Introduction. The stacking fault (SF), a planar, atomically thin defect, is one of the most common extended defects in zinc-blende, wurtzite, and diamond semiconductors. A fundamental understanding of the SF potential is important for determining how the defect affects semiconductor device performance [1–2], engineering heterostructures based on crystal phase [3–5], and providing a new two-dimensional (2D) platform for fundamental physics [6, 7]. Here we report on excitons bound to large-area, single SFs in high-purity GaAs, a unique system where SFs are easily isolated with far-field optical techniques. The atomic smoothness of the potential and extreme perfection of the surrounding semiconductor result in ultra-high optical homogeneity (≤80 µeV). This enables optical resolution of the SF exciton fine-structure and thus direct measurement of the giant built-in dipole moment (≥e⋅10 nm) via the magneto-Stark effect. These results indicate that the extremely homogeneous SF potential may be promising for studies of many-body excitonic physics, including coherent phenomena [8–10], spin currents [11], superfluidity [12], long-range order [13–17], and large optical nonlinearities [18–20].

Stacking fault photoluminescence. Figure 1(a) shows a spectrally resolved confocal scan of SF structures in a GaAs epilayer, excited with an above band-gap laser (1.65 eV, 1.5 K) [21]. The image is colored red, green or blue according to three characteristic emission bands shown in Fig. 1. The narrow-band PL at 1.493 and 1.496 eV originates from excitons, electron-hole pairs, bound to the 2D SF potential [22, 23]. The sample consists of a 10 µm GaAs layer on 100 nm AlAs on a 5 nm/5 nm AlAs/GaAs (10×) superlattice grown directly on a semi-insulating (100) GaAs substrate. Stacking fault structures nucleate near the substrate-epilayer interface during epitaxial growth [21].

The physical origin of the potential can be understood from the atomic structure of the SF defect: the lattice-plane ordering in the [111] direction of zinc-blende is modified by subtracting a layer (intrinsic SF, see Fig. 1(f)) or adding a layer (extrinsic SF). The intrinsic SF can be viewed as a monolayer of wurtzite (AB AB stacking) surrounded by zinc-blende (ABC ABC stacking) [3, 24]. Due to the band offset [25, 27] and spontaneous polarization at the stacking fault [28], electrons and/or holes are attracted to the SF plane. While useful for physical motivation, this bulk phase change model must be taken with caution when applied to atomically thin SFs, which can deviate from simple theory [29]. Here, however, we find that single SFs in bulk GaAs bind excitons, confirming that the potential is attractive for at least one carrier.

In the confocal scan in Fig. 1(a), most of the SF defects appear as single triangles, which we identify as a pair of nearby SFs [30, 31]. Because the binding energy of excitons to a pair of SFs depends on the distance between the SFs [32], the PL emission energy from excitons bound to these structures has a high variability of 10 meV between structures. Strikingly, this inhomogeneity disappears when four SFs grow in an inverted pyramid structure consisting of four well-isolated {111} SF planes [Fig. 1(b)], which we refer to as up, down, left and right [33]. The full width at half-maximum (FWHM) of the SF PL line in our sample is (77±19) µeV at zero magnetic field [21], somewhat narrower than excitonic lines associated with stacking faults in previous work [22, 34]. In comparison, the narrowest reported linewidth for a GaAs/AlGaAs quantum well is 130 µeV [35], while PL linewidths from analogous zinc-blende/wurtzite quantum discs in nanowires range from 0.6–10 meV [27, 36–38]. This unprecedented homogeneity allows us to resolve the SF-bound exciton fine structure.

Nature of hole in SF exciton. Experimentally, we determine that the SF exciton is composed of an electron and a heavy-hole using polarization resolved PL, consistent with the atomic-scale symmetry of the system [21]. For linearly polarized light incident from above (along the [001] axis), the largest overlap between the light polarization and the in-SF-plane heavy-hole dipole occurs when exciting and collecting along the H direction for the down SF [Fig. 1(d)], in agreement with our experimental data [Fig. 1(f)]. On the
other hand, the main dipole moment for the light-hole exciton is along the SF normal, which would give rise to a maximum signal at V polarization, contrary to what is observed. Further, we also note that no hole Zeeman splitting is observed for in-plane magnetic fields B up to 7 T (Fig. 2). This observation is fully consistent with our symmetry analysis, which finds that B-linear splitting in in-plane fields is forbidden for heavy-holes but allowed for light-holes [21]. The substantial separation of the heavy- and light-hole states prevents their magnetic-field induced mixing, in line with experiments on GaAs nanowires [21, 39].

