

THz induced selective catalytic CO oxidation on Ru: supplementary materials

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(a) Experimental setup

The THz beam in a quasi-half-cycle pulse was generated using Coherent Transition Radiation at the Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory[1-3]. In this process, electron bunches with a charge of 350 pC and a FWHM as short as 70 fs (or 21 μm), pass through a thin ($<10 \mu\text{m}$) beryllium (Be) foil. As the electrons approach and enter the metal surface, they produce broad band coherent transition radiation peaking at a wavelength longer than the bunch length of 21 μm (below 480 cm^{-1}). The radiation emitted is broadband, ranging from 2 to 20 THz, with a peak around 10 THz, as determined through interferometer measurements [2]. The transition radiation is emitted from the Be foil in a hollow cone with radial polarization. The THz beam operates at 120 Hz. At the focal point, the beam size is estimated to be 300-1000 μm by 300-1000 μm . The peak transverse electric field at the focus of the THz beam was estimated to be between 0.3 and 3 V/nm in a quasi-half-cycle pulse. This range was estimated using the focus size at the surface and comparing that to the peak electric field in reference [2], accounting for all intensity attenuations, such as the reflectivity loss of gold coated mirrors and the transmission loss of diamond windows.

A schematic of the overall experimental setup is shown in Fig. S1. Attached to the measurement chamber under UHV (5×10^{-10} torr) are a Hiden RGA mass spectrometer, Ne ion gun, and an X, Y, Z, Θ manipulator with a Ru(0001) crystal mounted on it. The Ru(0001) surface is in the plane of the THz focus. The spatial alignment between the THz focus and the Ru(0001) was determined to within a few hundred microns using a thermocouple . The maximum temperature rise of the thermocouple ($+ \sim 10 \text{ K}$) occurs when the spatial overlap of THz beam and the thermocouple junction is maximized.

(b) Sample preparation.

The Ru(0001) surface was cleaned using a predetermined method known to create atomically clean surface as follows: 5 minutes of Ne ion sputtering; heating the surface to 1100 K in an O_2

atmosphere of 2×10^{-7} Torr for 5 minutes; and 3 cycles of flashing the surface to ~ 1400 K in UHV to remove any O remaining on the surface. The CO/O/Ru(0001) surface[4] was prepared by introducing 10 Langmuirs of O₂ onto the surface between the temperatures of 500 and 900 K, which produces adsorbed atomic O, followed by the introduction of 10 Langmuirs of CO at 300 K.

(c) Coverage estimation

To estimate the coverage of CO and O, we recorded TPD traces of CO from the intensity of the mass 28 amu signal during a temperature ramp from 300 K to 600 K, which is sufficient to desorb all CO adsorbed on the surface[4]. The temperature ramping rate was 2.7 K/s over the temperature range of interest. We recorded TPD traces at these reaction stages: (1) without THz-irradiation of the CO/O/Ru(0001) surface, (2) after THz-irradiation of the CO/O/Ru(0001) surface, and (3) the surface after TPD measurement of (2) was re-dosed with a full coverage of CO at 300 K. The results from the RGA were corroborated with the pressure rise recorded during the TPD with an ion gauge. The CO coverage was determined by comparing the CO intensities of TPD traces (1) and (2). The O coverage was determined by comparing TPD trace (3) to the O coverage dependence of CO TPD from reference.[4]

References:

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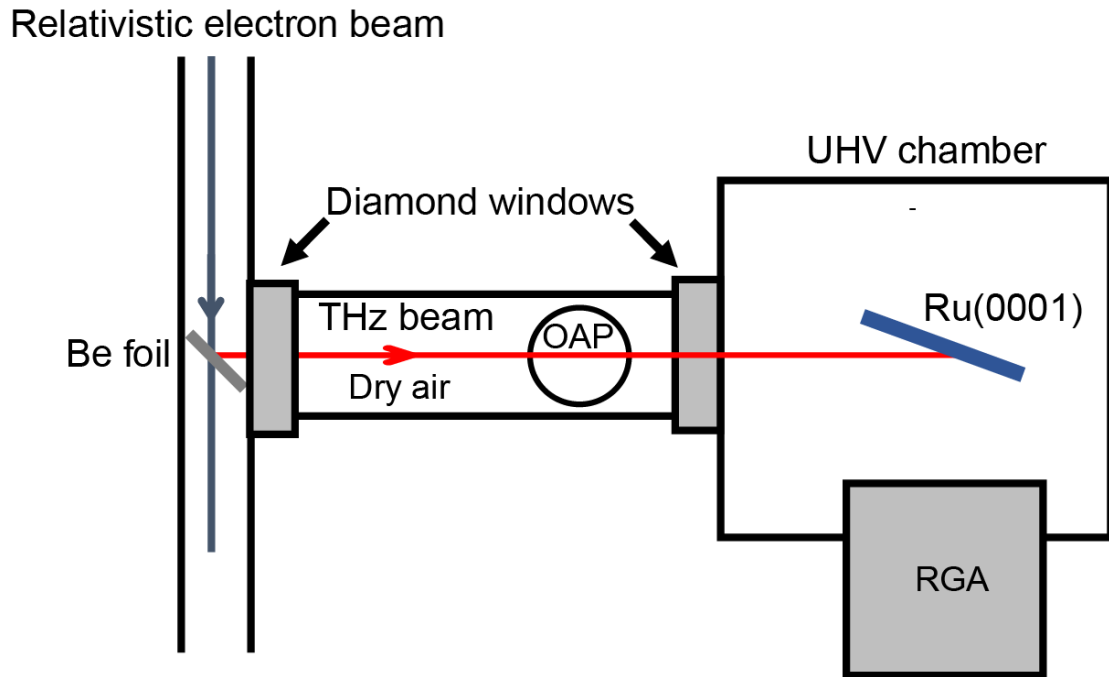


FIG. S1. Schematic of the experimental setup, showing the relativistic electron beam, THz beam and UHV chamber. The diffraction limited THz pulse is emitted from the Be foil through the transition radiation. The THz pulse is focused using an off-axis parabolic (OAP) mirror before passing through a diamond window into a UHV chamber. The THz beam path is enclosed in a dry air environment in order to minimize absorption by water.

