A terahertz near-field microscope for flakes of 2D materials

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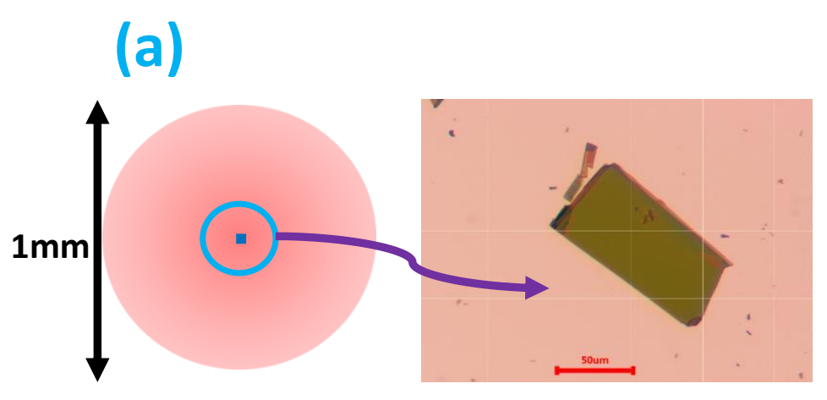
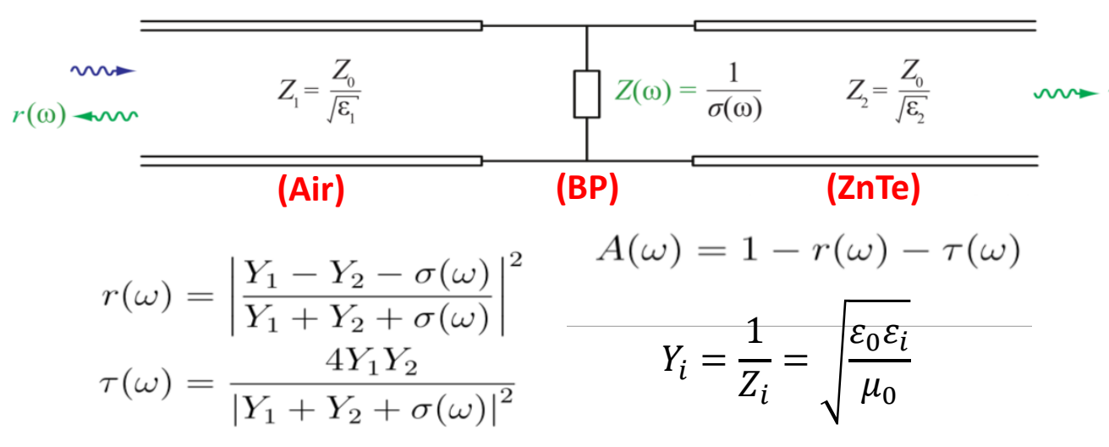
Introduction and Motivation

- Exfoliated 2D materials e.g. graphene and black phosphorus (BP) are of **µm-size**.
- THz spectroscopy is a powerful tool to investigate the **carrier dynamics** based the **Drude** absorption.
- The long wavelength (**300µm @ 1THz**), due to **small perturbation** precludes conventional spectroscopy (**spot size ~1mm**).

$\sigma(\omega)$	Conductivity
σ_0	DC conductivity
τ	Scattering time
q	Elementary charge
n_q	Carrier density
m_q^*	Carrier effective mass

$$\sigma(\omega)=\frac{\sigma_0}{1-i\omega\tau},\ \sigma_0=\frac{n_q q^2\tau}{m_q^*}$$





$$r(\omega)=\left|\frac{Y_1-Y_2-\sigma(\omega)}{Y_1+Y_2+\sigma(\omega)}\right|^2 \qquad A(\omega)=1-r(\omega)-\tau(\omega)$$

$$\tau(\omega)=\frac{4Y_1Y_2}{|Y_1+Y_2+\sigma(\omega)|^2} \qquad Y_i=\frac{1}{Z_i}=\sqrt{\frac{\varepsilon_0\varepsilon_i}{\mu_0}}$$

Equivalent Circuit Model (ECM) [1]

