Cavity-enhanced linear dichroism in a van der Waals antiferromagnet

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Optical birefringence is a fundamental optical property of crystals widely used for filtering and beam splitting of photons. Birefringent crystals concurrently possess the property of linear dichroism (LD), which allows asymmetric propagation or attenuation of light with two different polarizations. This property of LD has been widely studied from small molecules to polymers and crystals but has rarely been engineered on demand. Here we use the newly discovered spin-charge coupling in the van der Waals antiferromagnetic insulator FePS3 to induce large in-plane optical anisotropy and consequently LD. We report that the LD in this antiferromagnetic insulator is tunable both spectrally and in terms of its magnitude as a function of the cavity coupling. We demonstrate near-unity LD in the visible–near-infrared range in cavity-coupled FePS3 crystals and derive its dispersion as a function of the cavity length and FePS3 thickness. Our results hold wide implications for the use of cavity-tuned LD as a diagnostic probe for strongly correlated quantum materials and offer new opportunities for miniaturized, on-chip beam-splitters and tunable filters.

Optical anisotropy plays a crucial role in light manipulation for photonic and optoelectronic applications. Although giant out-of-plane optical anisotropy has recently been observed in the visible to mid-infrared wavelength range in perovskite chalcogenides, hexagonal boron nitride crystals and molybdenum or tungsten chalcogenides1, in-plane optical anisotropy has scarcely been explored in low-dimensional van der Waals crystals. This has inspired an ongoing search for giant in-plane optical anisotropy among natural and artificial materials. To achieve in-plane anisotropy a material must possess some asymmetry in its structural or electronic order along one direction. Often, this is achieved via the anisotropic, one-dimensional structure of the material itself, such as in liquid crystals, polymers2 or carbon nanotubes. An effective way to quantify this anisotropy is via linearly polarized light spectroscopy, which is often used to obtain the LD (that is, the difference in reflected light intensities along two different polarizations)3 or the birefringence (the difference in the refractive indices (Δn) along two different polarizations), which represent a measure of the anisotropically optical response. The property of LD has been very useful in producing versatile optical components4,4, including polarizers, wave plates, mirrors and phase-matching elements. Despite its great fundamental and technological importance, LD and in-plane LD in novel materials have rarely been studied systematically; moreover, even less is known about the tuning or engineering of LD in low-dimensional optical materials. Among known in-plane anisotropic materials such as black phosphorus (BP) and rhenium disulfide (ReS2), LD has recently been measured with a magnitude of around 20–40% (refs. 5–8), where the LD primarily emanates from the in-plane asymmetry of the atomic arrangement or crystal structure. However, these are static values and no investigations on the engineering/tuning of LD have been reported so far. A notable approach to tuning/engineering optical anisotropy involves the use of artificially designed metamaterials and metasurfaces that comprise patterned nanostructures, and these have been shown to exhibit large birefringence values9. Therefore, an in-depth study of LD and birefringence, particularly the ability to tune and enhance LD, is notably lacking. This ability to tune the LD would be particularly important for wavelength-tunable beam splitters, waveguides and detectors.

