

SUPPORTING INFORMATION

High-performances “fueled” photodetector based on few-layered 2D ternary chalcogenide NiGa₂S₄

*Marco Serra, ^{*a,f,g} Nikolas Antonatos, ^a Jan Luxa, ^a Luc Lajaunie, ^{b,c} Josep Albero, ^d Agata Sabik, ^e Wojciech M. Linhart, ^e Hermenegildo Garcia, ^d Robert Kudrawiec, ^e David Sedmidubský ^a and Zdenek Sofer ^{*a}*

^a Department of Inorganic Chemistry, University of Chemistry and Technology, Prague, Technická 5, 16228, Prague, Czech Republic

E-mail: soferz@vscht.cz, serram@vscht.cz

^b Departamento de Ciencia de los Materiales e Ingeniería Metalúrgica y Química Inorgánica, Facultad de ciencia, Universidad de Cádiz, Campus Río San Pedro S/N, Puerto Real, 11510, Cádiz, Spain

^c Instituto Universitario de Investigación de Microscopía Electrónica y Materiales (IMEYMAT), Facultad de Ciencias, Universidad de Cádiz, Campus Río San Pedro S/N, Puerto Real 11510, Cádiz, Spain.

^d Instituto de Tecnología Química, Universitat Politècnica de Valencia – Consejo Superior de Investigación Científicas (UPV-CSIC), Universitat Politècnica de València, Avda. de los Naranjos S/N, 46022, Valencia, Spain.

^e Department of Semiconductor Materials Engineering, Wrocław University of Science and Technology, Wybrzeże Wyspiańskiego 27, 50-270, Wrocław, Poland

^f Centro H2-MORE, Università di Modena e Reggio Emilia, 41125 Modena, Italy

^g Dipartimento di Scienze Chimiche e Geologiche, Università di Modena e Reggio Emilia, Via G. Campi 103, 41125 Modena, Italy

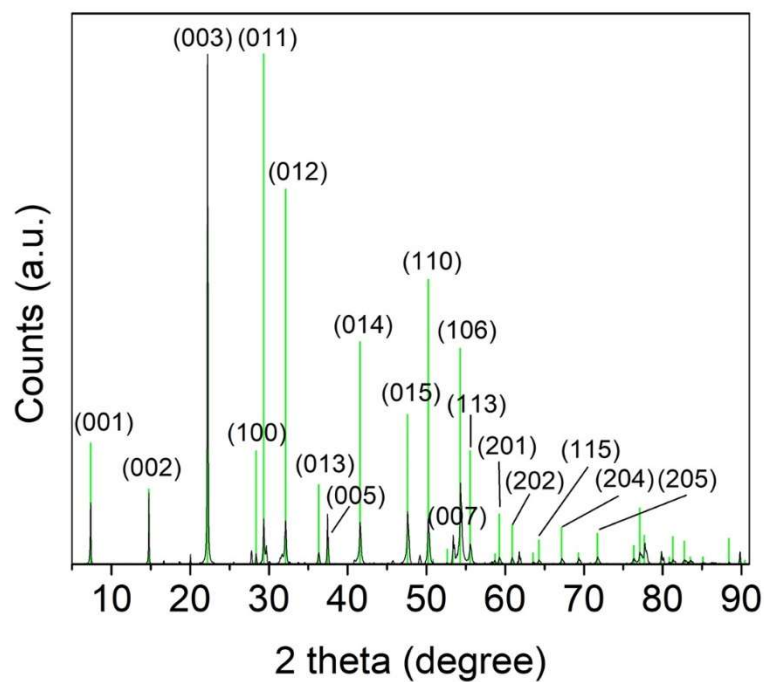


Figure S1. Indexed XRD pattern of the NiGa₂S₄ crystalline powder.

