Engineering ultrafast spin currents and terahertz transients by magnetic heterostructures


1. Sample details. The glass/Fe(10nm)/Au(2nm) and glass/Fe(10nm)/Ru(2nm) heterostructures are grown in an ultrahigh-vacuum chamber (base pressure $5 \times 10^{-10}$ mbar) by electron-beam evaporation at room temperature. Prior to evaporation, the glass substrates are cleaned using KOH, propanol, and acetone to remove carbon contaminants.

The films grow in a polycrystalline, textured (or columnar) manner. Using scanning tunneling microscopy, the surface roughness of the Fe/Au and Fe/Ru structures is determined to 0.3 and 0.6nm root mean square (RMS), respectively (Figs. S1a,b). These values correspond to 1 and 2 monolayers and 2.5 and 5% of the total film thickness. Measurements on additionally grown glass/Fe(10nm) and glass/Ru(2nm) samples reveal that about 0.2nm of the surface roughness originate from the Fe film (Figs. S1c,d), whereas the remaining roughness arises from the cap layer. In particular, we do not observe Au island formation. (Au clustering occurs at elevated growth temperature and may lead to enhanced THz emission from Au alone, which is not observed as shown below.)

We note in passing that the observed film roughness is state of the art for spin-transport devices. For example, with the same growth method, we routinely fabricate giant-magnetoresistance (GMR) trilayer structures with a switching contrast of as large as 2.7 to 3.3% (Ref. S1).

Optical transmission and reflection measurements show that the Fe/Au and Fe/Ru heterostructures absorb 60% and 61%, respectively, of the incident pump power at an 800nm wavelength. These values compare well to a calculated absorption of 55% and 57%, respectively, obtained by a transfer matrix formalism and literature data of the optical constants of the materials involved. Such good agreement is indicative of an optically homogeneous metal film.

The magnetic properties of the heterostructures are characterized by mapping their in-plane sample magnetization as a function of an in-plane external magnetic field using a magnetooptic Kerr effect (MOKE) setup. We find nearly rectangular hysteresis loops (Fig. S2) with a coercive field of about 5mT. The switching characteristics are typical of polycrystalline Fe thin films.

Supplementary Figure S1 | Sample surface characterization. a-d Scanning tunneling micrographs of various film systems (bottom) along with typical line scans (top). Note that the surface roughness of the Fe/Ru heterostructure (see b) approximately equals the sum of those of the single layers (see c, d).
2. Terahertz emission: magnetization state, isolated cap layers, directional dependencies. In our experiment, we detect the THz pulse emitted from the laser-excited heterostructures by means of electrooptic sampling\textsuperscript{S4,S5}. Figures 3a (main text) and S3 display typical signal waveforms $S(t)$ as a function of time $t$. Here, $S$ equals the $x$ component of the transient THz field $E(t)$ directly after the sample (Fig. 1a main text), convoluted with the response function of our setup\textsuperscript{S5}.

Figure S3 compares THz waveforms obtained from a Fe/Au heterostructure in a saturated (external field 80mT, much larger than the coercive field) and in a remanent (zero external field) magnetization state. We observe almost identical THz waveforms, which implies identical pump-induced charge currents in both cases. Note, however, that the signal from the remanent sample has a factor of 1.9 smaller amplitude than that from the saturated sample. A similar reduction of the MOKE signal was recently observed on ferrimagnetic Gd$_{23.1}$Fe$_{71.9}$Co$_{5.0}$ and assigned to the irreversible formation of randomly oriented magnetic domains in the pump-illuminated volume\textsuperscript{S6}. Comparable behavior of the THz emission is observed for the Fe/Ru system as well (not shown).

Figure S3 also shows THz signals obtained for single, 2-nm thin Au and Ru films on glass under external conditions identical to those for the heterostructures in the saturated magnetization state. We do not observe any THz emission above the noise floor of our experimental setup. Therefore, THz currents in isolated Au and Ru films make a negligible contribution to the THz radiation emitted by the heterostructures.

Finally, Fig. S4a shows the THz peak signal as a function of the direction of the linearly polarized pump pulse (with respect to the $y$ axis in Fig. 1a main text), whereas the impact of the magnetization direction (in plane, with respect to the $y$ axis) is displayed in Fig. S4b. The cosine-type behavior observed (Fig. S4b) is consistent with the directional dependence expected from the inverse spin Hall effect (ISHE) [see equation (1) main text]. Figure S4b furthermore implies that the THz pulse is linearly polarized within the accuracy of our experiment.

