Optical spin transfer in ferromagnetic semiconductors

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Circularly polarized laser pulses that excite electron-hole pairs across the band gap of (III,Mn)V ferromagnetic semiconductors can be used to manipulate and to study collective magnetization dynamics. The initial spin orientation of a photocarrier in a (III,V) semiconductors is determined by the polarization state of the laser. We show that the photocarrier spin can be irreversibly transferred to the collective magnetization, whose dynamics can consequently be flexibly controlled by suitably chosen laser pulses. As illustrations we demonstrate the feasibility of all optical ferromagnetic resonance and optical magnetization reorientation.

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I. INTRODUCTION

The collective magnetization dynamics of a single-domain ferromagnet can be dramatically modified when spin polarized quasiparticles are injected into the system. For instance, in a metallic ferromagnet current-carrying, non-equilibrium quasiparticles exert a torque on the collective magnetization which, at sufficiently high current densities, can produce a complete reversal of the magnetization orientation. This phenomenon, called spin transfer (ST), was predicted by Slonczewski[1] and Berger[2] and has been confirmed experimentally in ferromagnetic multilayer systems by a number of groups[3]. Although the microscopic mechanism is not completely settled and possibly not absolutely universal, it is clear that ST in itinerant electron ferromagnets is a consequence of irreversible transfer of magnetization between non-equilibrium quasiparticles and the collective magnetization. In ST, a spin-polarized injection current provides a non-conservative driving force which can either deliver or extract energy from the collective magnetic degree of freedom.

(III,Mn)V ferromagnetic semiconductors like GaAs:Mn combine ferromagnetism with familiar semiconductor properties similar to those of the parent semiconductor[4]. Most practical applications of GaAs and other III-V compounds are related to their optical properties. Unlike Si and Ge, III-V materials are optically active and therefore respond strongly to a laser field with a frequency close to the band gap. A well known property of III-V semiconductors is optical orientation[5] in which a laser generates a population of photocarriers strongly *spin polarized* along a direction which depends on the polarization state of the laser field. In this paper we predict that circularly polarized laser pulses which excite spin polarized electron-hole pairs across the bandgap of (III,Mn)V ferromagnets can control the magnetization dynamics through the spin transfer phenomenon. We outline a theory of optical spin transfer in ferromagnetic semiconductors and discuss some of the many possible applications of this phenomenon. In particular, by numerically solving Landau-Lifshitz equations that include a spin transfer term, we show that laser pulses with suitably chosen durations, intensities, and propagation directions enable all-optical ferromagnetic resonance, and nanosecond time scale switching between magnetic easy axes.

The rest of this paper is organized as follows. In section II we briefly review aspects of the electronic structure of (Ga,Mn)As relevant to our proposal. In section III we present our theory of optical spin transfer, which adds to the Landau Lifshitz equations (LLE) that describe collective magnetization dynamic an additional term that accounts for the irreversible exchange of angular momentum with the photocarriers. In section IV we present results of the numerical solution of the extended LLE in a number of different cases. In section V we discuss the relationship between our proposal and some recent experimental results[6,7]. We have relegated a description of some of the technical considerations that underly our theory to appendices A and B.

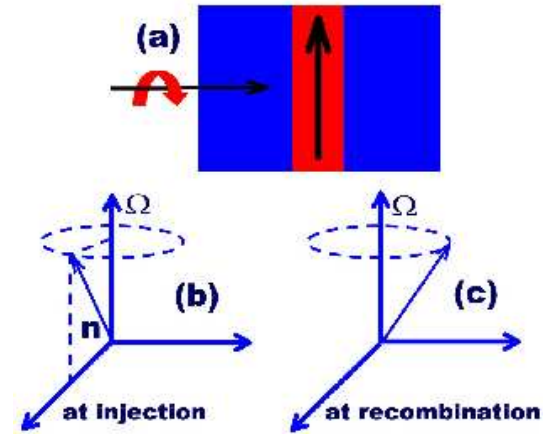

FIG. 1: Schematic summary of optical spin transfer theory

II. ELECTRONIC STRUCTURE OF (GA,MN)AS

The Fermi energy of intrinsic GaAs lies in the middle of a gap of approximately 1.5 eV. GaAs can be doped with up to 10 percent of Mn which, under suitable growth conditions, replaces Ga. Substitutional Mn acts as a relatively shallow single acceptor so that each Mn injects one hole in the GaAs host. In most of the samples there is some degree of compensation, due primarily to interstitial Mn ions, so that the hole density is smaller than density of magnetic impurities. Samples in which the density of interstitial Mn ions has been reduced by annealing tend to have higher carrier densities and metallic transport properties, indicating that the holes are delocalized. These relatively itinerant holes interact via a Kondo-like interaction with the localized d orbitals of the Mn ions. There is broad agreement that ferromagnetism is due to carrier mediated interactions between the Mn ions. Strong magneto-transport effects[8] and tunneling magnetoresistance[9] support this point of view and demonstrate that the valence band holes are strongly coupled to the Mn ions.

