Reconstruction of Bloch wavefunctions of holes in a semiconductor

A central goal of condensed-matter physics is to understand how the diverse electronic and optical properties of crystalline materials emerge from the wavelike motion of electrons through periodically arranged atoms. However, more than 90 years after Bloch derived the functional forms of electronic waves in crystals (now known as Bloch wavefunctions), rapid scattering processes have so far prevented their direct experimental reconstruction. In high-order sideband generation, electrons and holes generated in semiconductors by a near-infrared laser are accelerated to a high kinetic energy by a strong terahertz field, and recollide to emit near-infrared sidebands before they are scattered. Here we reconstruct the Bloch wavefunctions of two types of hole in gallium arsenide at wavelengths much longer than the spacing between atoms by experimentally measuring sideband polarizations and introducing an elegant theory that ties those polarizations to quantum interference between different recollision pathways. These Bloch wavefunctions are compactly visualized on the surface of a sphere. High-order sideband generation can, in principle, be observed from any direct-gap semiconductor or insulator. We thus expect that the method introduced here can be used to reconstruct low-energy Bloch wavefunctions in many of these materials, enabling important insights into the origin and engineering of the electronic and optical properties of condensed matter.

High-order sideband generation

Here we present a direct experimental reconstruction of Bloch wavefunctions of holes in bulk gallium arsenide (GaAs) using high-order sideband generation (HSG). In HSG, a relatively weak near-infrared (NIR) laser with frequency \( f_{\text{NIR}} \) and a strong laser with terahertz (THz) frequency \( f_{\text{THz}} \) simultaneously interact with a semiconductor, resulting in the emission of sideband photons with frequencies \( f_{\text{SB}} = f_{\text{NIR}} + nf_{\text{THz}} \) where \( n \) is an integer. If the band structure is symmetric under inversion, as in the (001) plane of GaAs studied here, \( n \) must be even. HSG experiments have been conducted with both extremely narrow-band quasi-continuous-wave fields with \( f_{\text{THz}} < 1 \) THz, which have enabled the resolution of sidebands with \( n > 60 \) (ref. 7), and broader-band pulsed fields with \( f_{\text{THz}} > 10 \) THz, which have enabled time resolution of the recollision process even in materials with dephasing times of less than 10 fs (ref. 7). A quasi-continuous-wave HSG spectrum from bulk GaAs at 60 K is shown in Fig. 1a. In the experiment, a 100-mW NIR laser, and 2.01 ± 0.13 mJ, 40 ns, 0.447 ± 0.001 THz pulses generated by the University of California, Santa Barbara (UCSB) millimetre-wave free-electron

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laser were linearly polarized and collinearly focused on a 500-nm GaAs epilayer (Fig. 1a, inset). The THz electric field strength in the epilayer was $70 \pm 2 \text{ kV cm}^{-1}$ (Methods). In GaAs, HSG can be described by the following three-step process (Fig. 1b, Methods). First, electrons (E) and two species of hole—light holes (LH) and heavy holes (HH)—are created by the NIR laser. Second, the E–LH and E–HH pairs are driven apart and then back towards each other along the direction defined by the THz field (Fig. 1b). Crucially, during this acceleration phase, the Bloch waves associated with the E–LH and E–HH pairs interfere with each other. Third, they recollide with significant kinetic energy and emit sidemband photons.

Information about the Bloch wavefunctions sampled by electrons and holes on their journeys through the Brillouin zone is imprinted on the polarization state of each sidemband, which we measured by Stokes polarimetry (Methods). As light from a particular sidemband can be associated with a quasi-momentum that is controlled by the THz field, its polarization will be different from the incoherent light emitted at the same energy in the absence of a strong THz field (photoluminescence), which is a superposition of emission from electron–hole pairs with all quasi-momenta satisfying energy conservation.

The linear orientation angle, $\alpha$, and the ellipticity angle, $\gamma$, for each sidemband are shown in Fig. 1c for four different NIR polarizations. The polarizations of sidebands depend on the sidemband index $n$ and the NIR polarization in a manifestation of dynamical birefringence. Although sidemband intensities have a highly nonlinear dependence on THz power, they are proportional to the NIR power if it is sufficiently small. All data reported here were taken in this regime of linear NIR response. In this linear regime, the sidemband polarization can be mapped onto the polarization state of the NIR laser by a dynamical Jones matrix $T$ (ref. 4), defined in a basis of circularly polarized fields $\sigma^\pm$ (with helicity $\pm 1$) as

$$
\begin{pmatrix}
E_{+n} \\
E_{-n}
\end{pmatrix} = \begin{pmatrix}
 T_{++n} & T_{+-n} \\
 T_{-+n} & T_{--n}
\end{pmatrix} \begin{pmatrix}
 E_{+n}^\text{NIR} \\
 E_{-n}^\text{NIR}
\end{pmatrix},
$$

where $E_{+n}$ and $E_{-n}^\text{NIR}$ denote the $\sigma^+$ components of the electric field associated with the $n$th sidemband and NIR laser, respectively, and $T_{\sigma\sigma'}$ denote the dynamical Jones matrix elements associated with the $n$th sidemband. $T$-matrix elements were determined by measuring the sidemband polarizations for four different linear NIR laser polarizations.