Supplementary Figure S2. Hysteresis loop of the Fe/Au heterostructure as obtained with a MOKE magnetometer in longitudinal geometry. A nearly identical curve is found for the Fe/Ru system (not shown).

Supplementary Figure S3 I Test measurements. The curves represent THz signals $S(t)$ from Fe/Au heterostructures in case of saturated and remanent Fe magnetization as well as signals from single, 2-nm thin Au and Ru films on glass.

Note that these test data were taken with a different setup than that used to obtain the data of all other figures. Compared to Fig. 3a (main text) or Fig. S6a, the THz signals shown here are substantially distorted, owing to a reduced time resolution of 50fs and absorption by water vapor that is present in ambient air.
3. *From electrooptic signals to electric fields.* In the frequency domain, the transient THz field $E_x$ directly after the sample (see Fig. 1a main text) and the electrooptic signal $S$ are connected by the relationship

$$S(\omega) = H(\omega)E_x(\omega) \quad (S1)$$

where $H(\omega) = H_{prop}(\omega)H_{det}(\omega)$ is the response function of our setup at frequency $\omega/2\pi$. In equation (S1), $H_{prop}$ accounts for the propagation of the THz pulse from the sample to the THz detector (including focusing), and $H_{det}$ quantifies the electrooptic detection of the THz pulse by the 10fs, 800nm gate pulse in the 250µm thick GaP(110) crystal.

We calculate $H_{prop}$ using the geometrical parameters of our spectrometer (distances, focal lengths and apertures of the parabolic mirrors) and the theory of Ref. S7, whereas $H_{det}$ is calculated using Ref. S8 and the optical constants of GaP published in Ref. S9. As first pointed out in Ref. S10, the dispersion of the electrooptic coefficient has also to be taken into account. Figure S5 shows the modulus of $H_{prop}(\omega)$, $H_{det}(\omega)$, and of the resulting total response function $H(\omega)$. The zero of $H$ at about 8 THz (Fig. S5c) arises from the GaP electrooptic coefficient, which also shows up in the signal spectra (Fig. S6b).

Figures S6a-d detail the steps taken to extract $E_x$ from $S$. The relationship between $E_x$ and its source $\langle j_c, x \rangle$ is derived in the next section.

4. *From fields to currents.* As within our sample, the beam diameter is much larger than the sample thickness ($d=12$nm) and the wavelengths involved (~100µm), plane-wave propagation along the $z$ axis (see Fig. 1a main text) is assumed. The charge current density $j_c$ resulting from the laser-driven spin current $j_s$ and the ISHE [equation (1) main text] generates an electromagnetic wave with electric field $E(z,t)$. In the frequency domain, the dynamics of this field is governed by the wave equation\textsuperscript{111}

$$\left[ c_s^2 + k^2(z, \omega) \right] E(z, \omega) = Z_0 \omega j_c(z, \omega)/i\epsilon. \quad (S2)$$

### Supplementary Figure S4 | Directional dependencies of THz emission.

**a** Peak of the THz signal $S(t)$ versus polarization direction of the normally incident linearly polarized pump pulse. **b** Peak $S(t)$ versus direction of the external magnetic field together with a cosine curve. The field is applied within the sample plane.

### Supplementary Figure S5 | Calculated transfer functions of the THz emission setup.

**a** Propagation from the sample to the electrooptic detection crystal. Focusing of the THz pulse leads to a typical linear increase of $H_{prop}$ with increasing frequency. **b** Response function of the electrooptic detector consisting of a 250µm thick GaP(110) crystal in conjunction with a 10fs, 800nm sampling pulse. **c** Total detector response.
Supplementary Figure S6 | Work steps for the extraction of the transient charge current from the detected THz signal. a Raw data: electrooptic signal S(t) of THz radiation emitted from the Fe/Au heterostructure. b Amplitude spectrum |S(ω)| obtained by a Fourier transformation (FT) of S(t). c Spectrum |E_x(ω)| of the THz field after the sample, obtained by using equation (S1). d Electric field transient E_x(t) directly after the sample as obtained by an inverse Fourier transformation (IFT) of E_x(ω). e Spectrum of the transient charge current, obtained by using equation (2). f Transient z-averaged charge current ⟨j_z(t)⟩ as obtained by an IFT of ⟨j_z(ω)⟩.