The recent emergence of van der Waals chalcogenides with long-range magnetic order has offered a unique opportunity for the tuning of light–matter interactions as a function of magnetic phase transitions or spin-ordering. Magnetic phase transitions can induce spin-charge couplings that can result in breaking the symmetry of the in-plane optical response, which leads to the observation of LD. Magneto-optical effects, such as the magneto-optical Kerr effect (or MOKE)10,11, are commonly used to detect such magnetic phase transitions and also spin configurations or ordering in ferromagnets. Compared with ferromagnets, however, the detection of antiferromagnetism is much more difficult due to the lack of a net magnetic moment. Earlier studies have used neutron scattering8,9, Raman spectroscopy12–15 and second harmonic generation16–18 to detect the symmetry of spin ordering in antiferromagnets, based on the expansion of the unit cell, magnetoelastic coupling, and inversion symmetry breaking phenomena, respectively. Here we show that in the van der Waals antiferromagnetic (AFM) insulator iron phosphorus trisulfide (FePS3) a strong spin-charge coupling can induce large in-plane anisotropy leading to extraordinarily high LD values. These LD values can be further tuned by coupling them with a simple optical-cavity medium, which results in enhancement of the LD to near-unity values. In addition, the tuning of the cavity allows the spectral tuning of this large LD response. Our results suggest that van der Waals AFM semiconductors and insulators are outstanding candidates for the tuning and amplification of in-plane LD in the visible to near-infrared (NIR) range and open the door to novel, multi-spectral, miniature nanophotonic components by virtue of spin-charge coupling phenomena.
Results
We report the observation of near-unity, tunable and cavity-enhanced LD in a broad-band visible–NIR region of the spectrum in thin layers of FePS₃, a van der Waals AFM material. FePS₃ belongs to a class of transition metal phosphorus trichalcogenides (MPX₃, M = Fe, Mn or Ni; X = S or Se), which are van der Waals AFM insulator materials with bandgap values ranging from 1.3 eV to 3.5 eV (refs. 12,13,19–21). Within individual FePS₃ layers, iron atoms are arranged in a honeycomb lattice structure. The spins pointing out-of-plane along a chosen row of iron atoms aligned in the zigzag direction (shown along the x axis) are opposite to the spins in the adjacent spin chain of iron atoms (the in-plane structure; Fig. 1a), forming a zigzag AFM order22. The blue and red iron atoms are aligned in the +z and –z direction, respectively. The interlayer coupling between adjacent layers is AFM, as shown in Fig. 1a (out-of-plane structure). Raman spectroscopy12 and magnetic susceptibility23,24 measurements from the literature have shown that the Néel temperature \( T_N \approx 118 \text{ K} \) in bulk FePS₃. Below the Néel temperature, the strong spin-charge correlation results in high optical anisotropy, which is locked into the zigzag direction of the FePS₃ crystal22,25.

To demonstrate this, linearly polarized optical reflectance measurements were performed on multilayer FePS₃, directly exfoliated on a silicon/silicon dioxide (Si/SiO₂) substrate (Fig. 1b; SiO₂ thickness, 90 nm). The samples show varying colours with varying thicknesses, suggesting strong optical interference effects. We use a polarizer placed in the reflected light path to the detector to detect the polarized reflectance and the LD from the sample (Fig. 1c). Within this multilayer FePS₃ structure, the LD can be enhanced from the cavities formed vertically inside the material, which is indicated by the light purple oval marked in Fig. 1c.

We define LD as \( \frac{R_\perp - R_\parallel}{R_\perp + R_\parallel} \), where \( R_\parallel \) (\( R_\perp \)) is the peak intensity of the horizontally (vertically) polarized optical reflection (Methods)26. The above expression can also be understood as the difference in reflection between the two axes divided by the total unpolarized reflection. Figure 1d shows the high degree of optical anisotropy with LD up to 98.6% for measurements performed at 30 K at the wavelength of 751 nm. The LD is observed to be a strong function of the temperature, as shown in Fig. 1e. The transition of LD from ~10% to ~100% begins at 130 K and is complete by 110 K, coinciding with a precipitous drop at the Néel temperature \( T_N = 118 \text{ K} \). The LD magnitude approaches the highest level of 98.6% below the Néel temperature, following the AFM phase transition. This temperature dependence is similar to earlier reports on temperature-dependent Raman spectroscopy12,13 and magnetic susceptibility23, which indicates spin-charge coupling due to the AFM ordering. This further supports our evidence of LD induced by the AFM transition and spin ordering along the zigzag crystallographic direction.