**Dynamical Jones matrices**

To understand the physics underlying each $T$-matrix element, it is necessary to consider the spins of electrons and holes. The four recollision pathways from the excitations generated by the $\sigma^\text{NIR}$ component of the NIR laser are shown in Fig. 2. The electrons have spin 1/2, whereas the HHs and LHs have total spin 3/2. Driven by the THz field from time $t$ to $t'$, an electron–hole pair acquires a dynamic phase

$$
A_{\text{HH(LH)}}(t', t) = \int_t^{t'} \! dt'' \, (E[k(t'')] - E_{\text{HH(LH)}}[k(t'')]) / h,
$$

where $E_{k}$ and $E_{\text{HH(LH)}}$ are the energies associated with the E and HH (LH) bands, respectively, shown schematically in Fig. 1b. $k$ is the quasi-momentum and $h$ is the reduced Planck’s constant (Methods, Supplementary Discussion). The spin ±1/2 of the electron does not change during acceleration. As the Bloch wavefunctions in both the HH and LH bands are superpositions of states with spin ±1/2 and spin ±3/2, the $\sigma^\text{NIR}$ component can generate sidembands with either $\sigma^\text{HSG}$ or $\sigma^\text{LSG}$ while satisfying angular-momentum conservation, giving rise to dynamical Jones matrix elements $T_{\sigma\sigma'}$ and $T_{\sigma'\sigma}$, respectively. Similar recollision pathways follow from the excitations generated by the $\sigma^\text{NIR}$ component, giving rise to $T_{-\sigma\sigma'}$ and $T_{-\sigma'\sigma}$ (Supplementary Discussion, Extended Data Fig. 4).

The properties of dynamical Jones matrices can be derived from the Luttinger Hamiltonian, which describes the physics of the HH and LH states at the relatively small energies and quasi-momemta probed in this experiment. We tune the NIR laser just below the bandgap and direct the THz field to propagate along the z axis to ensure the electrons and holes have no $z$ component of quasi-momentum $k$. In this case the Luttinger Hamiltonian takes a block diagonal form

$$
H(k) = \frac{h^2 k^2}{2m_0} \left[ \gamma_0 - 2\gamma_n \sigma \cdot \mathbf{r} \right]
$$

where $\gamma_n$ is the identity matrix, $\mathbf{r}$ is the vector of Pauli matrices, $\theta$ is the angle between the THz field and the [110] crystal direction (Fig. 3d, inset), $\gamma_0$, $\gamma_1$, and $\gamma_2$ are the scalar Luttinger parameters, $m_0$ is the electron rest mass, and $\mathbf{n}$ is

$$
\mathbf{n} = \left( \frac{3}{2} \sin(2\theta) \pm \frac{3}{2} \frac{\gamma_1}{\gamma_2} \cos(2\theta) \right)
$$

The Bloch wavefunctions are found by diagonalizing equation (1) and only depend on the $\mathbf{n} \cdot \mathbf{r}$ term because the first term is proportional to the identity. As $\mathbf{n}$ depends only on the crystal angle $\theta$ and $\gamma_1/\gamma_2$, an experimental measurement of $\gamma_1/\gamma_2$ allows the reconstruction of the Bloch wavefunctions. Even for $k \neq 0$, although the Luttinger Hamiltonian is not block diagonal, knowing $\gamma_1/\gamma_2$ is still sufficient to reconstruct the Bloch wavefunctions.

**Reconstruction of Bloch wavefunctions**

We use ratios of $T$-matrix elements to check the validity of the theory and measure $\gamma_1/\gamma_2$. As the diagonal elements of equation (1) are real, when sidemband and NIR laser polarizations are the same, for each pathway producing a $\sigma^\text{HSG}$ photon there is an equivalent pathway producing a $\sigma^\text{LSG}$ photon (Fig. 2, Supplementary Discussion, Extended Data Fig. 4) through states related by time-reversal symmetry. Therefore, the ratio of diagonal dynamical Jones matrix elements for all sidemband indices and crystal angles is

$$
\frac{T_{\sigma\sigma}(\theta)}{\overline{T}_{\sigma\sigma}(\theta)} = \xi_\sigma(\theta) = 1
$$

As the off-diagonal elements of equation (1) are complex, when sidemband and NIR laser polarizations are different, for each pathway producing a $\sigma^\text{HSG}$ photon there is an equivalent pathway producing a $\sigma^\text{LSG}$ photon with a complex-conjugated phase factor (Supplementary Discussion). Therefore, the ratio of off-diagonal dynamical Jones matrix elements for all sidemband indices is

$$
\frac{T_{\sigma\sigma'}(\theta)}{\overline{T}_{\sigma\sigma'}(\theta)} = \xi_{\sigma\sigma'}(\theta) = \frac{\gamma_1 \sin(2\theta) - i\gamma_2 \cos(2\theta)}{\gamma_1 \sin(2\theta) + i\gamma_2 \cos(2\theta)}
$$

The magnitude of $\xi_\sigma(\theta)$ in equation (4) is 1 for all angles, but the argument depends on $\gamma_1/\gamma_2$ and $\theta$.

