SUPPLEMENTARY INFORMATION: Optical Spin Polarization by Coherent Magnetoabsorption Generation

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Supplementary information 1 | High-resolution XRD for determining Sb content

The X-ray diffraction (XRD) measurements were performed using the Philips X'pert MRD diffractometer equipped with a four-bounce Ge(220) monochromator, 1.8 kW Cu Kα1 X-ray tube, and a Pixel detector.

The XRD analysis of the samples structure was performed by measuring the symmetrical ω/2θ scans in the vicinity of (004) of GaAs substrate, which are then compared with simulated diffraction curves based on dynamical theory. In Fig. S1, the strongest peak on the XRD profile corresponds to the bulk GaAs substrate, and the peak related to the $GaAs_{1-x}Sb_x$ QW is seen as a broad shoulder at the lower angles from the substrate peak. The dense oscillation on the XRD profile reflects the thickness of the GaAs top layer.

Assuming the absence of extended defects, our fitting indicates that the shape of the $GaAs_{1-x}Sb_x$ peak is greatly influenced by the QW layer thickness. Additionally, due to the pseudomorphic growth,

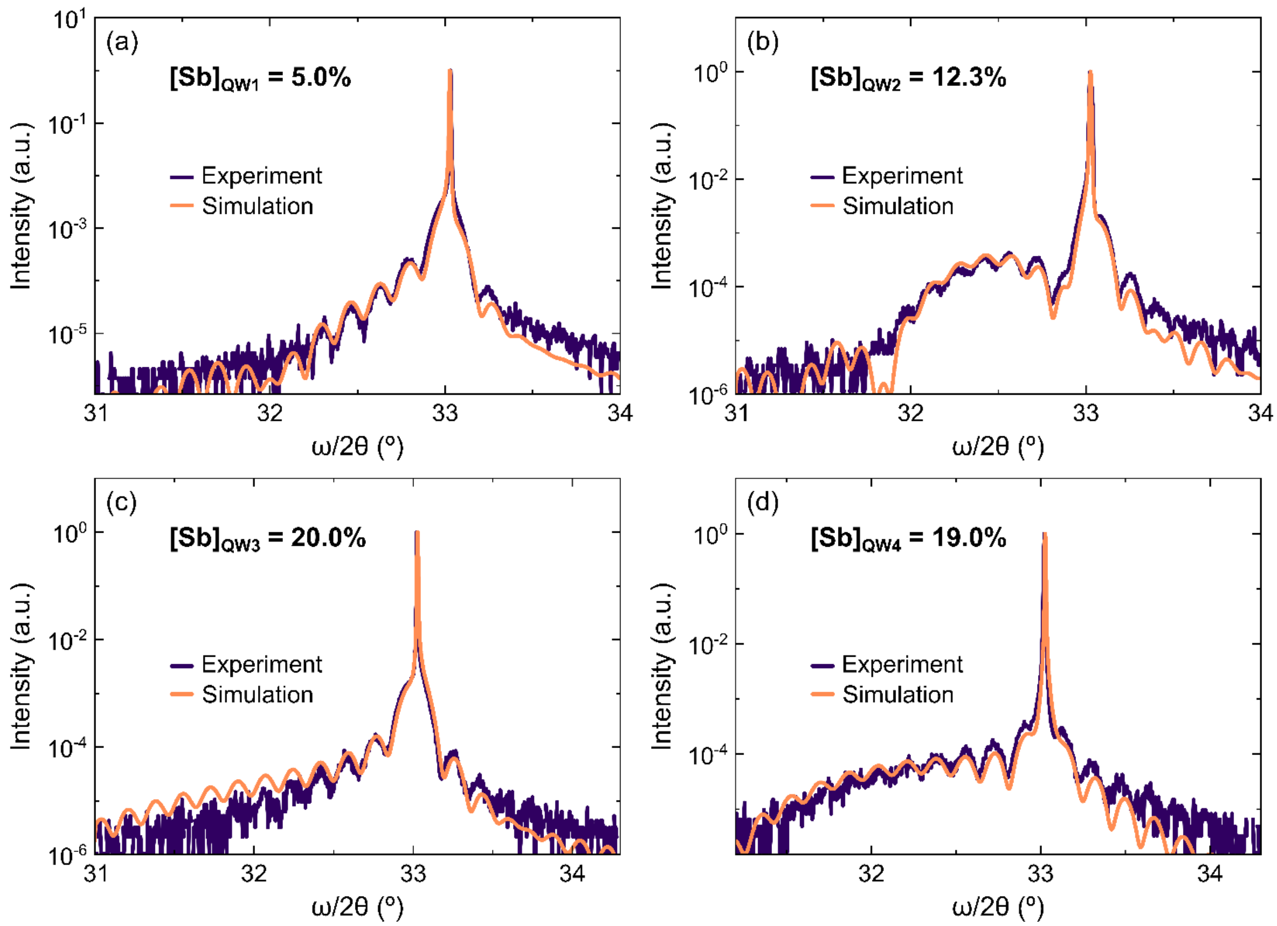

FIG. S1. ω/2θ curves from X-ray diffraction and respective simulations for QW1-4 to determine the Sb content in the $GaAs_{1-x}Sb_x$ layer.

the position of the peak $GaAs_{1-x}Sb_x$ is solely determined by fitting the Sb content. Finally, we estimate the thickness of the top GaAs layer by fitting the separation between the densely spaced oscillations.