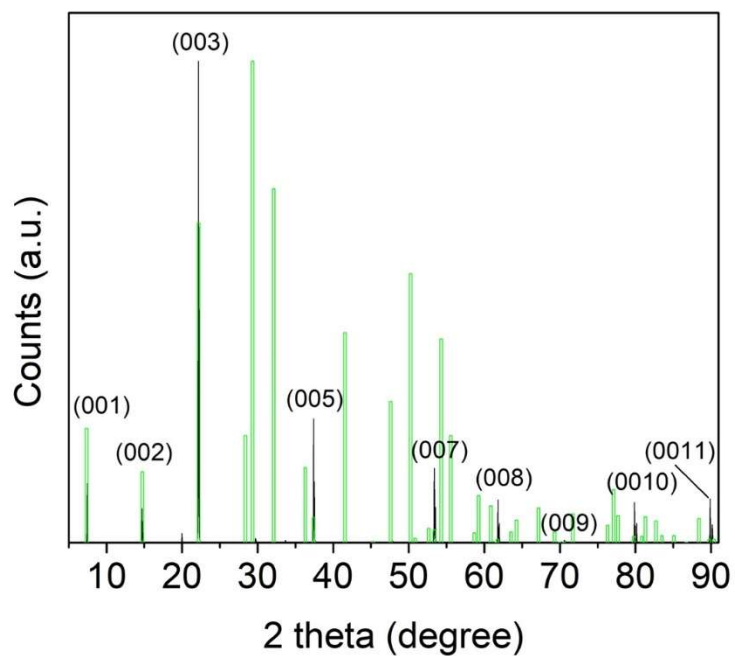


Figure S2. Indexed XRD pattern of a bulk NiGa₂S₄ crystal.

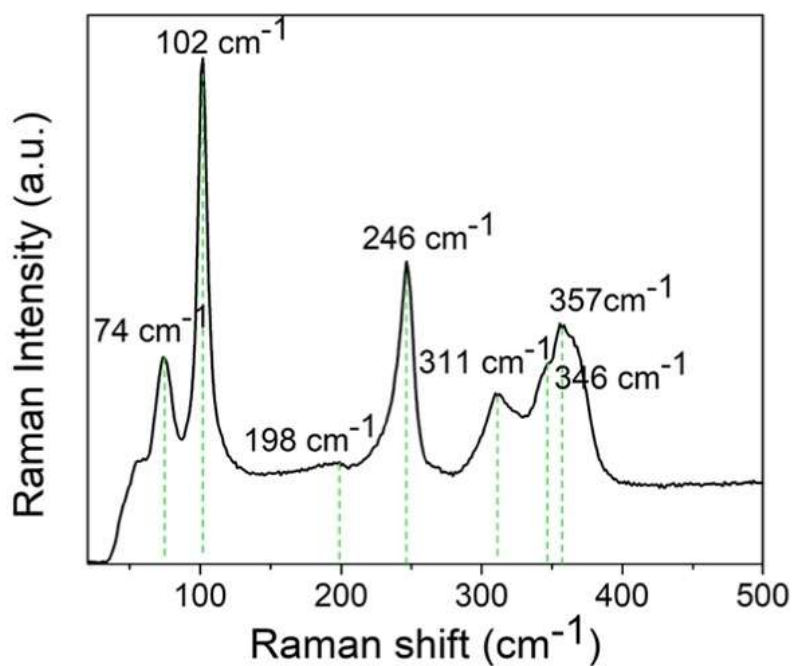


Figure S3. Raman spectra of NiGa₂S₄ crystal prepared by CVT technique.

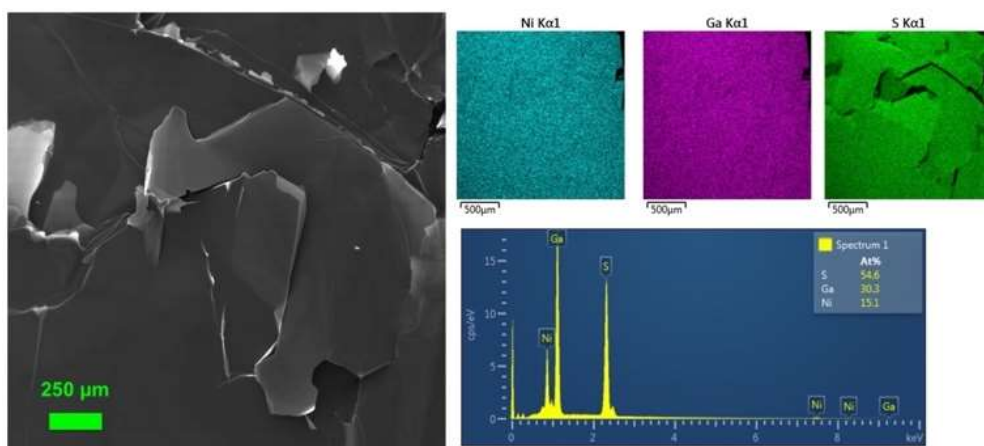


Figure S4. SEM image of a scotch tape exfoliated NiGa₂S₄ crystal prepared via CVT technique and the corresponding EDS elemental mapping, spectrum, and semi-quantitative analysis.