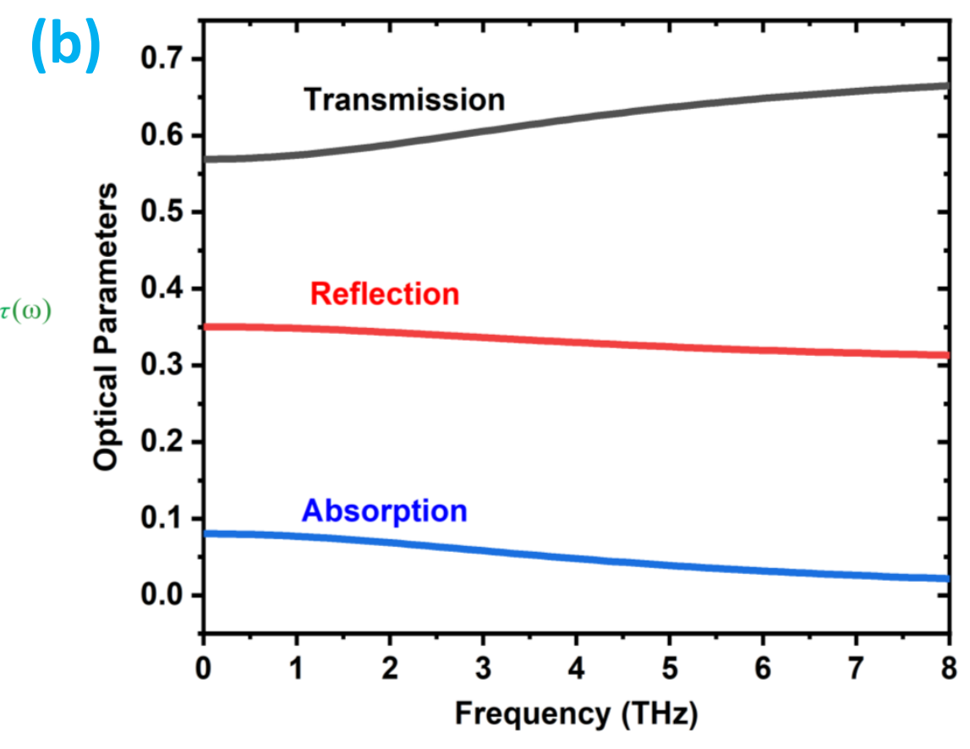
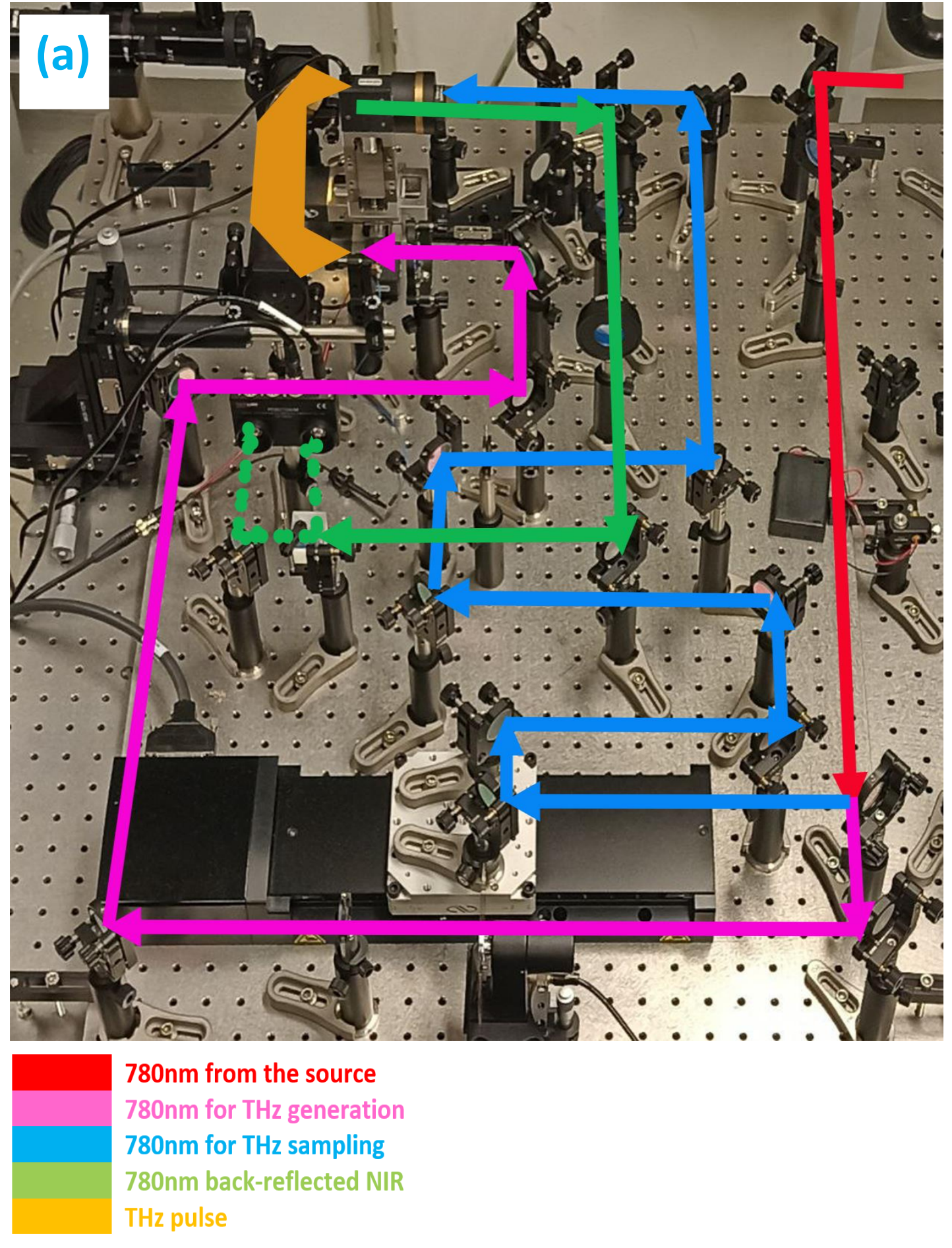
