

D. Janoschka<sup>1</sup>, P. Dreher<sup>1</sup>, A. Neuhaus<sup>1</sup>, A. Rödl<sup>1</sup>, T. Davis<sup>1,2,3</sup>,  
B. Frank<sup>2</sup>, H. Gießen<sup>2</sup>, F.-J. Meyer zu Heringdorf<sup>1</sup>

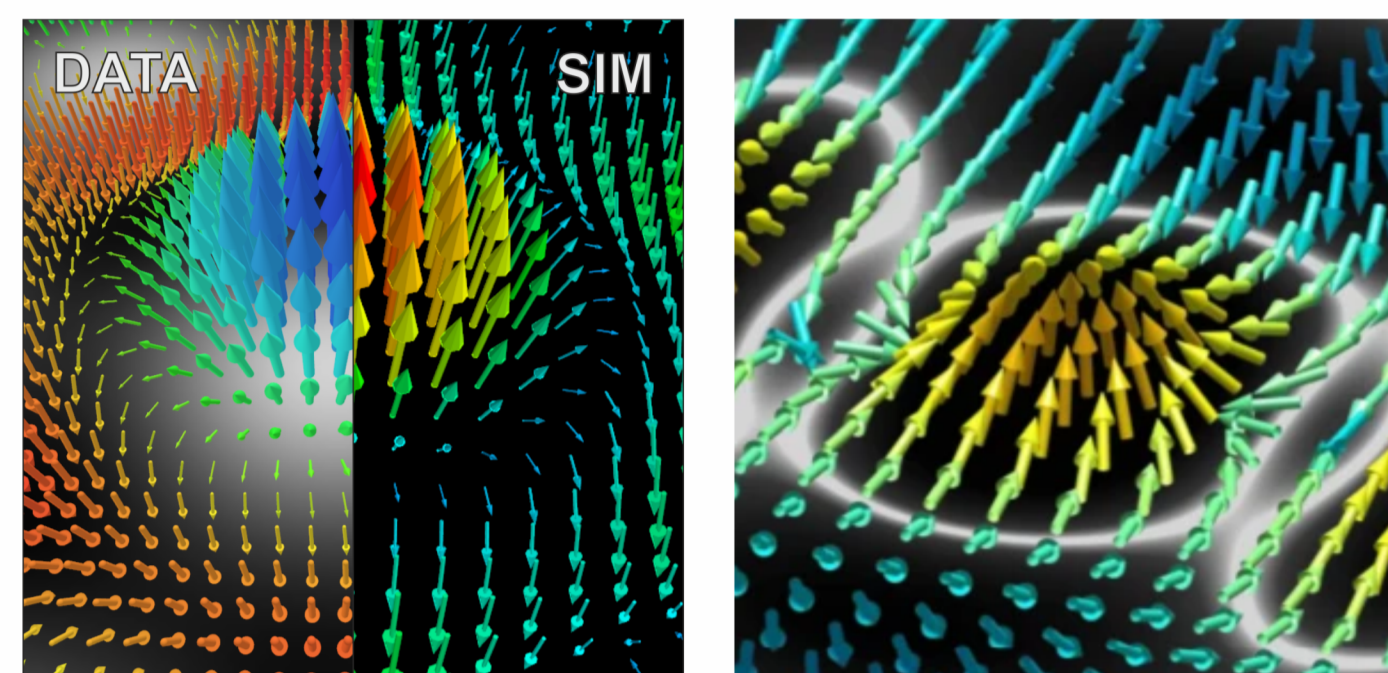
<sup>1</sup> Department of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany

<sup>2</sup> 4<sup>th</sup> Physics Institute, University of Stuttgart, 70569 Stuttgart, Germany

<sup>3</sup> School of Physics, University of Melbourne, Parkville, Victoria 3010, Australia

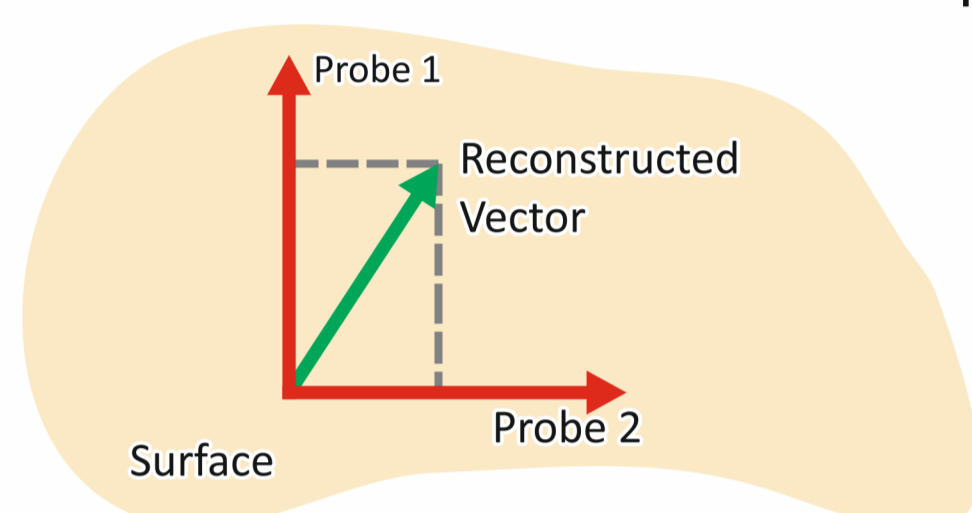
## Introduction

- Investigating near fields of Surface Plasmon Polaritons (SPPs) with sub-wavelength resolution on a femtosecond timescale is challenging.
- Time-resolved two-photon photoemission microscopy measures the projection of the SPP field onto a probing laser pulse.
- Performing the experiment with different probe polarizations allows reconstructing the SPP's electric field vector in time and space.