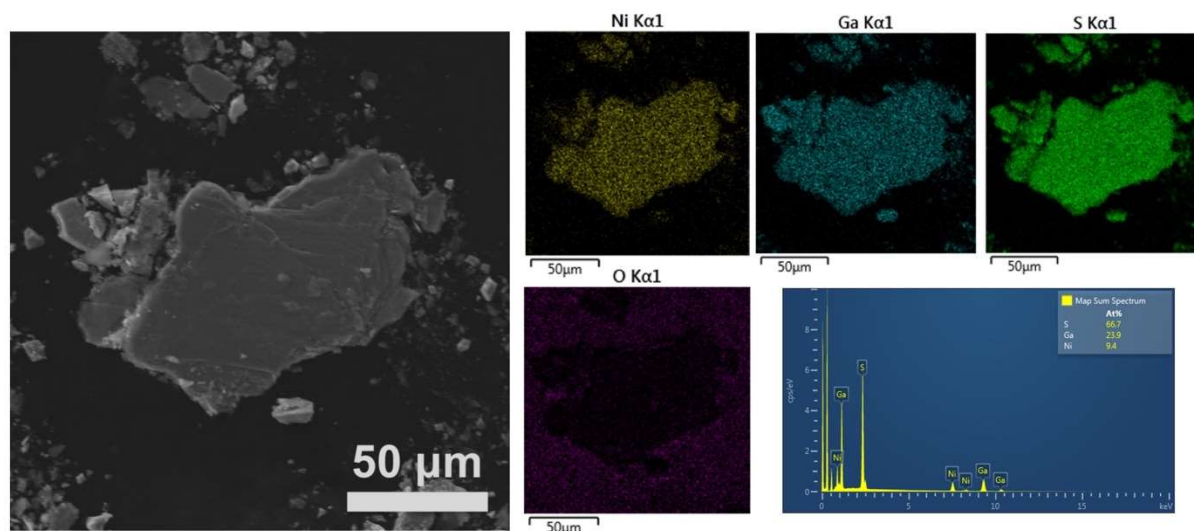


Figure S5. SEM image of a powder obtained grinding NiGa_2S_4 crystal exposed to air for a period of 3 months and the corresponding EDS elemental mapping, spectrum, and semi-quantitative analysis.

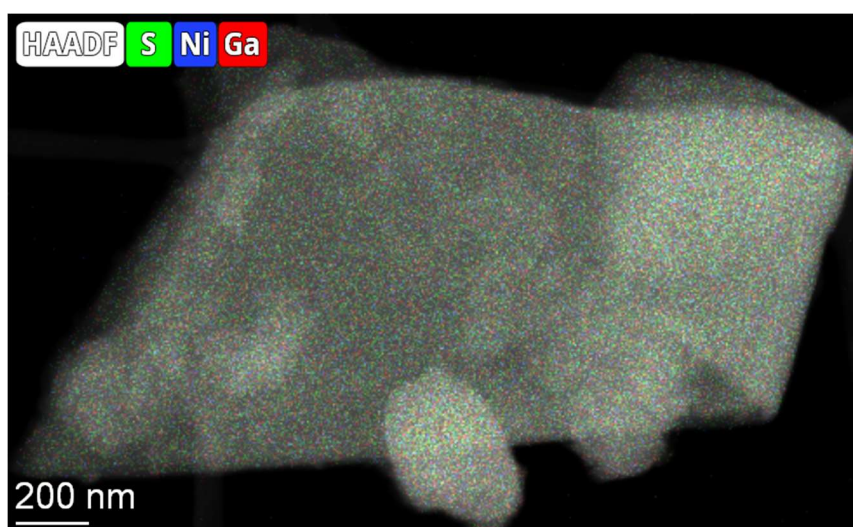


Figure S6. - EDS Map of a typical NiGa_2S_4 crystalline flake.

Table S1. Elemental composition obtained from STEM-EDS analysis.

