Femtosecond Spin Current Pulses Generated by the Nonthermal Spin-Dependent Seebeck Effect and Interacting with Ferromagnets in Spin Valves

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Using the sensitivity of optical second harmonic generation to currents, we demonstrate the generation of 250-fs long spin current pulses in Fe/Au/Fe/MgO(001) spin valves. The temporal profile of these pulses indicates ballistic transport of hot electrons across a sub-100 nm Au layer. The pulse duration is primarily determined by the thermalization time of laser-excited hot carriers in Fe. Considering the calculated spin-dependent Fe/Au interface transmittance we conclude that a nonthermal spin-dependent Seebeck effect is responsible for the generation of ultrashort spin current pulses. The demonstrated rotation of spin polarization of hot electrons upon interaction with noncollinear magnetization at Au/Fe interfaces holds high potential for future spintronic devices.

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Optimization and control of spin currents (SC) and their interaction with magnetic constituents in heterostructures on a femtosecond time scale is key for future terahertz spintronics applications. Although electronic transport through a ferromagnet (FM), as described by Mott’s two current model [1], generates a spin-polarized current, its density is intrinsically limited by Joule losses. The discovery of the spin-dependent Seebeck effect (SdSE), where thermal gradients over a bulk FM [2] or across an interface to a normal metal [3] generate SCs, opened a path towards overcoming such limitations. Indeed, short-lived thermal gradients can produce short (~100 ps) SC pulses at densities exceeding the static Joule limit, as recently demonstrated upon laser excitation of spin-valve structures [4].

Creating highly energetic electrons [5–9], femtosecond laser excitation is promising for SC pulse generation on subpicosecond time scales, before the electron-electron [9] and electron-lattice [8] equilibration is reached. Superdiffusive transport of laser-excited spin-polarized hot carriers on a femtosecond time scale [10,11] was evidenced in a plethora of experiments [12–18]. However, unraveling the underlying microscopic picture requires understanding the influence of the electron dynamics in a FM, the scattering in metallic layers, and the properties of interfaces on the SC pulse. For this, a direct SC detection is highly desirable yet challenging in the presence of laser-induced magnetization dynamics [19].

In this Letter we argue that under nonequilibrium conditions, the SdSE at interfaces can lead to the efficient generation of femtosecond SC pulses paving the way for future THz spintronics. Employing a nonlinear-optical approach, we demonstrate the generation of these pulses at the Fe/Au interface and Stern-Gerlach-like spatial spin separation upon their interaction with an orthogonal FM magnetization. The latter facilitates the development of nondissipative metallic spin polarizers and rotators for ultrafast spintronics.

In SdSE, spin transport is determined by gradients of the electron distribution function \( \nabla f(E) \), which are drastically enhanced at interfaces. The difference \( \Delta f(E) \) across the interface enables the flux of electrons \( e \) at energy \( E > E_F \) or holes \( h \) at \( E < E_F \), where \( E_F \) is the Fermi energy. A charge current \( j \) emerges when the total fluxes of \( e \) and \( h \) are unequal due to an asymmetry (with respect to \( E_F \)) of \( \Delta f(E) \), conductance [20], or the interface transmittance \( T \), known as the Seebeck effect. If \( j_\uparrow \neq j_\downarrow \) in the majority and minority subbands of FM, the resulting \( j = j_\uparrow + j_\downarrow \) is accompanied by the SC \( j^S \propto j_\uparrow - j_\downarrow \).

Now we analyze SdSE at the Fe/Au(001) interface for thermal and nonthermal \( f(E) \). From ab initio quantum transport calculations [21] we have obtained the momentum-averaged interface transmittance \( T_{FA} \) for spin-\( \uparrow \) and -\( \downarrow \) carriers moving from Fe to Au [Fig. 1(a)]. In the thermal case, the carriers at energies within \( k_B T_e < 100 \) meV around \( E_F \) are responsible for the transport [4,20], where \( T_e(z) \) is the electron temperature. Therefore, the thermal SdSE originates from the slope of transmittance \( \nabla z T_{FA}(E_F) \) [Fig. 1(a)] and \( \Delta f(E) \) determined by the temperature gradient \( \nabla z T_e \) across the interface.

