light crystals, and their diffraction angle. In the case of spatial coincidence between the refractive and the absorptive parts of the crystals we obtain symmetric diffraction, as shown in Fig. 1(b). This corresponds to the normal situation in solid state crystals, where Friedel's law is obeyed. However, a violation of Friedel's law is demonstrated in Figs. 1(a) and 1(c), where Bragg diffraction is dominant in one direction. There, the absorptive and refractive index parts of the crystals are arranged such that they are out of phase by $\pm\pi/2$.

In the remainder of this Letter, we will show that this diffraction asymmetry can be understood by evaluating the Fourier composition of the resulting *complex* potential, and employing dynamical diffraction theory. This means specifically that the effect of the total crystal potential cannot be separated into the individual actions of its components. However, in the weak diffraction limit a

more intuitive picture can be justified, which is presented in the following.

The asymmetry can be understood as an interference effect between diffraction at refractive and absorptive "subcrystals" spatially displaced with respect to each other (Fig. 2). Generally, there is a $\pi/2$ phase shift even between waves diffracted at *coinciding* refractive and absorptive structures. This results from the Kramers-Kronig relations [6], i.e., ultimately from causality. This phase difference is the same for diffraction into the two symmetric orders (Fig. 2, situations 1 and 2). Thus, if the refractive and the absorptive structures coincide, diffraction is symmetric and Friedel's law is obeyed.

An additional spatial displacement of the two gratings by (\pm) a quarter of a grating constant yields an additional phase shift of $\pm\pi/2$ which changes sign for the two Bragg diffraction orders (situations 3 and 4). Then, depending on the diffraction order, the two scattered components are

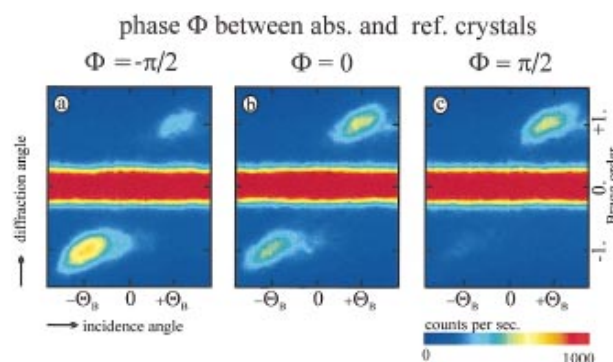


FIG. 1(color). Scattering of atoms at a superposition of absorptive and refractive crystals of light for three different spatial phases between the absorptive and refractive components. The atoms are registered as a function of their incidence and their diffraction angles. The red bar in the middle represents to transmitted atoms (0 order), whereas the two yellow islands on either side of the center bar represent the +1 and the -1 order diffracted atoms. If the two crystals are in phase (b), the intensities in the two diffraction orders are nearly identical. However, the symmetry is clearly broken in (a) and (c), where the refractive and the absorptive crystal are $\pm\pi/2$ out of phase. This asymmetry is a violation of Friedel's law.

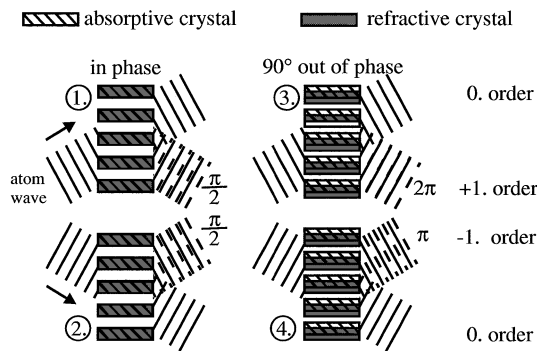


FIG. 2. “Intuitive” picture for the appearance of asymmetric diffraction in conjugate Bragg orders as an interplay between two phase contributions. The waves diffracted at the refractive and absorptive gratings are represented by the solid and the broken lines indicating the wave fronts. In the case where the two crystals are spatially in phase (situations 1 and 2), there is always a $\pi/2$ phase difference between the solid and broken lines, and the diffraction is symmetric. However, if the crystals are spatially displaced by one quarter of a grating period, interference between the two components leads to increased diffraction into one order (situation 3, solid and broken lines in phase), and to reduced diffraction in the conjugate order (situation 4, solid and broken lines π out of phase).

either in phase or π out of phase, resulting in constructive or in destructive interference, and consequently in enhanced or in suppressed diffraction, respectively.

In the following we will first show how to build a complex periodic potential with an arbitrary spatial phase between its real and imaginary components, then discuss our experiments in detail, and finally give a comprehensive discussion of our results.