To further understand the nature of this optical response, we carried out polarized spectral reflectance measurements in the visible–NIR range to estimate the LD spectrum. Figure 2a shows an optical micrograph and the corresponding atomic force topography of the FePS₃ crystal sample. The line cut (white line) shows the step edge of the sample corresponding to a thickness of 268 nm. As shown in the polarization-angle-dependent reflectance spectra (Fig. 2b), the extinction (reflectance) is highly dependent on the incident light polarization and, consequently, also the LD (Fig. 2c). Several resonant absorption (reflection) peaks are visible with values corresponding to near-unity absorption, particularly at ~750 nm. These resonant reflectance peaks suggest that the reflected light is strongly attenuated in the layered material due to the thin-film interference effect27. Briefly, a film with a thickness of \( \frac{\lambda}{4n_{eff}} \) (where \( p \) is an integer and \( \lambda \) is the incident light wavelength and \( n_{eff} \) is the effective refractive index of the combined group of several vertically stacked thin films (Si/SiO₂/FePS₃, stacked layers in this case), which is used to define the occurrence of the cavity mode due to the phase-matching condition) will form an optical cavity where light (that is, photons) of specific wavelengths will be trapped inside the film and thereby result in enhancement of the absorption28,29.
Hence, the resonant absorption (reflectance) peaks in Fig. 2b correspond to different orders of Fabry–Perot cavity-mode resonances. The high anisotropy is also observed in the polarized reflectance spectra. The absorption resonances vary at different polarization angles, illustrating the in-plane birefringence ($\Delta n$) of FePS$_3$, since the cavity-resonance mode is highly dependent on $n_{\text{eff}}$. The lattice anisotropy axes can be determined by charting the polar plot of the reflectance intensities, which also depends on the wavelength, as is shown in Fig. 2c. The polarization angle of 340° (70°) corresponds to the long (short) axis of lattice anisotropy. We also performed temperature-dependence reflectance measurements along the long (340°; Fig. 2d) and short (70°; Fig. 2e) axes. It is evident from these reflectance measurements that the cavity resonance at 765 nm blue-shifts (red-shifts) while the cavity resonance at 523 nm red-shifts (blue-shifts) at the 340° (70°) polarization angle as FePS$_3$ goes through AFM ordering below its transition temperature. This indicates that the birefringence originates from the AFM order, and the birefringence is also spectrally dispersive. The reflectance at 751 nm (862 nm) is the highest (lowest) at a polarization angle of 340° whereas it is the lowest (highest) at 70°. Figure 2f shows the temperature dependence of the LD spectrum as derived from Fig. 2b. It is evident from the plot that the LD intensity variations extend broadly in the visible–NIR spectrum. Multiple peaks are observed in the LD spectrum, which correspond to multiple cavity modes formed by the crystal in the reflectance spectrum in Fig. 2b. The largest LD peak reaches to 98.6%, corresponding to the near-unity cavity-resonant absorption peak at 751 nm, and the second-largest LD peak reaches to 80.0%, corresponding to the absorption peak at 862 nm in the reflectance spectrum. Further LD peaks at shorter wavelengths have a lower intensity and correspond to the higher order resonant-cavity modes. The absorption losses at these cavity resonances correspond to absorption in both the FePS$_3$ as well as the underlying Si/SiO$_2$. The temperature dependence of the LD spectra shows a drastic reduction in the magnitude of the LD above 118 K, matching well with the Néel temperature of FePS$_3$. More reflectance and LD measurements were performed on multiple samples with varying thicknesses ranging from 80 nm to 270 nm, as shown in the Supplementary Information. The cavity-enhanced LD peaks are observed similarly in various flakes, and multiple LD peaks are also observed at different wavelength ranges due to the various thicknesses of the samples (see Supplementary Fig. 1 for more details).

To better understand the cavity-enhanced LD of this structure and its thickness dependence, we performed electromagnetic wave calculations using the transfer matrix method (TMM) to estimate the thickness-dependent reflectance and the LD intensity. To make a correct estimate from TMM calculations we measured the dielectric function of FePS$_3$ via spectroscopic ellipsometry (see Supplementary Fig. 2 for the fitting details). The ellipsometry results are based on a large bulk crystal (~1 mm size) measured at room temperature where the original refractive index values $n$ and $k$ are derived without AFM ordering, which are primarily...
isotropic. To derive the birefringence of FePS₃, with AFM ordering, a low-temperature polarized reflectance measurement with normal incident light was performed on the bulk crystals (of 10 µm thickness, thereby avoiding any cavity-enhancement effects) and the LD spectrum for these is shown in Fig. 3a. The refractive index from the two anisotropic axes $n_1$ and $n_2$ can be derived from the reflectance data (Supplementary Fig. 3) by applying the Fresnel equation

$$R = \frac{\tilde{n} - 1}{\tilde{n} + 1}^2$$  \hspace{1cm} (1)$$

where $\tilde{n} = n + ik$ is a complex value.