The experimentally measured values $\chi_\sigma(\theta)$ and $\xi_\sigma(\theta)$ at various $\theta$ are compared with the predictions of equations (3) and (4) in Fig. 3 and Extended Data Fig. 5 using values for $\gamma_1$ and $\gamma_2$ recommended in ref. 13. Within experimental error, $\chi_\sigma(\theta)$ and $\xi_\sigma(\theta)$ are 1, as predicted by equations (3) and (4) (Fig. 3a, Extended Data Fig. 5b, c). The arguments of $\chi_\sigma(\theta)$ for eight different $\theta$ are independent of $n$ (Fig. 3b), lying within $22^\circ$ of the constant values predicted by equation 4 (dashed lines) for all $\theta$ except $-45^\circ$. The values of $\chi_\sigma(\theta)$ are $\equiv \chi(\sigma\sigma)$, and $\xi(\sigma\sigma')$ are independent of $\theta$, with a value of 1, as predicted by equations (3) and (4) (Fig. 3c). The argument of $\chi(\theta)$ is plotted with respect to $\theta$ in Fig. 3d, and is close to the prediction provided by...
equation (4). Averaging the argument of \( \chi(\theta) \) over experimentally sampled \( \theta \) gives \( \gamma_3/\gamma_2 = 1.47 \pm 0.48 \), within experimental error of the value 1.42 recommended in ref. 13. We attribute the deviations in measured \( \chi_n(\theta) \) and \( \xi_n(\theta) \) from theoretical predictions, as well as much of the experimental error in the determination of \( \gamma_3/\gamma_2 \), to small inhomogeneous strain in the GaAs membrane (Methods, Extended Data Fig. 2).

From \( \gamma_3/\gamma_2 \), we reconstruct the Bloch wavefunctions of the Luttinger Hamiltonian in GaAs. For two coupled bands, the Bloch wavefunctions can be represented as spinors on a Bloch sphere\(^{26}\). In the \( k_z = 0 \) plane,
Thus, in the photon with helicity $+3/2 - 1/2 = +1$. The interference of the result in similar pathways to produce $\text{Arg}()$ produces the dynamical Jones, with helicity $\pm 1$. (A photon excites either a spin-up electron and hole of spin $-3/2$ or a spin-down electron and hole of spin $-1/2$.) Driven by the THz field, an electron–hole pair accumulates dynamic phase $A_{\text{HH}}$ or $A_{\text{LH}}$, depending on the band of the hole state (HH or LH). The electron spin is unchanged, whereas the hole states originating from the spin $-3/2$ state are superpositions of spin $-3/2$ and $+1/2$ states and the states originating from the spin $-1/2$ state are superpositions of spin $-1/2$ and $+3/2$ states. Upon recollision, either $A_{\text{HH}}$ or $A_{\text{LH}}$ photons are produced following angular momentum conservation—for example, a spin $+3/2$ hole recombining with a spin-down ($-1/2$) electron produces $A_{\text{HH}}$, photon with helicity $+3/2 - 1/2 = +1$. The interference of the evolution pathways from $A_{\text{HH}}$ to $A_{\text{HH}}$ $(A_{\text{HH}})$ produces the dynamical Jones matrix element $T_{\text{HH}}$. (T.). Photons with $A_{\text{HH}}$ result in similar pathways to produce $T_{\text{HH}}$ and $T_{\text{LH}}$. (Supplementary Discussion, Extended Data Fig. 4).

Each block of the Luttinger Hamiltonian is a two-by-two matrix, whose eigenfunctions—the Bloch wavefunctions—depend on $\theta$ but not on $|k| = k$. Thus, in the $k_z = 0$ plane, for any $\theta$, a single point on the Bloch sphere represents the Bloch wavefunctions for arbitrary $k$. The closed black curves in the northern and southern hemispheres of the Bloch sphere in Fig. 4 represent the most likely Bloch wavefunctions consistent with our measured $y_j/y_i$ for the LH and HH, respectively. The north and south poles represent the states with spin $-3/2$ and $+1/2$, respectively. The Bloch wavefunctions for the degenerate partners of those represented in Fig. 4 are related by time-reversal symmetry.

**Discussion**

The complete electronic structure of a crystalline solid should include both its band structure and Bloch wavefunctions. We have reconstructed low-energy Bloch wavefunctions of holes in GaAs from polarimetry of high-order sideband spectra. GaAs is one of the most widely studied semiconductors, and the consistency of our results with the vast body of complementary previous work validates the novel method presented here. HSG can, in principle, be observed from any direct-gap semiconductor or insulator, and has been observed in semiconductor quantum wells\(^2,3,6,7\) and both monolayer and bulk semiconducting transition metal dichalcogenides\(^3,8,9\). Thus, we expect polarimetry of high-order sidebands can be measured from a large class of bulk and nanostructured materials. As a probe of electronic structure, the sensitivity of HSG to the bulk of electrically insulating materials has the potential to complement ARPES, which works best on surfaces.

Holes in GaAs are an interesting special case because the Luttinger Hamiltonian is one of the simplest non-trivial, low-energy effective Hamiltonians in solids. However, HSG spectra contain a wealth of information about the portions of the Brillouin zone explored during the acceleration phase, and straightforward extensions of the work presented here will enable reconstruction of Bloch wavefunctions from a wide range of low-energy Hamiltonians. Each peak in a HSG spectrum can be thought of as the output of an interferometer for Bloch waves. During acceleration by the THz field, Bloch wavepackets generally accumulate two types of phase: dynamic phases $A_{\text{HSG}}$, which have been extensively discussed here, and geometric phases (also called Berry phases)\(^6\). Dynamic phases depend on only the time-dependent energy eigenvalues of electrons and holes during acceleration. Geometric phases accumulate if the Bloch wavefunctions of electrons or holes change along their trajectories. The Luttinger model predicts...
that, along special trajectories that are straight lines through the Γ point oriented along the constant angle θ defined by the linearly polarized THz electric field, electron–hole pairs acquire dynamic phases θ defined by the linearly polarized THz electric field and NIR excitation with k = 0. These trajectories are represented on the Bloch sphere in the x–z plane.

