Time-delayed beam splitting with energy separation of x-ray channels

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We introduce a time-delayed beam splitting method based on the energy separation of x-ray photon beams. It is implemented and theoretically substantiated on an example of an x-ray optical scheme similar to that of the classical Michelson interferometer. The splitter/mixer uses Bragg-case diffraction from a thin diamond crystal. Another two diamond crystals are used as back-reflectors. For energy separation the back-reflectors are set at slightly different temperatures and angular deviations from exact backscattering. Because of energy separation and a minimal number (three) of optical elements, the split-delay line has high efficiency and is simple to operate. Due to the high transparency of diamond crystal, the split-delay line can be used in a beam sharing mode at x-ray free-electron laser facilities. The delay line can be made more compact by adding a fourth crystal.

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Complex nanoscale dynamics in condensed matter can be studied in a broad dynamic range by x-ray photon correlation spectroscopy (XPCS) using coherent x-rays from x-ray free-electron lasers (XFELs) [1]. The split-pulse technique is one of the promising approaches to access dynamics from femtosecond to nanosecond regimes. In this technique, each x-ray pulse is split into reference and delayed pulses of equal intensity, arriving at the sample separated in time. The scattering from the reference and delayed pulses is then collected during the same exposure of an area detector.

The principle optical scheme of the split-pulse technique was discussed in [1–3]. The main components are a splitter and mixer crystals in Bragg diffraction, and two additional Bragg reflecting crystals guiding the delayed pulse and controlling the delay. To ensure very short (femtosecond length) and even negative delays, an advanced scheme was proposed and realized [4–6]. It has additional four crystals (total eight) designed to guide the reference pulse. The large number of optical elements inevitably complicates alignment and operations, and also compromises the split-delay line efficiency.

Here we introduce and study theoretically the performance of a split-delay x-ray scheme with the minimal amount of crystals, three, as shown in Fig. 1. The scheme resembles the classical Michelson interferometer [24]. Crystal C₀ plays the roles of both the splitter and the mixer. Crystals C₂ and C₁ are set to reflect x-rays in almost exact Bragg backscattering geometry, and guide the reference (red) and delayed (blue) pulses to sample S through C₀, or by reflecting from C₀, respectively. The delay τ = 2L/c is varied by changing the crystal C₂ spatial position along the beam, i.e., a delay path L.

The central problem in the design of a split-delay line is how to split the incident beam and how to bring together the reference and delayed beams on the sample. In the traditional scheme, which we term as single-energy splitting (SES), photons of the same energy are divided between the reference and delayed beams. Equal intensity splitting can be realized by using Bragg diffraction

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FIG. 1: Optical scheme of a three-crystal C₀, C₁, C₂, split-delay line in a Michelson-interferometer-type configuration using dual-energy splitting (see text for details). The reference (red) and delayed (blue) beams are shifted parallel to each other by δx, provided the angular deviations from exact backscattering δθ₁ = δθ₂. The parallel beams are focused on sample S by focusing system F. The shift δx, see Eq. (1), is due to a finite thickness d of crystal C₀, and a delay path length L determining the nonzero x-ray pulse delay τ = 2L/c.
The splitter/mixer crystal $C_0$ reflects x-ray photons in an energy bandwidth $ΔE_0$ into the delayed pulse channel, shown in blue in Fig. 1. The reflectivity is close to 100% if the crystal thickness $d \gg Λ$, see Fig. 2(a). The incidence angle $θ_0$ is such that the central photon energy $E_0$ of the spectral distribution coincides with the central energy $E_0^c$ of the reflection bandwidth from crystal $C_2$ set into backscattering, see Fig. 2(b). The bandwidth $ΔE_2$ of crystal $C_2$ is chosen to be close to the bandwidth $ΔE_0^c$ of crystal $C_0$. The x-rays back-reflected from $C_2$ are transmitted through $C_0$ and steered onto the sample $S$, provided the angular offset $δθ_2$ is larger than the angular width $Δθ_0 = (ΔE_0/E_0) \tan θ_0$ of the Bragg reflection from $C_0$. This can be easily realized, since the angular width of back-reflection $Δθ_2 = 2 \sqrt{ΔE_2/E_2}$ is much larger.

X-ray photons with energies outside the energy bandwidth $ΔE_0$ are transmitted through $C_0$, see Fig. 2(d), and guided into the reference pulse channel by back reflection from $C_1$, as shown in red in Fig. 1. Crystal $C_1$ is equivalent to $C_2$. However, it is maintained at a different temperature $T_1 = T_2 + δT_1$ to reflect x-rays transmitted through $C_0$ in a bandwidth centered at $E_1$, which is shifted from $E_0$ by $δE_{01} = E_0 - E_1 > (ΔE_0 + ΔE_1)/2$, see Fig. 2(e). Photons back-reflected from $C_1$ will be reflected from $C_0$, see Fig. 2(f), and directed onto the sample, provided the angle of incidence to $C_0$ is $θ_0 + δθ_1$, where according to Bragg’s law $δθ_1 = (δE_{01}/E_0) \tan θ_0$. The same angular deviation $δθ_1$ is required in backscattering from $C_1$, see Fig. 1.