Here, c is the vacuum speed of light, Z_0 = 377Ω, and k(ω)=n(ω)ω/c is the wavevector of the THz wave of frequency ω/2π. The frequency-dependent refractive index n(ω) is taken from Refs. S12, S12, S13, and S14 for Fe, Au, Ru, and glass, respectively. For layered media (such as the heterostructures used in this work), equation (S2) can be solved with the Green’s function approach presented in Ref. S15. For example, if we assume for the moment that the two metal films on the substrate have the same refractive index, the electric field directly after the sample reads

\[ E(\omega) = C(\omega) \int dz j_c(z, \omega) \left[ e^{-i k z} + r_{21} e^{i k z} \right]. \]  

(S3)

Here, \( C(\omega) = -Z_0 Re^{i k d}/(n_2^2 + n_3) \), \( k_2 = n_2 \omega/c \), \( n_{ij} = (n_i - n_j)/(n_i + n_j) \) with \( n_i(\omega) \) being the refractive index of medium \( i \) where \( i=1,2,3 \) denotes substrate, metal film, and N_2 gas, respectively. The factor \( R = 1/(1 + r_{12} r_{23} e^{2 i k d}) \) accounts for all multiple reflections between the two metal-dielectric interfaces. We now make the electric dipole approximation (\( e^{\pm i k z} \approx 1 \) because \( |kd| \ll 1 \) in the frequency range considered) and exploit the fact that the THz refractive index of metals is much larger than that of the substrate and of gaseous N_2. After some algebra, equation (S3) delivers

\[ \int_0^d dz j_c(z, \omega) = -E(\omega) \int_0^d dz \sigma(z, \omega). \]  

(S4)

that is, the equation (2) of the main text. Here, the conductivity \( \sigma \) is related to the refractive index \( n \) by \( \sigma = (n^2 - 1)\omega/4\pi Z_0 \). Although equation (S4) was derived for a single metallic layer between two half spaces (glass substrate and N_2 gas), it remains valid for the heterostructures of this work (Fig. 1a main text) because the total metal thickness (\( d=12nm \)) is much smaller than the THz wavelength (~100μm) and the THz penetration depth into metals (~100nm). This result can be shown using the optical Green’s function for two films between two half spaces.S15
Note that equation (S4) can also be justified in a simpler way based on a quasi-static argument: All THz radiation generated within the metal film will be reflected between the metal-dielectric interfaces many times because their reflectivity is approximately one and propagation loss is relatively low. The various reflection echoes add up constructively, leading to an approximately constant electric field at all positions \( z \) within the metal region. Thus, we have \( \partial_z E(z, \omega) \approx 0 \), which still holds across the metal-dielectric interfaces because the electric field is continuous there. Then, integrating equation (S2) from \( z = 0 \) to \( d \) and translating refractive index into conductivity yields equation (S4).

Finally, Figs. S6e,f show the steps that lead us from \( E_x \) to its source \( \langle j_{x,x} \rangle \).

5. **Magnetic dipole radiation.** Besides spin transport, the pump pulse also induces an ultrafast reduction of the sample magnetization \( M \), which leads to the emission of magnetic dipole radiation \(^{S16}\). In order to estimate this contribution to the emitted THz field, we substitute the charge current density \( j_x = \partial_z M_y \) arising from \( M \) into equation (S3). This current distribution has vanishing electric dipole moment, and the magnetic dipole contribution is obtained by expanding \( e^{i k z} \approx 1 + i k z \) in the integrand of equation (S3), which finally leads to an emitted field

\[
E_x(\omega) = -i \omega n_s \langle M_y(\omega) \rangle / c(\sigma(\omega)).
\]  

\[ (S5) \]

Here, angular brackets again denote a \( z \) average over the thickness of the whole metal film. By comparing our excitation conditions to those of Ref. S17, we find the ground-state magnetization of the Fe film is reduced by 7% in a temporally step-like manner. Using this \( M_y(t) \) in equation (S5) yields spectral amplitudes \( |E_x(\omega)| \) of the electric field after the sample that are an order of magnitude smaller than those measured (Fig. S7).