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A much broader nonthermal $f(E)$ is formed by 1.5 eV laser excitation of Fe [shaded areas in Fig. 1(a)] [14]. High optical reflectance ensures the absence of excited carriers in Au providing much larger $\Delta f$ than in the thermal case. Excited into the $sp$ band of Fe [14,24], $e^\uparrow$ have much larger transmittance [Figs. 1(a),1(b)] than other carriers in the $d$ band which poorly matches the $sp$ band of Au. Moreover, much higher velocity of $e^\uparrow$ in Fe [25] provides their better transport towards the interface. Thus, an efficient injection of $e^\uparrow$ into Au [Figs. 1(b),1(c)] results in SC pulses with the spin component of $j^z_{s,p}$ along $\hat{z}=\hat{x}$, $\hat{y}$ set by the Fe magnetization and the average electron velocity $v$ along $\hat{z}$ [Fig. 1(c)]. Noteworthy, a similar spin-dependent interface transmittance can be expected in any itinerant FM/noble metal bilayer [26] facilitating demagnetization of the FM by superdiffusive spin transport [10,11].

In this nonthermal SdSE, the collective displacement of “cold” non-spin-polarized electrons near $E_F$ in Au partially screens the charge but not the spin component of the $e^\uparrow$ transport. Therefore, compared to the thermal case, SC pulses of much larger amplitude can be expected while the net charge transfer is suppressed. Furthermore, the pulse duration here will be primarily determined by the electron thermalization time in FM $\tau_{th}^{FM}<0.5$ ps [9,27]: once the nonthermal $f(E)$ [Fig. 1(a)] is relaxed to the Fermi-Dirac distribution, the remaining SC is of purely thermal origin. The thus expected generation of ultrashort and intense SC pulses is demonstrated below.

To achieve steady control of propagation direction and spin polarization of SC pulses, we combine the spin-valve concept with the pump-probe approach developed in Ref. [14]; see Fig. 1(c). The two Fe layers in the epitaxial Fe/Au/Fe/MgO(001) stack [Fig. 2(a)] are magnetically decoupled and can be optically excited and probed independently owing to the 50 nm-thick Au spacer. The samples were fabricated following Ref. [14] and examined with transmission electron microscopy, see Figs. 2(a)–2(c). The excellent epitaxial quality and flatness of interfaces, are essential for comparison with the ab initio calculations performed for an atomically sharp interface.

Magnetoinduced second harmonic generation (mSHG) [28,29] is a promising tool for direct SC detection owing to its high temporal resolution and sensitivity to the current-driven symmetry breaking, as demonstrated in GaAs [30,31] and multilayer graphene [32]. We performed all-optical experiments in either the front or back pump-probe scheme shown in Fig. 1(c). The 800 nm, 14 fs, 1 MHz output of a cavity-dumped Ti:sapphire oscillator (Mantis, Coherent) was split at a power ratio 4:1 into pump and probe pulses both focused into an ~10 µm spot resulting in a pump fluence of ~10 mJ/cm². The magneto-optical Kerr effect (MOKE) was measured with a balanced two-photon diode detector while the second harmonic signal was registered by a photomultiplier [14].
laser excitation, the propagating electrons break the applied along the two easy axes [Figs. 2(d)]. In the pump-probe experiments, the mSHG intensity was observed in Fe behind the Au/Fe interface [cf. (b) and Eq. (2)]. (b) Hot electron behavior at the Au/Fe interface in orthogonal (N and NL) configurations. (c) Calculated momentum-averaged transmittance of the Au/Fe interface for majority ($T^\uparrow_{AF}$) and minority ($T^\downarrow_{AF}$) carriers moving from Au to Fe [36].

![Figure 3](image365x630 to 432x704)

**FIG. 3.** Variations of nonmagnetic and current-induced (a) and magnetization- and SC-induced (b) components of the second harmonic electric field observed in p-in, p-out polarization geometry. SC vs M configurations for the front (F) and back ($P, A, N$) pumping are shown in the legend [cf. Fig. 1(c)]. The solid line represents the common background due to heating-induced demagnetization.

The second harmonic field $E_{2\omega} = E_e + E_m$ consists of “electronic” $E_e$ and magnetoeinduced $E_m$ terms which are, respectively, independent of and proportional to the magnetization $M$ [28]. In this work ($x, z$) is the plane of incidence and we use p-in, p-out polarization geometry. Then, $E_m$ is proportional to $M_z$, the transversal magnetization component [14,28] while the MOKE is sensitive to the longitudinal $M_z$ [33]. In order to set the magnetization of collector $M_{col}$ and emitter $M_{em}$ [Fig. 1(c)], transversal ($H^T||\hat y$) and longitudinal ($H^L||\hat x$) magnetic fields were applied along the two easy axes [Figs. 2(d)–2(g)] [34]. In the pump-probe experiments, the mSHG intensity was measured in transversal and longitudinal geometries for positive and negative $M_z$ and $M_{col}$, respectively. From these data, we retrieved $E_{c0}, E_{m0}$ for each pump-probe delay $t$ and $E_{c0}, E_{m0}$ in the absence of pump [35]. In the following, we discuss relative variations $\Delta E_c/E_{c0}$ and $\Delta E_m/E_{m0}$ which are shown in Figs. 3, 4.