Based on the C3v point symmetry of the SF and time reversal invariance, the effective Hamiltonian for an exciton moving in the presence of an in-SF-plane magnetic field B is

\[ \mathcal{H}_{KB} = \frac{g_e}{2}\mu_B(\sigma_xB_x + \sigma_yB_y) + \beta B^2 + \beta' [K \times B], \]

where \( g_e \) is the electron g-factor, \( \mu_B \) is the Bohr magneton, \( \sigma_{x,y} \) are the electron spin Pauli matrices, \( \beta \) is a parameter describing the excitonic diamagnetic shift, and \( \beta' \) is a constant responsible for the non-reciprocal effect [21]. In Eq. (1) we only retain 1st- and 2nd-order terms in B and use a frame of axes related to the SF plane: \( z \parallel [11\bar{2}] \) is the SF normal, \( x \parallel [11\bar{1}] \) and \( y \parallel [\bar{1}10] \). Each symmetry-derived term in Eq. (1) manifests itself in the energetic shift of the SF PL lines with magnetic field (Fig. 3). The first term is the electron Zeeman effect and gives rise to the doublets visible at \( \pm 7 \) T, since an electron with a particular spin projection can recombine with the corresponding hole. The second term is the exciton diamagnetic shift, arising from the magnetic-field-induced shrinking of the exciton wavefunction [41]. The last term is the magneto-Stark effect, which, as we show below, quantitatively explains the non-reciprocal PL spectra.

The experimental geometry, Fig. 4(b), is such that only light emitted normal to the sample surface is collected. For
a high quality 2D potential, in-plane exciton momentum is transferred to the photon during recombination, as depicted in Fig. 3(a). This conservation of momentum implies

\[ K_x = \frac{\omega n}{c} \sin \theta'' , \]  

(2)

where \( \theta'' \) is the angle between the SF normal and the emitted photon momentum inside the semiconductor, Fig. 3(a), \( \omega \) is the photon frequency, \( n \) is the refractive index and \( c \) the speed of light. Thus, the collected SF PL arises only from excitons with a specific center of mass momentum \( \mathbf{K} \). The last term in Eq. (1) provides, for a fixed \( K_x \) (Eq. 2), an odd in \( B_y \) contribution to the overall PL energy shift, giving rise to a magnetic non-reciprocity effect. It is worth noting that the up and down SFs are related by a mirror reflection in the (110) plane and such a reflection is accompanied by \( B_y \rightarrow -B_y \), resulting in the opposite behavior of up and down PL spectra observed in Fig. 2(a).

**Magnetostark effect.** The physical origin of the non-reciprocal PL is the magneto-Stark effect, the interaction of a moving exciton’s electric dipole moment with a magnetic field \( \mathbf{B} \). The effect can be understood with a relativistic argument: motion with velocity \( \mathbf{v} = (\hbar K_x/M)\mathbf{z} \) through a magnetic field \( \mathbf{B} = B_y\mathbf{z} \) gives rise to an electric field \( \mathbf{E} = \hbar K_x B_y/M \mathbf{z} \) in the moving frame of reference, where \( M \) is the exciton mass in translational motion and \( c \) the speed of light. Since for the SF, \( \mathbf{z} \propto [111] \) and \(-\mathbf{z}\) directions are not equivalent, the SF-bound exciton has a non-zero dipole moment \( \mathbf{p} = e\hbar c \mathbf{z} \), where \( e = |e| \) is the elementary charge, and \( \hbar c \) is the average separation between the hole and electron along the \( z \)-axis. The Stark effect \( H_S = -\mathbf{p} \cdot \mathbf{E}_{\text{eff}} \) in the exciton’s reference frame thus becomes the magneto-Stark effect:

\[ H_S = -\frac{\hbar}{M} e\hbar c K_x B_y , \]  

(3)

in agreement with Eq. (1) with \( \beta' = e\hbar c/(M \hbar c) \), see Ref. [21] for formal derivation.

Physically, the dipole moment of a SF bound exciton is a consequence of symmetry breaking and spontaneous polarization similar to that in zinc-blende/wurtzite heterostructures [23-25]. The hole in the exciton is presumably localized in the SF plane while the electron is weakly bound via the Coulomb interaction. The spontaneous polarization shifts the electron cloud to one side of the SF, resulting in a giant excitonic dipole moment.

Equations (1)-(3) predict that the asymmetric energy shift of exciton PL is linearly related to the in-plane wavevector \( \mathbf{K} \). Since the angle of light collection determines the exciton momentum [Eq. (2)], we test the applicability of the model by recording spectra of the up and down SFs as a function of the collection angle \( \theta \) and magnetic field \( B_y \) [Fig. 3(b)]. The collection angle is related to the emission angle from the up/down SF by \( \sin \theta = n \sin \theta' = \pm n \sin (\theta'' - \theta_{\text{SF}}) \), where \( \theta_{\text{SF}} \) is the angle the SF normal \( \mathbf{z}[111] \) makes with [001] [Fig. 3(c)].