Here, the imaginary part ($k \approx 0.2$) is much smaller than the real part ($n \approx 3$) at the peak position wavelength range (Supplementary Fig. 2). Therefore, we have neglected the imaginary part of the refractive index in equation (1) to derive the birefringence and simplify our analysis. From equation (1), we can obtain $n_1$ and $n_2$ with all real values:

$$n_1 = \frac{1 + \sqrt{R_1}}{1 - \sqrt{R_1}}$$

$$n_2 = \frac{1 + \sqrt{R_2}}{1 - \sqrt{R_2}}$$

and the birefringence is derived from $\Delta n = n_1 - n_2$. In addition, the differences in extinction coefficients ($\Delta k$) for the two anisotropic axes is not considered in this case due to the limitation of our experimental setup for performing spectroscopic ellipsometry measurements below the Néel temperature. The exact calculation details on the estimation and assumptions of the birefringence spectra are provided in Supplementary Fig. 3. This quantification at every wavelength results in the estimation of the birefringence ($\Delta n$) spectrum (Fig. 3b). The birefringence ranges from $-0.2$ to $0.075$ depending on the wavelength. The birefringence spectrum shows an interesting feature of crossing over from positive values below 650 nm to negative values above 650 nm, indicating the presence of an electronic band resonance in FePS₃ (ref. 30). This behaviour also explains the opposite directions of the shifts in the cavity-resonance transitions to a negative $\Delta n$ above 650 nm, indicating the presence of a wavelength-dependent birefringence of FePS₃, as shown in Fig. 3b. The inset shows the refractive index (real part) of FePS₃ measured via the spectroscopic ellipsometry method (298 K).

Fig. 3 | Simulation model of cavity-enhanced multilayer FePS₃. a. LD spectrum of bulk (10 µm thickness) FePS₃ crystal measured at 30 K. b. Birefringence spectrum at 30 K calculated from a. The spectrum shows a wavelength-dependent birefringence of FePS₃, indicating a positive $\Delta n$ below 650 nm that transitions to a negative $\Delta n$ above 650 nm. The inset shows the refractive index (real part) of FePS₃ measured via the spectroscopic ellipsometry method (298 K). c. Experimental LD spectra for specific FePS₃ thicknesses of 80, 125, 153, 252 and 268 nm. All measurements were performed at 30 K. d. TMM-calculated LD spectra for the same thicknesses as in c. The LD peaks match well in terms of their qualitative shape and spectral positions. e. Electric field profiles of the Fabry–Perot cavity-mode resonance peaks labelled in c. Triangles indicate the first-order cavity mode, whereas squares and stars indicate the second- and third-order cavity modes, respectively.

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reflectance measurements at low temperature (see Supplementary Fig. 4 for TMM calculation details). Figure 3c shows the experimental LD spectra of FePS₃ for various thicknesses. The corresponding simulated LD spectra (Fig. 3d) show a close qualitative match in terms of the resonances and features observed in the experimental spectra as indicated by the corresponding labels. This cavity model along with our simulations also explains and confirms the change of polarization axes from positive to negative at different wavelengths. There is, however, some quantitative mismatch between the simulation and experimental results due to two main reasons. First, the estimation of the birefringence is not perfect in this case since we are not considering the difference in extinction coefficients (Δε) for the two anisotropic axes. This will induce an intensity mismatch from simulation to experiments. Second, the AFM domains of FePS₃ may not be the same size and ideally matched in each FePS₃ unit layer, which means that the sampled thickness in the experiment may not be the ideal thickness for the calculations. To understand further the nature of these resonances, we also checked the electric field (E) distributions along the cross-section of FePS₃ crystal, that is, in the plane normal to the basal plane of the crystal, as shown in Fig. 3e. For thin-film interference, the cavity resonance occurs at \( d = \frac{l}{2n} \) (where \( p \) is the order of the mode), which is the approximate value of the resonant wavelength at the specific thickness (Fig. 3c-d). The peaks in the LD spectra are labelled using upright triangle, square and star symbols (Fig. 3c-e) and correspond to the first-, second- and third-order cavity modes, respectively, formed inside the FePS₃ crystal (Fig. 3e). The field profiles again verify that the LD is enhanced due to the cavity modes.