The physics of samples with metallic-like conductivity can be reasonably[10] described with a simple mean field theory: the valence band of the parent compound (GaAs), described with a $\vec{k}\cdot\vec{p}$ Hamiltonian, is occupied by holes which interact with an effective field that is the mean field representation of the exchange interaction with the array of Mn atoms[11,12,13]. Ferromagnetism occurs at low temperatures, when the paramagnetic energy gained by the degenerate Fermi gas formed by the holes is larger than the reduction of entropy of the magnetic atoms entailed by the spontaneous order. Importantly, the spin orbit interaction is properly included in the $\vec{k}\cdot\vec{p}$ Hamiltonian. As a result of spin orbit interaction, both the $\vec{k}\cdot\vec{p}$ valence bands $\epsilon_\nu(\vec{k})$ and the total energy $\mathcal{E}\left(\vec{\mathcal{M}}\right)$ depend on the relative orientation of the collective magnetization $\vec{\mathcal{M}}$ and the crystallographic axis. This magnetic anisotropy compares well with experimetal results[14].

The conduction band does not play an important role in equilibrium (Ga,Mn)As and remains empty unless electrons are generated there by optical excitation, as is the case of our proposal. We describe the bottom of the conduction band with a simple parabolic band model. The mean field spin splitting is given by $\Delta_c \equiv J_{sd}c_{Mn}$, where J_{sd} is the exchange coupling between the conduction band electrons and the Mn d electrons and c_{Mn} is the density of Mn ions. To the best of our knowledge, the conduction band exchange coupling constant J_{sd} has not been measured experimentally in (Ga,Mn)As. In analogy with the case of (II,Mn)VI[15], we *assume* that J_{sd} is five times smaller than J_{pd}, the exchange interaction between valence band holes and Mn ions. The latter can be inferred from transport and magneto-optics experiments. We take $J_{sd} = 11eV\ \AA^3$. (Our conclusions do not depend strongly on the numerical value of J_{sd}.)

III. OPTICAL SPIN TRANSFER

In equilibrium, the magnetization $\vec{\mathcal{M}}$ of a sample of (Ga,Mn)As with a density p of holes, lies along an easy axis in order to minimize $\mathcal{E}\left(\vec{\mathcal{M}}\right)$. In this paper we study the dynamics of $\vec{\mathcal{M}}$ when the material is photoexcited so that a density of extra holes, δp and extra conduction band electrons $\delta n = \delta p$ is injected in the system. We only consider the situation when $\delta n = \delta p << p$. The initial spin orientation of these photocarriers, $\hat{n}$, is determined by the polarization state of the laser, according to the selection rules of the material which depend ultimately on an interplay between angular momentum conservation and spin-orbit interactions. A circularly polarized laser propagating along the $\hat{z}$-direction with an energy equal to the band-gap creates photocarriers that are strongly spin polarized along the propagation direction[5].

The Mn spin dynamics of (III,Mn)V ferromagnetic semiconductors differs qualitatively from that of the paramagnetic (II,Mn)VI semiconductors which have been studied extensively[16,17] in interesting earlier work. In that case, laser pulses have been used both to trigger and to detect[16,17] the dynamics of substantially independent magnetic moments. In ferromagnets, moments behave collectively and many elegant and technologically important properties follow from the, often complex, behavior of the magnetization-orientation collective coordinate. In the case of (III,Mn)V ferromagnets[18] the underlying magnetic degrees of freedom are Mn ion $S = 5/2$ local moments and holes in the semiconductor valence band[10]. The magnetization-orientation dynamics is governed by the following equation:

$$\frac{\partial\vec{\mathcal{M}}}{\partial t} = \vec{\mathcal{M}}\times\left[-\gamma\frac{\partial\mathcal{E}(\vec{\mathcal{M}})}{\partial\vec{\mathcal{M}}} + \vec{\Gamma}_{\rm damping} + \vec{\Gamma}_{\rm ST}\right] \qquad (1)$$

where $\mathcal{E}(\vec{\mathcal{M}})$ specifies the relationship between energy and magnetization, $\vec{\mathcal{M}}$. The dissipative processes by which the collective coordinate relaxes towards the minimum of $\mathcal{E}(\vec{\mathcal{M}})$ are represented by $\vec{\Gamma}_{\rm damping}$. Choosing the Landau-Lifshitz form, the damping term is given by

$$\vec{\Gamma}_{\rm damping} = -\frac{\gamma\alpha}{\mathcal{M}_s}\vec{\mathcal{M}}\times\frac{\delta\mathcal{E}(\vec{\mathcal{M}})}{\delta\vec{\mathcal{M}}}$$

where $\mathcal{M}_s = |\vec{\mathcal{M}}|$ and $\gamma = \frac{e}{mc}$ is the gyromagnetic ratio. We define the unit vector $\vec{\Omega} \equiv \frac{\vec{\mathcal{M}}}{\mathcal{M}_s}$; note that since $d(\vec{\mathcal{M}}\cdot\vec{\mathcal{M}})/dt = 0$, these equations attempt to describe only the dynamics of the magnetization orientation $\vec{\Omega}$.