Fig. 4 | Reconstruction of the Bloch wavefunctions for kx = 0. The Bloch wavefunctions of HH and LH bands associated with $H$ in equation (1) are plotted as black lines. The orange shaded area corresponds to the uncertainty in the wavefunction associated with one standard deviation in the measurement of $y$. For a given θ, each wavefunction is represented by a point on the Bloch sphere. The arrows within the Bloch sphere point from the origin to the LH and HH Bloch wavefunctions for the values of θ defined by their Miller indices in the inset below. The poles correspond to the spin –3/2 and spin +1/2 states. The wavefunctions for $H$ are paths reflected across the x–z plane on a Bloch sphere with poles representing the spin +3/2 and spin –1/2 states.

Online content
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Methods

Fabrication of GaAs sample

A 500-nm-thick GaAs epilayer was grown via molecular beam epitaxy and then transferred onto a sapphire substrate through van der Waals bonding. The sapphire was transparent to both NIR and THz radiation. The fact that the thermal expansion coefficients of sapphire and GaAs are closely matched ensures relatively small strains in the GaAs epilayer upon thermal cycling. To make the strain as small and homogeneous as possible, the GaAs epilayer was etched to be circular. A layer of indium tin oxide (ITO)—which reflects THz radiation while transmitting NIR radiation—was grown on the sapphire surface that was opposite to the GaAs epilayer to create a low-quality-factor cavity that enhanced the THz field in GaAs at selected THz frequencies. At the 447 GHz frequency used in this study, the THz field is enhanced by a factor of 1.5 from the ITO layer (Extended Data Fig. 1). A silicon dioxide (SiO2) anti-reflection coating was grown on top of the ITO to minimize its NIR reflection and avoid NIR Fabry–Perot oscillations in the sideband spectra. See Supplementary Methods for a step-by-step fabrication procedure.

The absorbance spectra of the GaAs epilayer were measured in a cryogenic chamber as a preliminary characterization on strains, as well as the excitation gap, which motivated our choice of NIR laser wavelengths for HSG experiments. Extended Data Fig. 2 shows an absorbance spectrum measured at a sample temperature of 60 K using a white light source, and calculated as \( A = -10 \log(\text{Transmitted power with sample in cryostat/Transmitted power with cryostat (and sample) removed})\). The sharp peaks are assigned to exciton resonances associated with band-edge states with different angular momenta. These peaks are separated by 2.6 meV. A recent study has associated a similar splitting with a strain of order 0.1% (ref. 3). The absorbance spectra in the immediate neighbourhood of the illuminated spot chosen for the HSG experiments in this study showed little variation.

Optical methods

The NIR laser was generated from an M Squared SolTiS titanium:sapphire laser, with a 7-W, 532-nm Nd:YAG laser as the pump. The M Squared cavity is tuneable via piezoelectric response, with a precision of 0.01 Å output NIR wavelength, measured in real time by a WS6-600 wavemeter. The linewidth of the SolTiS is less than 5 MHz, enabling excitation of electron–hole pairs with very well defined energy and contributing negligibly to sideband linewidth, which is determined primarily by pulse-to-pulse fluctuations in the free-electron-laser (FEL) frequency. An acousto-optic modulator was used to direct the NIR laser onto the sample for 1 μs at a 0.0001% duty cycle, synchronized with the THz output pulse from the FEL. After the modulation, only the first-order beam propagated through the rest of the optical elements. The polarization of the NIR laser beam incident onto the GaAs epilayer was set with a quarter-wave plate and a half-wave plate, and measured by a Thorlabs PAX polarimeter. The NIR beam was focused down to about 500 μm at a quarter-wave plate and a half-wave plate, and measured by a Thorlabs PAX polarimeter, which was impractical for Stokes polarimetry of the sidebands because it is optimized for use with a continuous-wave laser beam at a single frequency. The intensity of each sideband was measured either by a photomultiplier tube or a charge-coupled device (CCD), each coupled to a dedicated monochromator. The photomultiplier tube measured the lowest-order sidebands, while the CCD imaged many higher-order sidebands simultaneously. To optimize the efficiencies of the diffraction gratings, a half-wave plate was placed after the Stokes polarimeter to rotate the sideband polarizations.

All measurements were performed at 60 K, which was the base temperature of the cryostat during this experimental campaign. HSG polarimetry spectra recorded at lower temperatures were similar to those reported here.