Given that the resonance magnitude and spectral position in LD is tunable as a function of the FePS₃ thickness, this system provides a unique opportunity for exploring the tunability of the LD spectrum. This can be done in two ways. Figure 4a shows the approaches adopted here to tune the optical-cavity sizes using both internal and external cavity media. For a fixed thickness of the substrate layer (that is, bulk Si with some fixed SiO₂ thickness on top), if the thickness, \( d \), of FePS₃ is much smaller than the incident wavelength \( \lambda \) (Fig. 4a, top left), then the light will pass through the FePS₃, leaving the material with minimal loss. Likewise, if the thickness is much larger (Fig. 4a, top right) then the light will be fully absorbed or reflected before reaching the bottom-most layers of FePS₃. In neither of these two cases will the FePS₃ layers form and support cavity modes. Only when the thickness of FePS₃ is at a comparable level to that of the incident light wavelength (Fig. 4a, bottom left) will the material be capable of supporting a stable cavity for the photons, forming a standing wave inside it. In this case, the photons are trapped in the FePS₃, which enhances the light–matter interaction and thus the LD. Similarly, one can achieve the spectral tuning of the LD peaks by changing the thickness of the external cavity, \( l \) (SiO₂: Fig. 4a, bottom right). The detailed electric field profiles of the cavity resonances formed by the internal cavity (FePS₃) and external cavity (SiO₂) are shown as a comparison in Supplementary Fig. 5. First, by sweeping the thickness of FePS₃, one can simply achieve the spectral tuning of the LD peaks by tuning the cavity size itself. Figure 4b shows a colour map of the LD spectra as a function of the FePS₃ thickness with the LD magnitude and sign (red positive and blue negative) shown on the colour scale. The star, square and triangular symbols correspond to the experimentally observed LD peaks in Fig. 3c. In this map, the linearly dispersive branches of the LD resonances correspond to the first-, second- and third-order Fabry–Perot cavity modes, respectively. The lowest order modes show the strongest LD contrast whereas the higher order modes show weaker contrast. The colour plot further shows that the desired thickness range for FePS₃ crystals on 90-nm-thick SiO₂ on Si is ~270 nm for observation of maximum magnitude of LD under reflectance in the visible wavelength range. The second way of tuning the LD is to tune the substrate SiO₂ thickness. For a fixed layer thickness of FePS₃ below 50 nm where the material is too thin to form any internal cavity to support a mode, we can use the external cavity (SiO₂) to enhance the absorption and LD of FePS₃, as seen in Fig. 4c. Here, the thickness of the FePS₃ layer is fixed at around 15 nm. Multiple orders of LD peak branches are observed and the symbols in the spectral map once again denote the experimental LD peak positions of first-order (triangles), second-order (square) and third-order (star) resonances. The corresponding full experimental spectra (line plots) are shown in Supplementary Fig. 6. Several FePS₃ crystal samples with roughly the same thickness (~15 nm) when measured on Si substrates with different SiO₂ thicknesses (50, 90 and 280 nm) show shifting of the LD resonance peak (Fig. 4c). This observation further verifies that the LD response is tunable via changing the optical-cavity size. Furthermore, through calculations, we also...
implemented a better closed optical cavity, which further enhances the absorption and LD in monolayer FePS$_3$ systems. The referenced model that uses gold/alumina (Au/Al$_2$O$_3$) layers as, respectively, mirror and dielectric layers encapsulating monolayer FePS$_3$, is shown in Supplementary Fig. 7. The closed optical cavity with metallic mirrors and Al$_2$O$_3$ on both sides can strongly couple the cavity photons into even a monolayer material with an enhanced absorption to a near-unity level. By controlling the size of the applied optical cavity, the monolayer FePS$_3$ can reach a near-unity absorption for a specific wavelength range. As a result, the near-unity LD can be generated due to the large absorption peak, even with a small birefringence in monolayers. This demonstrates that the enhancement of LD is possible at the monolayer limit.

