

## Observation of Sub-Bragg Diffraction of Waves in Crystals

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We investigate the diffraction conditions and associated formation of stop gaps for waves in crystals with different Bravais lattices. We identify a prominent stop gap in high-symmetry directions that occurs at a frequency below the ubiquitous first-order Bragg condition. This sub-Bragg-diffraction condition is demonstrated by reflectance spectroscopy on two-dimensional photonic crystals with a centered rectangular lattice, revealing prominent diffraction peaks for both the sub-Bragg and first-order Bragg conditions. These results have implications for wave propagation in 2 of the 5 two-dimensional Bravais lattices and 7 out of 14 three-dimensional Bravais lattices, such as centered rectangular, triangular, hexagonal, and body-centered cubic.

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The propagation and scattering of waves such as light, phonons, and electrons are strongly affected by the periodicity of the surrounding structure [1,2]. Frequency gaps called stop gaps, emerge for which waves cannot propagate inside crystals due to Bragg diffraction. Bragg diffraction is important for crystallography using x-ray diffraction [3] and neutron scattering [4]. Diffraction determines electronic conduction of semiconductors [1,2] and of graphene [5], and broad gaps are fundamental for acoustic properties of phononic crystals [6,7] and optical properties of photonic metamaterials [8,9].

Bragg diffraction is described in reciprocal space by the von Laue condition  $\vec{k}_{\text{out}} - \vec{k}_{\text{in}} = \vec{g}$ , where  $\vec{k}_{\text{out}}$ ,  $\vec{k}_{\text{in}}$  are the outgoing and incident wave vectors and  $\vec{g}$  is a reciprocal lattice vector. As a result, a plane exists in reciprocal space for which the von Laue condition is satisfied, called a Bragg plane. When the incident and outgoing wave vectors are located on a Bragg plane these waves are hybridized, thereby opening up a stop gap at the Bragg condition. The boundary of the Brillouin zone is formed by intersecting Bragg planes and therefore gaps open on this boundary [1]. When diffraction involves a single Bragg plane, we are dealing with well-known simple Bragg diffraction, which corresponds in real space with the well-known Bragg condition:  $m\lambda = 2d\cos(\theta)$ . Here  $m$  is an integer,  $\lambda$  is the wavelength inside the crystal,  $\theta$  is the angle of incidence with the normal to the lattice planes, and  $d$  is the spacing between the lattice planes. A stop gap is also formed when Bragg diffraction occurs on multiple Bragg planes simultaneously, which is called multiple Bragg diffraction [10], and is fundamental for band gap formation [2,11,12]. Wave propagation in crystals is described along high-symmetry directions [1]. Multiple Bragg diffraction has been recognized in high-symmetry directions at frequencies above the first-order simple Bragg-diffraction condition:

$m = 1$ ,  $\lambda = 2d$ , or  $\vec{k}_{\text{out}} = -\vec{k}_{\text{in}} = \frac{1}{2}\vec{g}$ . To our knowledge, multiple Bragg diffraction has not yet been observed at frequencies below simple Bragg diffraction [13].

In this Letter we show that for high-symmetry directions multiple Bragg diffraction can occur at frequencies below the first-order simple Bragg condition. As a demonstration, we have investigated diffraction conditions for two-dimensional (2D) photonic crystals using reflectance spectroscopy. A broad stop gap is observed below the simple Bragg condition, depending on the symmetry of the lattice. Our findings are not limited to light propagation, but apply for wave propagation in general, and therefore we anticipate similar diffraction for electrons in graphene [5], and sound in phononic crystals [6,7].