Extraction of \( y_1/y_2 \) from Stokes polarimetry

We characterize the polarization of each sideband using the four Stokes parameters defined as \( S_0 = I_0 \), \( S_1 = I_{pcos2acos2y} \), \( S_2 = I_{psin2acos2y} \), and \( S_3 = I_{psin2y} \), where \( I_0 \) is the total intensity, \( p \) is the degree of polarization, and the orientation angle \( \alpha \) and ellipticity angle \( \gamma \) are defined in the inset ofExtended Data Fig. 3b. After a sideband passes through the RQWP and horizontal linear polarizer, the intensity of the outgoing light, \( S_{out}(\phi) \), can be expressed as

\[
S_{out}(\phi) = S_0 \left[ \frac{1}{2} \right] \sin(2\phi) + \frac{1}{4} S_1 \cos(4\phi) + \frac{1}{2} S_2 \sin(4\phi)
\]

where \( \phi \) is the angle between the fast-axis of the RQWP and the horizontal. By measuring \( S_{out} \) as a function of \( \phi \), the four Stokes parameters can be extracted from the Fourier transform \( c_{m}\phi = \int_{-\infty}^{\infty} S_{out}(\phi) e^{-im\phi} d\phi/2\pi \):

\[
S_0 = 2C_0 - 4Re(C_{20}) , \quad S_1 = 8Re(C_{20}) , \quad S_2 = -8Im(C_{20}) , \quad S_3 = 4Im(C_{22}) .
\]

We sampled the intensities of each sideband at 16 different angles \( \phi \). We define plots of \( S_{out} \) as functions of the angle \( \phi \) as ‘polaragrams’ (see Extended Data Fig. 3a, c for examples). For each angle \( \phi \), four CCD scans were taken to establish the variance of the intensity \( S_{out} \). From the Stokes parameters of the \( n \)-th-order sideband, \( S_{0n} \), the polarization state of the sideband can be extracted by calculating the angles \( \alpha_n \) and \( \gamma_n \) from relations

\[
\tan(2\alpha_n) = \frac{S_{2n}}{S_{1n}}
\]

\[
\tan(2\gamma_n) = \frac{S_{3n}}{\sqrt{S_{1n}^2 + S_{2n}^2}}
\]

Examples of extracted polarization states of sidebands are shown in Extended Data Fig. 3b, d. To reconstruct the dynamical Jones matrices, the polarization states of the sidebands were measured for four different polarization states...
of the NIR laser. All polarizations of the NIR laser were linear \((\gamma_{\text{NIR}} = 0^\circ)\) with orientation angles \(\alpha_{\text{NIR}} = 0^\circ, \theta_{\text{NIR}} = 45^\circ, \phi_{\text{NIR}} = 90^\circ\) and \(\phi_{\text{NIR}} = 45^\circ\), respectively. Each dynamical Jones matrix \(J\) connects the electric fields of the NIR laser and a sideband through

\[
\begin{align*}
E_{x,n} & = \left( \begin{array}{c} J_{xx,n} \cr J_{yx,n} \end{array} \right) \equiv E_{x,\text{NIR}} \\
E_{y,n} & = \left( \begin{array}{c} J_{yx,n} \cr J_{yy,n} \end{array} \right) \equiv E_{y,\text{NIR}}
\end{align*}
\]

where

\[
\begin{align*}
E_{x,\text{NIR}} & = \left( \begin{array}{c} \cos \alpha_n - \sin \alpha_n \cr \sin \alpha_n \end{array} \right) \left( \begin{array}{c} \cos \theta_n \cr \sin \theta_n \end{array} \right) e^{i\phi_n} \\
E_{y,\text{NIR}} & = \left( \begin{array}{c} -\sin \alpha_n \cr \cos \alpha_n \end{array} \right) \left( \begin{array}{c} \cos \theta_n \cr \sin \theta_n \end{array} \right) e^{i\phi_n}
\end{align*}
\]

The deviation \(k = e^{i\delta} = 0\) is satisfied with the initial condition \(\eta = 0\) and the THz electric field of \(R\) measures the deviation \(k\) with the sideband index \(m\). Each dynamical Jones matrix \(J\) was obtained. An average over sideband index \(m\) was calculated as the mean and the standard deviation of the generated distribution, respectively. Note that the dynamical Jones matrix elements are complex valued, and we set \(J_{xx,n} = 1\) in this study. Extended Data Fig. 6c shows the distributions of the dynamical Jones matrix elements produced from the distributions of \(a_{\text{NIR}}\) and \(y_{\text{NIR}}\) in Extended Data Fig. 6b.

**Effective Hamiltonian and low-energy Bloch wavefunctions**

In the basis consisting the eigenvectors of the position operator \(r\), a Bloch wavefunction with band index \(N\) and quasi-momentum \(k\) has the form \(\langle \psi_n | k \rangle = e^{ikr}u_n(r)\), where \(u_n(r)\) is a periodic function with the periodicity of the crystal. The eigenvalue problem of the Bloch wave functions \(H|\psi_n | k \rangle = E_n|\psi_n | k \rangle\) can be equivalently stated as an eigenvalue problem of the form \(H|\psi_n | k \rangle = E_n|\psi_n | k \rangle\), where \(H(k)\) is a \(3 \times 3\) Hermitian matrix. According to the \(\mathbf{k} \cdot \mathbf{p}\) method, in cases where the excited Bloch waves are located in energy bands that are relatively isolated and their quasi-momenta are restricted in a small portion of the Brillouin zone, a finite number of states \([u_n(k_r)]\) around quasi-momentum \(k_r\) can be approximately taken as a complete basis. On this finite basis, the Hamiltonian \(H(k)\) can be represented as a finite-dimensional matrix—the effective Hamiltonian, whose eigenfunctions, the low-energy Bloch wavefunctions, are linear combinations of the states \([u_n(k_r)]\). Determination of the effective Hamiltonian does not rely on the exact representations of the states \([u_n(k_r)]\) in real-space coordinates but their symmetry properties. The Luttinger Hamiltonian is an effective Hamiltonian with the basis chosen as four valence-band-edge states (Supplementary Discussion).