In equilibrium, $E_e = E^{\text{int}}_e$ and $E_m = E^{\text{int}}_m$ originate from the interface dipole polarization [37] $P^{2\omega}_i = \chi^{(2)}_{ijk\omega}P_{E_{k\omega}}M_{i\omega} + \chi^{(2,m)}_{ij\omega}P_{E_{k\omega}}^{E_{m\omega}}M^{\text{col}}_{i\omega}$, where $i, j, k = x, z$. However, after fs-laser excitation, the propagating electrons break the inversion symmetry. Thus, bulk dipole current- and SC-induced polarizations $\chi^{(2,C)}_{ijk\omega}E_{j\omega}^{E_{k\omega}}, \chi^{(2,SC)}_{ijk\omega}E_{j\omega}^{E_{k\omega}}$ appear [39]. They contribute to $E_{2\omega}$ as

$$E^C_e \propto j_z \propto v_z, \quad E^{SC}_m \propto j^{S}_{z,y} \propto v_z s_y, \quad (1)$$

where $j^{S}_{z,y}$ is the SC tensor component with the electron velocity $v_z$ and spin polarization $s_y$ [40]. Thus, in addition to the modulation of the interface contributions, the total pump-induced variations $\Delta E_{c,m}$ contain bulk terms (1) which provide direct access to the charge and spin currents [41]:

$$\Delta E_c = \Delta E^{\text{int}}_e + E^C_e, \quad \Delta E_m = \Delta E^{\text{int}}_m + E^{SC}_m. \quad (2)$$

This is demonstrated in Fig. 3 where we excite currents from the front (F) or back side of the sample. In the latter case, we consider parallel ($P$), antiparallel ($A$), and non-collinear ($N$) alignment of $M_{em}$ with respect to the transversal $M_{col}$. First, we discuss the pronounced characteristic features at $t < 0.5 \text{ ps} \sim r_{\text{FM}}^{\text{AF}}$. In the $F$ configuration, we observe $\Delta E^{\text{F}}_c > 0$ [Fig. 3(a)] and $\Delta E^{\text{F}}_m < 0$ [Fig. 3(b)]. Then we reverse $v$ keeping the direction of $s$ (determined by $M_{em}$) and find in this $P$ configuration $\Delta E^{\text{P}}_c < 0$ and $\Delta E^{\text{P}}_m > 0$, in agreement with the sign change of $j_z$ and $j^{S}_{z,y}$ in Eq. (1). Reversing $j^{S}_{z,y}$ (but not $j_z$) by keeping $v_z$ but changing the sign of $s_y$ ($A$ configuration) only slightly affects $\Delta E_c$ [Fig. 3(a)] while $\Delta E_m$ changes its sign: $\Delta E^{A}_m < 0$ [Fig. 3(b)]. Lastly, we rotate $s$ by $90^\circ$ ($N$ configuration) to set $j^{S}_{z,y} = 0, E^{SC}_m = 0$ [cf. Eq. (1)] and
obtain no sizable $\Delta E_N^m$. Since the major part of $\Delta E_m$ changes its sign with reversal of $v_\perp$ or $s_z$ (i.e., of $J_{\perp}^m$), we conclude on the dominant role of $E_m^{2N}$ in the mSHG response at short delays. Thus, we unambiguously observe ultrashort SC pulses in Fig. 3(b).