Supplementary information 2 | Zeeman splitting from ground-state emission in type-II QW

The energy splitting from the circular components, σ_+ and σ_-, of QW1 PL emission versus magnetic field is shown in Fig. S2, where we obtain an effective *g*-factor of -2.3 by fitting the data with the linear Zeeman splitting: $E_Z = \mu_B g_{eff} B$.

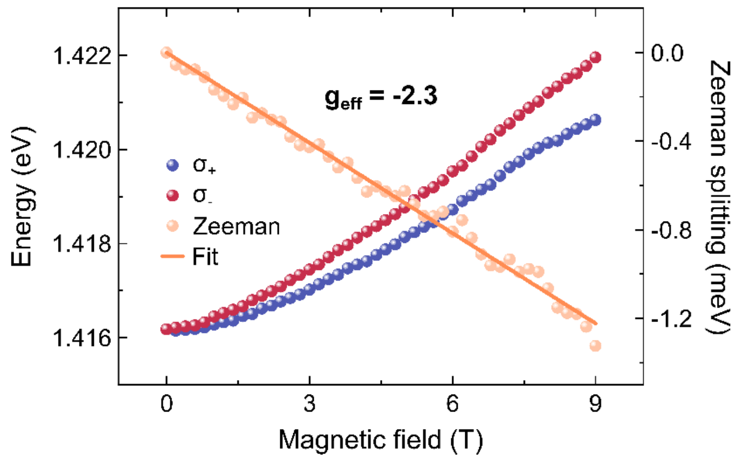

FIG. S2. Energies (left) from the circular components, σ_+ and σ_-, of QW1 PL emission versus magnetic field and respective Zeeman splitting (right) with linear fit at 3.7 K, 1.6 μW, and 1.579 eV excitation.

Supplementary information 3 | Light absorption in the $GaAs_{1-x}Sb_x$/GaAs QWs

In order to investigate the influence of each region (barriers and QW) on light absorption, PL measurements for QW1 with excitation energies above and below the GaAs bandgap are shown in Fig. S3. The results demonstrate a significant decrease of the PL intensity by changing the excitation line from 1.579 to 1.494 eV, supporting that the GaAs barriers dominate light absorption in our structure while the $GaAs_{1-x}Sb_x$ QWs play a minor role. Besides the intensity variation, no changes on the PL shape are observed.

Supplementary information 4 | Spin polarization model without magnetoabsorption

Our theoretical approach for determining the degree of circular polarization (DCP) in the ground-state optical recombination in the type-II $GaAs_{1-x}Sb_x$/GaAs QW is described by terms linked to spin-dependent generation rates, P_+ and P_-. In the main text, we make use of these terms to connect the spin polarization to magnetoabsorption features. Then, assuming non-conservation of the spin

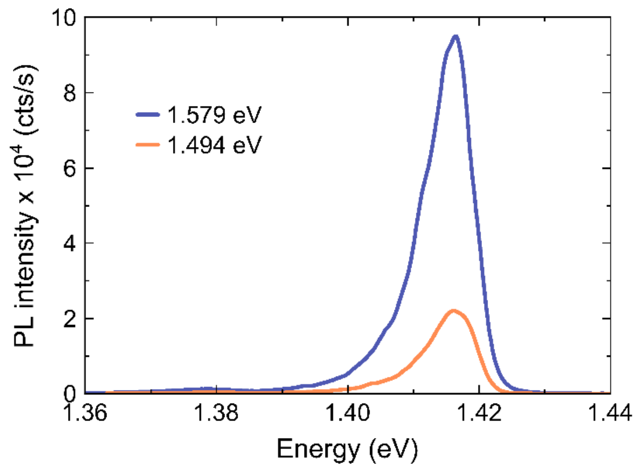

FIG. S3. PL spectra for QW1 at 3.7 K, power of 1.6 μW and 1.579 and 1.494 eV excitation energies.

projection, P_+ and P_- should be equal and we may reproduce the results in the cases without magnetoabsorption spin polarization, as shown in Fig. S4.

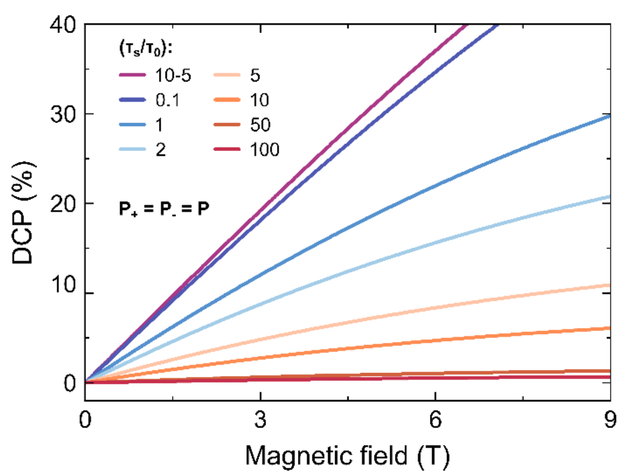

FIG. S4. Theoretical DCP for different ratios between spin-flip and optical recombination times.

Here, the DCP comes only from the thermalization between the Zeeman split states and changing the ratio between spin-flip and optical recombination times only modifies the magnitude of the spin polarization.