

X-Ray Resonance in Crystal Cavities: Realization of Fabry-Perot Resonator for Hard X Rays

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X-ray back diffraction from monolithic two silicon crystal plates of 25–150 μm thickness and a 40–150 μm gap using synchrotron radiation of energy resolution $\Delta E = 0.36$ meV at 14.4388 keV clearly show resonance fringes inside the energy gap and the total-reflection range for the (12 4 0) reflection. This cavity resonance results from the coherent interaction between the x-ray wave fields generated by the two plates with a gap smaller than the x-ray coherence length. This finding opens up new opportunities for high-resolution and phase-contrast x-ray studies, and may lead to new developments in x-ray optics.

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Lasers of long wavelength, ranging from visible spectra to soft x rays, have had a great impact on the development of sciences and led to diverse applications in physics, chemistry, biology, material sciences, engineering, etc. In the case of shorter wavelength hard x rays, x-ray lasers, including free-electron lasers, have long been anticipated to provide a coherent x-ray source for probing structures of matter, periodic and nonperiodic, in subatomic scales, including the structures of nanoparticles, quantum dots, and single biological molecules. One of the components for making lasers is the optical resonator, such as the Fabry-Perot resonators [1–3]. However, an x-ray resonator for x-ray lasers has never been realized, although it has long been proposed [4–6] and pursued [7–14] from time to time for more than three decades. In fact, the difficulty in realizing x-ray resonators with observable resonance fringes arises mainly from the required experimental conditions on coherence being not easily attainable, aside from the many well documented theoretical studies of x-ray cavity resonance [3–12] reported in the literature. Recent experiments of Liss *et al.* [13] and Shvyd'ko *et al.* [14] have observed storage of x-ray photons in a few tens of back-and-forth reflection cycles in two-plate crystals with continuously decaying reflectivities in time-resolved experiments. The latter have also shown the beating of two Pendellösung fringes [15] on the tails of diffraction profiles in photon energy scans, mainly due to crystal thickness effects. Although these results are useful for x-ray optics and time-resolved experiments, they do not show observable cavity resonance fringes, mainly because the required coherence conditions were not satisfied and the insufficient energy resolution used washed out resonance fringes (see later discussion).

An unambiguous way to demonstrate the cavity resonance is, under appropriate coherent conditions, to show resonance interference fringes inside the total-reflection range and inside the energy gap of a back diffraction [7,11,12]. Moreover, images of concentric interference

rings are expected to be seen in an x-ray resonator as in an optical Fabry-Perot resonator [1]. For an optical Fabry-Perot resonator, the expected resonance interference fringes in an energy scan should show well resolved fringes with a separation $E_d = hc/(2d)$, the so-called free spectral range [1,2], between fringes, where h , c , and d are the Planck constant, the speed of light, and the effective gap between the two mirrors. If the spectral width of the fringe is Γ and the energy resolution of the incident photon beam is ΔE , the required experimental criteria in energy scans for observing well resolved resonance fringes are as follows: (a) $\Delta E < E_d$, (b) $\Delta E < \Gamma$, and (c) $\Gamma \leq E_d$. By taking the reciprocal of (a) and the uncertainty principle $(\Delta t)(\Delta E) \cong \hbar$, the required criterion (d) for time scans is $\Delta t (= \hbar/\Delta E) > t_f/2\pi (= \hbar/E_d = 2d/c/2\pi)$, where Δt is the coherent time and t_f the circulation time of photons between the two mirrors. That means, within the Δt of the incident beam, photons in the cavity can interact coherently. From criterion (d) and the definition of longitudinal coherence length $l_L = \langle \lambda^2/\Delta\lambda \rangle = \langle \lambda/(\Delta E/E) \rangle$, it is easy to show that $l_L > 2d$ [criterion (e)], where $\langle \dots \rangle$ means “average.” For a normal distribution the average equals to 1/2. From criteria (a)–(e) and the relations between (a) and (d) and (e), $\Delta E/E$ and d are evidently the key parameters, relating to the temporal coherence, which determine whether resonance fringes can be observed. For an x-ray cavity, shown schematically in Fig. 1, the effective distance $d = d_g + t$, where d_g is the gap and t the thickness of the crystal plate.