In this work the irreversible transfer of angular momentum and energy from non-equilibrium quasiparticles to the collective magnetization is described by $\vec{\Gamma}_{\rm ST}$. The expression that we use for $\vec{\Gamma}_{\rm ST}$ is based on the following

physical picture. After photon absorption, the photocarriers are spin polarized along the direction $\hat{n}$ determined by the polarization state and the propagation direction of the laser (figure 1-b). Due to their mutual exchange interaction[18], the spin of a photocarrier and the collective magnetization will then precess around each other at rates defined by their mean-field interactions. The precession involves periodic *reversible* transfer of angular momentum back and forth between the collective magnetization and an individual photocarriers, which can be described either classically or quantum mechanically. The precession is abruptly interrupted by one of the following two processes: spontaneous emission of a photon (photocarrier recombination) or photocarrier spin decoherence. As we discuss below, the latter process will usually involve spontaneous emission of a spin wave. Importantly, the distribution of values for the component of the photocarrier spin *perpendicular* to $\vec{\mathcal{M}}$, $\hat{n}_\perp$, is uniform at the instant of decay, provided that the decay time is much larger than the precession time (see figure 1-c). Therefore, when summing over all the photocarriers, there is a *net loss of quasiparticle spin* along the $\hat{n}_\perp$ direction *which must be transferred to the collective magnetization* because of the conservation of total spin during precession[1]. Therefore, the stochastic nature of the spin decoherence event, whether due to carrier recombination or spin wave emission, leads to an irreversible spin transfer from the photocarriers to the collective magnetization. This argument implies that:

$$\vec{\Gamma}_{\rm ST} = \frac{\mu_B \mathcal{P}(t)}{\mathcal{M}_s^2} \vec{n} \times \vec{\mathcal{M}} \qquad (2)$$

where $\mathcal{P}(t)$ is the rate per unit volume at which quasiparticles are injected into the system and μ_B is the Bohr magneton. In appendix A we present a mathematical derivation of eq. (2). A term similar to eq. (2) has been proposed previously on the basis of similar arguments in the context of current induced magnetization switching in ferromagnetic metals[1,2].

The argument leading to Eq. (2) assumes that the photocarriers precess many times around the effective field created by the collective magnetization before they loose spin coherence. The major source of spin decoherence in the case of the electrons in the conduction band in non magnetic p-doped GaAs at low temperatures is exchange with holes[19]. The measured[20] electron spin relaxation time in p-doped samples is well above 100 ps at low temperatures. To the best of our knowledge, the conduction band electron spin relaxation rate has not been measured in(Ga,Mn)As. Using the standard master equation approach with Fermi Golden rule rates we have derived an expression for the conduction band spin decoherence time. The details are outlined in appendix B. Whereas the static component of the magnetic environment produces the conduction band spin splitting, Δ_c, the fluctuating magnetic environment provided by the spin waves of the Mn-hole system results in the following T_2:

$$\frac{1}{T_2} = \left(1 + e^{-\beta_L \Delta_c}\right) S \, \frac{J_{sd}^2 (k_B T_e)^2}{4\pi^2 \hbar} \left[\frac{2m}{\pi \hbar^2 \mathcal{D}}\right]^{3/2} \mathcal{F}(y) \quad (3)$$

where

$$\mathcal{F}(y) = \int_0^\infty \sqrt{x} e^{-x} \int_0^{z_D} \sqrt{x + y - \frac{T_e}{T_L} z} \times \\ \times \sqrt{\frac{T_e}{T_L} z} \, \left[1 + n_B\left(\frac{T_e}{T_L} z\right)\right] dz dx \qquad (4)$$