At $t > \tau_{\text{th}}^{\text{FS}}$, we expect a reduction of $E_m^{\text{int}}$ by thermalized electrons heating up the collector: $\Delta E_m^{\text{int}} \propto \Delta M_{\text{col}}^m$. This agrees with the observed $\Delta E_m^{p,A,N}(t)$ coinciding at $t > 2$ ps [Fig. 3(b)]. Subtracting this trend (solid line) from $\Delta E_m^p$, we obtain a unipolar trace [Fig. 4(a)] with a sharp onset in contrast to the bipolar behavior of $\Delta E_m^{A,N}$ [Fig. 3(b)]. To understand this difference and its relation to the SC profile [41], we discuss the interaction of spin-polarized electrons with the Au/Fe interface [Fig. 4(b)] determined by $T_{\text{AF}}$ [Fig. 4(c)]. After considering open ($P$) and closed ($A$) states of the spin valve important for GMR devices, we turn to $N$ and $NL$ (noncollinear, longitudinal collector) configurations with $M_{\text{em}} \perp M_{\text{col}}$ having high potential functionality for spin transfer torque applications [42].

Averaging the calculated interface transmittance over the energy of emitted electrons we obtain $T_P = \langle T_{\text{AF}}^P \rangle \approx 0.95$ and $T_A = \langle T_{\text{AF}}^A \rangle \approx 0.25$. Therefore, in the $A$ configuration, the reflected SC modifies $E_m^{2\text{SC}}$ [43] leading to bipolar $\Delta E_m^A(t)$ similar to that measured at the Au surface [14]. In the $P$ configuration, the SC is absorbed by the collector [44] and $\Delta E_m^p(t)$ in Fig. 4(a) represents the temporal profile of incoming SC pulse. For ballistic electrons traveling in Au with the Fermi velocity $1.4$ nm/fs [45] and random angular distribution the average propagation time $\tau_b$ is $70$ fs. The maximum of $\Delta E_m^p(t)$ observed at this delay thus suggests ballistic transport [14] with the length $\lambda_{\text{Au}} \gtrsim 100$ nm for electrons at $0.3 < E - E_F < 1.5$ eV [46]. We conclude that $\Delta E_m^p(t)$ closely reproduces the dynamics of electron emission. The similarity of its decay time of $250$ fs [Fig. 4(a)] to the electron thermalization time $\tau_{\text{th}}^{\text{FS}} \approx 200$ fs [47] strongly corroborates the nonthermal SdSE mechanism considered above. Its high efficiency is evident from the negligibly small contribution of thermal SdSE [4], which is within the noise level here.

We now turn to the $NL$ configuration where the longitudinal $M_{\text{col}}$ excludes demagnetization effects in $E_m^{\text{int}}$ while the transversal $M_{\text{em}}$ provides $E_m^{2\text{SC}} \neq 0$. The striking similarity of $\Delta E_m^p$ and $\Delta E_m^{NL}$ at $t < 1$ ps [Fig. 4(a)] indicates a negligible contribution of the spin polarization of reflected electrons. Within the single-particle approach, we treat an electron with spin $s \perp M_{\text{col}}$ representing its wave function $\langle \Psi_s \rangle = \langle | \Psi_{\downarrow} \rangle + | \Psi_{\uparrow} \rangle \rangle / \sqrt{2}$ as a superposition of the spin eigenfunctions [48] corresponding to $s \uparrow M_{\text{col}}$ and $s \downarrow M_{\text{col}}$ which were discussed above for $P$ and $A$ configurations. Considering diagonal transmission operators, we obtain that the transmittance for electrons with $s \perp M_{\text{col}}$ is the average of transmittance for electrons with $s \uparrow M_{\text{col}}$ and $s \downarrow M_{\text{col}}$. A limiting case of $T_P = 1$ and $T_A = 0$ is illustrated in Fig. 4(b): only the spin-up (down) component of the incoming wave is transmitted (reflected). In other words, all electrons with orthogonal spins interacting with FM rotate their spins into parallel and antiparallel states with equal probabilities. Subsequently, electrons with parallel spins are transmitted and antiparallel reflected; i.e., the two spin components are separated in space like in the Stern-Gerlach experiment. This rotated spin polarization does not contribute to $\Delta E_m^{NL}$, which explains the similarity of the latter to $\Delta E_m^p$.