In this experiment, we modified the collection angle by mounting the sample at different angles. Since the sample was removed from the cryostat to change the angle, different SF pyramids were used at different angles. This does not introduce artifacts because of the extreme similarity of different SFs, which have a standard deviation of line-center energies of only 57 \( \mu \)eV, less than the linewidth. Spectra were acquired with \( B_y \) ranging from \(-6.5 \) T to 6.5 T on the up and down SFs. We fit the spectra to one or a sum of two Voigt function(s) depending on whether the electron Zeeman splitting is resolved. The singlet or doublet line center is denoted \( E_{\text{up/down}}(B_y) \). The part of the exciton energy odd with magnetic field is found by computing

\[ \Delta E_{\text{up/down}}(B_y) = E_{\text{up/down}}(B_y) - E_{\text{up/down}}(-B_y) \]  

(4)

It follows from Eq. (3) that the asymmetric shift is

\[ \Delta E_{\text{up/down}}(B_y) = \frac{2n\hbar e d_{he}}{M} c \sin(\theta_{\text{SF}} \pm \theta') B_y . \]  

(5)

Thus the proportionality constant of \( \Delta E_{\text{up/down}} \) vs. \( B_y \) provides a measurement of the SF exciton’s built-in dipole moment. The experimental values and first-order theory for \( \Delta E \) are shown in Fig. 3(e)-(g). Further, the ratio

\[ r(\theta) = \frac{|\Delta E_{\text{up}}| - |\Delta E_{\text{down}}|}{\frac{1}{2} (|\Delta E_{\text{up}}| + |\Delta E_{\text{down}}|)} \]  

(6)

depends (to first order in \( B_y \)) only on the experimental geometry and the index of refraction: \( r(\theta) \) vanishes for collection angle \( \theta = 0 \) and increases as a function of \( \theta \) [Fig. 3(h)]. We obtain good agreement between \( r(\theta) \) calculated experimentally from the \( B = 0 \) slope of \( \Delta E \) without any fit parameters [Fig. 3(h)].

Further, by fitting \( \Delta E_{\text{up/down}}(B_y) \) with a \( B_y \)-linear function, we can estimate the dipole moment of the exciton \( p = e d_{he} c \cdot (17-20) \) nm. The main uncertainties result from the accuracy of the \( B_y \)-linear fit and the value of the in-(111)-plane heavy-hole mass, which depends on the details of the SF potential [21]. The exciton mass can be roughly estimated as 0.17 \( m_o \), the sum of the bulk-GaAs in-(111)-plane heavy-hole mass and the isotropic electron mass, where \( m_o \) is the free electron mass. In addition, we note the magneto-Stark induced splitting saturates at high fields [Fig. 3(f,g)], possibly due to a decreased exciton dipole moment from the magnetic-field-induced shrinking of the exciton wavefunction. Future work will investigate exciton confinement potentials consistent with the observed dipole moment, diamagnetic shift and saturation of the magneto-Stark effect.

A microscopic understanding of the confinement potential may enable predictions for the binding potential and excitonic dipole moment for SFs in other semiconductors.

**Conclusion.** We have shown that SFs in GaAs are an almost perfect 2D potential which binds heavy-hole excitons. These excitons freely propagate in the SF plane, a conclusion confirmed via the magneto-Stark effect. Further, an asymmetry of the SF potential induces a giant dipole moment of the SF-bound exciton. Such excitons could be useful for studying the many-body physics of interacting dipoles. In conventional excitonic systems, typical electron-hole separations are on the order of several nm [6, 46], whereas the SF-bound exciton has a gigantic electron-hole separation of 10 nm and the possibility to modify this value with an
FIG. 3. (a) Because of conservation of in plane momentum during exciton recombination, the angle of light emission depends on the exciton wavevector. Collecting different angles probes different exciton momenta. The SF has a built in potential that creates a zero-field dipole moment for the SF exciton. In the exciton frame of reference, the in-plane magnetic field becomes an out-of-plane electric field, leading to the magneto-Stark effect. (b) Spectra of up and down SFs as a function of θ and in-plane magnetic field $B_y$. (c) Light from the up SF originates from excitons with larger $K_x$ than light from the down SF (for $\theta > 0$). (d-e) Spectra of up and down SF at positive and negative $B_y$ for $\theta = 0^\circ$ and 43°. At $\theta = 0^\circ$, $\Delta E_{up}$ and $\Delta E_{down}$ have the same magnitude, while for $\theta = 43^\circ$, the magnitude of $\Delta E_{up}$ is larger than $\Delta E_{down}$. (f-g) Splitting $\Delta E_{up/down}$ as a function of magnetic field. Data are obtained from Voigt fits to spectra similar to those shown in d-e. Solid lines are a fit to $\Delta E = aB$ for the first three data points. (h) The ratio of $B = 0$ slopes, Eq. (6), depends only on geometrical constraints. The theory (solid line) has no adjustable parameters. Data for other angles in 21.