In this expression we distinguish between the Mn temperature T_L and the temperature of the photocarriers, T_e, which is taken as a parameter to describe their excess energy. Here $\mathcal{D} \equiv \frac{2A}{c_{Mn} S}$ is the spin stiffness[21] in the spin wave spectrum $\Omega = \mathcal{D} q^2$, m^* is the conduction band effective mass, $z_D = \beta k_B^2 \left(6\pi^2 c_{Mn}\right)^{2/3}$ is the normalized spin wave Debye cutoff[22] and $y = \Delta_c / T_e$. Since and $y >> z$ the argument of the square root in eq. (4) is always positive. The physical mechanisms underlying equation (3) are the spontaneous emission and absorption of spin waves with the corresponding spin flip of the conduction band electron. Since the spin wave gap is neglected in the derivation of equation (3), T_2 is probably somewhat underestimated. T_2 is a decreasing function of the hot carrier temperature T_e. If we take $S = 2.5$, c_{Mn} =1.1×10^{21} cm^{-3} (x=0.05), $A = 0.2pJ\ m^{-1}$ (according to reference 21), $m^* = 0.067$,$T_L = 1meV$ and $J_{\rm sd}$ = 11 eV Å^3, then we have $T_2 \simeq 20$ ps for $T_e = 100$ meV, and even longer T_2 for smaller values of T_e. This time is long compared to the precession period for photoelectrons which is $\sim h/J_{sd} S c_{\rm Mn} \sim 0.15$ ps, where $c_{\rm Mn}$ is the Mn concentration, and J_{sd} is the exchange interaction between Mn moments and conduction band electrons. The photocarrier spin-orientation randomization assumption that underlies Eq. (2) is therefore valid for electrons in the conduction band. Radiative recombination time, by which one photo-electron and one photo-hole annihilate by emission of a photon, lies in the range between 2 and 25 ps[6,7]. We expect that both carrier recombination and spin wave emission will contribute significantly to the spin decay of photoelectrons.

Unlike photoelectrons, photoholes experience a strong momentum-dependent effective field $\frac{1}{3}\Delta_{so}\vec{L}$, due to their spin-orbit interaction with the atomic orbital angular momentum, $\vec{L}$. In GaAs, for example, $\Delta_{\rm so}$ = 340 meV, larger than the valence band exchange mean field, $\Delta_{\rm pd} = S J_{pd} c_{\rm Mn}$. Scattering between Bloch states causes $\vec{L}$ to fluctuate strongly so that the valence band spin decoherence time is on the same order as the momentum scattering time[23]. In (Ga,Mn)As the momentum scattering time is of the order of 10 femtoseconds[24], shorter than or comparable to the band quasiparticle spin-precession time $\sim h/\Delta_{\rm pd} \sim 30$ fs. These numerical values in combination with our picture of optical spin transfer suggest that the contribution of photo-holes will be strongly suppressed. We neglect this contribution in the rest of the

paper. The effect of spin orbit interaction in spin transfer has been studied theoretically by two of us[28], for a model without orbital degeneracy. We believe that this issue calls for further work, especially in the light of recent experiments in which current driven magnetization switching is achieved at a much smaller than expected current density[29].

IV. OPTICAL SPIN TRANSFER DYNAMICS IN (GA,MN)AS

In the remainder of this paper we explore the collective magnetization dynamics of (Ga,Mn)As driven by polarized laser pulses, as described by Landau Lifshitz equations that include the spin transfer term (eq. 1). In the model of valance-band-hole mediated ferromagnetism[11,12], the magnetic anisotropy is due to the spin orbit interaction of the holes and is sensitive to lattice mismatch strains. The anisotropy energies we consider can be fit to the form

$$\mathcal{E}(\vec{\Omega}) = k_1\Omega_x^2\Omega_y^2 + k_2\Omega_z^2 - k_3\Omega_z^4 + k_4\left(\Omega_x\Omega_y\Omega_z\right)^2 + k_u\Omega_x^2$$

The uniaxial term proportional to k_2 is a consequence of lattice-matching strains and favors magnetization orientations in the $\hat{x}-\hat{y}$ plane, while the much weaker uniaxial term proportional to k_u is obtained by fitting to recent experiments[14] which demonstrate that the kinematics of the MBE growth process lowers the symmetry of the layer-by-layer growth planes in a way which influences the magnetic anisotropy energy. The following set of values $k_1 = 0.025$ meV/nm^3, $(k_2, k_3, k_4, k_u) = 1.34, 1.05, -1.25, 0.08)$ in units of k_1 follow from the numerical mean field calculation[12], for $(Ga_{0.95},Mn_{0.05})As$ with a density of holes $p = 3.5\ 10^{20}$ cm^{-3}. These values are in good agreement with the experimental results reported by Tang *et al.*[14] and can be taken as typical for GaAsMn. We have verified that the results reported below are robust with respect to modifications in the functional $\mathcal{E}(\vec{\Omega})$.

Writing $\mathcal{E}(\vec{\Omega}) = k_1 e_0(\vec{\Omega})$, where e_0 is dimensionless, we define a typical time scale $t_0 \equiv \left|\frac{\mathcal{M}_s}{\gamma k_1}\right| \sim 30\ ps$ for a Mn fraction x=0.05. Defining the dimensionless quantities $\hat{t} = \frac{t}{t_0}$ and $\hat{\mathcal{P}} \equiv \frac{\mathcal{P}(\hat{t})t_0}{Sc_{\rm Mn}}$, the Landau Lifshitz equation can then be written as