Discussion and outlook
The above experiments, calculations and results have several important implications, both at a fundamental level and in terms of the potential for future applications. LD spectroscopy is a valuable technique for probing anisotropic spin structures in AFM materials. Since the optical anisotropy axis is locked to the zigzag direction in real space along which the spin chains are aligned, polarized LD can also serve as a probe in detecting or imaging AFM domains (Fig. 5a,b). The previously shown FePS$_3$ flakes all have a more uniform LD signal across the whole sample. The exact polarized LD that results at several specific points is shown in Supplementary Fig. 8. To deliberately induce inhomogeneity, we exfoliated thin samples on the soft polydimethylsiloxane (PDMS) substrate and then transferred them onto a gold-coated Si substrate (the optical image of one sample is shown in Fig. 5a). As shown in Fig. 5b, the LD mapping is performed across the thin sample (~20 nm), where the direction and length of each segment denote the LD direction and magnitude, respectively. The inhomogeneous LD distribution, which indicates the coexistence of multiple zigzag domains, is distinctly different from the uniform LD response obtained from previous samples that were directly exfoliated on the Si/SiO$_2$ substrate, and this is probably due to the inhomogeneous sample distortion induced in the process of the PDMS-assisted sample exfoliation. The results not only demonstrate LD mapping as a powerful probe for detecting AFM zigzag domains but also suggest the easy tunability of the zigzag domains using external in-plane anisotropic stimuli.

Unlike other van der Waals materials with lattice in-plane anisotropy, the birefringence from FePS$_3$ is induced by strong spin-charge coupling from the AFM zigzag spin chains, which indicates that the birefringence of FePS$_3$ can be manipulated simply by controlling the magnetic states. The tunable large birefringence or LD provide a wonderful platform for various potential applications in photonics, which is not possible with other materials of latticed-induced anisotropy. A summary of the possible tunability and applications of AFM-coupled birefringence is shown in Fig. 5c. First, the zigzag orientation can be tuned and switched when the applied in-plane magnetic field (Fig. 5c(i)) is large enough to flip the AFM zigzag domain state$^{31}$. An out-of-plane external magnetic field can also affect the intrinsic birefringence as shown in Supplementary Fig. 9. Second, the spin-chain orientation can also be tuned through the external strain (Fig. 5c(ii))$^{32}$. Therefore, the direction of LD can be controlled by the competition between the strain and the original AFM order along the zigzag direction. This is also illustrated from Fig. 5b with an inhomogeneously oriented LD direction in a thin FePS$_3$ flake transferred by the soft PDMS substrate. Third, the birefringence tunability can also be controlled via various nanophotonic structures (Fig. 5c(iii)). By tuning the AFM orders in both magnitude and orientation using different external stimuli, AFM insulators allow the optical anisotropy of the medium to be controlled in multiplexed ways, which opens new directions in
nanophotonic design such as integration with Bragg mirror-based cavities and patterning of subwavelength resonators (Fig. 5c(iii)). The cavity-coupled enhancement of LD demonstrated in this work is an example of a simple fabrication-free photonic structure. It is worth noting that the AFM-coupled LD of FePS3 originates from a phase transition from a parametric phase to an AFM phase, which corresponds to structural symmetry breaking. This will also pave the way for opportunities in tunable topological photonic metasurfaces comprised of AFM insulators. Furthermore, the tunability of birefringence in antiferromagnets can also be expanded to include tunability applications via temperature control, the gating control of monolayers, the doping level in materials and so on. Finally, it is also worth noting that, although the critical temperature (Tc) values of van der Waals antiferromagnets and ferromagnets are cryogenic, it is well known that the Tc value can be elevated through doping and nanostructuring, thereby making these materials more suitable for realistic applications.

In summary, the extremely high LD (98%) demonstrated in FePS3 is among the largest in most two-dimensional anisotropic materials and makes FePS3 an attractive candidate for future anisotropic optical elements. In addition, the ability to tune the magnitude and orientation of AFM-coupled birefringence both spectrally and spatially, through varying external stimuli such as the magnetic field, strain and artificial nanostructuring, opens new opportunities in nanophotonic component design, particularly for ultrathin and highly birefringent materials for on-chip filtering and beam splitting, as well as for non-linear optical components.