We have studied light propagation in 2D silicon photonic crystals [16]. Figure 1(a) shows a scanning electron microscope image of one of these crystals from the top view. The centered rectangular unit cell has a long side  $a = 693 \pm 10$  nm and a short side  $c = 488 \pm 11$  nm. The pores have a radius of  $r = 155 \pm 10$  nm and are approximately  $6 \mu\text{m}$  deep. The photonic crystals are cleaved parallel to either the  $a$  side or  $c$  side of the unit cell. The cleavages define two directions of high symmetry,  $\Gamma M'$  and  $\Gamma K$ , in the Brillouin zone, see Fig. 1(b). If light travels parallel to these directions, one expects simple Bragg diffraction from the lattice planes in real space [dashed lines in Fig. 1(a)]. A stop gap should appear that is seen in reflectivity as a diffraction peak. Because both directions are of high symmetry, one naively expects simple Bragg diffraction to give the lowest-frequency diffraction peak.

We have identified the diffraction conditions of our 2D photonic crystals along the  $\Gamma M'$  and  $\Gamma K$  directions using reflectance spectroscopy [17]. The photonic crystals are illuminated with a supercontinuum white light source

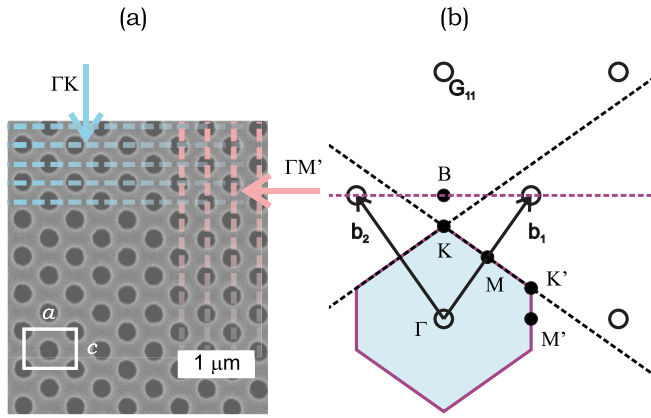


FIG. 1 (color online). (a) Scanning electron microscope image of a 2D photonic crystal with a centered rectangular lattice. The white rectangle marks a unit cell with  $a = 693 \pm 10$  nm,  $c = 488 \pm 11$  nm, and  $r = 155 \pm 10$  nm. The arrows mark two directions of high symmetry  $\Gamma K$  and  $\Gamma M'$ . The dashed lines mark real space lattice planes whose lowest-frequency simple Bragg diffraction occurs along the  $\Gamma K$  and  $\Gamma M'$  directions. (b) Reciprocal space of the centered rectangular lattice (circles). The filled area is the first Brillouin zone,  $b_1$  and  $b_2$  are primitive vectors.  $\Gamma$ ,  $K$ ,  $K'$ ,  $M$ , and  $M'$  are points of high symmetry. The dashed lines are Bragg planes.

(Fianium SC-450-2). TE-polarized light is focused on the crystal using a gold-coated reflecting objective (Ealing X74) with a numerical aperture of 0.65, resulting in a spectrum angle-averaged over  $0.44\pi \pm 10\%$  sr solid angle in air. By assuming an average refractive index ( $n = 2.6$ ), the angular spread inside the crystal is only  $14^\circ$ , corresponding to  $0.06\pi \pm 10\%$  sr solid angle. The diameter of the focused beam is estimated to be  $2w_0 = 1 \mu\text{m}$ . Reflected light is collected by the same objective, and the polarization is analyzed. The spectrum is resolved using Fourier transform infrared spectroscopy (BioRad FTS-6000) with an external InAs photodiode. The spectral resolution was  $15 \text{ cm}^{-1}$ , corresponding to about  $10^{-3}$  relative resolution. For calibration, spectra are normalized to the reflectance spectra of a gold mirror.