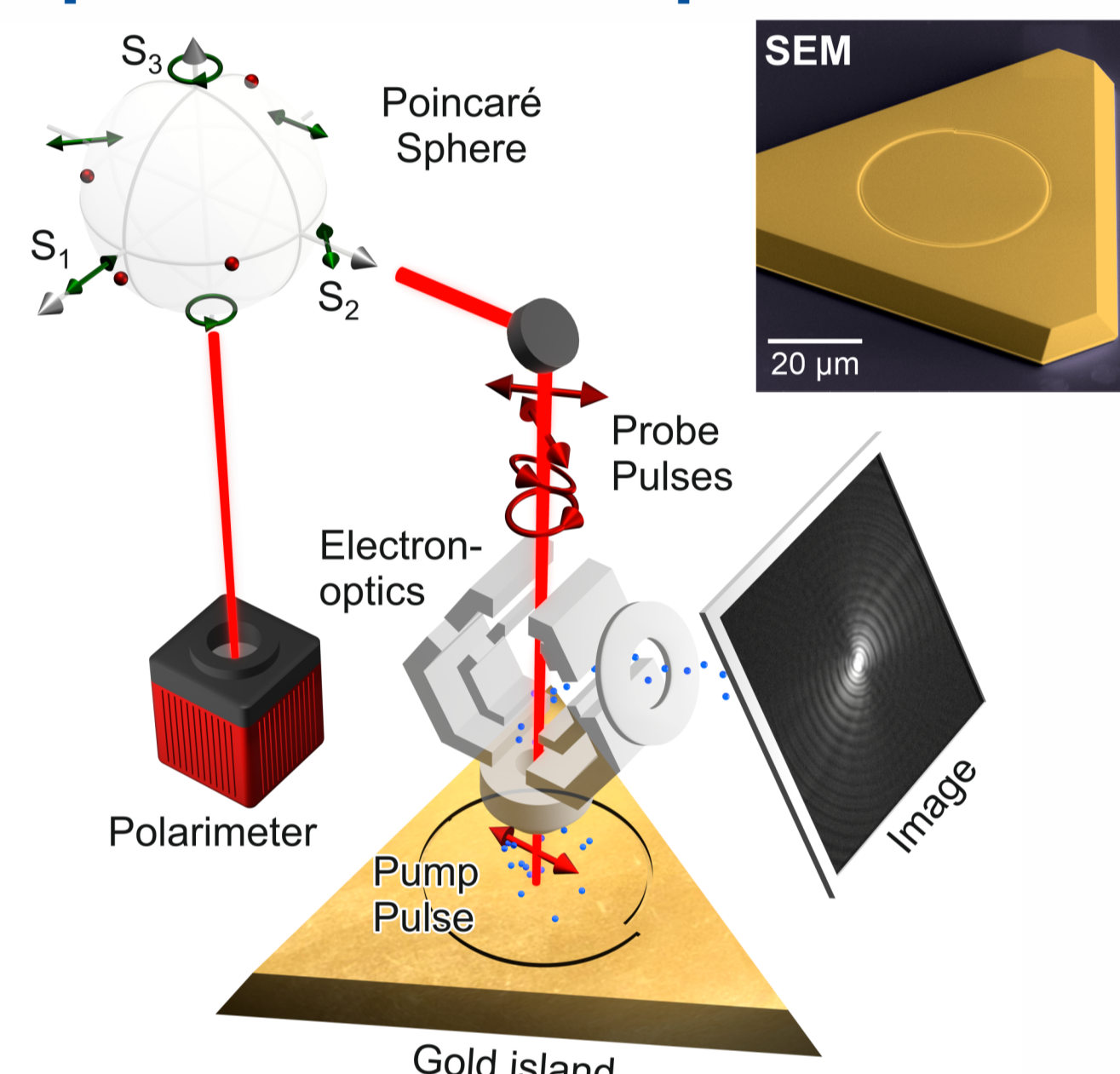
T. Davis et al., Science 368, eaab6415 (2020) Y. Dai et al., Nature 588, 616 (2020)

- Recent work: Reconstruction of a SPP skyrmion lattice by vector microscopy.
- Here: Determination of the electric field vectors of topological SPP fields by vector polarimetry. Analysis of the spin angular momentum using topological concepts.