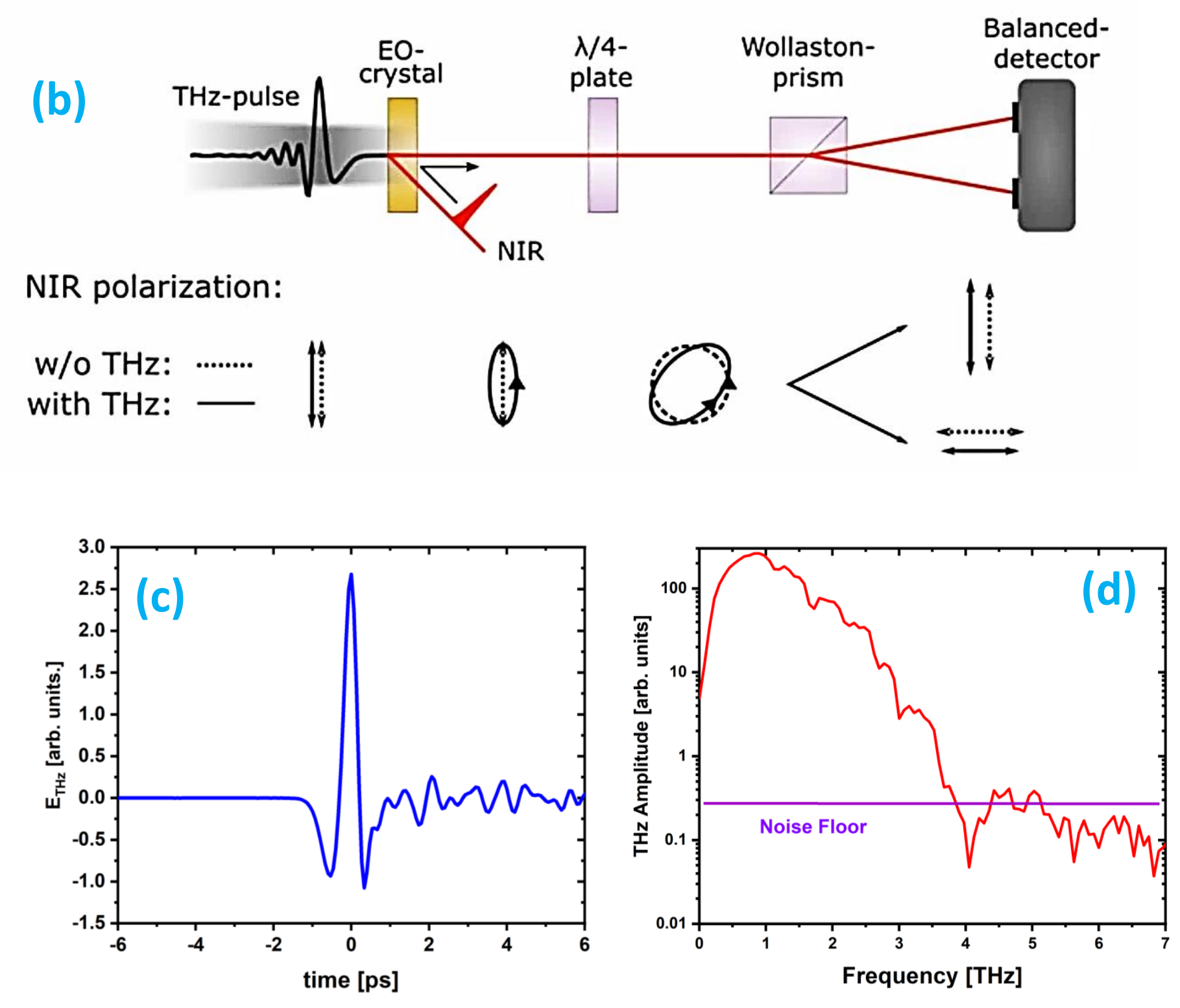