6. **Spin pumping.** When the magnetization of a ferromagnetic thin film is modulated, spin angular momentum is transferred into an adjacent normal metal film \(^{S18}\). To estimate the spin current resulting from such spin pumping in our experiment, we use Ref. S19, where the spin current is given by

\[
j_{sx}(\omega) = g \omega \sin^2 \theta / 4 \pi.
\]  

\[ (S6) \]

Here, \( g \approx 10 / \text{nm}^2 \) is the spin-mixing conductance of the Fe/Au interface \(^{S20}\), \( \omega/2\pi \sim 10 \text{THz} \) is the mean frequency of the spin dynamics, and \( \theta \) is the magnon opening angle that is approximated by the relative magnetization change of \( |\Delta M| / |M| \approx 0.1 \). We obtain \( j_{sx} \sim 10^{30} \text{ electrons/s m}^2 \), which is two orders of magnitude smaller than the spin current density predicted by our superdiffusion simulations (see Fig. 4b main text).

**Supplementary Figure S7 | Expected emission due to demagnetization.** Spectral amplitude of THz transient field after the sample as measured for Fe/Ru and Fe/Au heterostructure (blue and red curves). The estimated contribution of the magnetic dipole radiation arising from the ultrafast Fe demagnetization is also shown (black dashed curve).
7. Simulation of superdiffusive spin transport. A detailed description of the treatment of the superdiffusive spin transport, the numerical techniques used to solve the superdiffusion equation, and the expression of the magnetization flux can be found in Ref. S21.

8. Calculation of spin Hall angles. The spin Hall conductivity (SHC) is calculated as a function of electron energy by means of the Kubo formula within density-functional theory (DFT) using the full-potential linearized augmented plane-wave (FLAPW) program FLEUR\(^{S22}\). We employ the generalized gradient approximation of the exchange correlation potential, a plane-wave cutoff at a wavevector of 85\(\text{nm}^{-1}\), and the experimental lattice constant of 2.866\(\text{Å}\). In order to facilitate the efficient computation of the SHC, a set of 18 first-principles Wannier functions\(^{S23,S24}\) was constructed to describe the electronic structure up to 10eV above the Fermi energy. Further details on the computational method are given in Ref. S25.

In Fig. S8a, the calculated SHC of ferromagnetic Fe is plotted as a function of energy. At roughly 0.33eV above the Fermi energy \(E_F\), the sign of the SHC changes from plus to minus. The hot non-equilibrium electrons excited by the femtosecond laser pulse are approximately at 0.5eV above the Fermi energy, where \(\sigma^{\parallel}_{xy}=-157\text{S cm}^{-1}\). To obtain the spin Hall angles, we divide the SHC by the experimentally determined conductivity of 1.1\(\times\)10\(^5\)S cm\(^{-1}\) of Fe (Ref. S26).

It is noteworthy that the SHC sign change versus energy in Fe (Fig. S8a) occurs in an opposite manner as the SHC sign change versus band filling of paramagnetic transition metals\(^{S27}\). Indeed, Fig. S8b shows that the SHC of paramagnetic Fe exhibits only a sign change from minus to plus as the d-shell is filled. The “anomaly” seen in Fig. S8a results from the nonlinear nonmonotonous energy dependence of the SHC of paramagnetic Fe and exchange-split majority- and minority-spin bands of ferromagnetic Fe. This effect is illustrated by Fig. S8c which shows the SHC of majority and minority electrons as calculated for paramagnetic Fe yet energetically shifted to enforce the correct numbers of majority and minority conduction band electrons. Adding both curves yields the approximate SHC of ferromagnetic Fe. Comparison of Figs. S8a and S8c shows that our two-current model reproduces the qualitative features of the \textit{ab initio} calculations quite well.

Supplementary Figure S8 I Inverse spin Hall Effect in Fe. a Spin Hall Conductivity \(\sigma^{\parallel}_{xy}\) (SHC) of ferromagnetic Fe (magnetization in \(z\) direction) as a function of energy \(E\) above the Fermi energy \(E_F\). The spin Hall angles are obtained by dividing the SHC by the experimentally determined Fe conductivity of 1.1\(\times\)10\(^5\)S cm\(^{-1}\) (Ref. 26). b SHC of paramagnetic Fe as a function of \(E-E_F\). c Two-current model of SHC for ferromagnetic Fe constructed from the data in b.
References


Also see Parge, A. Current-Induced Excitations in Ferromagnetic Single Layer and Trilayer Nanodevices (Dissertation, University of Göttingen, 2007, online http://webdoc.sub.gwdg.de/diss/2007/parge/)

S2. Yeh, P. Optical waves in layered media (Wiley, 2005).


S22. See http://www.flapw.de