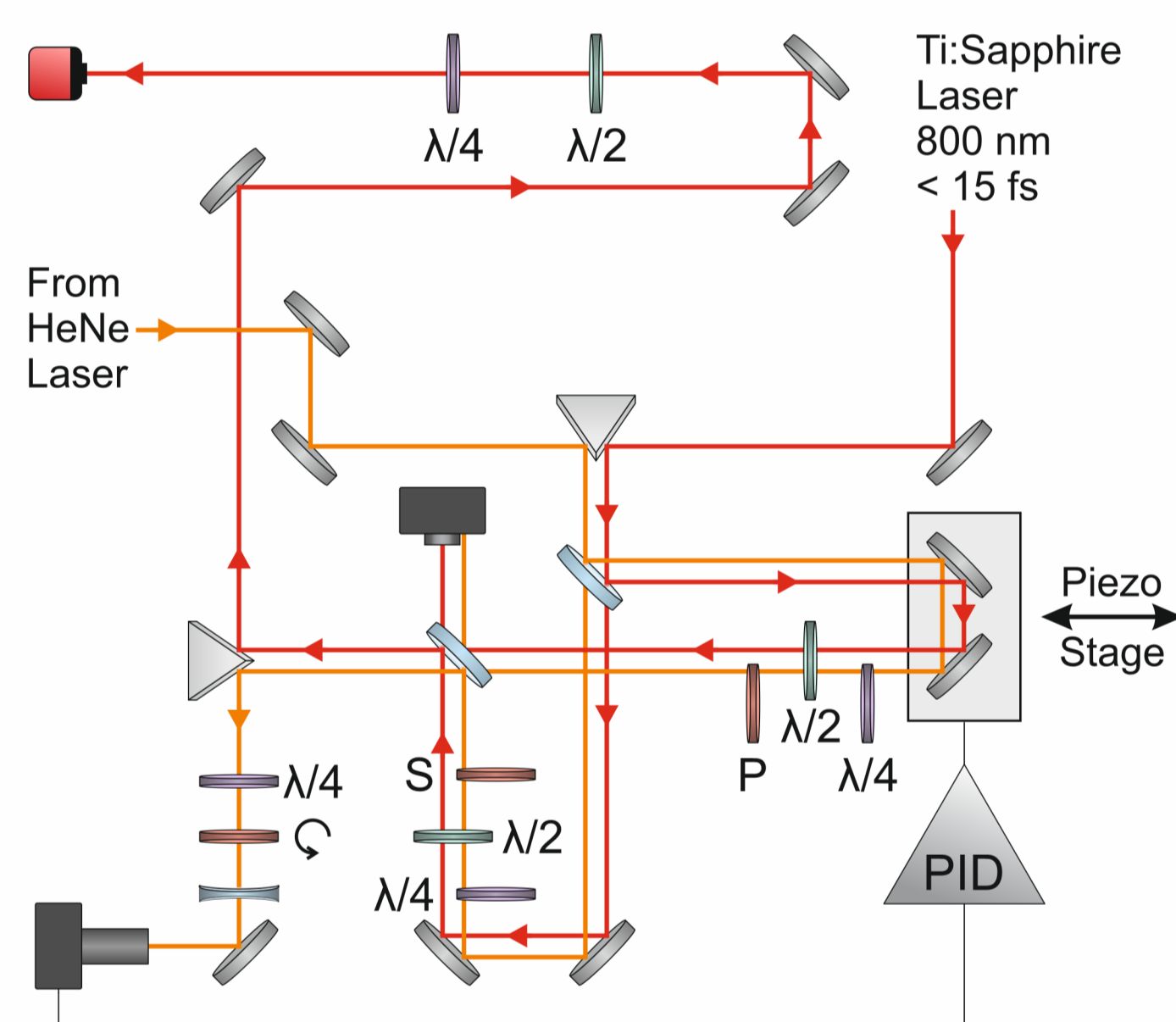


## Experimental Details & General Idea

### Experimental Setup



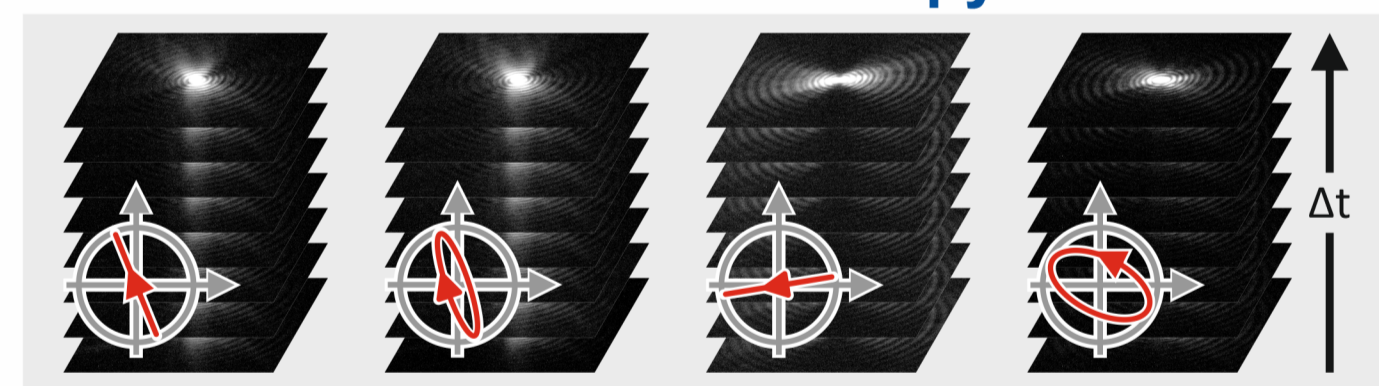
- Femtosecond laser pulses of controlled polarisation impinge on an atomically flat Gold island.
- SPPs are excited at grooves in the shape of Archimedean spirals that are cut into the island using focused ion beam milling.
- Photoemitted electrons are imaged in an ELMITEC Low Energy Electron Microscope upgraded with a TVIPS F216 detector.



- Time-resolution well below one femtosecond is achieved in a pump-probe setup using a Pancharatnam's phase-stabilized Mach-Zehnder interferometer.
- By inserting wave plates into the two interferometer arms, different polarization states for pump- and probe- pulses are prepared.
- All polarizations are verified using a polarimeter in an equivalent sample plane.