The interaction between a light field and a two level atom with an additional decay channel of the excited state can be described by the complex optical potential [7]

$$V(x, y) = \frac{1}{\hbar} \frac{d^2 E^2(x, y)}{\Delta + i\gamma/2}. \quad (1)$$

Here, Δ is the detuning of the light frequency from an electronic transition with a dipole matrix element d of the atom, $E(x, y)$ is the electric field of the light wave, and γ is the loss rate from the excited level to a noninteracting state. A standing light wave creates a periodic potential for atomic de Broglie waves—a crystal made of light. By scanning the light frequency it is possible to vary between an effectively real ($|\Delta| \gg \gamma$) and an imaginary ($\Delta = 0$) periodic potential.

A significant advantage of crystals of light is that one can, in principle, realize a huge variety of potential structure by the superposition of light fields. In our experiments, we use this to create a periodic potential with an arbitrary spatial phase between its refractive (real) and absorptive (imaginary) parts.

Our experiments (Fig. 3) are performed with a beam of metastable argon atoms produced in a dc gas discharge

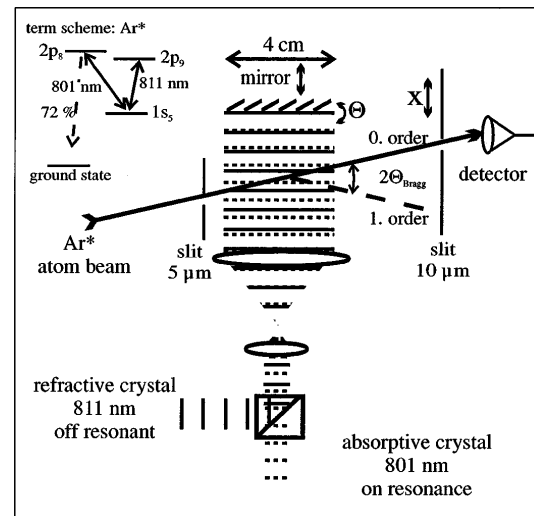


FIG. 3. Experimental setup (not to scale): A collimated beam of metastable atoms crosses a crystal made of two light wavelengths 801 and 811 nm. In the far field behind the light crystal, the atoms are registered by a channeltron detector which is only sensitive to metastable atoms. The incidence angle of the atoms at the crystal can be varied by tilting the retroreflection mirror, used to set up the standing light waves. The phase between the two crystals can be controlled by adjusting the distance between the mirror surface and the atomic beam. The inset shows the level scheme of metastable argon.

[8]. The atoms are thermally distributed with an average velocity of $v = 600$ m/s ($\lambda_{dB} = mv/h = 17$ pm), and 65% (FWHM) velocity spread. This is in the range of the velocity acceptance of our light crystal (55%) at Bragg incidence, as determined by the limited crystal length (3.5 cm). Before passing the light crystal, the beam is collimated transversely with two slits (10 and $5 \mu\text{m}$) separated by 1.4 m (divergence $5 \mu\text{rad}$, corresponding to one-fourth of the Bragg diffraction angle Θ_{Bragg}). At a distance of 1.4 m behind the interaction region, the atoms are registered using a “channeltron” detector, which is only sensitive to atoms in the metastable state.

Argon offers a level scheme (see inset of Fig. 3) which allows a straightforward realization of an arbitrary complex potential. There are two transitions starting from the $1s_5$ metastable state (lifetime about 40 s [9]). The first, with a wavelength of 801 nm to the $2p_8$ state ($A_{ik} = 9.6 \times 10^6 \text{ s}^{-1}$), is open and decays with a probability of 72% to the ground state, which is not detected with our channeltron. Using this transition on resonance (detuning $< 0.1\gamma$, intensity 0.3 mW cm^{-2} , $\Omega_{\text{Rabi}} = 500 \text{ kHz}$), we obtain an effectively absorptive (imaginary) potential. For this purpose we use a diode laser which is locked by saturation spectroscopy at a gas cell. Additionally, we use the closed transition to the $2p_9$ state ($A_{ik} = 36.6 \times 10^6 \text{ s}^{-1}$) with a wavelength of 811 nm. Far-detuned light (detuning 10 GHz, intensity 45 mW cm^{-2} , $\Omega_{\text{Rabi}} = 15 \text{ MHz}$) of a Titanium/Sapphire laser realizes the phase crystal. To form the complex potential the two light fields are

overlapped at a beam splitter and enter the vacuum chamber collinearly (Fig. 3). The light crystal is set up by retroreflecting the central part (3.5 cm) of a collimated Gaussian laser beam (FWHM ca. 3.5 cm) at a mirror in the vacuum.