Fig.1 (a) Scale of the flakes compared to the THz beam, (b) Drude based calculation of the optical parameters in the THz frequency range

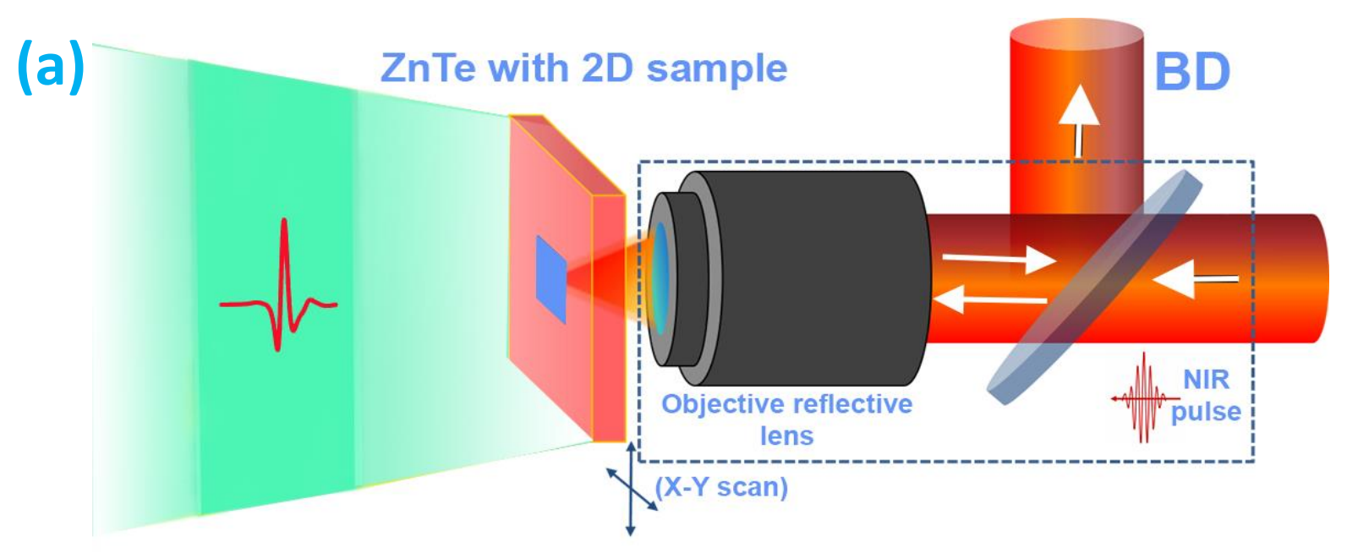
THz Setup and Working Principles





- **ZnTe** is used as the Electro- Optic (EO) crystal for the detection.
- THz field induces **birefringence** in the EO crystal. Then the **polarization change** of the **NIR** beam due to the birefringence will be measured.

Fig.2 (a) Overview of the current setup. Arrows indicate the different optical beam pathes, (b) schematic view of the EO sampling and the summary of the data extraction process by using the balance detector (BD). (c, d) Time-domain transient and amplitude spectrum, respectively