If $θ_0 = δθ_2$, the two beams propagate to the sample parallel to each other with a small offset

$$δx = 2d_0 \cos θ_0 - L \deltaθ_1. \tag{1}$$

For $d_0 = 50 \, \mu m$ the first term in Eq. (1) is about 60 $\mu m$, while the second term varies from zero for $L = 0$ to $-30 \, \mu m$ for $L = 1.5 \, m$. As a result, $δx$ varies from 60 $\mu m$ to 30 $\mu m$, respectively. Given, a usual XFEL beam size of $\approx 300 - 700 \, \mu m$, such shift is insignificant. Despite the shift, focusing system F brings all the photons to the same point on the sample.

Spectral dependencies of reflection from and transmission through each crystal optical element of the split-delay line are presented in Fig. 2 numerically calculated according to 14 using equations of the dynamical theory of x-ray diffraction. Spectral distribution of x-rays arriving on the sample in the reference and delayed channels are shown in Fig. 3(a) by the red and blue lines, respectively. The calculations are performed assuming that the XFEL is working in a self-seeding mode providing x-rays in a 400-meV bandwidth (black line) 12, and with an angular spread of 2.5 $μrad$ (FWHM).

The calculations presented in Figs. 2 and 3 are performed with crystal parameters given in Table I for device 1. The choice of the crystals is not unique. In this particular example, diamond crystals are chosen for all three optical elements for several reasons. First, the photoabsorption in diamond is much less than in Si, and therefore the efficiency of diamond optics is higher. Second, for the same photon energy, the spectral bandwidth of

FIG. 2: Spectral dependencies of reflection ($R$) from and transmission ($T$) through individual crystals of the split-delay line, calculated in the framework of the dynamical theory of x-ray diffraction, with crystal parameters of device 1, given in Table I.
TABLE I: Crystal elements $C_n$ ($n = 0, 1, 2$) of the split-delay lines schematically presented in Fig. 1 and $C_n$ ($n = 0, 1, 2, 3$) of the split-delay lines schematically presented in Fig. 4. Crystal, Bragg reflection parameters, and incident radiation polarization states are given as used in all dynamical theory calculations: $(hkl)$ - Miller indices of Bragg reflections; $T_n$ - crystal temperature, $d_n$ - crystal thickness, $\theta_n$ - glancing angle of incidence; $E_n$ - photon energy at the reflection curve center; $\Delta E_n$ and $\Delta \theta_n$ are Bragg’s reflection spectral width, and angular acceptance, respectively. $\varepsilon_n$ and $\varepsilon_n^{(abs)}$ are the relative and absolute efficiencies of respective channels.

| device | polarization | $C_n$ | material | $(hkl)$ | $T_n$ [K] | $E_0$ [keV] | $\Delta E_n$ [meV] | $\theta_n$ | $\Delta \theta_n$ [\mu rad] | $d_n$ [\mu m] | $\varepsilon_n$ | $\varepsilon_n^{(abs)}$ |
|--------|--------------|-------|----------|---------|-----------|----------------|----------------|----------|----------------|---------------|-------------|-----------------|
| 1      | $\sigma$     | $C_0$ | diamond  | (004)   | 300       | 8.51389       | 72             | 54.7359° | 12             | 50            | –           | –               |
|        |              | $C_1$ | diamond  | (224)   | $T_2 + \delta T_{12}$ | $E_0 - \delta E_{01}$ | 44             | $90^\circ - \delta \theta_2/2$ | $\delta \theta_1 = 20$ \mu rad | 3675        | 50           | 94.9            |
|        |              | $C_2$ | diamond  | (224)   | 300       | $E_0$      | 44             | $90^\circ - \delta \theta_2/2$ | $\delta \theta_1 = 0$ \mu rad | 3675        | 50           | 93.8            |
| 2      | $\pi$        | $C_0$ | diamond  | (220)   | 300       | 8.51389       | 66             | 35.2641° | 5.5            | 50            | –           | –               |
|        |              | $C_1$ | diamond  | (224)   | $T_2 + \delta T_{12}$ | $E_0 - \delta E_{01}$ | 44             | $90^\circ - \delta \theta_2/2$ | $\delta \theta_1 = 10$ \mu rad | 3675        | 50           | 85.6            |
|        |              | $C_2$ | diamond  | (224)   | 300       | $E_0$      | 44             | $90^\circ - \delta \theta_2/2$ | $\delta \theta_1 = 0$ \mu rad | 3675        | 50           | 84.5            |

FIG. 3: Spectral distribution of x-rays arriving on the sample in the reference (red line) and delayed (blue line) channels, calculated using the dynamical theory of x-ray diffraction, with crystal parameters of device 1, given in Table 1. Black line shows the spectrum of incident x-rays from a seeded XFEL. The magenta line shows the spectral distribution of the photons, transmitted through crystals $C_0$ and $C_2$. (a) Calculations for the dual-energy splitting case. (b)-(c) Calculations under similar conditions, but with $C_0$ functioning in the single-energy splitting mode in Bragg-case and Laue-case diffractions, respectively.