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Supplemental Materials: Fundamental properties of 2D excitons bound to single stacking faults in GaAs

S1. STACKING FAULT FORMATION

Stacking fault (SF) structures can grow from the substrate-epilayer interface during epitaxial growth. In the present work, SFs form in a 10 µm GaAs layer grown by molecular beam epitaxy with room temperature electron density $n \sim 1.9 \times 10^{14} \text{ cm}^{-3}$ and mobility $\sim 7400 \text{ cm}^2/\text{Vs}$. The entire structure consists of the 10 µm GaAs layer on 100 nm AlAs on a 5 nm/5 nm AlAs/GaAs (10×) superlattice grown directly on a semi-insulating (100) GaAs vertical gradient freeze substrate (Wafer Technology Ltd), started with AlAs. The sample was grown at a pyrometer temperature of 600°C with the relatively low As$_4$ beam equivalent pressure of $8 \times 10^{-6}$ Torr, measured by a flux tube. The growth rate was 0.7 ML/s for the GaAs and 0.35 ML/s for the AlAs. Oxide removal before growth was performed at 620°C under As flux. This growth procedure resulted in oval defects in the sample surface, a feature commonly associated with stacking faults [22, 33, 47, 48]. We observe two types of stacking fault defects, a SF pyramid and a SF pair defect, shown in Fig. S4. The size of the pyramid structure (14.1 µm top edge in Fig. 1a) is consistent with stacking faults [111] planes through the 10 µm thick epilayer.

![SF Pyramid and SF Pair Defect](image)

**FIG. S4.** Diagram showing the types of defects visible in the sample. The SF pyramid consists of four stacking faults arranged in a pyramid shape. The SF pair defect is a set of two SFs with a small ~nm separation.

S2. PHOTOLUMINESCENCE SPECTROSCOPY SYSTEM

The sample is excited with a continuous wave Sirah Matisse Ti:Sapphire laser. The laser is focused to a spot size of ~1 µm on the sample using an aspheric lens (numerical aperture 0.77) mounted inside a liquid helium immersion cryostat (Janis). Coarse positioning of the sample was performed with slip-stick positioners (Attocube). For confocal scanning, spatial selectivity is achieved with a pinhole in the intermediate image plane and a scanning mirror to raster the excitation and collection spot over the sample. The photoluminescence (PL) is imaged on a spectrometer (Andor).

S3. STACKING FAULT PHOTOLUMINESCENCE LINEWIDTH

![Graph showing high resolution PL spectrum of SF](image)

**FIG. S5.** High resolution PL spectrum of up SF. The FWHM of the line is (77 ± 19) µeV from a weighted Lorentzian fit, taking into account the instrument resolution. Before deconvolution the FWHM is (94 ± 13) µeV. Excitation at 1.65 eV, 1.44 K, 1.4 nW.

Figure S5 shows a high resolution PL spectrum of the SF. In order to extract the true PL linewidth, we need to take into account the spectral resolution of our setup. The spectrometer instrument resolution is found by taking a spectrograph of a narrow band Ti:Sapphire laser and fitting to a Voigt function, see Fig. S6(a). We find that neither a Gaussian nor a Lorentzian accurately describe the spectral point spread function, so we use a Voigt fit. For spectral lines that are nearly as narrow as the spectrometer FWHM (full width at half maximum), the measured FWHM will be wider than the true FWHM. Figure S6(b) shows the FWHM of a line obtained by the convolution of a Lorentzian or Gaussian spectral lineshape with the spectrometer response function. For example, a measured linewidth of 94 µeV corresponds to a true linewidth of 72 or 82 µeV, depending on whether the true lineshape is assumed to be Lorentzian or Gaussian. Hence, to evaluate the intrinsic linewidth of the SF emission we fit the spectrum in Fig. S5 with a weighted Lorentzian and use the deconvolution procedure [Fig. S6(b)] to obtain an intrinsic PL linewidth of only (77 ± 19) µeV. Here, the uncertainty combines the original fit uncertainty and the uncertainty of the deconvolution procedure.

S4. MAGNETO-STARK HAMILTONIAN

Microscopically, the magneto-Stark effect can be derived from the Hamiltonian for an electron and hole in a magnetic field $\mathbf{B} = B_y \hat{y}$:

$$H = \frac{\hbar^2}{2m_e} \left( \mathbf{k}_e + \frac{e}{\hbar c} \mathbf{A} \right)^2 + \frac{\hbar^2}{2m_h} \left( \mathbf{k}_h - \frac{e}{\hbar c} \mathbf{A} \right)^2,$$

where $\mathbf{k}_e$ ($\mathbf{k}_h$) is the wavevector of the electron (hole), $m_e$ ($m_h$) is the effective mass of the electron (hole) and the vector potential is $\mathbf{A} = B_y z \hat{x}$. The Coulomb interaction...
FIG. S6. (a) Spectrum of the narrow band Ti:Sapphire laser used to determine the spectrometer instrument resolution. The best fit is a Voigt function with a 32.7 µeV FWHM. The Voigt lineshape is the convolution of a Lorentzian and a Gaussian with best fit widths provided as an inset in the figure. (b) Convolution of the spectrometer instrument response in a with a Lorentzian or Gaussian fit is a Voigt function with a 32 µeV FWHM. By interpolating backwards, the deconvoluted FWHM of a spectral line can be found.