Figure 2(a) shows the band structure calculated using a plane wave expansion method [18] and reflectivity measured along the  $\Gamma M'$  direction (black solid line). The broad lowest-frequency measured reflectivity peak between 4700 and  $7300 \text{ cm}^{-1}$  agrees well with the calculated stop gap. This reflectivity peak is caused by simple Bragg diffraction on the lattice planes indicated in the cartoon above, corresponding to the vertical lattice planes in Fig. 1(a). One can also approximate the lowest-frequency simple Bragg-diffraction condition from the dispersion with a constant effective refractive index ( $n_{\text{eff}}$ ), obtained from the low-frequency limit [19]. This estimation is marked by the dashed vertical line and agrees well with the calculated stop gap. The two measured peaks between 9800 and  $11100 \text{ cm}^{-1}$  agree well with a higher-frequency stop gap marked by a second blue area, caused by multiple Bragg

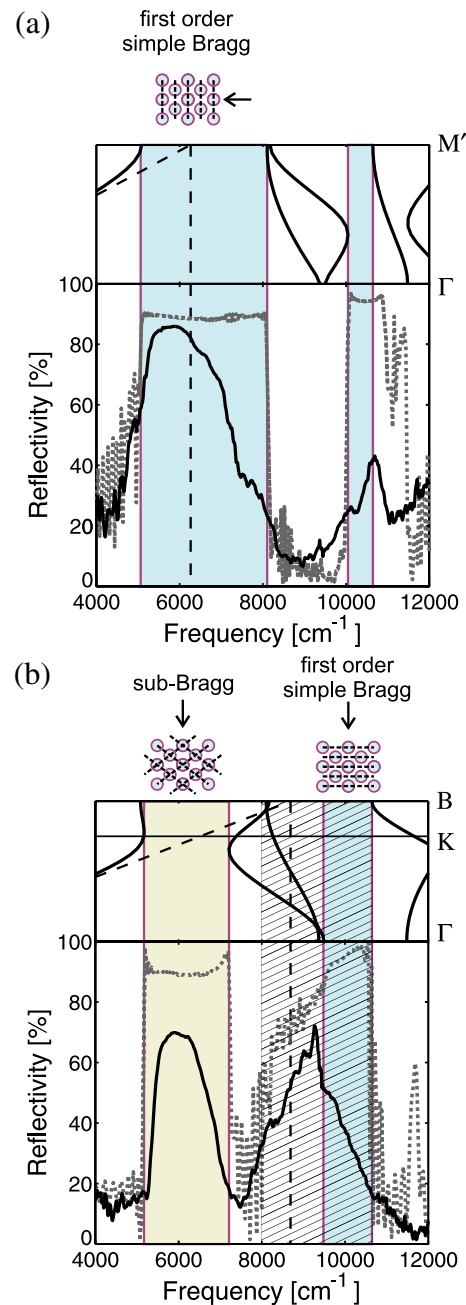


FIG. 2 (color online). Measured (black, solid line) and simulated (grey, dashed line) reflectivity spectra, and calculated band structures for TE-polarized light of a 2D photonic crystal along directions of high symmetry. (a) The measured and simulated lowest-frequency diffraction peaks in the  $\Gamma M'$  direction match a calculated stop gap that occurs at the simple Bragg-diffraction condition. (b) The measured and simulated lowest-frequency diffraction peaks in the  $\Gamma K$  direction match a calculated stop gap and is caused by multiple Bragg diffraction that occurs at a lower frequency than simple Bragg diffraction.

diffraction. The peaks appear at higher frequency than simple Bragg diffraction, as expected. The reflectivity of an incident plane wave on a finite size structure has been simulated with finite difference time domain (FDTD)

simulations [20] (grey, dashed line). The agreement between the simulated and measured reflectivity is gratifying.