$$\frac{d\vec{\Omega}}{d\hat{t}} = \vec{\Omega}\times\left[-\frac{\partial e_0}{\partial\vec{\Omega}} - \alpha\left[\vec{\Omega}\times\frac{\partial e_0}{\partial\vec{\Omega}}\right] + \hat{\mathcal{P}}(\hat{t})\left[\hat{n}\times\vec{\Omega}\right]\right] \quad (5)$$

where the damping coefficient α, estimated using the ferromagnetic resonance experiments of reference[25], is $\alpha \simeq 0.07$. We use this value for the calculations that follow, although our conclusions are not particularly sensitive to this parameter. We assume that the magnetization lies along the $\vec{\Omega}_0 = \hat{x}$ easy axis before the laser pulse is applied and consider two different situations: *i)* Weak circularly polarized laser pulses which propagate along $\hat{z}$ and initiate a free induction decay from which a ferromagnetic resonance spectrum can be obtained without the need for a time dependent magnetic field and *ii)* Intense circularly polarized laser pulses, propagating either parallel or perpendicular to $\vec{\Omega}_0$ which drive $\vec{\Omega}$ away from $\hat{x}$ and switch the magnetization direction.

A. All optical Ferromagnetic Resonance

We first discuss the possibility of performing a time resolved pump and probe experiment which yields information similar to that of a ferromagnetic resonance (FMR) experiment. We refer to this as all optical FMR. In standard FMR experiments, the dynamics of the magnetization is triggered by an AC magnetic field, whereas detection of the magnetization dynamics can be done using different methods, including optical ones. Here, we show that the motion of the collective magnetization can be both triggered and detected with pump and probe laser pulses. The pump laser, propagating in the $\hat{z}$ direction, is circularly polarized, so that the photocarriers are spin polarized perpendicular to the equilibrium magnetization ($\hat{x}$). Because of the spin transfer term, the magnetization can be tilted away from $\hat{x}$ (see inset of figure 2) by the pulse. The energy per pulse considered in figure 2 is $E = 0.1mJ\ cm^{-2}$ and the laser duration is $3ps$, corresponding to a laser power of $W = 33MW\ cm^{-2}$. Very similar results are obtained for pump pulse widths between 0.2 and 20 picoseconds, keeping the laser power constant: it is primarily the laser power that controls the the spin transfer effect on the magnetization, as implied by equation (2).

The subsequent free induction decay process can be probed by measuring the Faraday rotation of a second linearly polarized probe laser pulse. The Faraday rotation is proportional to $M_z(t)$, which[17] oscillates at the FMR frequency. In Fig. 2-b we show a normalized Fourier transform of $M_z(t)$ for two different values of the damping constant, α. It is apparent from Figure 2-b, both the precession frequency, ω_0 and the linewidth $\Delta\omega$, can obtained from such a procedure.

Although the experimental procedure we propose is similar to the one used in reference[17] for diluted paramagnetic II-VI-Mn, the spin transfer mechanism is quite different. In the case of II-VI-Mn quantum wells, the spin of the photoinduced heavy holes points along the growth direction, creating a transient effective field $J_{pd}\ p_{\rm holes}\ \hat{z}$ which tilts the Mn spins away from their equilibrium orientation[17,27], determined by an external magnetic field along $\hat{x}$. After recombination the transient field is absent and the Mn spins follow free induction decay dynamics.

B. Laser induced magnetization switching

In the case of all optical FMR, the magnetization orientation is weakly perturbed. By increasing the laser

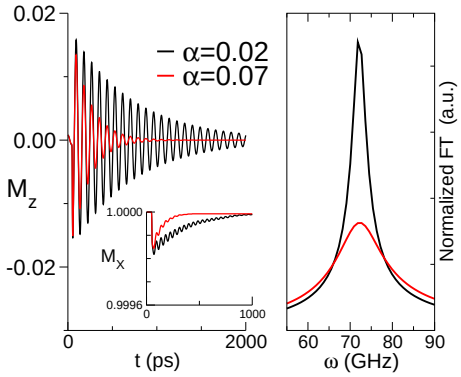

FIG. 2: All optical FMR signal. The energy per laser pulse for the illustrated simulation is $E = 0.1mJ\ cm^{-2}$ and the laser duration is $t_L = 3$ ps. The density of photocarriers corresponding those parameters and a extinction coefficient $\alpha_L = 10^4$ cm^{-1}[26] and recombination time $t_R = 2$ps[6] is 1.2×10^{18} cm^{-3}. Similar results are obtained for pump pulse widths between 0.2 and 20 picoseconds; it is primarily the laser power that determines the spin transfer effect on the magnetization, as implied by equation (2).