Based on this consideration, we combine a small gap cavity of 40–150 μm prepared by the microelectronic technique and an energy resolution of $\Delta E = 0.36$ meV at 14.4388 keV of x-ray synchrotron radiation to fulfill the required coherence conditions for cavity resonance and succeed for the first time in observing resonance fringes inside the energy gap in energy scans and inside the total-reflection range of angle scans near or at the (12 4 0) reflection position for silicon crystal cavities, thus effec-

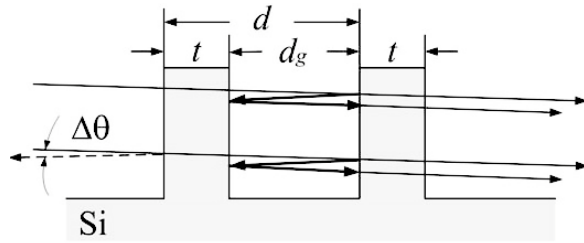


FIG. 1. Schematic of resonance interference in a Fabry-Perot-type two-plate cavity of silicon for an incident x-ray beam deviated $\Delta\theta$ from the Bragg angle ($=90^\circ$) for back diffraction. Multiple reflections take place within the crystal gap and generate forward-transmitted and back-reflected beams. The coherent interaction among the transmitted and the reflected beams inside the crystal plates and within the gap leads to cavity resonance.

tively realizing an x-ray resonator. Images of interference rings are also observed, which again confirm the x-ray resonance in crystal cavity for hard x rays and near γ rays. The experimental results are reported in this Letter.

An x-ray Fabry-Perot resonator consists, in principle, of two crystal plates as reflecting mirrors. Cavity resonance occurs when an incident x-ray beam is reflected back and forth coherently between the two plates (Fig. 1), thus generating interference fringes. Each reflection is a back diffraction from a set of atomic planes with a Bragg angle very close to 90° . In this study, several two-plate and multiplate crystal cavities with a plate thickness t ranging from 25 to 150 μm and a gap of 40–150 μm between them were prepared from a 4 in. Si (001) crystal wafer by using the microelectronic lithography process. The patterned photoresist and mask were made on the wafer surface according to the width and thickness of the crystal plate and the gap d_g between adjacent crystal plates. The patterned surface was then dry etched by reactive ions and cleaned afterwards. The width w and height h of crystal plates are 800 and 200 μm (Fig. 2). Cavities with plate numbers up to 8 were manufactured. Sharp surfaces of each plate were seen from an optical microscope.

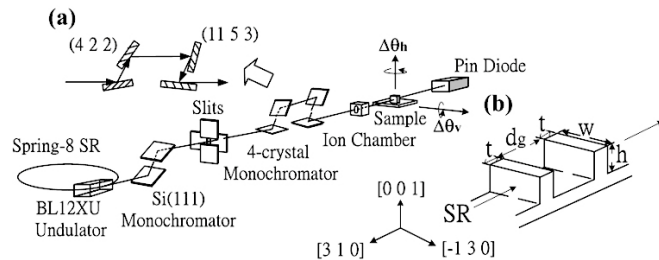


FIG. 2. Experimental setup: (a) the side view of the four-crystal high-resolution monochromator; (b) the crystallographic orientation of the two-plate cavity. $\Delta\theta_v$ and $\Delta\theta_h$ are the vertical and horizontal tilting angles of the cavity crystal. The transmitted diffracted and the back-reflected beams are monitored by the pin diode and the ion chamber, respectively. SR stands for storage ring.