**Interference of Bloch waves**

We consider the case where the photon energy of the NIR laser lies just below the bandgap and assume that the sideband amplitudes are dominantly determined by electron–hole pairs created at \(k = 0\). Under an approximation of free electrons and holes, the amplitude of the nth sideband can be written as (Supplementary Discussion)

\[
P_n = \sum_{n=1}^{N_0} \frac{1}{2\pi\hbar} \int_0^{2\pi/\Omega} dt \int_0^{2\pi/\omega} d\epsilon \left( e^{\Omega(t-n)t} \right) R_n(t)^* R_n(t')
\]

where \(\omega\) and \(\Omega\) are the angular frequencies of the THz field and the NIR laser, respectively, \(V\) is the volume of the material, \(E_{\text{NIR}}(t) = F_{\text{NIR}}e^{i\omega t}\) is the electric field of the NIR laser under the rotating wave approximation, the two components of \(D_{\text{NIR}} = -d(\mathbf{a}, \mathbf{a}, /3)^3 \) and \(D_{\text{LH}} = -d(\mathbf{a}, \mathbf{a}, /3)^3\) are dipole matrix elements with spin-down (spin-up) electron and hole states with spin +1/2 (−1/2) and spin −1/2 (+1/2), respectively (\(d\) is a constant dipole matrix element), and \(R_n\) is a two-by-two unitary matrix that diagonalizes the hole Hamiltonian \(H_H(k_r)\) through \(R_n^*(\mathbf{n} \cdot \mathbf{t} - \gamma_{\text{NIR}}(\mathbf{n})\mathbf{t})\) with \(\mathbf{n}_i = \mathbf{n}_i/|\mathbf{n}_i|\). The first and second column of \(R_r(\mathbf{R}, \mathbf{R})\), respectively, represent the wavefunction of HH and LH on the basis of hole states with spin +1/2 (−1/2) and −1/2 (+1/2). The first (second) component of the quantity \(R_{\text{NIR}}^* \mathbf{D}_{\text{NIR}}(\mathbf{n}_i, \mathbf{t})\) represents the dipole matrix elements between E and HH (LH) bands. The acceleration process is described by the dynamic phase \(A_{\text{NIR}}(\gamma_{\text{NIR}}(\mathbf{n})\mathbf{t})\), which contains the quasi-momentum \(k_r(t') = F_{\text{NIR}}e^{i\omega t'}\) satisfying the initial condition \(k_r(t') = 0\) indicated by the subscript \(t'\) and \(\hbar\), \(\mathbf{e}_H, \mathbf{a}\), \(\mathbf{t}'\) = 0, with \(e\) being the elementary charge and \(E_{\text{NIR}}(t) = F_{\text{NIR}}e^{i\omega t}\) the THz electric field.
The three-step process of HSG can thus be described as interference of the following recollision pathways: a Bloch wave associated with an electron–hole pair $E\textnormal{–}HH$ ($E\textnormal{–}LH$) is first created by the NIR laser with amplitude proportional to $\mathbf{\Omega}_{\text{HH}}(t)$, acquires a dynamic phase $A_{\text{HH}}(t', t)$ during the acceleration phase from $t'$ to $t$, and generates sidebands through the dipole vector $\mathbf{\Phi}_{\text{HH}}$. The major contribution to the sideband amplitudes comes from the recollision pathways around the saddle points $(t', t)$ given by the stationary-phase conditions:

$$
-h \frac{\partial A_{\text{HH}}(t', t)}{\partial t'} + h\Omega = 0
$$

$$
= \int_{t'}^{t} \frac{\partial k_{\text{HH}}(t'')}{\partial t'} \left( \langle \mathbf{\Omega}_{\text{HH}}(t'') \rangle - \langle \mathbf{\Omega}_{\text{HH}}(t') \rangle \right) = 0
$$

$$
-h \frac{\partial A_{\text{HH}}(t', t)}{\partial t'} = \mathbf{E}_{\text{HH}}(t') - \mathbf{E}_{\text{HH}}(t) = h\Omega + nh\omega
$$

We have used the condition $\mathcal{E}(\mathbf{O}) - \mathcal{E}_{\text{HH}}(\mathbf{O}) = E_{\mathbf{F}} = h\Omega$, where $E_{\mathbf{F}}$ is the bandgap. Substituting the energy dispersion relations $\mathcal{E}_{\mathbf{F}}(k) = E_{\mathbf{F}} + \frac{\hbar k^2}{2m_{\mathbf{F}}}$ (mass of the effective mass of the conduction band), $\mathcal{E}_{\text{HH}}(k) = \frac{\hbar k^2}{2m_{\text{HH}}}$, and $\mathcal{E}_{\text{HH}}(k) = \frac{\hbar k^2}{2m_{\text{HH}}}$ with $m_{\text{HH}} = m_{\text{EHH}}(1 + 2y_1|\mathbf{n}_1|^2)^{-1}$ and $m_{\text{LH}} = m_{\text{EHH}}(1 + 2y_2|\mathbf{n}_1|^2)^{-1}$ are the effective masses of the HH and LH bands, respectively) into the stationary-phase conditions, we obtain

