



# Simultaneous lasing of Ni $K\alpha$ and Cu $K\alpha$ lasers from an alloy foil irradiated with an intense X-ray free-electron laser pulse

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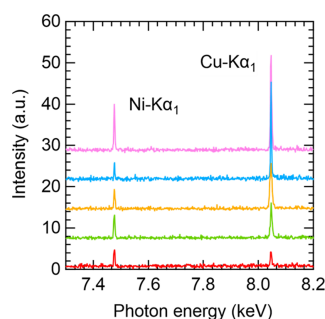
**Keywords:** X-ray free-electron laser;  $K\alpha$  laser; single-shot X-ray spectroscopy.

We report the simultaneous lasing of two distinct  $K\alpha$  emissions at photon energies of 7.48 keV (Ni  $K\alpha_1$ ) and 8.05 keV (Cu  $K\alpha_1$ ). This was achieved by a population inversion induced through intense X-ray free-electron laser (XFEL) irradiation of a Cu–Ni alloy foil. This demonstration of multi-color X-ray lasing using a single XFEL source is expected to contribute significantly to the future development of X-ray lasers and their applications.

## 1. Introduction

Brilliant femtosecond X-ray free-electron laser (XFEL) pulses (Emma *et al.*, 2010; Ishikawa *et al.*, 2012) have played an important role in opening up new fields of X-ray science (Chapman *et al.*, 2011; Suga *et al.*, 2015; Kim *et al.*, 2020). Lasing via amplified spontaneous emission (ASE) of  $K\alpha$  emission has been successfully achieved by generating population inversion through high-intensity XFEL irradiation of various materials (Rohringer *et al.*, 2012; Yoneda *et al.*, 2015; Kroll *et al.*, 2018; Kroll *et al.*, 2020; Zhang *et al.*, 2022; Doyle *et al.*, 2023; Linker *et al.*, 2025). If ASE lasing at different wavelengths can be simultaneously induced using a single XFEL pulse, multi-color X-ray lasers could be realized. Such a capability is expected to greatly advance the development of X-ray laser systems and enable novel applications, including laser oscillators (Halavanau *et al.*, 2020) on multi-color X-rays and nonlinear X-ray spectroscopic techniques (Tanaka & Mukamel, 2002; Sun *et al.*, 2010), which require synchronized multi-color X-ray sources.

When a material is irradiated with X-rays, various characteristic emissions such as  $K\alpha_1$ ,  $K\alpha_2$  and  $K\beta$  lines are simultaneously generated. In spontaneous emission, the intensity of  $K\alpha_1$  is typically about twice that of  $K\alpha_2$ . However, in the case of stimulated emission, such as in ASE X-ray lasers, the transition from the  $2p_{3/2}$  to the  $1s$  state (which has a significantly larger stimulated emission cross section than the  $2p_{1/2}$  to  $1s$  transition responsible for  $K\alpha_2$ ) dominates. As a result, only  $K\alpha_1$  typically undergoes lasing. When excited by extremely intense X-rays,  $K\alpha_2$  lasing may occur very weakly like a satellite of  $K\alpha_1$  (Yoneda *et al.*, 2015). Similarly,  $K\beta$  ASE lasing is nearly impossible due to strong competition with  $K\alpha$  transitions. These constraints make the generation of multi-color ASE X-ray lasers from a single-element sample extremely challenging. To overcome this, we propose using a sample composed of different atomic species to achieve



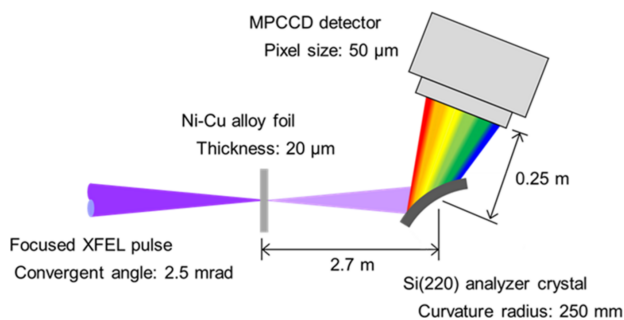
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simultaneous multi-color ASE lasing. Since ASE lasing occurs independently in each atomic species, simultaneous oscillation of distinct  $K\alpha_1$  lines becomes possible. In this study, we report the simultaneous generation of 7.48 keV Ni  $K\alpha_1$  and 8.05 keV Cu  $K\alpha_1$  ASE lasers using a foil of Ni–Cu alloy.

## 2. Experiment and discussion

The experiment was conducted at SACLA BL2 EH3. A schematic of the experimental setup is shown in Fig. 1. XFEL pulses with a photon energy of 10 keV were focused using Kirkpatrick–Baez (KB) optics (Inubushi *et al.*, 2025). The beam sizes were measured to be 150 nm (horizontal) and 220 nm (vertical) in full width at half-maximum (FWHM). Typically, a pulse energy of 110  $\mu\text{J}$  was achieved at the focal point. The pulse duration was 7 fs (Inubushi *et al.*, 2012; Inubushi *et al.*, 2017), resulting in an intensity of  $2.5 \times 10^{19} \text{ W cm}^{-2}$ . The XFEL pulses were focused onto a 20  $\mu\text{m}$ -thick constantan foil, an alloy composed of 45% Ni and 55% Cu. Given that the  $K$ -absorption edges of Ni and Cu are 8.33 keV and 8.98 keV, respectively, the 10 keV XFEL pulses were capable of ionizing the  $K$ -shell electrons of both elements. The constantan foil, placed in air, was moved after each shot to ensure irradiation on a fresh surface. The XFEL intensity was