The experiment was carried out at the Taiwan undulator beam line BL12XU at the SPring-8 synchrotron radiation facility in Japan. The storage ring was operating at 8 GeV and 100 mA. As shown in Fig. 2, the incident radiation was monochromatized first by a Si (111) double crystal and then by a four-crystal ultrahigh resolution monochromator for the energy $E = 14.4388$ keV. The four-crystal monochromator consisted of two pairs of (422) and (11 5 3) asymmetric reflections in a (+ - - +) geometry similar to the reported arrangement [16]. This monochromator yields the energy resolution $\Delta E/E = 2.5 \times 10^{-8}$ at 14.4388 keV, i.e., $\Delta E = 0.36$ meV, such that the longitudinal coherent length, $l_L = 1717$ μm , is much longer than the crystal gap (100 μm). Criterion (e) for observing cavity resonance is satisfied. Energy scans were performed by tuning together the Bragg angles of the third and fourth crystals with a minimum step of 0.005 arc sec, equivalent to 58.548 μeV in energy. The crystal cavity with the [001] direction along the vertical direction (Fig. 2) was placed on a goniometric head at the center of a Huber 8-circle diffractometer 1.5 m away from the last (11 5 3) crystal. The incident beam was in the $[\bar{3} \bar{1} 0]$ direction normal to the large face of the crystal plates. The beam size, limited by the slits and monochromators, was 0.05 mm vertical and 0.2 mm horizontal at the crystal cavity. The rotations $\Delta\theta_h$ and $\Delta\theta_v$ of the crystal cavity around [001] and $[\bar{1}30]$, respectively (see Fig. 2), with a minimum step of 0.0005°, were performed with the diffractometer, and the sample height was adjusted such that the incident beam hit only the cavity.

The (12 4 0) reflection was chosen as the back diffraction for $E = 14.4388$ keV. Both the forward-transmitted (000) and the back-reflected (12 4 0) beams were monitored by a pin-diode detector and an ion chamber, respectively (Fig. 2). Because of the high energy x ray used and the symmetry of the diamond structure, 24 reciprocal lattice points (r.l.p.), including those of (12 4 0) and (000) reflections, lie on the surface of the Ewald sphere at this energy, thus generating 24 diffracted beams from the center of the Ewald sphere towards these 24 r.l.p. [17,18]. This 24-beam (simultaneous) diffraction consists of the nine coplanar diffractions indicated in the legend of Fig. 4, each of which has the same zone axis. A two-plate cavity of 70 μm thickness with a 100 μm gap was aligned for the 24-beam diffraction at 14.4388 keV by adjusting the $\Delta\theta_h$ and $\Delta\theta_v$. According to the dynamical theory of x-ray diffraction [15,18], the angular $\Delta\theta_h$ and $\Delta\theta_v$ scans of the forward-transmitted beam (000) show broad dip profiles with an averaged flat bottom [see Fig. 3(a)] at the photon energies close to the exact 14.4388 keV. Figure 3(b) shows the $\Delta\theta_v$ scan of the back-reflected (12 4 0) beam. Similar profiles were observed for $\Delta\theta_h$ scans (not shown). The region of broad width is the total-reflection region, which corresponds to the energy gap in energy scans through the relation $\Delta E/E = \Delta\theta^2/2$, where $\Delta\theta = \sqrt{\Delta\theta_h^2 + \Delta\theta_v^2}$ and

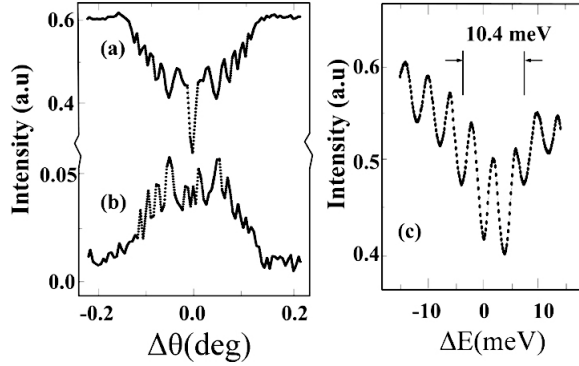


FIG. 3. $\Delta\theta_v$ scans of (a) the forward-transmitted (000) beam and (b) the back-reflected (12 4 0) beam of the two-plate cavity at $\Delta E = 9$ meV and $0.002^\circ/\text{step}$. (c) The energy E scan. The rather noisy profile of panel (b) is due to the contribution of the incident beam and the poor resolution of the ion chamber. The crystal cavity was kept at room temperature with fluctuation less than 0.1°C .