between the electron and the hole as well as the SF potential are omitted in Eq. (S7) for brevity. We use the standard definition of center of mass (COM) and relative coordinates

\[ k_e = \frac{m_e}{M} K + k \]
\[ k_h = \frac{m_h}{M} K - k \]

where \( M = m_e + m_h \). With these substitutions, Eq. (S7) becomes

\[ H = \begin{pmatrix}
\frac{\hbar^2 K^2}{2M} + \frac{\hbar^2 k^2}{2\mu}
+ \frac{e \hbar}{Mc} (z_h - z_e) K_y B_y \\
\text{Orb. Zeeman}
+ \frac{e \hbar}{2c} \left( \frac{z_h}{m_h} + \frac{z_e}{m_e} \right) k_y B_y + \frac{e^2 B^2 y}{2c^2} \left( \frac{z^2}{m_e} + \frac{z^2_h}{m_h} \right)
+ \frac{e^2 B^2 y}{2c^2} \left( \frac{z^2}{m_e} + \frac{z^2_h}{m_h} \right)
\end{pmatrix} \]

where \( \mu = (m_e^{-1} + m_h^{-1})^{-1} \). The first term is the COM kinetic energy of the exciton. The second term is the kinetic energy associated with the electron-hole relative motion. The third term describes the magneto-Stark effect for the exciton COM motion. The fourth term describes the orbital Zeeman effect. The fifth term is the harmonic potential created by the magnetic field which produces the diamagnetic shift and would produce Landau quantization for higher magnetic fields. The COM magneto-Stark term is the same as Eq. (4) in the main text, derived from relativistic arguments.

S5. HEAVY HOLE – LIGHT HOLE SPLITTING

The possible carrier spin states entering into the SF exciton can be predicted on general symmetry considerations. Due to spin orbit coupling, the valence band in bulk GaAs splits into the heavy-hole/light-hole \((j = \frac{3}{2})\) bands and split-off \((j = \frac{1}{2})\) band. At the \( \Gamma \) point, the heavy-hole (HH) and light-hole (LH) bands are degenerate and transform according to the four-dimensional \( \Gamma_8 \) irreducible spinor representation of the \( T_d \) point symmetry group [49], see Refs. [50, 51] for notations.

A SF oriented in one of the \{111\} planes possesses the lower \( C_{3v} \) point symmetry, as illustrated in Fig. S7. The \( C_{3v} \) symmetry group contains six symmetry operations: \( E \) (identity), \( C_3 \) (rotation by \( 2\pi/3 \) about \( z \)), \( C_3^{-1} \) (rotation by \( -2\pi/3 \) about \( z \)), \( \sigma_v \) (reflection \( y \rightarrow -y \)), \( \sigma_{v'} \) \((\sigma_{v'} = \sigma_v C_3^{-1})\), and \( \sigma_{v''} \) \((\sigma_{v''} = \sigma_v C_3)\). These operations are depicted in Fig. S8. We note that the SF symmetry group is different from the \( C_{6v} \) symmetry of wurtzite [52].

When a SF is introduced into a zinc-blende crystal, the degeneracy of the valence and conduction band edges could

FIG. S7. GaAs crystalline lattice. Stacking fault plane is a \{111\} plane, shown as a triangle. The mirror reflection plane for a SF sends \( y \rightarrow -y \). Note that \( x \rightarrow -x \) is not a mirror reflection plane.
be lifted. The compatibility analysis shows that the conduction band edge transforms according to the two-dimensional irreducible representation $\Gamma_4$ of the $C_{3v}$ point group, i.e. the conduction band does not split. On the other hand, the degeneracy of the valence band is lifted: the $\Gamma_8$ irreducible representation of $\Gamma_d$ decomposes into $\Gamma_4 \oplus \Gamma_5 \oplus \Gamma_6$ of $C_{3v}$. The two degenerate states $\Gamma_4$ transform as the spinors $|3/2,1/2\rangle, |3/2,-1/2\rangle$ (or $|1/2,1/2\rangle, |1/2,-1/2\rangle$), where $|j,m_j\rangle$ signifies the basic function for angular momentum $j$ and angular momentum $z$-component $m_j$. The $\Gamma_5$ and $\Gamma_6$ hole states transform as linear combinations of the spinors $|3/2,3/2\rangle$ and $|3/2,-3/2\rangle$, see Table I. At $B = 0$ and $K = 0$, the two states $\Gamma_5$ and $\Gamma_6$ are guaranteed to be degenerate by time reversal symmetry \textsuperscript{53,55}. It follows that the levels of excitons bound to a SF are split into sublevels of symmetry $\Gamma_4^x \times \Gamma_4^y$ and $\Gamma_4^X \times (\Gamma_5^v \oplus \Gamma_6^v)$ where the superscripts $c, v$ refer to the conduction and valence bands.