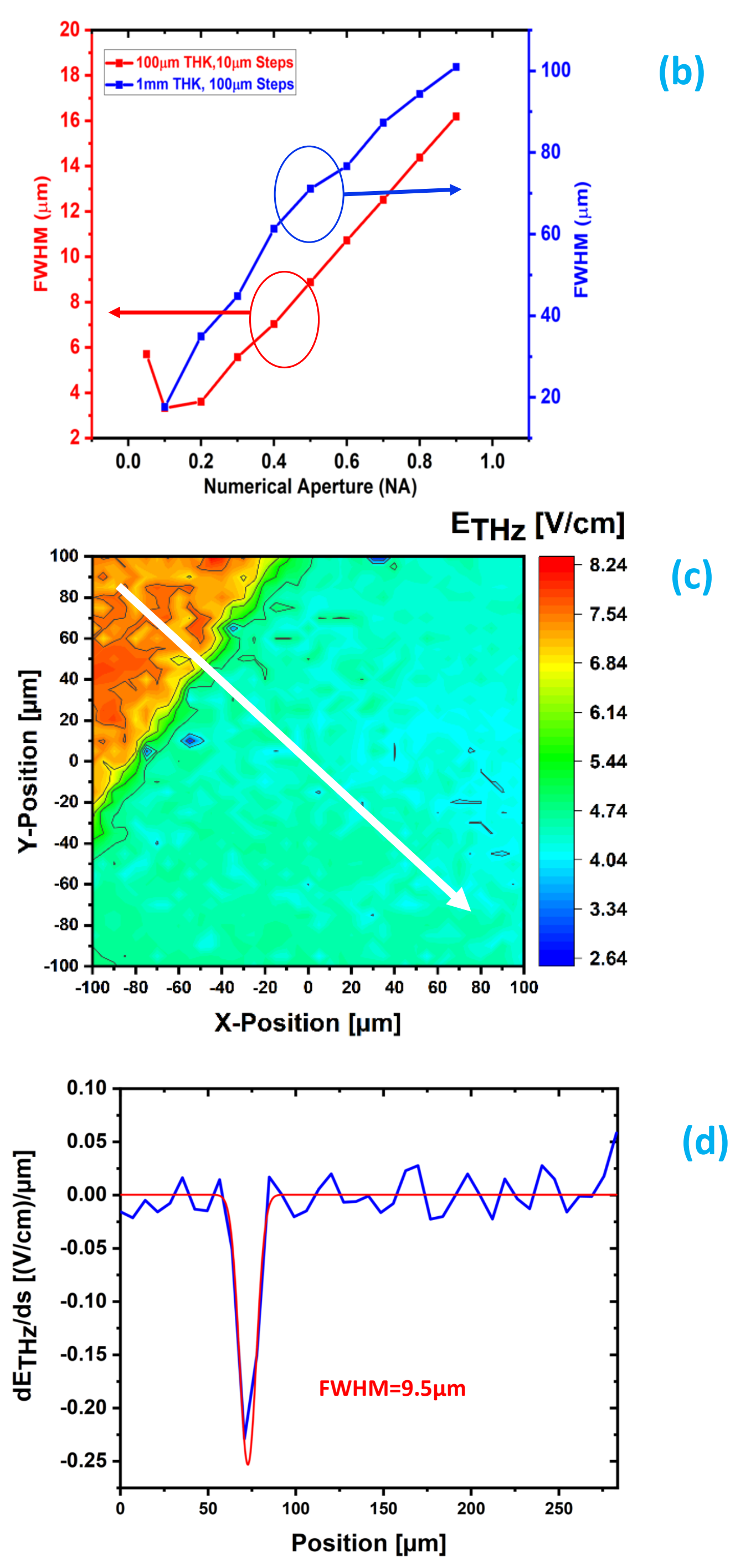


Fig.3 (a) Direct detection of the signal from the sample in the vicinity of the EO crystal, (b) modeling of the effect of the thickness of the EO crystal on the spatial resolution (NIR beam propagation), (c) Experimental verification of the spatial resolution and the homogeneity of thin EO crystals, (d) Variation of the measured THz electric field in the diagonal direction

- Quantitative time-domain detection at the **µm-scale**
- Spatial resolution depends on:
  1. The **thickness** of the EO crystal
  2. The **diffraction** of the THz beam within the EO crystal
  3. The **focusing** of the near infrared (NIR) sampling beam

- With the same numerical aperture(0.5 for our setup), the **thinner** the EO crystal, the **higher** the spatial resolution of a NIR beam propagating the crystal (Fig.3b)
- **Homogeneity** of the spatial resolution of the EO crystal, guarantees the calculated spatial resolution while scanning the surface of the EO crystal (Figs.3c & 3d).

References

[1] Nonlinear Terahertz Absorption of Graphene Plasmons, MM. Jadidi, *et al.*, **Nano Lett., Vol. 16, 4 (2016)**.

[2] Electron-doped phosphorene: A potential monolayer superconductor, D. F. Shao , *et al.,* **Europhysics Letters, Vol. 108, No. 6, 7004 (2014)**.

[3] Fast-Fourier-transform based numerical integration method for the Rayleigh–Sommerfeld diffraction formula, F. Shen and A. Wang, **Applied Optics Vol. 45, Issue 6, 1102 (2006)**.

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Materials and Sample Preparation Methods