$\Delta E = E - 14.4388$ keV. The minimum intensity in the middle of Fig. 3(a) was due to the 24-beam diffraction. Distinct interference fringes due to cavity resonance are clearly seen in the transmitted (000) beam [Fig. 3(a)] and the back-reflected (12 4 0) beam [Fig. 3(b)]. Figure 4(a) shows a three-dimensional intensity plot of a $\Delta\theta_h - \Delta\theta_v$ mesh scan of the transmitted beam, and Fig. 4(b) is the two-dimensional projection. The three-dimensional plot revealing the interference intensity distribution is like a Roman amphitheater [Fig. 4(a)]. The two-dimensional fringes show concentric rings, i.e., $\Delta\theta = \text{const}$, of alternating maxima and minima and the straight lines are due to the coplanar diffractions mentioned. The calculated 2D image, Fig. 4(c), agrees qualitatively with Fig. 4(b). At $\Delta\theta_v = 0.03^\circ$ and $\Delta\theta_h = -0.02^\circ$, slightly off the normal incidence position ($\Delta\theta_v = 0^\circ$, $\Delta\theta_h = 0^\circ$), an energy scan was performed. The obtained profile near the energy gap of 10.4 meV is shown in Fig. 3(c). The gap corresponds to the angular width approximately equal to $3.6\sqrt{\chi}$ for the exact (12 4 0) back diffraction in angle scans for this crystal thickness, $\chi \sim 0.4 \times 10^{-6}$ being the electric susceptibility of (12 4 0). Again, interference fringes due to cavity were observed. The fringe spacing, namely, the E_d , is about 3.60 meV, in agreement with the calculated value 3.65 meV from the relation $E_d = hc/2d$. This is also consistent with the calculations based on the dynamical theory [11,12,15,18]. Since $\Delta E = 0.36$ meV is smaller than $E_d = 3.60$ meV, criterion (a) is fulfilled. The effective resonance distance $d = d_g + t$ can be considered as the distance between the two electric fields, the x-ray wave fields, generated by each crystal plate in one back-and-forth reflection period. Also, $2(d_g + tn_x) \approx 2dn_x \approx 2d$ is the optical path length of x rays which gives a phase shift of 2π , n_x being the x-ray index of refraction and $n_x = 1 - \delta$ with $\delta = 2.3 \times 10^{-6}$. The spectral width $\Gamma = 1.60$ meV