Element	Atomic Fraction (%)	Atomic Error (%)
S	58.2	2.4
Ni	13.4	1.8
Ga	28.4	2.7

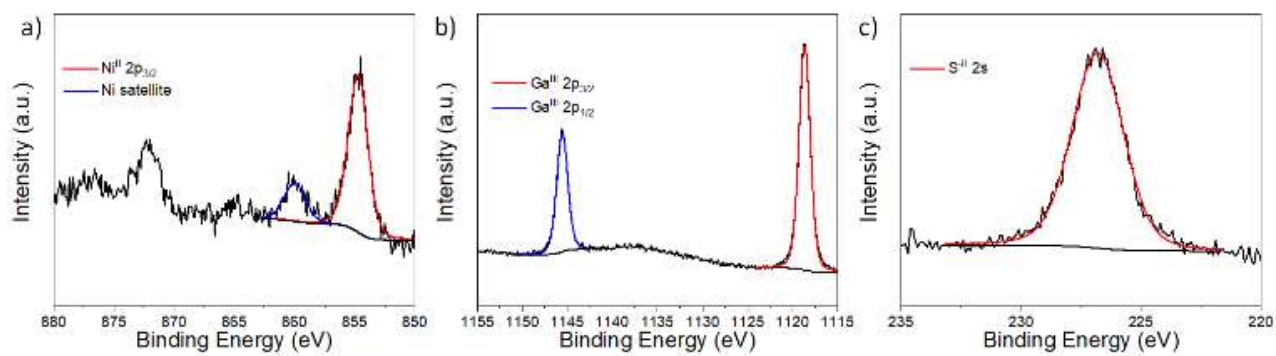


Figure S7. High-resolution core-level spectra of **a)** Ni 2p region; **b)** Ga 2p region and **c)** S 2s region.