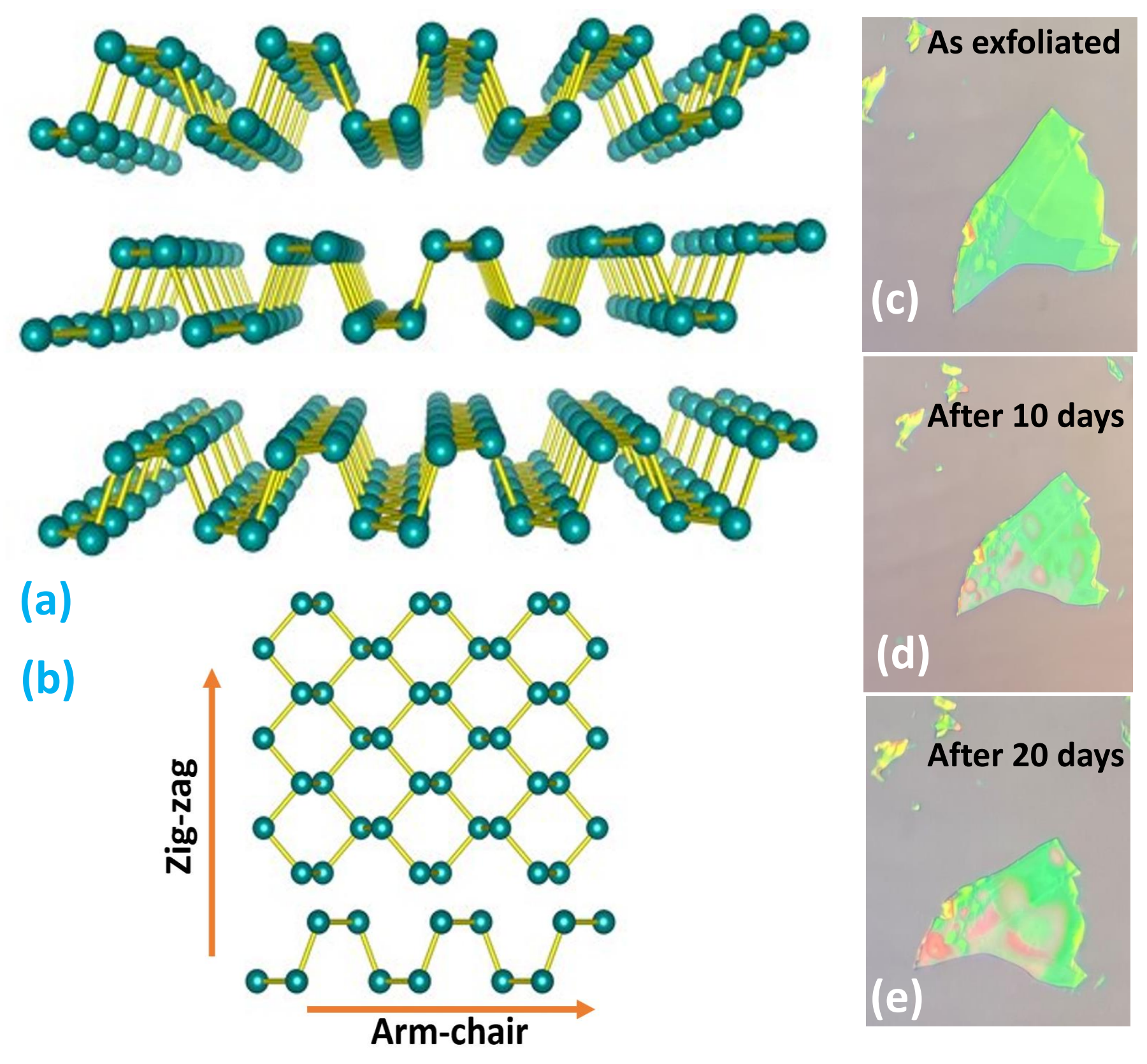


Fig.4 (a & b) Anisotropic nature of the black phosphorus (BP) from the side and top view, respectively [2], (c-e) degradation steps of BP, (f) BP crystal used for the exfoliation

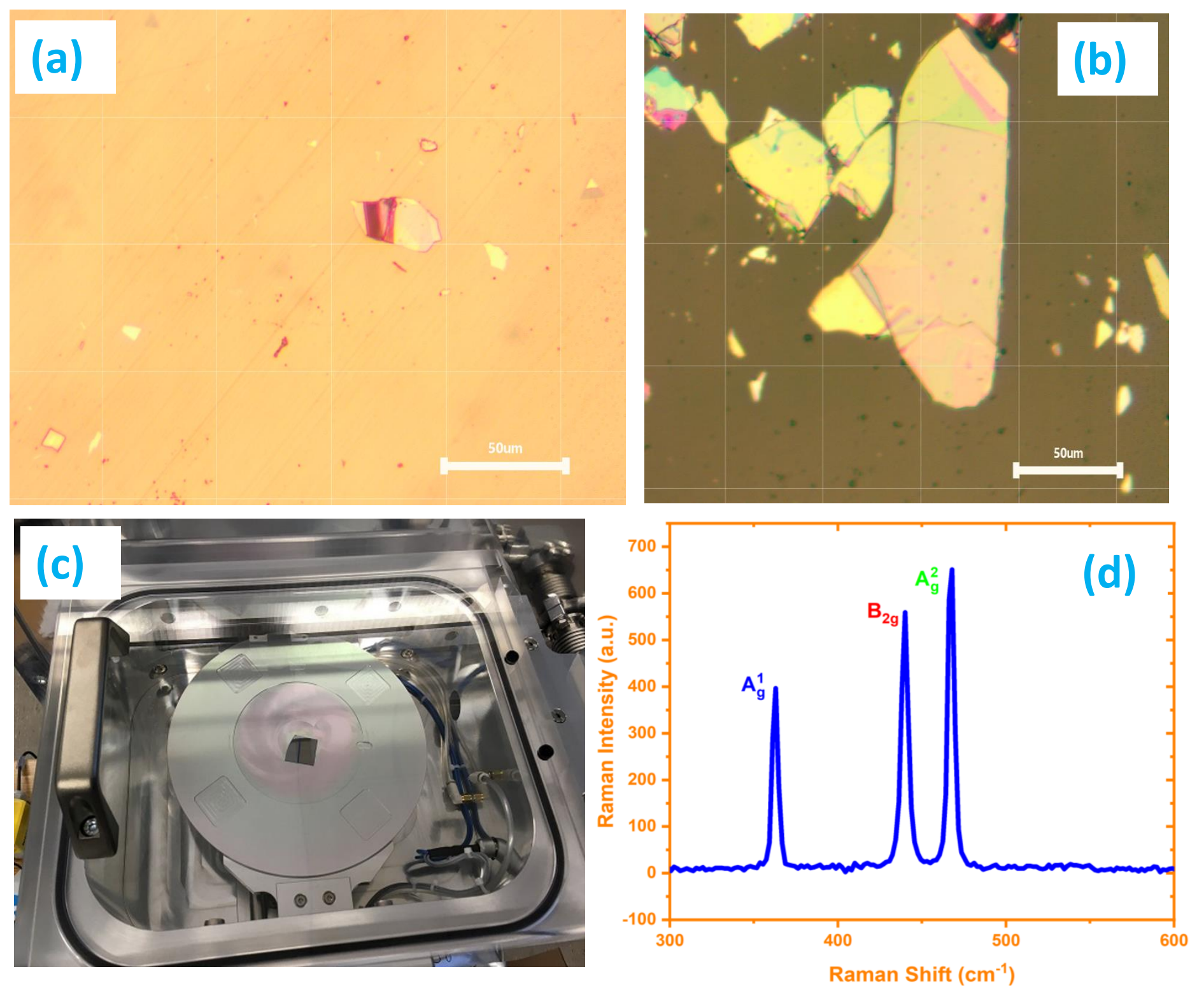


Fig.5 (a) Exfoliated and passivated BP flake on **150µm ZnTe** for the near-field setup, (b) Exfoliated and passivated BP flake on **150µm glass slide** for the auto-correlation pump-probe measurement, (c) **ALD** chamber used for the passivation of **Al_2O_3** layer(30nm), (d) Raman modes of the exfoliated and passivated BP flakes