For convenience, the former and latter excitonic states are called the light- and heavy-hole excitons, or the LH and HH excitons, irrespective of the relation between the exciton binding energy and the splitting $\Delta_{HL}$ between the HH and LH sublevels.

The experimental data imply that the in-plane hole $g$-factor in the ground exciton state is negligible. This is exactly true for the HH exciton $\Gamma_4^x \times (\Gamma_5^v \oplus \Gamma_6^v)$ at zero magnetic field $B_y$ and zero wavevector $K_y$. This implies that the splitting $\Delta_{HL}$ is high enough to prevent the mixing between the LH and HH sublevels induced by the finite value of $K_y$ realized in the experiment and by the applied magnetic field, $|B| \leq 7$ T.

In order to confirm that the SF splits HH and LH states, we can estimate whether the HH-LH splitting is much greater than interactions which mix HH and LH. The magnetic-field-induced HH-LH mixing can be estimated by using the hole Zeeman Hamiltonian

$$\mathcal{H}_Z = -2\mu_B \varepsilon (\mathbf{J} \cdot \mathbf{B}),$$

(S10)

where $\mu_B$ is the Bohr magneton, $\varepsilon$ is the magnetic Luttinger parameter on the order of unity, $\mathbf{J} = (J_x, J_y, J_z)$ are the matrices of the angular momentum 3/2 operator, and we neglect a weak cubic anisotropy \textsuperscript{56}. According to Eq. (S10), the magnitude of the Zeeman $\Gamma_4$-coupling matrix element at the maximum field of 7 T is $< 1$ meV. As for the HH-LH mixing caused by the nonzero value of $K_y$, we note that the maximum exciton wavevector measured in our system is 5 $\mu$m\textsuperscript{-1}. In this case the terms of the Luttinger-Kohn Hamiltonian that couple the HH and LH excitons have magnitudes less than 2 $\mu$eV \textsuperscript{49}. Thus, we conclude that no significant mixing occurs if the splitting $\Delta_{HL}$ is greater than a few meV. The lack of significant HH-LH mixing in our experiments is in agreement with the HH-LH splitting of 16 meV estimated for interface excitons in polytypic zincblende/wurtzite GaAs nanowires \textsuperscript{39}.

In order to determine the dipole moment of the SF exciton using Eq. \textsuperscript{5} it is necessary to estimate the in-plane exciton effective mass, the sum of the electron and hole in-plane effective masses. While the electron mass is isotropic in GaAs, the effective in-plane hole mass depends on the detailed nature of the stacking fault potential. We note here that HH-LH mixing can affect the in-plane HH effective mass \textsuperscript{58}. Using second-order perturbation theory, we obtain:

$$\frac{m_0}{m_{hh,||}} = \gamma_1 + \gamma_3 + (4\gamma_2^2 + 2\gamma_3^2)\frac{\hbar^2}{m_o} \sum_n \frac{|(LH,n)|\hat{k}_z|HH,\rangle^2}{E_{HH} - E_{LH,n}}$$

(S11)

where the summation goes over all the light-hole states $n$ (both bound and continuum), $|HH,\rangle$ denotes the ground subband HH envelope function along the $z$ axis and $|LH,\rangle$ denote the LH envelopes and $\hat{k}_z = -\partial / \partial z$ and $\gamma_1, \gamma_2, \gamma_3$ are the Luttinger parameters \textsuperscript{49}. The sum in Eq. (S11) is sensitive to the details of the HH and LH envelope functions as well as to the energy positions of the size-quantized levels \textsuperscript{58,59}.

The estimate of the exciton dipole moment involves the exciton’s effective mass, $M = m_h^* + m_e^*$, see Eq. \textsuperscript{5}, where $m_h^*$ and $m_e^*$ are the in-plane effective masses of the hole and electron. The electron effective mass is isotropic and therefore its in-plane value is $m_e^* = 0.067 m_o$. Due to the anisotropy of the hole effective mass and the unknown extent of HH-LH coupling, determining the correct value of $m_h^*$ requires a complex calculation, which we do not perform here. However, we can estimate the HH effective mass using some simple arguments. If HH-LH coupling is neglected, the in-plane heavy hole mass is that of a heavy-hole in the (111) plane, $m_h^* = 0.10 m_o$ \textsuperscript{60}. Another estimate of the in-plane heavy hole mass can be made from the spatially averaged heavy-hole mass of 0.45 $m_e$. We will use the lower value to estimate the dipole moment, and the higher value to estimate the error in our measurement of the dipole moment. This conservative procedure yields the minimum possible value of the dipole moment.