At the metallic mirror surface the two light fields are in phase. However, as a function of distance from the mirror surface, their relative spatial phase oscillates with a beating period of 32 μm , due to their wavelength difference [10]. This means that by controlling the distance between the atomic beam (5 μm wide) and the mirror surface we can vary the spatial phase between the two superposed crystals of light.

To study asymmetric diffraction our experiments were performed at 12 different distances from the mirror surface corresponding to 12 different relative phases between the absorptive and refractive index gratings. For each position, the incidence angle of the atoms was scanned between $-1.5\Theta_{\text{Bragg}}$ and $+1.5\Theta_{\text{Bragg}}$ ($\Theta_{\text{Bragg}} = k/2G = 17 \mu\text{rad}$) by changing the mirror angle in 40 steps (x axis in Fig. 1) using a piezoactuator. For every step the far-field diffraction pattern was measured by scanning a 10 μm slit in front of the detector (y axis in Fig. 1). Thus, the mirror angle (x axis) and the detection slit position (y axis) were proportional to the angle of incidence and to the diffraction angle of the atoms, respectively. Data for three different distances between the mirror and the atomic beam, corresponding to the three relative phases ($-\pi/2, 0$, and $+\pi/2$) between the absorptive and the refractive index crystals, are shown in Fig. 1. The red bar in the middle of each picture corresponds to atoms which pass the crystal without being diffracted [11]. For incidence angles near the Bragg angles, there is an additional peak of Bragg diffracted atoms (yellow island). Picture 1(b) corresponds to the normal case obeying Friedel's law. Diffracted atoms appear at symmetric incidence angles at different sides of the peak of undeflected atoms. On the other hand, pictures 1(a) and 1(c) show very clearly an asymmetric diffraction into the two Bragg orders. There, one dominant peak of diffracted atoms on either side of the center bar is observed [12].

For a more detailed analysis of the asymmetric diffraction behavior, we have to analyze the structure of the crystal of light: The potential is proportional to the light intensity, and is therefore a periodic sinusoidal function for our standing light wave. A superposition of a far-detuned (refractive) and a resonant (absorptive) standing wave with an arbitrary spatial phase leads to a complex potential [13],

$$V(x) = a[1 + \cos(Gx)] + ib[1 + \cos(Gx + \phi)]. \quad (2)$$

Here, the parameters a and b describe the strength of the real and the imaginary potential and are proportional to the intensities of the detuned and resonant light fields, respectively, and ϕ is the spatial phase between the two fields. In our case of two counterpropagating light waves,

the grating vector \vec{G} is given by $\vec{G} = 2\vec{k}_L$, where \vec{k}_L is the wave vector of the light [10].

A very basic principle in diffraction theory is that the Fourier transform of the potential determines the scattering amplitude in the corresponding diffraction peaks. In our case, the Fourier transform of the potential is

$$\begin{aligned} V(k) &= (a + ib)\delta_{0,k} + (a + ibe^{i\phi})\delta_{G,k} \\ &\quad + (a + ibe^{-i\phi})\delta_{-G,k} \\ &= F(0)\delta_{0,k} + F(G)\delta_{G,k} + F(-G)\delta_{-G,k}. \end{aligned} \quad (3)$$

Kinematic diffraction theory, which is applicable in the weak scattering limit, states that the squared absolute value of the Fourier amplitudes is proportional to the efficiency of diffraction at an order with the corresponding momentum transfer ($0, \hbar G, -\hbar G$). Therefore, the values of $|F(G)|^2$ and $|F(-G)|^2$,

$$\begin{aligned} |F(G)|^2 &= a^2 + b^2 - 2ab \sin \phi, \\ |F(-G)|^2 &= a^2 + b^2 + 2ab \sin \phi, \end{aligned} \quad (4)$$

are proportional to the intensities of the respective Bragg peaks. Friedel's law states that $|F(G)|^2 = |F(-G)|^2$ (symmetric diffraction) [14]. However, this is only true for $\phi = 0, \pi, 2\pi, \dots$, corresponding to the cases where the two crystals are either exactly in phase or π out of phase. This situation appears, for example, in the case of a cosine potential, since $\cos(Gx) \sim e^{iGx} + e^{-iGx}$. On the other hand, a maximal difference between $|F(G)|^2$ and $|F(-G)|^2$ arises if the phase difference is $\pi/2$ or $3\pi/2$. For $a = b$ the potential is proportional to either e^{iGx} or e^{-iGx} , and thus a momentum transfer is possible only in the $+G$ or $-G$ directions, respectively. Consequently, the diffraction efficiency for conjugate diffraction orders is maximally asymmetric.