- **ALD-Al_2O_3 passivation: Department of Physics, Uni. Bielefeld, Prof. Dr. G. Reiss & J. Biedinger**
- **Raman measurements: Department of Physics, Uni. Duisburg-Essen, AG Schleberger , S. Sleziona**
- **Ion irradiation of the samples for the defect engineering: Department of Physics, Uni. Duisburg-Essen, AG Schleberger , Y. Liebsch**

THz and NIR Beam Within the EO Crystal

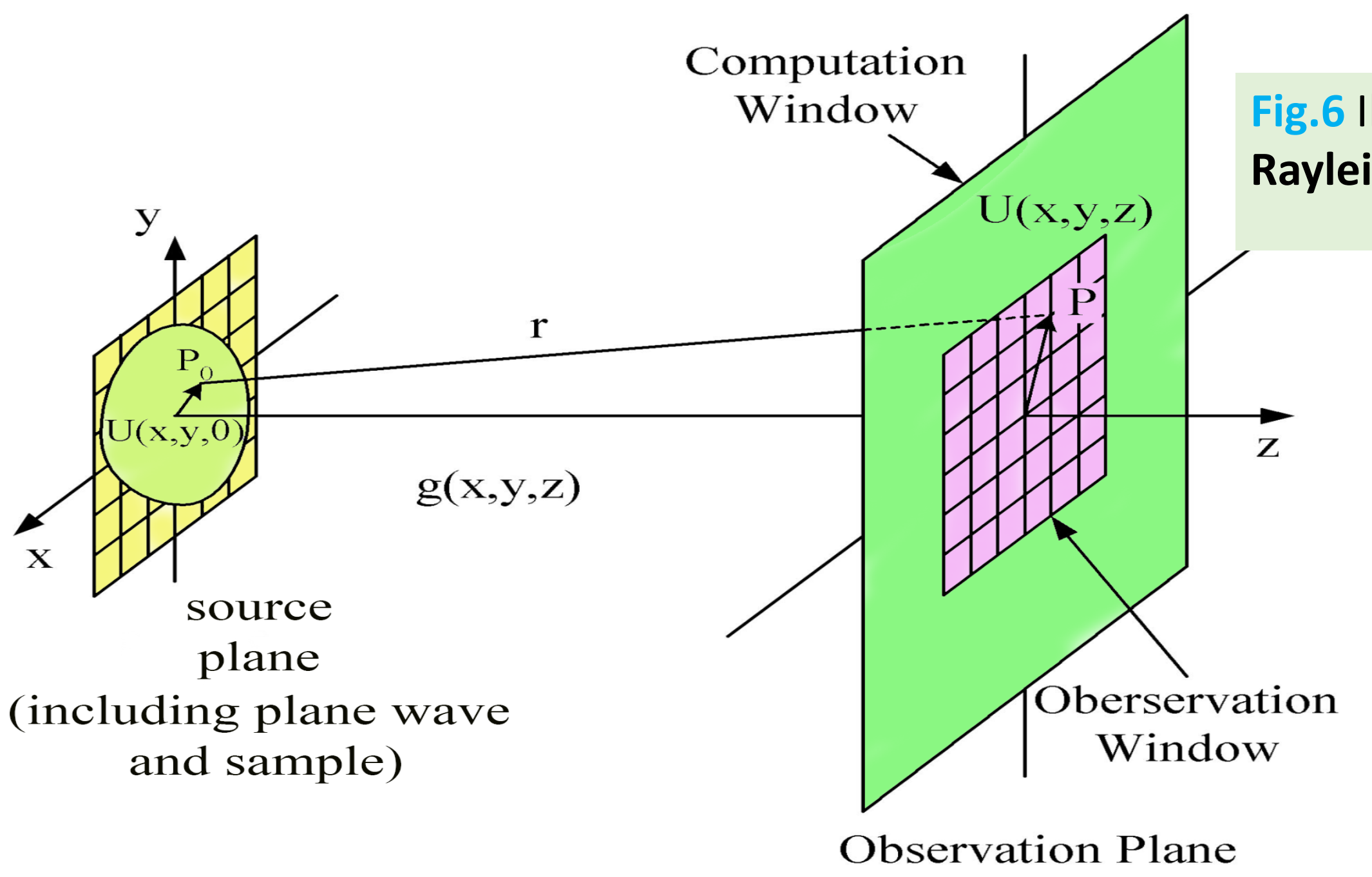


Fig.6 Illustration of the coordinate system of the **Rayleigh-Sommerfeld** diffraction integral [3].