In Fig. 2(b) we show the calculated band structure and measured reflectivity along the  $\Gamma B$  direction, where  $K$  is located on the edge of the Brillouin zone and  $B$  is located on the Bragg plane between  $\Gamma$  and reciprocal lattice vector  $G_{11}$ . Two significant broad measured reflectivity peaks are visible. The lowest-frequency peak between 5400 and 6900  $\text{cm}^{-1}$  agrees well with a calculated stop gap marked by the yellow area. This peak is caused by multiple Bragg diffraction on the lattice planes indicated in the cartoon above the calculated stop gap, and is part of the two-dimensional band gap for TE-polarized light. The second reflectivity peak between 8100 and 10000  $\text{cm}^{-1}$  agrees with a second calculated stop gap (blue area). The flat bands in the dispersion relation, causing an impedance mismatch of coupling light into the crystal [9,21], likely broaden the observed peak (hatched area). This is supported by FDTD simulations of the reflectivity of an incident plane wave on a finite size structure (grey, dashed line). The agreement between the simulated and measured reflectivity peak is gratifying. The measured peak is probably rounded off as a result of the high numerical-aperture microscope objective. Note that band structure calculations and FDTD simulations neglect the dispersion of silicon. Scattering from surface imperfections becomes more important at higher frequencies, which could explain why the measured reflectivity peak is much lower near 10000  $\text{cm}^{-1}$ . At any rate, the frequency ranges of the measured and simulated peaks agree very well.

This second stop gap is caused by simple Bragg diffraction on the lattice planes indicated in the cartoon above the calculated stop gap, corresponding to the horizontal lattice planes in Fig. 1(a). The frequency of the simple Bragg-diffraction condition based on an  $n_{\text{eff}}$  is inaccurate because a broad stop gap is already present at lower frequencies. The observation of a prominent diffraction peak caused by multiple Bragg diffraction at a much lower frequency than simple Bragg diffraction is important. This result shows that even for high-symmetry directions such as the  $\Gamma K$  direction, there can be a diffraction condition below simple Bragg diffraction, which we address as *sub-Bragg* diffraction [22].

We have performed reflectivity measurements on photonic crystals with a range of  $\frac{c}{a}$ . Figure 3 shows the width of the diffraction peaks for the  $\Gamma M'$  (a) and  $\Gamma K$  directions (b). The areas correspond with calculated stop gaps, such as in Fig. 2. The dash-dotted line is the approximated frequency of lowest-frequency simple Bragg diffraction assuming a constant  $n_{\text{eff}}$ . Note the very good agreement between the measured frequencies of the diffraction peaks and the calculated stop gaps. We observe for the  $\Gamma K$  direction diffraction always appearing at a lower frequency than simple Bragg diffraction. This observation confirms the robustness of sub-Bragg diffraction [23].

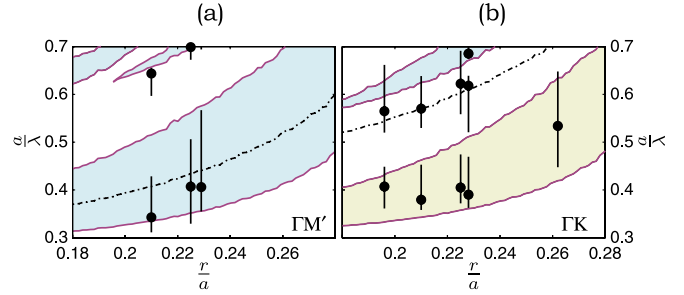


FIG. 3 (color online). Determined reduced width of the diffraction peaks (bars) and frequency of the maximum reflectivity (circles) for different  $\frac{c}{a}$ . The filled areas are calculated stop gaps, color coded as in Fig. 2. (a) Reduced frequency of the diffraction peaks for the  $\Gamma M'$  direction. (b) Reduced frequency of the diffraction peaks for the  $\Gamma K$  direction.