Bragg back-reflections from diamond crystals is larger, due to larger Debye-Waller factors (larger Debye temperature) [13][14]. We have chosen Bragg reflections with the largest bandwidth and therefore with the highest efficiency, applicable in a comfortable for XPCS experiments photon range of 8-9 keV. Given the very recent advancement in fabrication of high-quality diamond crystals and their use in high-resolution, low-loss x-ray optics [12][14][17], the proposed configuration with diamond crystal elements is deemed to be feasible.

Bragg back-reflection is very often accompanied by parasitic Bragg reflections [10][15][20], which may waste a significant number of useful photons and reduce the optics efficiency. The choice of back-reflection was actually dictated also by the requirement of the minimal amount of the parasitic reflections. In particular, the 422 back-reflection from $C_1$ and $C_2$ diamond crystals considered here is accompanied only by one pair of the parasitic Bragg reflections: 400 and 022. It is easy to suppress them. To do this, the crystal plane containing the (422), (400), and (022) reciprocal vectors has to be inclined by $\pm 100$ \mu rad to the 422 Bragg diffraction plane.

Efficiency is a figure of merit for a split-delay line. Relative spectral efficiencies for the reference and delayed channels are very high: $\varepsilon_1 \simeq 94\%$ and $\varepsilon_2 \simeq 93\%$, respectively. The relative spectral efficiency is defined as the number of photons in the relevant channel normalized to the number of incident photons within the channel bandwidth. Absolute spectral efficiencies are calculated by normalization to the total number of incident photons. For the 400-meV bandwidth of the XFEL radiation, they are equal to $\varepsilon_1^{(abs)} = 9.9\%$ and $\varepsilon_2^{(abs)} = 9.8\%$, respectively. The total absolute efficiency of the scheme is about 20\%.

The rest of the photons, $\simeq 60\%$, transmitted through both $C_0$ and $C_1$ can be utilized by the downstream experiment. Therefore, such a split-delay line can be used in a beam sharing mode at XFEL facilities.

Figures 3(b) and 3(c) show for comparison the results...
FIG. 4: By adding crystal C₂ with \( \theta₃ = \pi/2 - \theta₀ \), the delay line in Fig. 1 becomes more compact, with reference and delayed pulses propagating almost parallel to the incident beam.

of calculations of the spectral distributions for the same scheme, but, with C₀ functioning in single-energy splitting mode in Bragg-case (b) or Laue-case (c) diffraction, with \( d₀ = 6.4 \, \mu m \) and \( d₃ = 52.4 \, \mu m \), respectively. Single-energy splitting requires that \( \delta \theta₇ = \delta \theta₅ = 0 \). The values of efficiencies indicated in Fig. 3 show that the dual-energy splitting scheme is about three to four times more efficient than the single-energy splitting schemes.

If the vertical configuration is inconvenient, the same scheme could be used in the horizontal scattering geometry. In this case the incident radiation is in the \( \pi \)-polarization state. Device 2 in Table I presents an example of the split-delay line functioning in horizontal scattering plane and having very similar performance in terms of efficiency and other parameters.

The scheme could be made more compact, with the beams propagating to the sample parallel to the incident beam. This is achieved by additional Bragg reflection from an additional crystal C₃ with Bragg angle \( \theta₃ = \pi/2 - \theta₁ \), as show in Fig. 4. The efficiency of the four-crystal scheme is almost the same as that of the three-crystal scheme, provided a high-reflectivity diamond crystal C₃ is used, as suggested in Table I.

The split-delay lines presented here are also applicable at synchrotron radiation facilities. The larger angular divergence of the incident beam \( \approx 10 - 15 \, \mu rad \), however, result in a \( \approx 15 - 30\% \) reduction of the efficiency for the three-crystal scheme and a \( \approx 20 - 35\% \) reduction of the efficiency for the four-crystal scheme.

In conclusion, the three-crystal split-delay x-ray scheme for XPCS applications is introduced and studied theoretically. Application of the dual-energy splitting significantly increases the efficiency of the optical scheme. Due to the high transparency of diamond crystal, the split-delay line of such design can be used at XFEL facilities in the beam sharing mode. A four-crystal modification makes the scheme in-line and more convenient for XPCS experiments.

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[25] While working on the manuscript, it came to our attention that a similar idea of energy separation of the x-ray beams has been discussed in [7] for a very thin splitter without performing theoretical analysis or experiments.