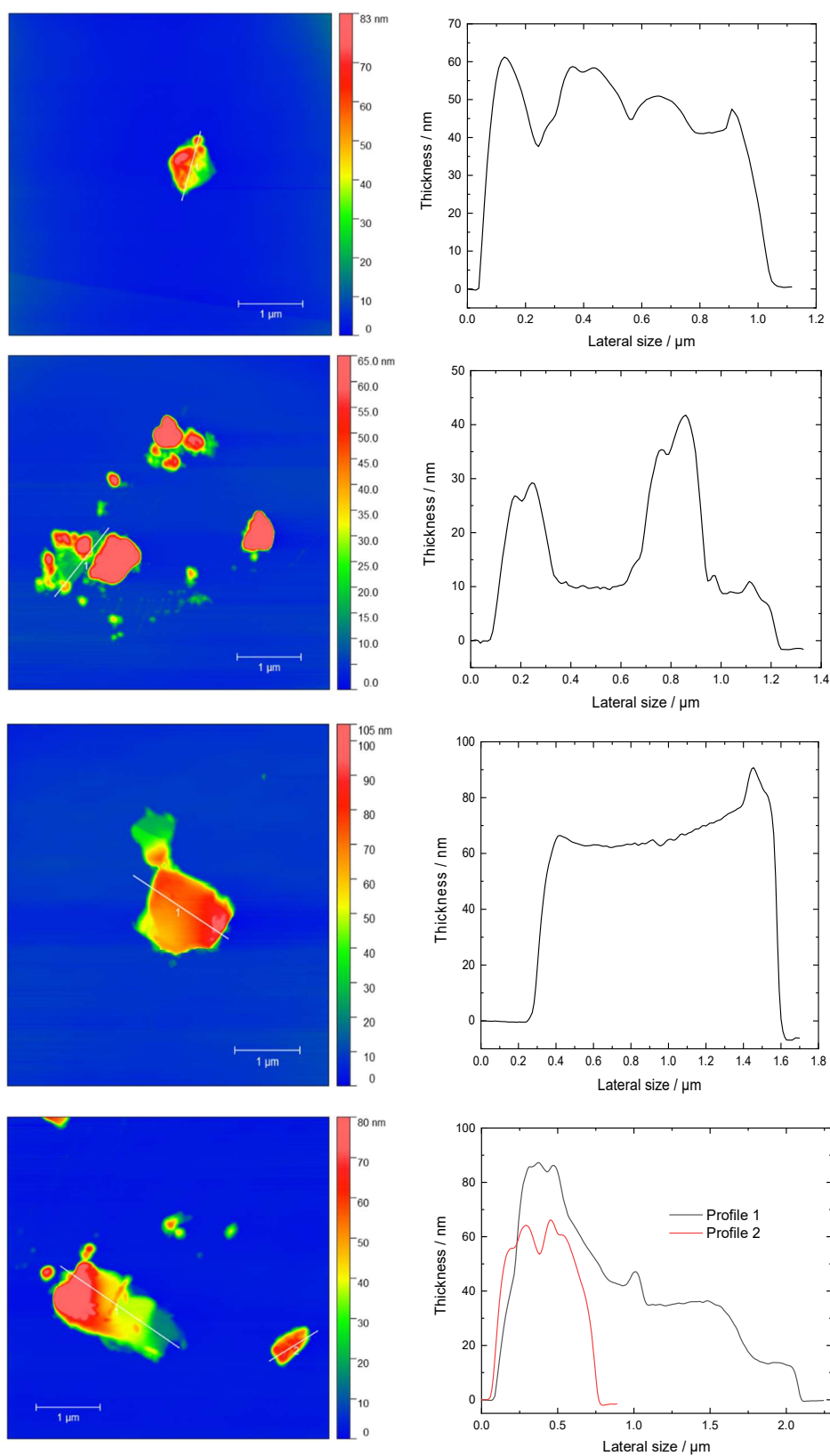


Figure S8. – Various AFM images of 2D-NiGa₂S₄ with their corresponding height profile. These flakes were deposited on ITO that were employed as the photodetector.

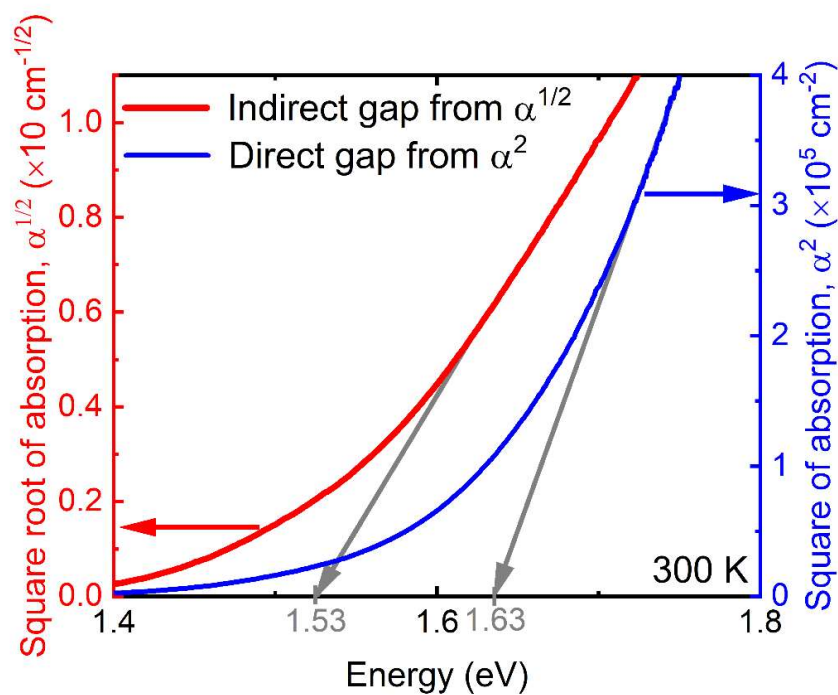


Figure S9. An exemplary analysis of room temperature optical absorption spectrum: square root of absorption (red line) typical for indirect absorption and square of absorption (blue line) typical for direct absorption.