$$
\int_{t'}^{t} \frac{\partial k_{\text{HH}}(t'')}{\partial t'} = 0
$$

$$
\frac{\hbar k_{\text{HH}}(t)}{2\mu_{\text{ch}}} = nh\omega
$$

where $\mu_{\text{ch}} = \left( m_{\mathbf{F}}^{-1} + \frac{\hbar}{m_0} \frac{1 + 2y_1|\mathbf{n}_1|^2}{m_0} \right)^{-1}$ is the reduced mass of the $E\textnormal{–}HH$ ($E\textnormal{–}LH$) pair. The first equation has the meaning that the electron–hole pair returns to the position where they are created. The second equation states energy conservation at recollision. For each sideband order $n$, these two equations can be solved for the saddle-points $(t', t)$, which determine $k$-space trajectories $k_{\text{HH}}(t')$, as well as classical real-space trajectories with the velocities of $E$, HH and LH given by $h\mathbf{k}_{\text{HH}}(t')/m_{\text{HH}}$, $-h\mathbf{k}_{\text{HH}}(t')/m_{\text{HH}}$, and $-h\mathbf{k}_{\text{HH}}(t')/m_{\text{HH}}$. Figure 1b shows the shortest trajectory for the 24th-order sideband, and parameters $m_{\text{HH}} = 0.067m_0$, $m_{\text{HH}} = 0.711m_0$, and $m_{\text{HH}} = 0.081m_0$ are used in the calculation.

**Representation of Bloch wavefunctions**

The wavefunctions of the Hamiltonian $\mathcal{H}_n(k)$ are eigenfunctions of $\mathbf{\hat{n}}_{\mathbf{F}} \cdot \mathbf{\tau}$, which is defined on the basis of spin $\pm 3/2$ and $\pm 1/2$ states. We define $\mathbf{\hat{n}}_{\mathbf{F}} = \sin\theta \cos\phi \mathbf{\hat{r}} + \sin\theta \sin\phi \mathbf{\hat{z}} + c\text{c} \mathbf{\hat{z}} + c\text{c} \mathbf{\hat{z}} \phi$ as a point on a Bloch sphere with polar angle $\theta$ and azimuthal angle $\phi$, and write the eigenfunctions of $\mathcal{H}_n(k)$ as:

$$
\mathcal{H}_n = \begin{pmatrix}
\cos\left(\frac{\theta}{2}\right) & e^{i\phi}\sin\left(\frac{\theta}{2}\right)
\end{pmatrix}
$$

$$
\mathcal{H}_n = \begin{pmatrix}
-\sin\left(\frac{\theta}{2}\right) & e^{i\phi}\cos\left(\frac{\theta}{2}\right)
\end{pmatrix}
$$

The point on the Bloch sphere with coordinates $\hat{n}_{\mathbf{F}} = (\hat{n}_{\mathbf{F}})$ represents the state $|\mathcal{H}_n\rangle$ for the HH (LH) band. The angles $\theta$ and $\phi$ are determined from the measured $y_1$, $y_2$, and angle $\phi$ through the definition $\mathbf{\hat{n}}_{\mathbf{F}} = \left( \frac{2}{7} \sin(2\theta), \frac{1}{7} \cos(2\theta), -\frac{1}{7} \right)$.

**Data availability**

The datasets generated and/or analysed during the current study are available in the Materials Cloud repository. Source data are provided with this paper.

**Code availability**

The codes used in the data analysis are available at Zenodo.

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Performing experiments, data collection and analysis: S.D.O. and J.B.C. Software: J.B.C., D.C.V. and S.D.O. Theory: Q.W. Conceptualization: M.S.S. and Q.W. Resources (GaAs epilayer growth): K.W.W. and L.N.P. Development of broad-band polarimetry: D.C.V. Software: J.B.C., D.C.V. and S.D.O. Theory: Q.W. Conceptualization: M.S.S. and Q.W. Resources (GaAs epilayer growth): K.W.W. and L.N.P. Development of broad-band polarimetry: D.C.V. Writing: M.S.S., Q.W., S.D.O. and J.B.C. Supervision, funding acquisition and project administration: M.S.S.

**Competing interests**

The authors declare no competing interests.

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**Additional information**

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Extended Data Fig. 1 | Field enhancement at the GaAs epilayer from the ITO-coated sapphire substrate. The field enhancement is calculated as $|1 + r(f_{THz})|$ with the complex reflection coefficient $r(f_{THz})$ measured by a Vector Network Analyzer.
Extended Data Fig. 2 | An absorbance spectrum of the GaAs epilayer mounted on the ITO-coated sapphire substrate. The measurement was taken at the spot illuminated by a white light source (left inset). The right inset shows a zoom-in of the spectrum, with the bandgap and the photon energy of the NIR laser denoted by dash-dot blue and red lines, respectively. The two peaks are strain-split exciton resonances associated with band-edge states with different angular momenta. The temperature was 60 K.
**Extended Data Fig. 3 | Stokes polarimetry with linearly polarized NIR laser ($\psi_{\text{NIR}} = 0^\circ$).**