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Methods
Sample preparation. FePS$_3$ layers were mechanically exfoliated from bulk crystal (2D Semiconductors) using Scotch Tape and transferred onto the Si/SiO$_2$ substrate with thermally grown oxide using a dry process. The Si/SiO$_2$ wafers with SiO$_2$ thicknesses of 50, 90 and 280 nm were commercially purchased. For sample-thickness-dependent measurements, samples were transferred onto the same substrate with an SiO$_2$ thickness of 90 nm. For dielectric-layer-dependent measurements, different FePS$_3$ crystal samples of the same thickness (~15 nm) were transferred onto the substrate with an SiO$_2$ thickness of 50, 90 and 280 nm.

Reflectance measurements. The normalized reflectance intensity ($R_{\text{normalized}}$) values from 0 to 1 were all achieved by subtracting the background reflectance intensity ($R_{\text{background}}$) from the sample reflectance intensity ($R_{\text{sample}}$) and normalizing it with the reflectance intensity from a silver mirror ($R_{\text{mirror}}$), using

$$R_{\text{normalized}} = \frac{R_{\text{sample}} - R_{\text{background}}}{R_{\text{mirror}} - R_{\text{background}}}$$

Linear dichroism measurements. An unpolarized halogen light source (AvaLight-HAL) was focused on the sample using a ×50 objective (Olympus SLMPLN; numerical aperture, 0.35). The reflected light was analysed using a linear polarizer and collected via a multi-mode optical fibre into a spectrometer with a charge-coupled device (CCD) camera. The polarization-dependent LD spectrum was calculated using $R_{\parallel} - R_{\perp}$, where $R_{\parallel}$ and $R_{\perp}$ are, respectively, the reflectance perpendicular and parallel to the polarization. The sample was loaded in a helium-free cryostat for these measurements.

Ellipsometry. Ellipsometry spectra were obtained at three angles of incidence (65°, 70° and 75°) using a VASE ellipsometer (J.A. Woollam). Since the sample size was small, a focusing-optics module with a spot size of ~100 μm was attached to the instrument. In addition, the instrument was equipped with an auto-retarder to monitor the depolarization caused mainly by non-specular reflections. Scans with high depolarization values (~10% or higher) were discarded as they were harder to model, and scans with only low depolarization values were used to obtain the dielectric function of the sample. More details of the dielectric function results are in Supplementary Fig. 2.

Magnetically tunable linear dichroism measurements. The LD magnetic-field dependence of the sample was measured using an attoDRY2100 cryostation with the sample held at 30 K. The magnetic field was swept from -9 T to +9 T in the Faraday geometry. White light was produced using a stabilized tungsten-halogen light source (SLS201L, ThorLabs) and focused onto the sample with a 0.82 numerical aperture in situ microscope objective. The reflected light was sent through a Glan-Thompson linear polarizer, dispersed in a SpectraPro spectrometer (SP-2750, Princeton Instruments) and collected using a CCD camera (400 BR-eXcelon, Princeton Instruments).

Calculations. All the calculations and simulations were carried out using the TMM. Details are discussed in Supplementary Fig. 4.

Data availability
All data needed to evaluate the conclusions in the paper are present in the paper and the Supplementary Information. Additional data are available from the corresponding authors upon reasonable request.

Code availability
The codes used in this study for plotting and modelling are available from the corresponding authors upon request.

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Author contributions
Z.N. discovered the large LD around 800 nm. D.J., H.Z. and Z.N. conceived the project. H.Z. and Z.N. made the samples, performed the linearly polarized reflectance measurements and atomic force microscopy characterization. H.Z. and Z.N. performed the calculation work. Under the supervision of L.W. Z.N. performed the LD imaging/spatial mapping. C.E.S. and J.R.H. performed the magnetic-field-tunable LD measurements. F.P. and A.B performed the ellipsometry measurements. With help from Z.N. and D.J. H.Z. analysed and interpreted the optical spectroscopy and simulation data. H.Z. and D.J. wrote the paper with input from all co-authors. D.J. supervised the entire study.

Competing interests
The authors declare no competing interests.

Additional information
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