The existence of sub-Bragg diffraction can be explained by considering the lattice in reciprocal space, see Fig. 1(b). For the  $\Gamma K$  direction we observe in reciprocal space two points of high symmetry:  $K$  and  $B$ .  $K$  is located on the Brillouin zone boundary, at the intersection of two Bragg planes corresponding to the von Laue conditions between  $\Gamma$  and  $G_{10}$ ,  $\Gamma$  and  $G_{11}$ . Thus, at  $K$  we have a multiple Bragg diffraction condition on both Bragg planes.  $B$  is located at the Bragg plane (dashed line) that satisfies the von Laue condition between  $\Gamma = G_{00}$  and  $G_{11}$  resulting in simple Bragg diffraction. Since  $B$  is located outside the Brillouin zone, the simple Bragg condition occurs at higher frequency than the sub-Bragg condition. From this figure we describe three conditions for sub-Bragg diffraction: (i) The diffraction condition corresponds to a point on a corner edge of the Brillouin zone, giving rise to multiple Bragg diffraction; (ii) the incident wave vector should be along a high-symmetry direction, which is satisfied by considering only reciprocal lattice vectors  $G_{khl}$ , for which  $|h|, |k|, |l| \leq 1$ , or equivalent; (iii) sub-Bragg diffraction can only occur at a lower frequency than the simple Bragg-diffraction condition.

Using these three conditions, it becomes evident that diffraction conditions for  $M$  and  $M'$  correspond to simple Bragg diffraction for  $G_{10}$  and  $G_{1\bar{1}}$  respectively.  $K'$  satisfies criteria (i) and (iii); however, it does not satisfy criterion (ii). This diffraction condition belongs to multiple Bragg diffraction in a direction of lower symmetry, similar to the observation in Ref. [11]. Therefore, sub-Bragg diffraction is only observed at  $K$ . In this case we have measured the reflectivity of photonic crystals that strongly interact with light. For our crystals, we find that for  $\frac{c}{a} > 0.07$  a stop gap opens at  $K$ , and for  $\frac{c}{a} \leq 0.07$  flat dispersion bands appear.

Up to now we have considered a centered rectangular lattice with long side  $a$ , short side  $c$ , and  $\frac{a}{c} = \sqrt{2}$ . However, sub-Bragg diffraction can be expected for any  $\frac{a}{c} > 1$  [24]. To illustrate this we have made an analytical model to explain the sub-Bragg-diffraction frequency. We calculate

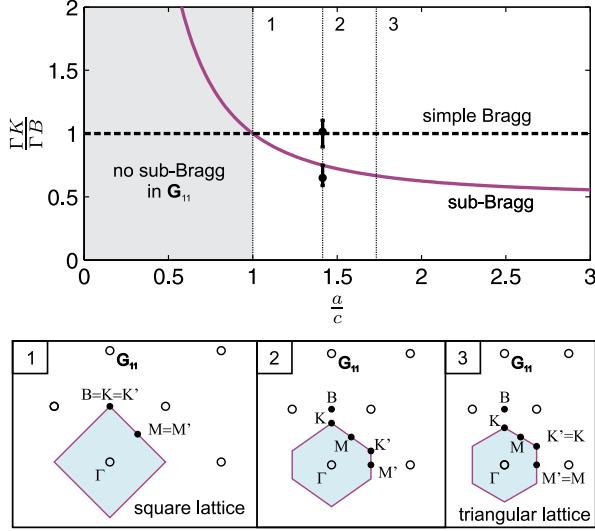


FIG. 4 (color online). Normalized sub-Bragg-diffraction condition (purple) and normalized simple Bragg-diffraction condition (black dashed line) as a function of  $\frac{a}{c}$ . The symbols mark the reflectivity peaks of Fig. 2(b), assuming identical  $n_{\text{eff}}$  for both diffraction conditions. Sub-Bragg diffraction is satisfied for  $\frac{a}{c} > 1$ . Labels 1, 2, 3 refer to  $\frac{a}{c}$  shown in the insets (bottom). Inset 1 shows the reciprocal lattice (circles) for  $\frac{a}{c} = 1$ , giving a square lattice and no sub-Bragg diffraction. Inset 2 shows the reciprocal lattice (circles) for  $\frac{a}{c} = \sqrt{2}$ , resulting in sub-Bragg diffraction at  $K$ . Inset 3 shows the reciprocal lattice (circles) for  $\frac{a}{c} = \sqrt{3}$ , resulting in sub-Bragg diffraction at  $K$  and  $K'$ .