In the general case, transmittances differ from 1 and 0 [Fig. 4(c)] resulting in nonzero residual ($s \perp M_{\text{col}}$) spin polarization $s_{\perp}$ in both reflected and transmitted currents [48], $|s_{\perp}^R|^2 \leq \sqrt{R^2 R^p} |s_{\perp}|^2$ and $|s_{\perp}^0|^2 \leq \sqrt{T^2 T^p} |s_{\perp}|^2$, where $R = 1 - T$. The rotated components ($s \parallel M_{\text{col}}$) are given by $s_{\parallel}^R = (R^p - R^A) |s_{\parallel}|^2 / 2$ and $s_{\parallel}^0 = (T^p - T^A) |s_{\parallel}|^2 / 2$. These $s_{\parallel}$ and $s_{\perp}$ are addressed in $N$ and $NL$ configurations, respectively. Using Eqs. (1)–(2), we obtain $\Delta E_m^{NL}(t) = (\Delta E_m^p + \Delta E_A)/2$, in line with Ref. [48]. Experimentally, this holds for both $\Delta E_m$ thus indicating the key role of $s_{\parallel}^R$ in the positive $\Delta E_m^p$ observed at $0.5 < t < 2$ ps. The negligible effect of the spin polarization of reflected electrons in $NL$ configuration agrees well with the calculated $R_A$, for which $|s_{\parallel}^R|$ does not exceed $|s_{\perp}| / 5$.

The angular momentum conservation upon interaction with the Au/Fe interface together with the quantum decoherence of the $|\Psi_{\uparrow} \rangle + |\Psi_{\downarrow} \rangle$ superposition ($s_{\perp}$ can be up to $s_{\perp}/2$) within the inelastic mean free path $\lambda_{\text{Au}} < 1$ nm [25], leads to the emergence of $\Delta M_{\text{col}} \perp M_{\text{col}}$ in the vicinity of the interface [Fig. 4(b)]. This ultrashort spatially confined spin transfer torque effect inducing $\Delta M_{\text{col}}$ at the interface is responsible for the small deviation of $\Delta E_m^{NL}$ from $\Delta E_m^p$ observed in Fig. 4(a) at $t > 0.5$ ps. Subsequently, several lowest standing spin wave modes at the frequencies up to $0.6$ THz are excited in the collector resulting in the nonmonotonic behavior of $\Delta E_m^{NL}$ at $t > 1$ ps [Fig. 4(a)], see Ref. [42].

Summarizing, using the high sensitivity of nonlinear-optical probe to the transient inversion symmetry breaking, we have demonstrated the generation of ultrashort spin current pulses in Fe/Au/Fe epitaxial multilayers. The measured pulse shape agrees with the proposed nonthermal spin-dependent Seebeck effect and indicates ballistic transport of spin-polarized electrons in Au. The pulse duration ($\sim 250$ fs) is determined by the electron thermalization time in Fe. We have shown the large difference in transmittance of the Au/Fe interface for the spin-polarized electrons with $E - E_F < 1.5$ eV. This results in a high spin rotation efficiency of 70% at the interface, where the transmitted (reflected) current loses its orthogonal spin component and becomes polarized parallel (antiparallel) to the Fe magnetization. These findings facilitate the development of metal-based sources.
of ultrashort spin current pulses and nondissipative reflective or transmissive spin polarizers and rotators for ultrafast spintronics.

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[21] We have calculated the full spin-dependent transmission matrix between all incoming states in Fe and the outgoing states in Au for the scattering region consisting of 12 atomic layers of bcc Fe(001) interfacing an analogous slab of fcc Au(001) using the nonequilibrium Green’s function technique implemented in the Smeagol code [22,23] at the level of LDA.
[26] At the moment we have experimental evidence for the generation of similar SC pulses at the hcp-Co/Au(111) interface.
Figures 2(d)–2(g) show the hysteresis loops measured vs \( H^T \). For \( H^L = 0 \), both \( M_{\text{col}} \) and \( M_{\text{em}} \) switch abruptly, although at different \( H^T \). For \( H^L \neq 0 \), they rotate in two 90° steps, which results in plateaus [Figs. 2(d),2(e)] corresponding to \( M_{\text{col},em} \uparrow \uparrow H^L \). At the same field \( H^T \), the MOKE ellipticity appears with its sign determined by the direction of \( H^L \) [Figs. 2(f),2(g)]. The step width determined by \( H^L \) is set such that we can realize all 16 possible combinations of in-plane \( M_{\text{col}} \) and \( M_{\text{em}} \) parallel to the easy axes. 

Because of the velocity sign reversal, contributions of incoming and reflected SC have opposite signs, cf. Eq. (1).

The current- and SC-induced contributions from the collector are neglected due to much smaller non-linearity (almost two orders of magnitude smaller mSHG intensity) and inelastic mean free path in Fe [25], as compared to Au.

This ballistic spin transport is solely defined by the interface transmittance and inelastic mean free path in Au, which can be rather long for single crystalline samples. In our earlier work [14], we have demonstrated the ballistic electron transport across 100 nm thick Au films.