### Vector Polarimetry Data Pipeline

#### Raw Photoemission Microscopy Data

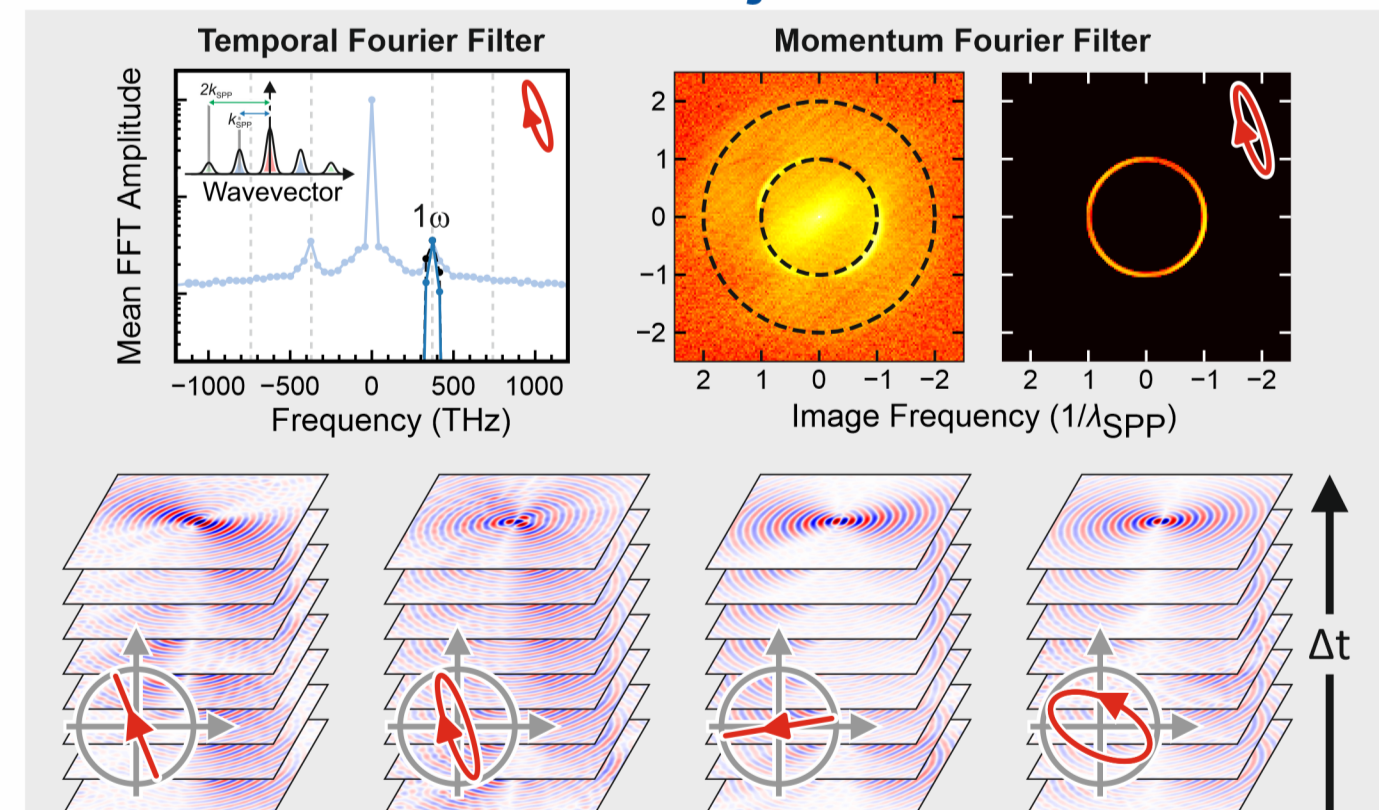


- A time-resolved PEEM dataset is recorded for the same pump- but for different probe-polarizations.
- The contrast in the images is rather complex. The average electron emission rate is described by

$$\Gamma_{2PPE} \propto |\vec{E}_{SPP}(\vec{r})|^4 + |\vec{E}_L(\vec{r})|^4 + 4|\vec{E}_L|^2 |\vec{E}_{SPP}|^2 + 4|\vec{E}_L|^2 \text{Re}\{\vec{E}_L^* \cdot \vec{E}_{SPP} \cdot e^{-i\omega t + i\vec{k}_{SPP} \cdot \vec{r}}\} + 4|\vec{E}_{SPP}|^2 \text{Re}\{\vec{E}_L^* \cdot \vec{E}_{SPP} \cdot e^{-i\omega t + i\vec{k}_{SPP} \cdot \vec{r}}\} + 2 \text{Re}\{\vec{E}_L^* \cdot \vec{E}_{SPP} \cdot e^{-2i\omega t + 2i\vec{k}_{SPP} \cdot \vec{r}}\}$$

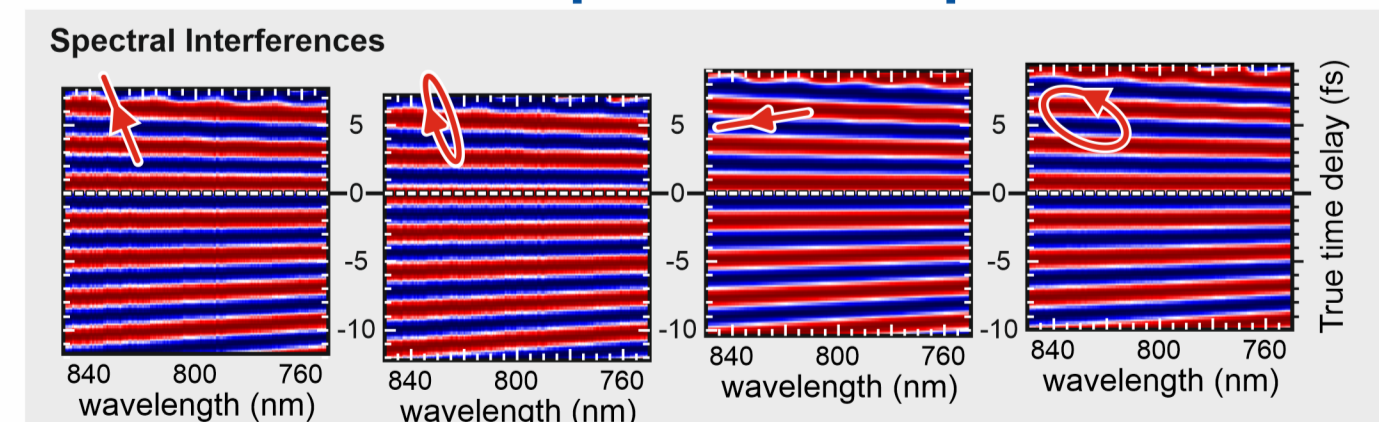
- Fourier-filtering to the  $1\omega$  and the  $1|k|$  component produces a signal that is linear in  $\vec{E}_{SPP}$ , as long as  $\vec{E}_{SPP}$  is weak compared to  $\vec{E}_L$ .
- Changing the probe-polarization in the interferometer affects the relative time-delay.