- **Rayleigh-Sommerfeld** diffraction integral is calculated numerically based on the Green's functions.
- **Initial condition**: Plane-wave with the **Gaussian** THz beam distribution at the front facet

$$U(x,y,z)=\iint_A U(s,\eta,0).\,g(x-s,y-\eta,z).\,\mathrm{ds}.\,\mathrm{d}\eta=\iint_A U(s,\eta,0).\frac{e^{-ikr}}{r}.\left(ik+\frac{1}{r}\right).\frac{z}{r}.\,\mathrm{ds}.\,\mathrm{d}\eta$$

$U(x,y,z)$: THz electric field at the target point

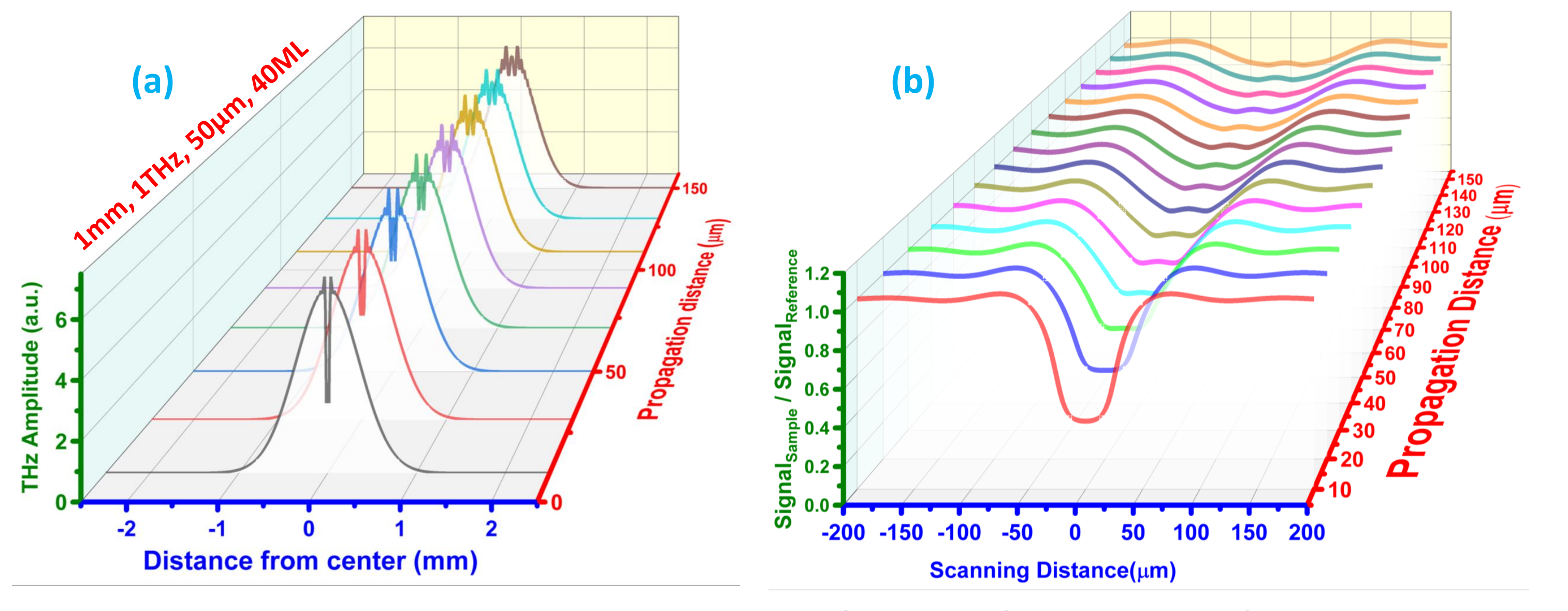
$U(s,\eta,0)$: Input THz electric field

z: horizontal distance, source and target plane

ds , $d\eta$: Sampling intervals

$g(x-s,y-\eta,z)$.: Rayleigh-Sommerfeld function

k: wave vector



$$\Delta\Phi=\frac{2\pi l}{\lambda_{NIR}}n^3 r_{41}E_{THz}=\arcsin\left(\frac{I_y-I_x}{I_y+I_y}\right)$$

$\Delta\Phi$: phase shift due to THz field

c: speed of light

l: crystal length

E_{THz}: THz electric field

λ_{NIR}: NIR beam wavelength

I_y, I_x: orthogonally detected signal

n: refractive index of the medium

r_{41}: electro-optic coefficient

Fig.7 (a) The **evolution** of the THz electric field within the ZnTe crystal (150µm), (b) The sampling of the THz electric field exploiting a **Gaussian NIR beam** focused on the sample which is directly placed on top of the EO crystal

Outlook

- Further extension of the current setup by using an additional 1.55µm pump beam to study the non-equilibrium dynamics of the charge carriers
- Auto-correlation and photo-current studies on passivated and irradiated samples
- Making new BP based opto-electronic devices for complementary measurements with and without the irradiation effect