Our experimental results for the Bragg diffraction efficiencies at various phases between the two crystals are summarized in Fig. 4. The two curves in the upper graph show the number of atoms diffracted at the $+1$ and -1 Bragg orders, respectively. At the intersection points, the diffraction is symmetric. These points are at the predicted spatial phase positions of $0, \pi, 2\pi$, where the absolute values of the Fourier components [Eq. (4)] are equal. Our simple argument with the Fourier coefficients predicts that the two graphs should be two sine waves which are π out of phase. As stated above, the description is only valid in the regime of weak scattering. For stronger scattering, as in our experimental situation, a full integration of the Schrödinger equation is more appropriate. This is usually done in the framework of the theory of dynamical diffraction, which treats the wave field in the periodic potential as a coherent superposition of all scattered wavelets [15]. The results of such a numerical calculation are shown in the lower graph of Fig. 4, and explain the deviations from the purely sinusoidal intensity oscillations as observed in our experiment.

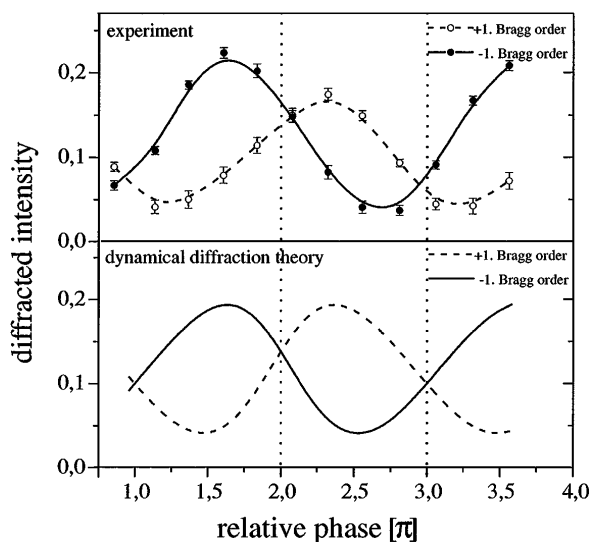


FIG. 4. Diffracted atoms in the two conjugate (+1) and (-1) Bragg orders as a function of the relative spatial phase between the two superposed light fields. The upper graph shows our experimental results (the solid and the broken lines used as guides for the eyes). The error of the relative phase determination (x axis) is less than $\pm\pi/30$. At relative phases of multiples of π , the diffraction is symmetric. For all other phases the number of atoms in the two orders is different. This indicates a violation of Friedel's law. The lower graph shows the results of an exact calculation using dynamical diffraction theory with our experimental parameters, reproducing the observed deviations from purely harmonic functions predicted by the simple kinematic theory. The different amplitudes in the two experimental curves are not reproduced in the calculations. They are probably due to a not completely parallel alignment of the collimation slits and the mirror surface.

In conclusion, we have shown that Friedel's law can be broken for atoms moving in crystals of light. Similarly to the case of light optics, the basic reason for this behavior results from the specific diffraction properties of absorptive and refractive index crystals. In atom optics as well as in photon optics, the scattering processes at absorptive and refractive index structures are fundamentally different. The diffraction at a phase structure cannot be modeled by any combination of absorptive structures alone, and vice versa. This demonstrates the analogy between matter wave diffraction and light wave diffraction down to a very fundamental level, which is based on the validity of the Kramers-Kronig relations. Our experiment shows that atoms moving in crystals of light are a very versatile model system to study in detail the propagation phenomena in periodic structures.

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- [11] The apparent diffraction efficiency is decreased by the fact that 15% of the atoms from the discharge are occupying a second metastable state, which does not interact with the light but contributes to the transmitted intensity.
- [12] We also looked for an effect of the light polarization, because the atoms are in a $J = 2$ state which has five sublevels. In our experiment, the two light fields were orthogonally polarized. However, there was no difference when the polarization of the crystals of light was aligned parallel. The reason is that the mixing of the states in the Earth's magnetic field is faster than the transit time through the light fields.
- [13] Here we neglect the residual imaginary contribution of a far-detuned light potential, and its ac Stark shift. These approximations are justified since the ratio between real and imaginary parts is $2\Delta/\gamma = 3000$ in our case of 10 GHz detuning (linewidth $\gamma = 6$ MHz).
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