ESRF	**Experiment title:** *COHERENT TWO X-RAY PHOTON EMISSION SPECTROSCOPY*	**Experiment number**: **MI-436**
Beamline: **ID-12**	**Date of experiment**: from: 31-OCT-2001 to: 06-NOV-2001	**Date of report**: 01-MAR-2002
Shifts: **18**	**Local contact(s)**: **A. ROGALEV and F. WILHELM**	*Received at ESRF:*

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1. NON-LINEAR X-RAY EMISSION SPECTROSCOPY

Non-Linear X-ray Optics is a formidable challenge for new synchrotron radiation sources. Since the pioneering work of Eisenberger and McCall [1] who reported in 1971 the 1rst parametric *down-conversion* of 17 keV photons ***non-linearly diffracted*** by a Be single crystal, several attempts have been made, *e.g.* at the Photon Factory and at the ESRF, to reproduce such a difficult experiment with Synchrotron Radiation. Unfortunately, the count rates remain so far depressingly low (few counts per hour). The aim of our project was to explore another type of *non-linear* effect which we expected to be more favorable since it concerns non-linear Resonant Inelastic X-ray Processes. We have proposed to transpose with modern Synchrotron Radiation instrumentation the experiment of Bannet and Freund [2] who produced already in 1982 some rather convincing evidence that one could detect the coherent emission of two X-ray photons associated with the ***single electron*** 2s $\rightarrow$ 1s *monopole* transition which is strictly forbidden by selection rules in conventional X-ray emission spectroscopy. As opposed to *Second Harmonic Generation* (SHG) which involves the 2nd order electric susceptibility -and thus requires *odd* space parity-, a 2 photon coherent emission is compatible with *even* space parity because it takes its origin in the ***3rd order*** electric susceptibility which is commonly involved in *Stimulated* Raman emission at optical wavelengths.

2. TWO PHOTON COHERENT EMISSION: *1rst ENERGY RESOLVED SPECTRUM*

We have reproduced in Fig. 1 the first 2-photon coherent emission spectrum recorded at the ESRF. Note that the experiment was performed in the low-current 16-bunch mode which was not particularly favorable for this photon-hungry experiment. The sample was a single crystal of Vanadium cut parallel to the planes (111) and inclined at 45°. The marked peak matches nicely the expected V (2s $\rightarrow$ 1s : ΔE = 4838.5 eV) transition if one assumes the coherent emission of two photons of 2420 eV. The linewidth is dominated by instrumental resolution. Excitation energy was 5500 eV.

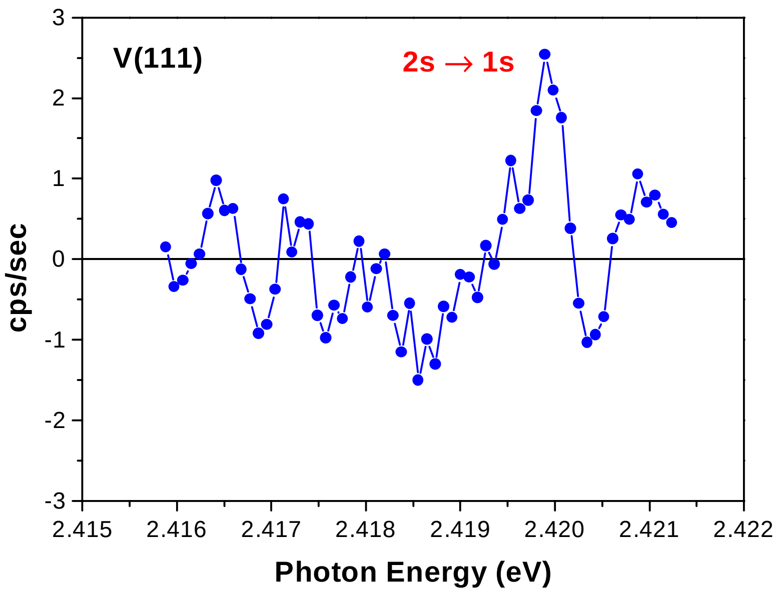

Figure 1
Two photon coherent emission spectrum of the V (2s $\rightarrow$ 1s) monopole transition. The energy resolution of the Si (111) crystal analyzer was typically 1.3 eV at a Bragg angle of 54.8°.
The detector was a standard Si photodiode operated in integrated current mode. The frequency is NOT a true event counting rate but simply the noise averaged output of a V$\rightarrow$F converter. A statistical background of 1427 c/s has been subtracted.
The spectrum displayed has been averaged over 80 scans of 60 data points. The duration of a single scan was 1800s.
The signal intensity is only 3.10^{-5} that of the Lβ_2 line of Mo at 2518.3 eV.

The emission spectrum was recorded using a new high-vacuum compatible, high resolution emission spectrometer designed by the ID12 team. Its key component is a wide angular acceptance spherically bent Si (111) crystal. The source was the new helical undulator HU-38 (Apple-type) installed on the ID12 straight section in 2001. The undulator was operated in the pure circular mode in order to minimize the emission of high energy harmonics on axis. It was our initial idea to expose the sample to the direct -fairly intense- undulator beam, *i.e.* upstream with respect to the two-crystal monochromator. Numerous practical difficulties, *e.g.* associated with the operation of the emission spectrometer in a UHV section of rather restricted access, led us to reconsider in detail the experimental requirements. Finally, the 1rst experiment was carried out under monochromatic excitation, *i.e.*in the most challenging *one photon-in / two photon-out* configuration. In this way, we could benefit of the combined rejection of harmonics by the monochromator and two additional SiC mirrors located downstream.

REFERENCES
[1]P. Eisenberger and S.L. McCall, *PRL* **26**, (1971), 684-688.
[2]Y. Bannet and I. Freund, *PRL* **49**, (1982), 539-542.