#### Extraction of Vector Projection



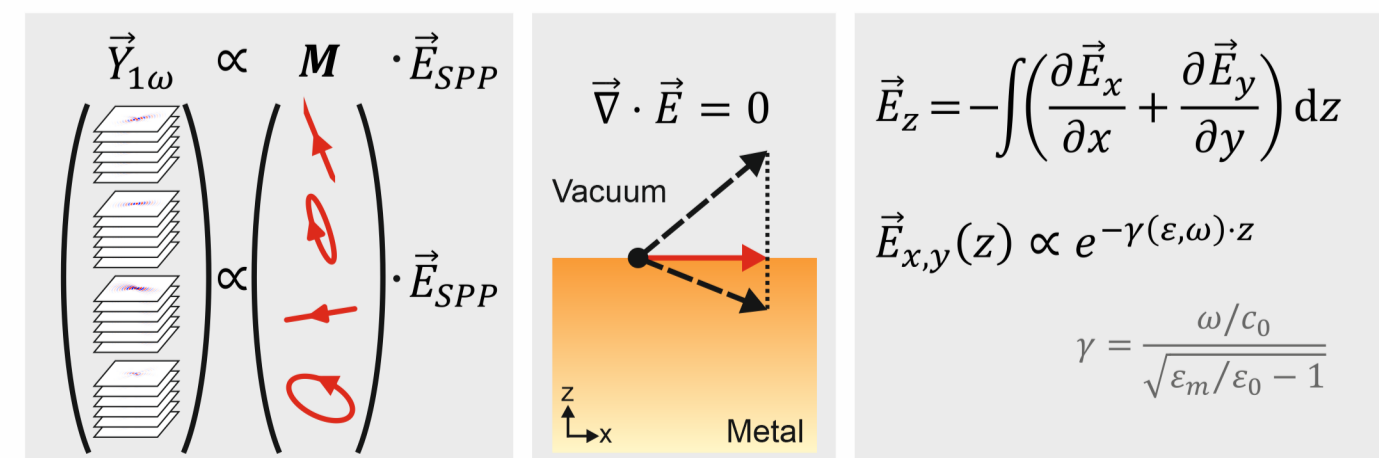
- Spectral interferences provide the phase of the Jones-vector of the probe pulses.
- The in-plane component of the field vectors can then be calculated from the data.

#### Correction of Temporal Overlap



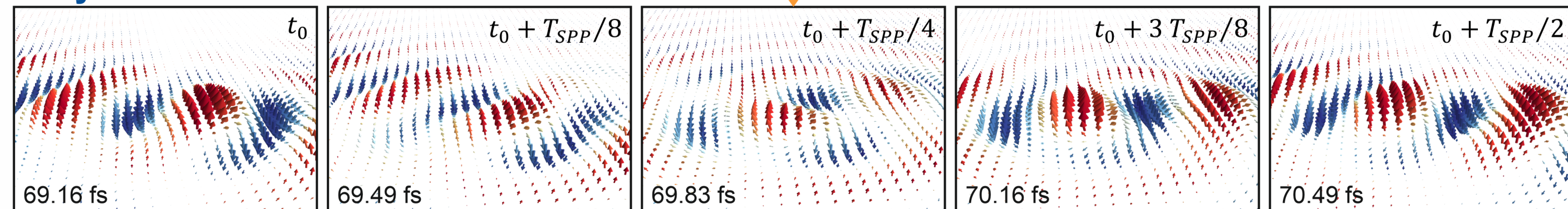
- The z-component of the field vector can be reconstructed (above the surface) using the Maxwell equation  $\vec{\nabla} \cdot \vec{E} = 0$ . The numerical integration is easily possible since the SPP is an evanescent wave.

#### Vector Reconstruction



- The result is a full vector reconstruction with sub-wavelength spatial and sub-femtosecond temporal resolution.