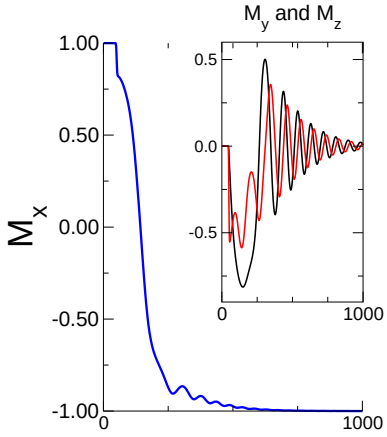

FIG. 3: Switching in the perpendicular configuration. Initially, the magnetization is pointing along $\hat{x}$. The sample is excited with a laser pulse of duration $t_L = 3$ ps and energy density $E = 4$ mJ/cm^{-2}. The density of photocarriers is $5 \times 10^{19}\ cm^{-3}$.

pulse intensity, the departure from the initial equilibrium orientation can be made large enough to drive $\vec{\Omega}$ to a *different* easy axis. We consider two geometries the spin of the photocarrier lying (a) perpendicular to $\vec{\Omega}_0$, and (b) parallel to $\vec{\Omega}_0$. These situations were also distinguished by Slonczewski[1]. For that case the geometry (a) the magnetization develops a precession around the easy axis. In the geometry (b) the effect of the spin transfer is to enhance or reduce the damping of the departures of $\vec{\Omega}$ from $\vec{\Omega}_0$. For a sufficiently high flux of non-equilibrium quasiparticles, the rate at which non-equilibrium quasiparticles deliver energy into the collective magnetization can overcome the rate at which the latter dissipates energy. When this happens, $\vec{\Omega}$ departs from $\vec{\Omega}_0$ and, depending on $\mathcal{E}(\vec{\Omega})$, it will evolve to a different easy axis orientation. We have explored the two geometries using laser pulses. A pulse that propagates perpendicular to the magnetization drives $\vec{\Omega}$ from $\hat{x}$. It is possible to tailor the laser pulse energy and duration so as to control which easy direction the magnetization decays to. In particular, the laser pulse can produce switching. In figure 3 we show an example of this. In the case of photocarriers injected with spin parallel to $\vec{\Omega}_0$, we have verified that the laser can control the rate at which the collective magnetization decays towards equilibrium. The damping can be made arbitrarily large, for lasers propagating anti-parallel to the magnetization, or arbitrarily small for lasers propagating parallel to the magnetization.

V. DISCUSSION

Ferromagnetic order of the microscopic degrees of freedom in (Ga,Mn)As, Mn d electrons and valence band holes, is described with the order parameter $\vec{\mathcal{M}}$. In equilibrium, $\vec{\mathcal{M}}$ lies along some easy axis. An external perturbation can trigger the motion of the order parameter, as described by the Landau Lifshitz equations. In this paper we describe a new type of external perturbation, the optical injection of photocarriers taht are polarized along a direction $\hat{n}$ different from the orientation of the collective magnetization$\vec{\mathcal{M}}$. Our proposal takes advantage of the selection rules for inter-band optical transitions in (II,V) semiconductors. Provided that the spin coherence time of the photocarriers is much longer than the precession time, we predict a spin transfer (c.f. 2) term in the LL equation. The spin-coherence time requirement is clearly met for conduction band electrons, while the situation for valence band holes is less clear and calls for further work. Numerical solutions of the LL equations with the spin transfer term show that the magnetization dynamics in (Ga,Mn)As can be controlled with laser pulses.

We now discuss the extent to which our theory can account for experiments recently reported by Oiwa *et al.*[7], in which the magnetization of a GaAs:Mn film excited by laser pulses of 120 fs duration and power below $5{\times}10^{12}$ photons/cm^2. For a central photon energy of 1.579 eV, the laser power per pulse is approaximately $6MW/\text{cm}^2$, comparable with the simulation of figure 2. Apart from the fact that both the experiment and our theory describe the departure of the magnetization from the equilibrium configuration induced by spin polarized photocarriers, there is a number of differences between the results of our model and those observed experimentally. The Kerr rotation signal reported by Oiwa *et al.*[7] is an exponentially decaying function with a time decay constant of less than 50 ps in contrast with our figure 2, in which M_z is an oscillating function whose amplitude decays in a time scale of 2000 ps. The Kerr signal ob-

served experimentally is overdamped, the decay constant is smaller than the precession period. In order to have an overdamped behaviour in our simulations, we would need to take a Gilbert damping coefficient of $\alpha \simeq 5$, two orders of magnitude larger than the value reported experimentally[25] At a more quantitative level, Oiwa *et al.*[7] claim that each photo-hole is able to rotate 100 Mn spins in the case GaMnAs excited with laser pulses and as much as 10^6 Mn spins in the case of cw experiments[6]. In our theory angular momentum is exchanged between photocarriers and Mn, and one photocarrier could not flip more than one Mn spin. We believe that, in order to account for these experimental results, a different physical mechanism which involves other degrees of freedom, possibly nuclear spins valence band hole angular momentum, might be needed.

