Femtosecond Laser Induced Emission of Coherent Terahertz Pulses from Ruthenium Thin Films

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Supporting information

Dependence of THz emission on incident laser polarization

Figure S 1(top) shows the peak-to-peak amplitude of the THz electric-field emitted by a 8 nm Ru sample as a function of the angle of incident (linear) polarization. This angle was varied by means of a half-wave plate. The measurements were carried out at normal incidence to exclude the dependence of the absorption on incident polarization at non-zero AOI. Figure S 1(bottom) shows the THz electric-field peak-to-peak amplitude as a function of the sample azimuthal orientation. These measurements were carried out in the same configuration and on the same sample as the ones reported in figure S 1(top). By comparing these two plots and the values of the fit parameter A (obtained via the procedure explained for figure 7(bottom), 0.79 ± 0.6 for figure S 1 (top), 22.7 ± 1.12 for figure S 1 (bottom)) it is clear that rotating the incident polarization has little to no effect on THz emission, while rotating the sample has an evident effect, as is also show in the main text (figure 8).
Figure S 1: (top) THz Pk-Pk amplitude as a function of the angle of the incident (linear) polarization. (bottom) THz Pk-Pk amplitude as a function of the sample azimuthal orientation.
X-Ray diffraction

To obtain information on the crystallinity of the sample, we performed 2D X-ray diffraction pattern contains information about the orientation of the crystallites because it captures different rotation angles of such crystallites. Each crystallite contributes to the diffraction intensity in one point of the surface of a cone, which is defined by the diffraction angle. If all crystallites were oriented in the same way with respect to the substrate, we would obtain a diffraction pattern consisting of spots. Only if there is a very large number of randomly oriented crystallites, as there is in a polycrystalline thin film, there is homogeneously distributed intensity across the full surface of the cone, and a 2D cut gives continuous rings without intensity variations in reciprocal space. This description can be clarified by looking at figures 2,3 and 4 of reference S [1]

Figure S 2 reports the grazing incidence X-Ray diffraction pattern obtained from a 20 nm Ru film. The black line corresponds to the experimental data, the red lines are peak position retrieved from the Materials project for Ru (mp-33) from database version v2022.10.28., DOI:10.17188/1206459. The diffraction pattern corresponds to polycrystalline ruthenium with no preferential crystal orientation. We can therefore exclude that the preferential direction observed in the THz polarization is caused by a preferential orientation of the crystallites in the sample.

*Figure S 2: Grazing incidence XRD pattern obtained from a 20 nm Ru film on a glass substrate (left) extracted from the 2D-diffraction pattern (right).*
AFM of Ru on sapphire

Figures S 3 are AFM images obtained from 8 nm Ru on Sapphire before annealing (a) and after annealing (b) at 350 °C in a $10^{-3}$ mbar O$_2$ atmosphere for 30 minutes. Comparing figure S 3(a) with the figure 10 (a) we notice a lower density of nanoprotusions, with a lower height for Ru on sapphire than on glass. Similarly, comparing figure S 3 (b) with figure 10 (b), corresponding to the annealed samples, Ru on sapphire shows less and lower protrusions than glass.

Since we observed azimuthal-angle dependence in THz emission from the sample after annealing at 350 °C but not before annealing, we believe that the height and/or density of nanoprotusions in the as-deposited sample is not sufficient to introduce the anisotropic surface-parallel component of THz emission. It is instead sufficient to introduce this polarization component after annealing.

![AFM images of an 8 nm Ru layer on sapphire. (a) As-deposited not-annealed sample. (b) Sample annealed at 350 °C in a $10^{-3}$ mbar O$_2$ atmosphere for 30 minutes.](image)
Bibliography