**Figure 1**

Schematic of the experimental setup. Focused 10 keV XFEL pulses irradiated a Ni–Cu alloy (constantan) foil. The resulting  $K\alpha_1$  lasers were simultaneously measured in a single-shot manner using a dispersive spectrometer consisting of a convex bent Si(220) analyzer crystal and an MPCCD detector. The central photon energy for the observation was set to 7.8 keV, corresponding to a Bragg angle of  $24.5^\circ$ . An energy observation range of 1.2 keV was achieved, with an energy resolution of 1.5 eV per pixel.

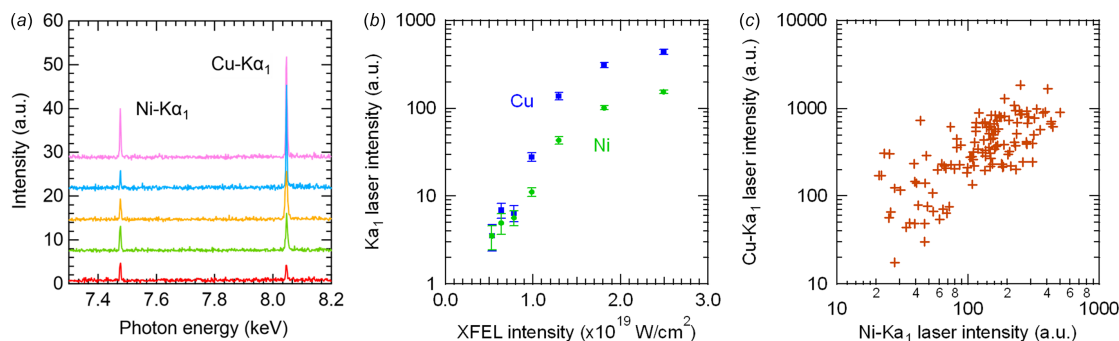
varied by adjusting the position of the foil along the beam path. In this context, the position where the maximum XFEL intensity was achieved is referred to as the ‘optimal position’.

To verify the simultaneous lasing of two  $K\alpha_1$  lines, a single-shot measurement with an energy observation range exceeding 600 eV is necessary, as the photon energies of Ni  $K\alpha_1$  and Cu  $K\alpha_1$  emissions are 7.48 keV and 8.05 keV, respectively. Here, we propose the combined use of a divergent X-ray beam and a bent crystal (Zhu *et al.*, 2012) to achieve a significantly broader energy observation range. In our experiment, a convex bent Si(220) crystal with a curvature radius of 250 mm was employed. A ray-tracing simulation was conducted to design and optimize the spectrometer configuration. Fig. 1 shows the experimental setup, including the parameters used in the ray-tracing simulation. As a result, an energy observation range of 1.2 keV was achieved. The MPCCD detector (Kameshima *et al.*, 2014) provided an energy resolution of 1.5 eV per pixel.

Fig. 2(a) shows the single-shot spectra measured at the ‘optimal position’. Distinct peaks are observed at 7.48 keV, corresponding to the Ni  $K\alpha_1$  line, and at 8.05 keV for the Cu  $K\alpha_1$  line, with variations in intensity. As illustrated in Fig. 2(b), both peaks exhibit nonlinear increases in signal intensity as a function of XFEL intensity, indicating successful simultaneous lasing of the Ni  $K\alpha_1$  and Cu  $K\alpha_1$  lines. At higher XFEL intensities, the Cu  $K\alpha_1$  laser becomes more intense than the Ni  $K\alpha_1$  laser. This is likely due to the higher Cu content in the alloy, the greater absorption coefficient of Cu for 10 keV X-rays and the higher transmittance of the Cu  $K\alpha_1$  emission through the sample. Furthermore, as shown in Fig. 2(c), a statistically significant positive correlation was observed between the Ni  $K\alpha_1$  and Cu  $K\alpha_1$  laser intensities (correlation coefficient  $r = 0.63$ ,  $p$  value  $p < 0.001$ ), indicating that the two lasing processes are not mutually competitive. This implies that increasing the XFEL beam intensity could lead to the simultaneous enhancement of both lasers, until saturation.

## 3. Summary and future perspectives

We successfully demonstrated the simultaneous generation of two  $K\alpha_1$  lasers by irradiating an alloy foil with an intense XFEL pulse. This achievement suggests the potential for producing multi-color X-ray laser pulses by incorporating



**Figure 2**

(a) Single-shot spectra observed at the optimal position. (b) Averaged intensities of the Ni  $K\alpha_1$  and Cu  $K\alpha_1$  lasers as functions of XFEL intensity. Error bars represent standard errors. (c) Correlation between the intensities of the Ni  $K\alpha_1$  and Cu  $K\alpha_1$  lasers at the optimal position.

multiple atomic species – not limited to alloys, but also including, for example, layered sample and mixed-atom droplets. Such multi-color X-ray lasers are expected to pave the way for future advancements in X-ray laser technology and enable novel applications across various scientific fields.

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