In summary, we have proposed the possibility of controlling the magnetization dynamics of (III,Mn)V ferromagnetic semiconductors by means of laser pulses. Our proposal is based on an optical spin transfer effect in which angular momentum is transferred from the laser to the collective spin magnetization by optically oriented photocarriers. We have argued that the efficiency in the spin transfer is close to one in the case of the photoelectrons, but smaller in the case of photoholes because of their rapid spin decoherence. Finally, we have proposed pump and probe experiments which can achieve all optical FMR and magnetization switching on ns time scales.

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APPENDIX A: DERIVATION OF THE SPIN TRANSFER TERM

Here we derive an expression for the influence of the photo-carrier spins on the dynamics of the collective magnetization. The Mn ion local moments see the conduction electrons through their mean-field interaction with its spin-density. At each instant in time $\hat{\Omega}$ precesses around an effective field with a photocarrier contribution $J\vec{s}$:

$$\left.\frac{\mathcal{M}_s}{\mu_B}\,\frac{d\hat{\Omega}}{dt}\right|_{pc} = \left(\hat{\Omega}\times J\vec{s}\right). \tag{A1}$$

The optical spin-transfer torque is specified by the time-dependent spin density which satisfies

$$\frac{d\vec{s}}{dt} = -J\hat{\Omega}\times\vec{s} + \mathcal{P}\hat{n} - \frac{\vec{s}}{\tau} \tag{A2}$$

Here J is the exchange energy between the quasiparticle and the collective magnetization, τ is the photo-carrier spin relaxation time, $\hat{n}$ is the initial spin polarization of the photo-carrier, and $\mathcal{P}$ is the photo-carrier generation rate, as defined in section III. We look for solutions of the form:

$$\vec{s}(t) = e^{-t/\tau}\vec{v}(t) + \vec{s}_0 \tag{A3}$$

where $\vec{v}(t)$ satisfies $\dot{\vec{v}}(t) = J\hat{\Omega}\times\vec{v}$. It follows that

$$\vec{s}_0 + \xi\hat{\Omega}\times\vec{s}_0 = \mathcal{P}\hat{n} \tag{A4}$$

where $\xi \equiv J\tau$ is the dimensionless (taking $\hbar = 1$) ratio between the photo-carrier decay time and precession period.Equation (A4) can be written as a matrix equation

$$A\vec{s}_0 = \tau\mathcal{P}\hat{n}$$

where $A_{ij} = \delta_{ij} + \xi\epsilon_{ijk}\Omega_k$. $\vec{s}_0 = A^{-1}\tau\mathcal{P}\hat{n}$ where

$$A^{-1}_{ij} = \frac{1}{1+\xi^2}\left(\delta_{ij} + \xi^2\Omega_i\Omega_j - \xi\epsilon_{ijk}\Omega_k\right). \tag{A5}$$

Since $\xi >> 1$ for conduction band photo-carriers we can drop $O(\frac{1}{\xi^2})$ terms to obtain:

$$\vec{s}_0 = \tau\mathcal{P}\left(\hat{\Omega}\cdot\hat{n}\right)\hat{\Omega} + \frac{\mathcal{P}}{J}.\hat{n}\times\hat{\Omega} \tag{A6}$$

Because of the high precession rate of the photo-carriers, the precessing contribution proportional to $\vec{v}(t)e^{-t/\tau}$ is unimportant even for t shorter than τ. Therefore, combining equations (A1,A3,A4,A6) we find that the contribution of the photocarriers to the collective magnetization dynamics is

$$\left.\frac{\mathcal{M}_s}{\mu_B}\,\frac{d\hat{\Omega}}{dt}\right|_{pc} = \mathcal{P}\hat{\Omega}\times\left(\hat{n}\times\hat{\Omega}\right). \tag{A7}$$

which leads to eq. (2). This contribution corresponds to the exchange field from the portion of the injected spin-density that is perpendicular to the magnetization. Note that in equation A4 we might have distinquished longitudinal and transverse spin-relaxation rates; this distinction would not have mattered in the end, essentially because only the transverse component produces a spin torque. This analysis leads to the same conclusion as the qualitative discussion in the main text, which appears superficially to follow a different line. The difference is simply one of bookkeeping. In this appendix we consider the photocarrier spin density at a given point in space and time, instead of following the history of a given photocarrier from the moment of generation to spin-decay.