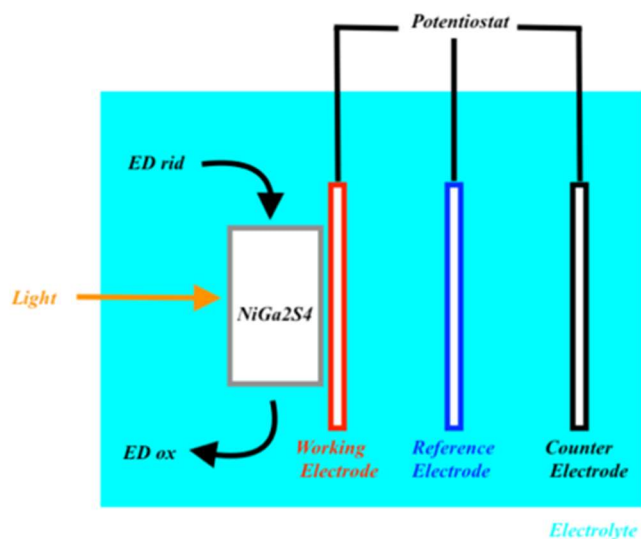


Figure S10. Scheme of the cell employed for the PEC tests.

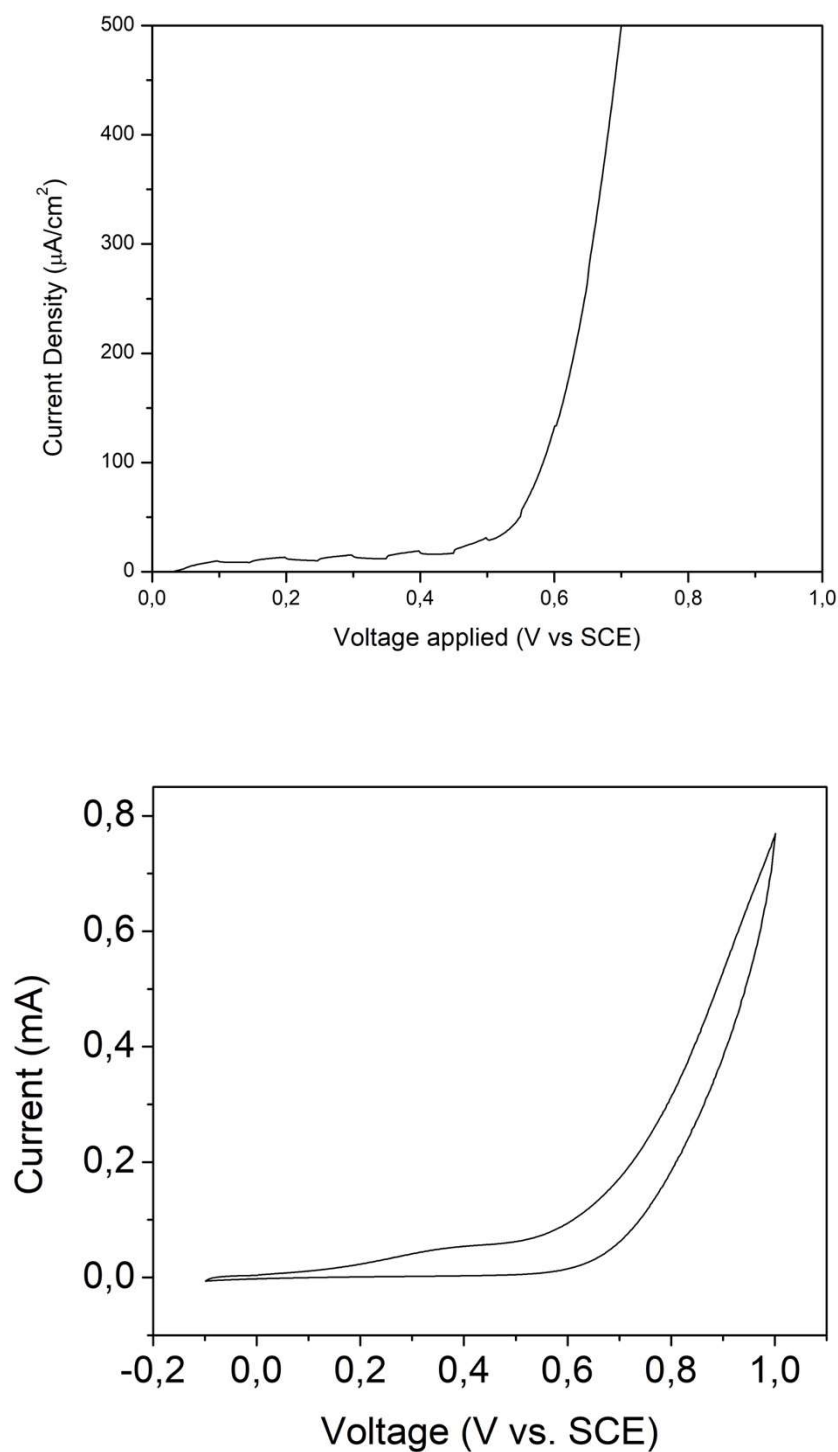


Figure S11. Linear Sweep Voltammetry (**top**) and Cyclic voltamperometric curve (**bottom**) of $\text{NiGa}_2\text{S}_4/\text{ITO}$. A 420 nm LED light source has been employed to investigate the photocurrent induced upon irradiation.