#### Fully Reconstructed Vectors

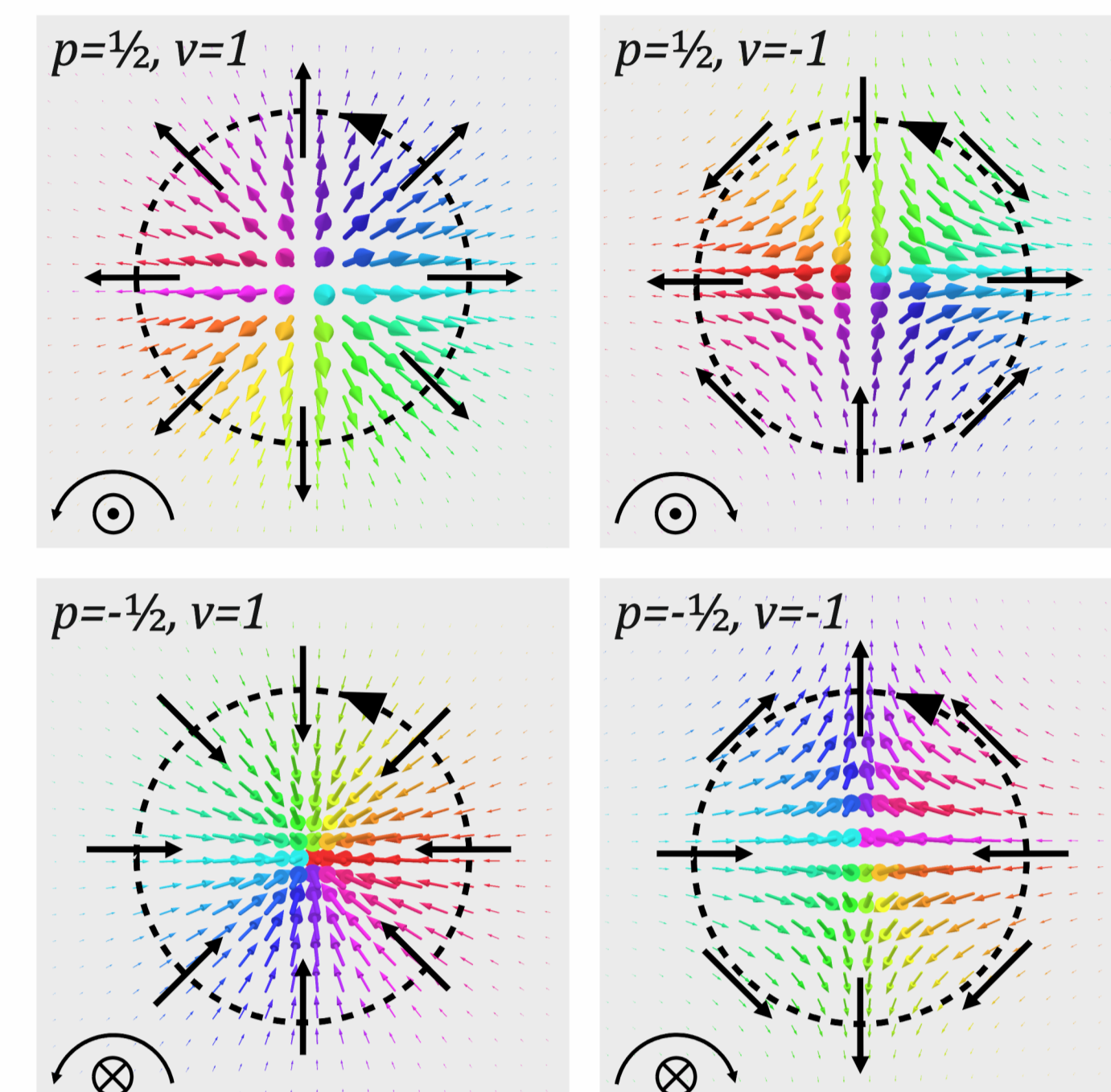


## Analysis of Topological Fields

- Nanoplasmonic topology can be described by C-points and L-lines in the spin-angular momentum density

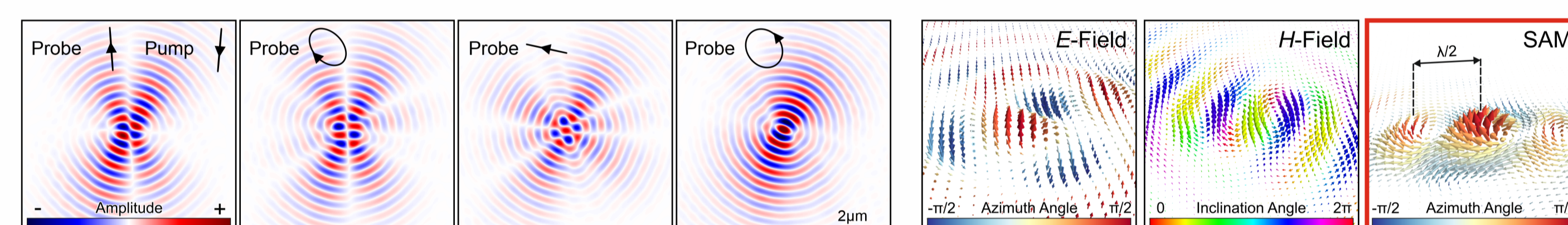
$$\vec{S} = \frac{1}{2\omega} \cdot \text{Im}(\epsilon \vec{E}^* \times \vec{E} + \mu \vec{H}^* \times \vec{H})$$

- C-points are isolated locations where  $\vec{S}$  points directly into ( $p=-1/2$ ) or out of the surface ( $p=+1/2$ ).
- L-lines are closed loops where  $\vec{S}$  lies within the surface plane.
- The vorticity  $v$  depends on how often and with what handedness  $\vec{S}$  rotates along an L-line. The vorticity is connected to the C-points within the region.
- The Chern number  $C = p \cdot v$  follows from both polarity and vorticity.