a. Polaragrams for sideband index $n = 12$ and orientation angle of the NIR laser $\alpha_{\text{NIR}} = 0^\circ$. b. The polarization state of the sideband extracted from the polaragrams in a. c. Polaragrams for sideband index $n = 24$ and orientation angle of the NIR laser $\alpha_{\text{NIR}} = 45^\circ$. d. The polarization state of the sideband extracted from the polaragrams in c. In a and c, the black dots show the measured polaragrams, with error bars showing the standard deviation over 4 measurements, and the red solid lines are the reconstructed polaragram through Fourier transform, with the red dotted lines showing the bounds. In b and d, the polarization states of the sidebands are represented as trajectories of the tips of the electric field vectors ($E_x, E_y$) over time. The orientation angle $\alpha$ and ellipticity angle $\gamma$ are defined in the inset in b.
Extended Data Fig. 4 | Quantum interference in three-step model of HSG leading to sideband polarization. A photon from the NIR laser is decomposed into components $\sigma_{\text{NIR}}$, with helicity ±1. a, A $\sigma_{\text{NIR}}^+$ photon excites either a spin-up electron and hole of spin $-3/2$ or a spin-down electron and hole of spin $-1/2$. A $\sigma_{\text{NIR}}^-$ photon excites either a spin-up electron and hole of spin $+1/2$ or a spin-down electron and hole of spin $+3/2$. b, Driven by the THz field, an electron-hole pair accumulates dynamic phase $A_{\text{HH}}$ or $A_{\text{LH}}$, depending on the band of the hole state (HH or LH). The electron spin is unchanged, while the hole states originating from the spin $-3/2$ state are superpositions of spin $-3/2$ and $+1/2$ states and the states originating from the spin $-1/2$ state are superpositions of spin $-1/2$ and $+3/2$ states. c, Upon recollision, either $\sigma_{\text{HSG}}^+$ or $\sigma_{\text{HSG}}^-$ photons are produced following angular momentum conservation—for example, a spin $+3/2$ hole recombining with a spin-down $(-1/2)$ electron produces a $\sigma_{\text{HSG}}^+$ photon with helicity $(+3/2 -1/2) = +1$. The interference of the evolution pathways from $\sigma_{\text{NIR}}^\pm$ to $\sigma_{\text{HSG}}^+$ ($\sigma_{\text{HSG}}^-$) produces the dynamical Jones matrix element $T_{++}$ ($T_{--}$).
Extended Data Fig. 5 | Additional data for ratios of Jones matrix elements, $\xi_n(\theta)$ and $\chi_n(\theta)$. a, The argument of $\xi_n(\theta)$. The dash-dot line marks the expected value of 0. b, The magnitude of $\xi_n(\theta)$. The dash-dot line marks the expected value of 1. c, The magnitude of $\chi_n(\theta)$. The dash-dot line marks the expected value of 1. All quantities are presented as functions of sideband index $n$ for eight values of angle $\theta$. Inset, The definition of $\theta$ by using the GaAs crystal lattice and the THz electric field.
Extended Data Fig. 6 | Monte Carlo simulation in calculating the dynamical Jones matrices. a, The polarization state of the n = 12 sideband (θ = 23°) for all 4 initial NIR polarizations (i-α_0IR = 0°, ii-α_0IR = 45°, iii-α_0IR = 90°, iv-α_0IR = −45°). The horizontal and vertical axes represent α and γ, respectively. Dashed ovals correspond to confidence intervals in the measurement of α and γ.

b, Histograms of α and γ for the 4 measured sidebands’ polarizations. Normal distributions of α and γ were sampled, with the central value and standard deviation of the distributions set by the measured values. In this figure, 1,000 iterations are shown, but the results of this paper are calculated from 10,000 iterations. c, The complex J-matrix elements resulting from the α and γ in b. The horizontal and vertical axes represent the real and imaginary part, respectively. Each red dashed line shows one standard deviation of the distribution of each J-matrix element resulting from the Monte Carlo simulation. All three plots have the same scale. The value of J_{xx,xx} is set as 1 in these calculations.
Extended Data Fig. 7 | Berry connection matrix element $A_{\text{HH}, \text{HH}}$, in the $k_z = 0$ plane of the Brillouin zone. The double-headed black dotted arrow represents a path of a hole accelerated by a linearly polarized THz field, which is perpendicular to the Berry connection (color arrows) at all points. The Berry connection is plotted in units of $a$, which is the lattice constant of GaAs.
Extended Data Fig. 8 | Effect of a biaxial strain on the valence band structure and non-Abelian Berry connection along the direction of quasi-momentum \( \mathbf{k} \) in the \( \mathbf{k}_z = 0 \) plane of the Brillouin zone. The strain is chosen as tensile along [001] direction to be consistent with the splitting of the exciton peaks in the absorbance spectrum (Extended Data Fig. 2). a, Valence band structures along \( k_x = k_z = 0 \) for unstrained (top) and strained (bottom) GaAs. The blue and orange curves represent the heavy-hole and light-hole bands, respectively. b, The magnitude of the diagonal Berry connection matrix element \( A_{HH,HH} \) along the direction of quasi-momentum for unstrained (top) and strained (bottom) GaAs. c, The magnitude of the off-diagonal Berry connection matrix element \( A_{HH,LM} \) along the direction of quasi-momentum for unstrained (top) and strained (bottom) GaAs. For the unstrained case, the Berry connection along the quasi-momentum is identically zero in the plots except for the singularity at \( k = \mathbf{0} \). The Berry connection is plotted in units of \( a \), which is the lattice constant of GaAs.