APPENDIX B: CALCULATION OF THE CONDUCTION BAND SPIN DECOHERENCE TIME

In this appendix we outline the calculation of the conduction band spin decoherence time T_2, which is done in the framework of 'system plus reservoir' master of equation (ME) approach[30]. The system is the conduction

band electron spin and the reservoir is formed by the spin waves of the Mn-hole system the orbital degrees of freedom of the conduction band electron. The Hamiltonian reads:

$$\begin{aligned}\mathcal{H}_{\rm cond} &= \mathcal{H}_{0s}+\mathcal{H}_{0r}+\mathcal{V}_{sr},\\ \mathcal{H}_{0s} &= \frac{\Delta_c}{2}\vec{\sigma}\cdot\vec{\mathcal{M}}_0\\ \mathcal{H}_{0r} &= \frac{p^2}{2m}+\sum_q \omega(q)b_q^\dagger b_q^\dagger\\ \mathcal{V}_{sr} &= \frac{J_{sd}}{2}\sum_I \delta\left(\vec{r}-\vec{R}_I\right)\vec{\sigma}\cdot\delta\vec{M}(\vec{R}_I)\end{aligned} \quad \text{(B1)}$$

where $\Delta_c = J_{sd}c_M$. The spin fluctuations $\delta\vec{M}(\vec{R}_I)$ and spin wave operators are related by[22]:

$$\begin{aligned}\mathcal{M}^z(\vec{R}) &\equiv S - b^\dagger(\vec{R})b(\vec{R})\\ \delta\mathcal{M}^{(+)}(\vec{R}) &= \mathcal{M}^{(+)}(\vec{R}) \simeq \sqrt{2S}b(\vec{R})\\ \delta\mathcal{M}^{(-)}(\vec{R}) &= \mathcal{M}^{(-)}(\vec{R}) \simeq \sqrt{2S}b^\dagger(\vec{R})\end{aligned} \quad \text{(B2)}$$

and

$$\begin{aligned}b(\vec{R}) &\equiv \frac{1}{\sqrt{\mathcal{N}}}\sum_{\vec{q}} e^{i\vec{q}\cdot\vec{R}}b_{\vec{q}}\\ b^\dagger(\vec{R}) &\equiv \frac{1}{\sqrt{\mathcal{N}}}\sum_{\vec{q}} e^{i\vec{q}\cdot\vec{R}}b^\dagger_{-\vec{q}}\end{aligned} \quad \text{(B3)}$$

The $\mathcal{H}_{0s}$ term is the interaction of the spin with the average magnetization which, in the master equation approach, is the 'system' Hamiltonian. $\mathcal{H}_{0r}$ accounts for the electron kinetic energy and free spin waves. They are the 'reservoir' Hamiltonian. The last term, the coupling between the system and the reservoir variables, comes from the exchange coupling of the conduction electron with the spin waves (spin fluctuations).

Using second order perturbation theory around $\mathcal{H}_{0s}+\mathcal{H}_{0r}$ it is possible to derive[30] a closed set of ME for the reduced density matrix of the conduction band spin, including the coupling with the reservoir degrees of freedom to second order in the exchange coupling. In this language, the above Hamiltonian describes elementary processes in which a spin wave is absorbed or emitted, and spin and momentum are exchanged between the spin wave and the photo-carrier. Let us denote by

$$\Gamma_{\sigma_i k_i sw_i,\sigma_f,k_f,sw_f} = \frac{2\pi}{\hbar}|\mathcal{V}_{if}|^2\delta(E_i-E_f) \quad \text{(B4)}$$

the Fermi Golden rule (FGR) transition rate for the process in which the photo carrier spin goes from σ_i to σ_f, the photocarrier momentum goes from k_i to k_f by emission or absorption of a spin wave. We now define

$$\Gamma_{\sigma_i,\sigma_f} \equiv \sum_{k_i,sw_i} P(k_i)P(sw_i)\sum_{kf,sw_f}\Gamma_{\sigma_i k_i sw_i,\sigma_f,k_f,sw_f} \quad \text{(B5)}$$

where $P(k_i)$ and $P(sw_i)$ are the equilibrium distribution functions for the inital photocarrier momentum and spin wave occupation respectively. Eq. (B5) involves both an average over initial and sum over final reservoir states of the Fermi Golden rule transition rates. It can be seen[30] that the spin decoherence time is

$$\frac{1}{T_2} = \Gamma_{\uparrow,\downarrow}+\Gamma_{\downarrow,\uparrow} \quad \text{(B6)}$$

since the so called non adiabatic contribution to T_2 is zero for Hamiltonian B1). After some work, the spin wave emission rate reads:

$$\Gamma_{\uparrow,\downarrow} = \frac{V}{Z}\frac{J_{sd}^2}{\hbar}2S\pi\int_{\mathcal{V}_D}\frac{d\vec{q}}{(2\pi)^3}\int\frac{d\vec{k}}{(2\pi)^3}$$
$$\int\frac{d\vec{k'}}{(2\pi)^3}e^{-\beta\epsilon_k}\left[1+n_B(\Omega(q))\right]\delta\left[\epsilon_k-\epsilon_{k'}+\Delta-\Omega\right] \quad \text{(B7)}$$

and a similar expression can be derived for the spin wave absorption rate. Here V is the volume of the sample, $\mathcal{V}_D$ is the Debye sphere and $Z = \frac{V\sqrt{\pi}}{8\pi^2}\left[\frac{2mk_bT}{\hbar^2}\right]^{(3/2)}$. Equation (3) is obtained from eq. (B7) after some extra changes of variables.

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