S6. Hamiltonian by symmetry

The form of the Hamiltonian describing the SF-bound exciton state can be derived based on the symmetry of the SF system. Specifically, we would like to find terms of the Hamiltonian that are odd with magnetic field and can thus explain the magnetic non-reciprocity data.

It is first necessary to find how the symmetry operations affect a vector, such as a position vector $\mathbf{r} = (x, y, z)$, and a pseudo-vector, such as magnetic field $\mathbf{B} = (B_x, B_y, B_z)$. In free space, $\mathbf{r}$ and $\mathbf{B}$ transform according to the $D^+_1$ and $D^+_1$ irreducible representations, where $\pm$ denotes parity with respect to space inversion. Making use of the compatibility tables for $C_{3v}$ point symmetry, one can readily check that $D^+_1 = \Gamma_1 \oplus \Gamma_3$, while $D^+_1 = \Gamma_2 \oplus \Gamma_3$. Taking into account that under rotations the components of polar and axial vectors transform identically and making use of Fig. S8 we find that $\mathbf{r}$ and $\mathbf{B}$ transform according to the irreducible representations $\Gamma_1$ and $\Gamma_2$, respectively, while the pairs $(x, y)$ and $(B_x, B_y)$ form equivalent bases of the two-dimensional $\Gamma_3$ irreducible representation of $C_{3v}$, see Table I.

To apply the method of invariants, we need to establish transformation rules for the basic $2 \times 2$ matrices acting in the spin subspaces of electrons and heavy-holes. We
introduce basic electron matrices \( I^c \) (the 2 \( \times \) 2 unit matrix) and \( \sigma^x = (\sigma^x_1, \sigma^x_2, \sigma^x_3) \) (Pauli matrices) acting in the basis of \( |s = \pm 1/2 \rangle \) electron spinors. The decomposition \( \Gamma_4 \otimes \Gamma_4 = \Gamma_1 + \Gamma_2 + \Gamma_3 \) indicates the ways in which the basic electron spin matrices transform. By calculating the effect of the \( C_{3v} \) symmetry operators on the matrices, one finds that \( I^c \) is invariant, \( \sigma^x_1 \) transforms according to \( \Gamma_2 \) and \( (\sigma^x_2, \sigma^y_2, \sigma^z_2) \) form a basis of the two-dimensional irreducible representation \( \Gamma_3 \) with \( \sigma^x_2 \) and \( \sigma^y_2 \) transforming equivalently to \( B_x \) and \( B_y \) respectively (Tab. 1). The basic matrices for the light-hole doublet transform in exactly the same way.

By contrast, the heavy-hole spin doublet transforms according to the reducible representation \( \Gamma_5 + \Gamma_6 \). The direct product

\[
(\Gamma_5 \oplus \Gamma_6) \otimes (\Gamma_5 \oplus \Gamma_6)^* = 2\Gamma_1 + 2\Gamma_2.
\]

indicates that among the four basic matrices, \( I^{hh}, \sigma^{hh} = (\sigma^{hh}_x, \sigma^{hh}_y, \sigma^{hh}_z) \) acting in the space \( | \pm 3/2 \rangle \), two are invariant and two transform as \( B_z \) \([53]\). Taking into account that at the mirror reflection \( \sigma_v[110] \) the matrix \( \sigma^{yy}_v \) does not change sign, we find that \( I^{hh} \) and \( \sigma^{hh} \) transform according to \( \Gamma_1 \) (note that \( \sigma^{yy}_v \) changes its sign under time reversal whereas \( I^{hh} \) does not), while \( \sigma^{hh}_x, \sigma^{hh}_z \) transform according to \( \Gamma_2 \), see Tab. 1 and Ref. [53].

Using the transformation properties of the relevant basis functions (Tab. 1), we can build the effective Hamiltonian. Any valid term of the Hamiltonian must transform as the identity representation \( \Gamma_1 \) and be even under time reversal. In order to know which combinations of basis functions transform as \( \Gamma_1 \), we use the product rules for irreducible representations. From this information, we can build the linear in \( B \) Hamiltonian. For the electron, this analysis produces the result that the in-plane and out-of-plane \( g \)-factors are potentially different:

\[
H_{Z,e} = \frac{1}{2} g_{zz,e} \mu_B B_z \sigma^e_z + \frac{1}{2} g_{yy,e} \mu_B (B_x \sigma^e_x + B_y \sigma^e_y)
\]

(S13)

For the heavy holes, the Hamiltonian takes the form [50]:

\[
H_{Z,hh} = \frac{1}{2} \mu_B B_z \left( g^{hh}_{zz} \sigma^{hh}_z + g^{hh}_{xx} \sigma^{hh}_x \right).
\]

(E14)

Equation [S14] implies that the heavy hole has a zero in-plane \( g \)-factor, and that an out-of-plane component \( B_z \) creates a tilted effective field with a spin precession vector lying in the \( xz \) plane. We note that the atomic-scale symmetry of the SF makes the two in-plane directions \( x \) and \( y \) inequivalent.