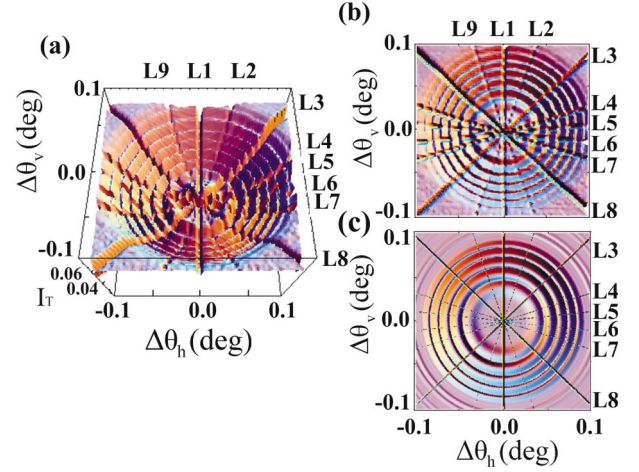


FIG. 4 (color). Intensity distributions of resonance interference at $\Delta E = 12$ meV: (a) angular ($\Delta\theta_h$, $\Delta\theta_v$) distribution of the transmitted (000) intensity I_T of the two-plate crystal cavity in a linear scale; (b) two-dimensional contour map of panel (a); (c) calculated map of (b) without angle integration. Multiple-beam interaction generates additional radial intensity lines for nine coplanar diffractions. Lines L1–L9 are related to the nine coplanar diffractions C1–C9, where C1: (040), (440), (480), (840), (880), and (12 0 0) reflections; C2: ($6\bar{4}2$) and (682) reflections; C3: (022) and ($12\bar{2}\bar{2}$) reflections; C4: (606) and (646) reflections; C5: (426) and ($82\bar{6}$) reflections; C6: ($42\bar{6}$) and (826) reflections; C7: (606) and (646) reflections; C8: (022) and ($12\bar{2}\bar{2}$) reflections; C9: (642) and (682) reflections. The (000) and (12 4 0) reflections are omitted in each of the coplanar diffractions listed (step width in $\Delta\theta_h$ and $\Delta\theta_v$: 0.003° ; counting time is 1 s).

of fringes gives the finesse [1,2,12] $F = E_d/\Gamma = 2.3$, which is less than the designed value $F = 4.0$ due to the real experimental situation, such as crystal surface roughness and inclination, and small lattice distortion due to the sample treatment and temperature effect. Various $\Delta\theta_h$, $\Delta\theta_v$, and energy scans were carried out for other two-plate and eight-plate cavities and similar interference fringes were observed. Here $\Delta E = 0.36$ meV $< \Gamma = 1.60$ meV, and $\Gamma = 1.60$ meV $\leq E_d = 3.60$ meV, so criteria (b) and (c) are also fulfilled.

The time t_f for x-ray photons traveling between the two crystal plates of 100 μm gap is 0.67 ps, much shorter than the time $\Delta t (= 1.8$ ps). Namely, $\Delta t > t_f/2\pi (= 0.1$ ps). Therefore, criterion (d) is satisfied. Moreover, as already stated, $l_L > 2d$, the longitudinal (temporal) coherence is maintained. The transverse coherence determined by the photon emittances is also retained for x rays very close to normal incidence. Hence both the spatial and the temporal coherence of the x-ray beams guarantee the realization of x-ray cavity resonance. In contrast, the experimental conditions of the previous work [14], $\Delta E (= 2$ meV) $> E_d (= 12.4$ $\mu\text{eV})$, $\Delta E (= 2$ meV) $> \Gamma (= 0.64$ $\mu\text{eV})$, $\Delta t (= 0.33$ ps) $< t_f/2\pi (= 53$ ps), and $l_L (= 0.31$ mm) $< 2d (= 100$ mm), do not fulfill the required criteria (a),

(b), (d), and (e). Similarly, these required conditions were also not satisfied by the work of [13]. Under these unfavorable circumstances, the expected resonance fringes spaced in $12.4 \mu\text{eV}$ intervals would certainly be smeared out by the energy resolution of 2 meV [14].

For different photon energies, different back diffractions together with different multiple reflections need to be employed. The presence of multiple diffraction is not a problem. Slight angular deviation of the crystal cavity from the multiple-beam positions can always be achieved without losing the resonance condition. Since the fixed phase relation between the forward-transmitted and the back-reflected beams and the narrow energy and angular widths of x rays from the cavity, this crystal cavity with a better finesse (i.e., thicker crystal plates with smaller surface roughness) can be used for phase-contrast and high-resolution x-ray optics, such as high-resolution monochromator using the back reflection and narrow-band filter with the transmission [7,11,13,14]. All of these can be applied for high-resolution x-ray scattering, spectroscopy, and phase-contrast microscopy in many physical, chemical, and biological studies, such as the investigation of the dynamics of solids, liquids, and biomolecules, precise measurements of wavelength and lattice constant, etc. [7,11,13,14,17,19]. Furthermore, improved crystal cavities of the present type with better finesse might be useful for the development toward hard x-ray (or γ -ray) lasers, if suitable lasing materials [20] could be developed.

In conclusion, we have successfully observed cavity resonance fringes in silicon crystal cavities and realized for the first time Fabry-Perot resonators for hard x rays (near γ rays). The required conditions for cavity resonance in normal incidence geometry are the criteria (a)–(e), with which the longitudinal coherence is retained for x-ray photons in the cavity. The quasicohherent and highly monochromatic x rays generated from the resonator provide new opportunities for x-ray optics, spectroscopy, and microscopy.

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