### Plasmonic Meron

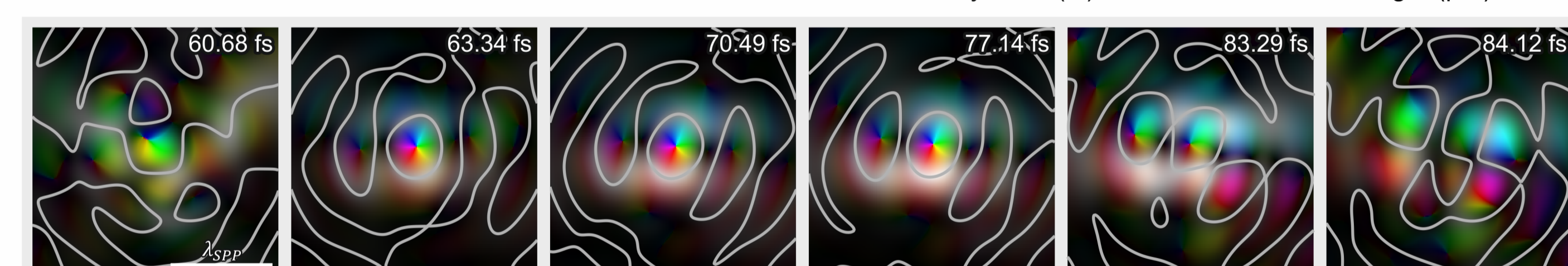
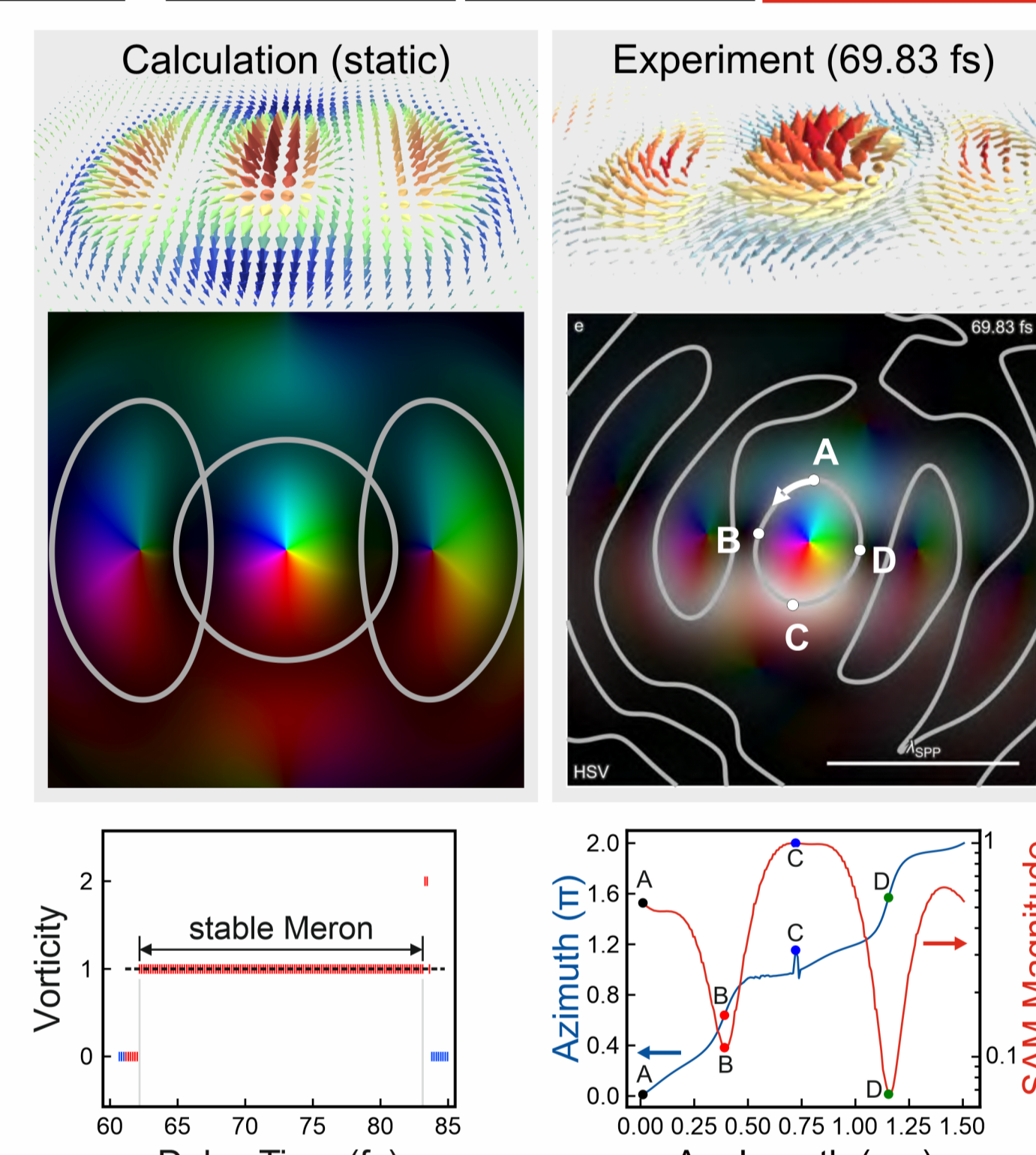
- Exciting an L=2 Archimedean spiral with linear polarized light yields a SPP Meron.
- 1 $\omega$ -filtering of data recorded with different probe polarizations yields the projection of the SPP on the probe pulse.



- To visualize the topology,  $\vec{S}$  is plotted in a HSV color scale:
- H: azimuth angle of  $\vec{S}$
- S: sine of polar angle ( $|\vec{S}_\perp| / |\vec{S}|$ )
- V: relative length  $|\vec{S}|$