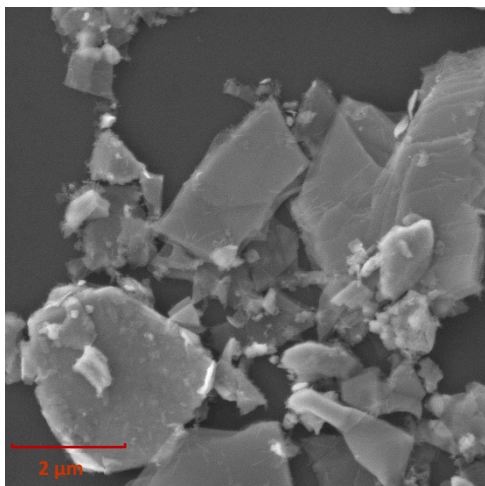


Figure S12. SEM image of 2D-NiGa₂S₄ flakes which were employed for the fabrication of the photodetector.

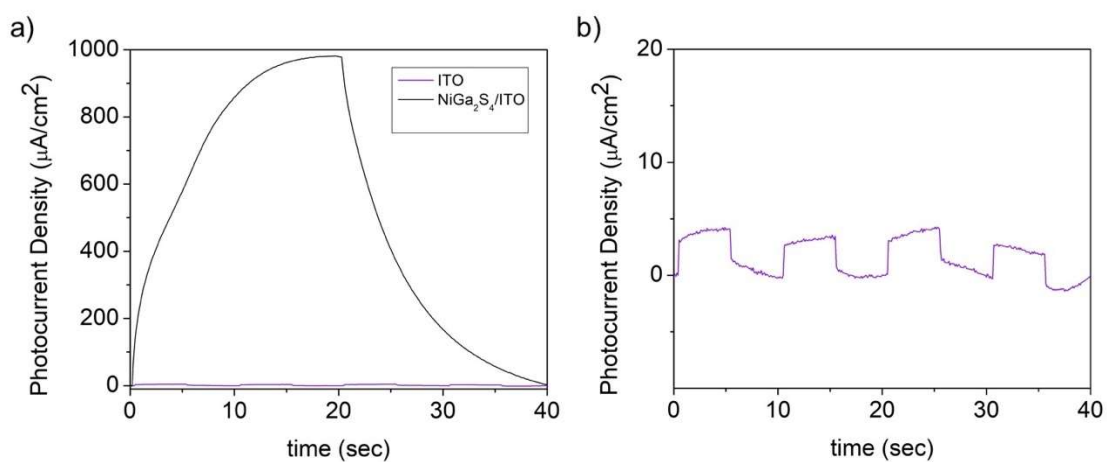


Figure S13. a) Comparison of the photocurrent density obtained using an ITO glass and a NiGa₂S₄/ITO photodetector in the presence of a 1 M KOH ethanol (25% v/v) water solution as electrolyte under irradiation with purple light (power = 1000 mW). **b)** Magnification of the photocurrent density using bare ITO glass as photodetector.