The magneto-Stark Hamiltonian can be derived by symmetry using a similar procedure. The combination of the two \( \Gamma_3 \) irreducible representations (\( K_x, K_y \)) and (\( -B_y, B_x \)) contains an invariant representation, leading to the Hamiltonian

\[
H_{\text{magneto-Stark}} = \beta'(K_x B_y - K_y B_x).
\]

Furthermore, the Hamiltonian describing the diamagnetic shift is derived from the invariants \( B_x^2 \) and \( B_x^2 + B_y^2 \), namely,

\[
H_{\text{dia}} = \beta_1 B_x^2 + \beta_2 (B_x^2 + B_y^2).
\]

For Eq. 1 of the main text, we take into account only an in-plane field effect and set \( \beta_2 \equiv 0 \).

We note for completeness that, besides \( B \)-linear, \( B \)-quadratic and \( K \)-linear terms, the effective Hamiltonian also includes \( K \) linear terms, \( \sigma^x_1 K_y - \sigma^y_1 K_x \), which arise from spin-orbit coupling. Our estimates show that these terms are not significant for the relevant wavevectors and do not lead to the non-reciprocal emission spectra to first-order in \( B \).
S7. SUPPLEMENTAL ANGLE RESOLVED DATA

For the angle resolved experiment that tests the magneto-Stark effect, PL spectra were acquired on multiple different SF pyramids positioned at various angles. The spectra of the up and down SF at ±6.5 T are shown in Fig. S9. We note that the PL spectra show a strong and a weak doublet. The origin of the weak doublet is unknown, but we tentatively attribute it to scatter of PL from other exciton populations.

We note that there are two types of SF pyramids: one where the left/right SFs show higher energy PL than the up/down SFs (type a), and a second type where the emission energies are swapped (type b). For example, using this naming scheme the SF pyramid shown in Fig. 1(d) is of type a. Here, the left/right directions refer to the direction of the majorities of the intrinsic/extrinsic pair defects, visible as single triangles in Fig. 1(a). We also define a standard orientation of the bulk crystal depending on whether the sample is mounted as it is in Fig. 1(a) (standard) or rotated by 90° about [001] (rotated).

The PL emission energy as a function of magnetic field for the 6 SFs is shown in Fig. S10. These plots show that the atomic structure of the majority of the intrinsic/extrinsic pair defects, visible as single triangles in Fig. 1(a). The majority of the defects has the same absolute value of momentum and hence $\Delta E_{\text{up}}$ and $\Delta E_{\text{down}}$ have the same magnitude [Fig. 3(d)]. When the angle is increased, $\Delta E_{\text{up}}$ and $\Delta E_{\text{down}}$ have different magnitudes [Fig. 3(e)].

In the linear in $B_y$ regime, the magnetic field is not expected to significantly perturb the exciton dipole moment, and so to first order $\Delta E$ should be proportional to $B_y$. Experimentally, we find the energy shift is linear with $B_y$ for $B_y < 1$ T, and we extract the proportionality constant using a fit to $\Delta E = aB_y$. The estimate of the dimensionless dipole moment $\alpha$ depends somewhat on the fitting method used to find the slope $\Delta E$ vs. $B_y$. The estimate of the dimensionless dipole moment $\alpha$ depends somewhat on the fitting method used to find the slope $\Delta E$ vs. $B_y$. The estimate of the dimensionless dipole moment $\alpha$ depends somewhat on the fitting method used to find the slope $\Delta E$ vs. $B_y$.

We note that the combination

$$ r(\theta) = \frac{|\Delta E_{\text{up}}| - |\Delta E_{\text{down}}|}{\frac{1}{2}(|\Delta E_{\text{up}}| + |\Delta E_{\text{down}}|)} $$

(S17)

depends only on given geometry and the index of refraction (to first order in $B$), and is independent of the exciton dipole moment. Using Eq. S15 and expanding about small $\theta$, we can produce a small-$\theta$ approximation for $r(\theta)$,

$$ r(\theta) \approx \frac{2\cot \theta_{SF}}{n} $$

(S18)

correct to within 10% for angles up to 45° and $n = 3.5$. We obtain good agreement between $r(\theta)$ calculated experimentally from the $B = 0$ slope of $\Delta E$ with Eq. S17 (Fig. 3h).

We emphasize that this model agrees with the data without any fit parameters.

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FIG. S9. Spectra at -6.5 T and 6.5 T for the up and down SF for six different SF pyramid defects. The difference between $\Delta E_{\text{up}}$ and $\Delta E_{\text{down}}$ is visible when $\theta \neq 0$. 1 $\mu$W, 1.5 K, excite at 1.65 eV, excite and collect H polarization.

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