$|\Gamma K|$  and  $|\Gamma B|$  as a function of  $\frac{a}{c}$ , where the frequency of the sub-Bragg condition is proportional to  $\frac{c_0}{n_{\text{eff}}} |\Gamma K|$  and the frequency of the simple Bragg condition is proportional to  $\frac{c_0}{n_{\text{eff}}} |\Gamma B|$ , where  $c_0$  is the vacuum velocity. The results are shown in Fig. 4. When  $\frac{a}{c} \rightarrow \infty$ , sub-Bragg diffraction occurs at  $\frac{|\Gamma K|}{|\Gamma B|} = \frac{1}{2}$ . Inset 1 shows the reciprocal lattice for  $\frac{a}{c} = 1$ , corresponding to the square lattice. In this case,  $|\Gamma K| = |\Gamma B|$  and therefore sub-Bragg diffraction and simple Bragg diffraction occur at the same frequency, violating condition (iii). Inset 2 shows the reciprocal lattice for  $\frac{a}{c} = \sqrt{2}$ , corresponding to the experimental conditions of the structures investigated by us. Inset 3 shows the reciprocal lattice for  $\frac{a}{c} = \sqrt{3}$ , corresponding to the triangular lattice. All three conditions for sub-Bragg diffraction at  $K$  are fulfilled. There is also a sub-Bragg-diffraction condition for  $K'$ . It may seem that condition (ii) is violated because the  $\Gamma K'$  direction corresponds to  $G_{2\bar{1}}$ . However, because of the rotational symmetry of the Brillouin zone,  $K = K'$  and the diffraction conditions in the  $G_{2\bar{1}}$  direction are identical to the  $G_{11}$  direction, and therefore condition (ii) is satisfied. In a similar experiment performed by [25], a diffraction peak was observed at  $K$ . However, these excellent experiments were compared with band structures between  $\Gamma K$ , since it was not recognized that there is also a diffraction condition at  $B$ . For the centered rectangular

lattice, one must calculate band structures between  $\Gamma B$  to accurately estimate the width of the stop gaps. This is evident from the band structures in Fig. 2(b) by comparing the width of the stop gaps when one would consider only  $\Gamma K$  instead of  $\Gamma B$ .

In the case of three-dimensional (3D) crystals, if a Bravais lattice has a planar cross section that can be described by a centered rectangular lattice along a direction of high symmetry, sub-Bragg diffraction will occur. For 2D Bravais lattices sub-Bragg diffraction can occur for 2 out of 5 Bravais lattices: centered rectangular or triangular (which is a special case of centered rectangular), see Fig. 4. There are 7 out of 14 3D Bravais lattices that have a planar cross section that can be described by a centered rectangular lattice in a direction of high symmetry: body-centered cubic, body-centered tetragonal, base-centered orthorhombic, body-centered orthorhombic, face-centered orthorhombic, base-centered monoclinic and hexagonal. We predict that sub-Bragg diffraction can occur for these 7 Bravais lattices.

Sub-Bragg diffraction is valid for any kind of wave propagation in structures that fulfill the symmetry conditions. Therefore we predict that for x-ray spectroscopy on crystals a sub-Bragg-diffraction peak can be observed. As multiple Bragg diffraction is required for photonic band gap formation, hence sub-Bragg diffraction can affect band gap formation [14]. Indeed, the sub-Bragg-diffraction condition is part of the 2D TE-band gap in triangular lattices [9]. For elastic wave diffraction a propagation gap is formed at the sub-Bragg condition and therefore also for phonons and for relativistic electrons, such as the case of graphene, which has a triangular lattice.

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