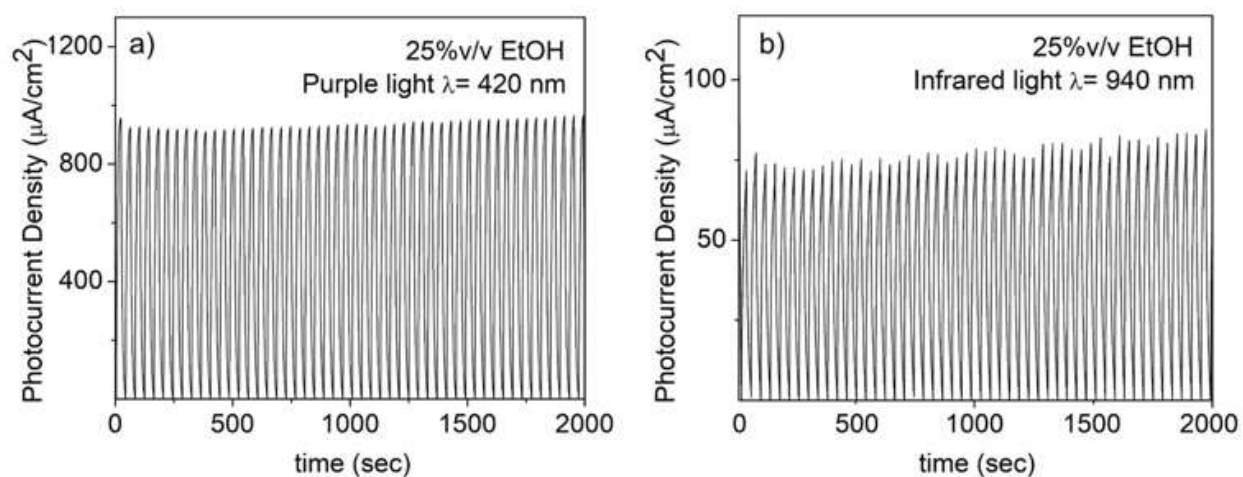


Figure S14. Long term cycle stability test of NiGa₂S₄-based photodetector in 1 M KOH ethanol (25% v/v) water solution at 0.5 V vs SCE under purple and infrared light, with irradiance power of 800 and 300 mW respectively.

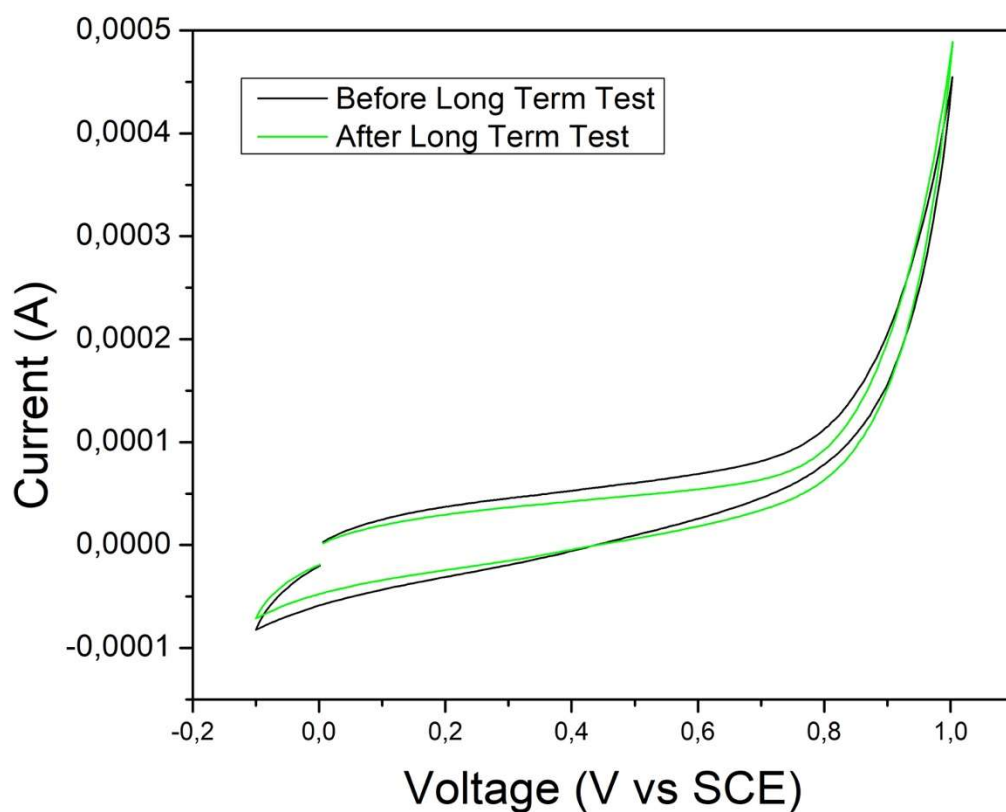


Figure S15. Cyclic voltammetry diagram of the NiGa₂S₄/ITO electrode before and after the long-term ON-OFF tests.