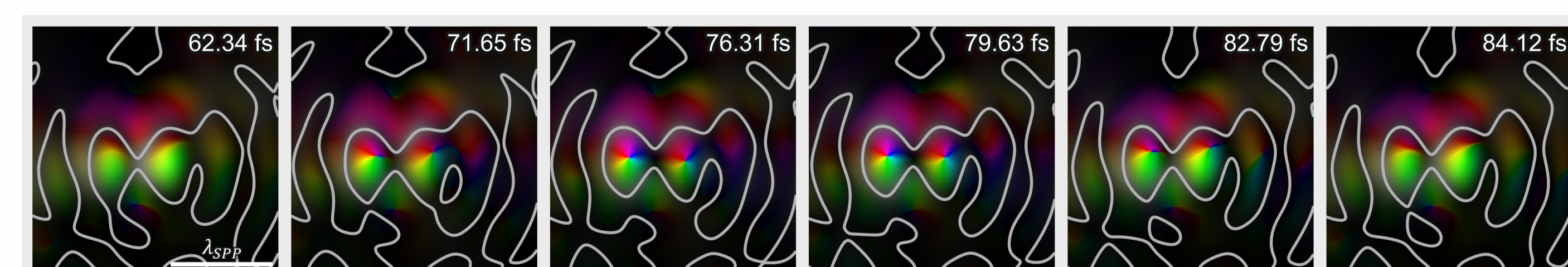
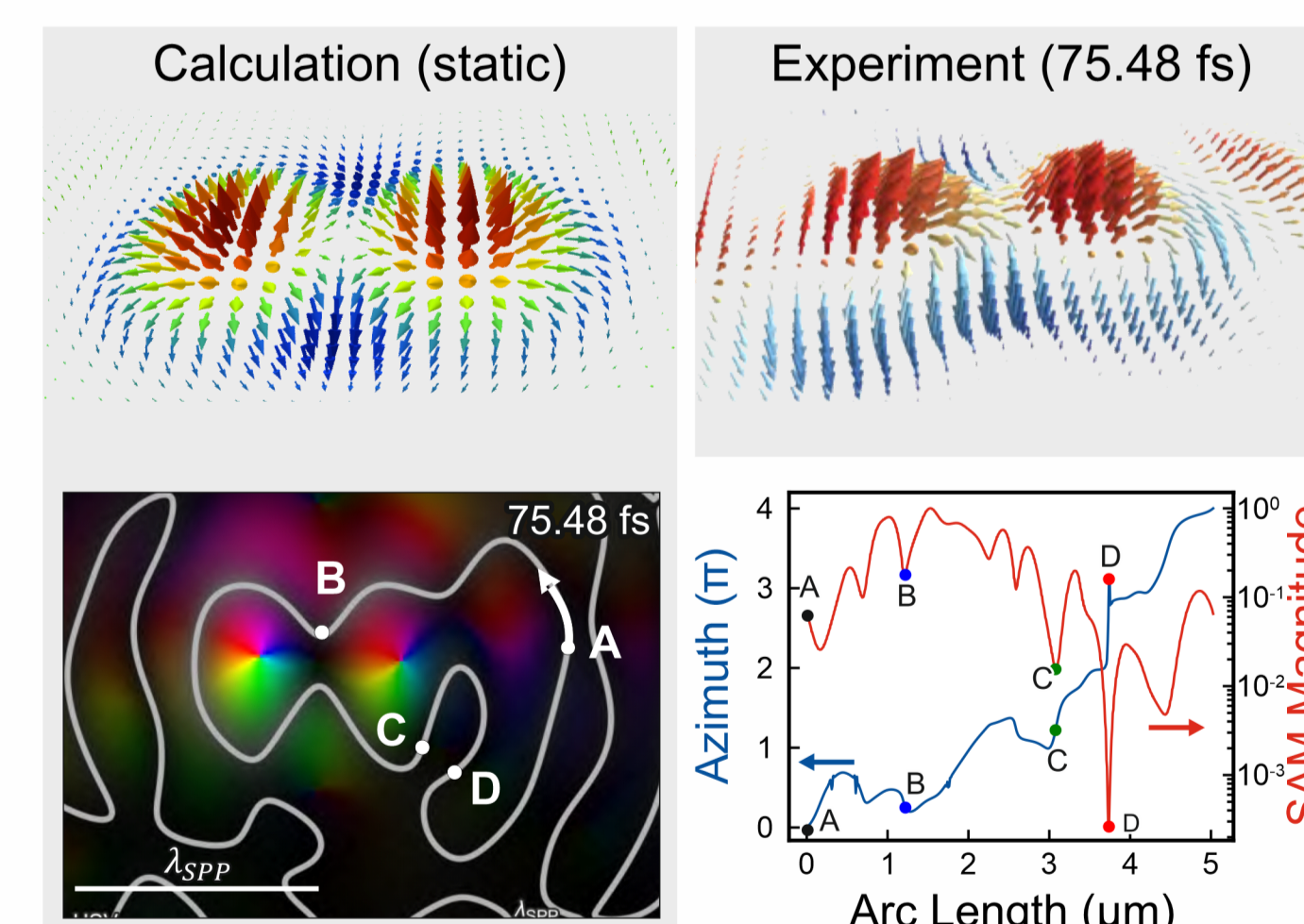


- C-points, L-lines and the relative azimuth of  $\vec{S}$  can be determined experimentally.
- With a polarity  $p=+1/2$  and a vorticity  $v=1$  the Meron's Chern number is  $C=0.5$ .
- The Meron is stable for more than 20 fs.



### Plasmonic Meron Pair

- The same analysis can be applied to a SPP Meron pair, where two Merons with the same polarity and vorticity are next to each other.
- Such Meron pair can be created by exciting an L=1 spiral with linearly polarized light.
- The polarity is  $p=+1/2$  for both Merons comprising the Meron pair.
- The total vorticity is  $v=2$  resulting in  $C=1$ .
- The Meron pair is stable for more than 10 fs.



## Conclusion

- Vector polarimetry is an excellent tool to reconstruct the electric and magnetic field of SPPs.
- Using vector polarimetry it is possible to measure the spin angular momentum of an SPP even in complex field situations with femtosecond time and deep-subwavelength spatial resolution.
- After calculating the spin angular momentum it is possible to determine L-lines and C-points for SPP Merons and Meron pairs and verify experimentally that they are topologically different.