A Solid State Paramagnetic Maser Device Driven by Electron Spin Injection

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In response to an external, microwave-frequency magnetic field, a paramagnetic medium will absorb energy from the field that drives the magnetization dynamics. Here we describe a new process by which an external spin-injection source, when combined with the microwave field spin pumping, can drive the paramagnetic medium from one that absorbs microwave energy to one that emits microwave energy. We derive a simple condition for the crossover from absorptive to emissive behavior. Based on this process, we propose a solid-state, paramagnetic device in which microwave amplification by stimulated emission of radiation is driven by spin injection.

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In spin electronics ("spintronics"), the spin degree of freedom of the electron is used as a new means of storing, manipulating, and transferring signals and information [1]. Since the prediction that a spin-polarized current can excite the (stimulated) emission of spin waves at a ferromagnetic interface [2], it has been demonstrated in magnetic multi-layer systems that spin-polarized currents can drive microwave-frequency magnetization dynamics in the ferromagnetic layers [3–5]. More recently, it has been proposed that the magnetization dynamics of a ferromagnet can be used as a "spin pump" to drive a spin current into an adjacent, nonmagnetic metallic layer [6]. In all of the above, the dynamics of the magnetization in the ferromagnetic layer is of primary importance.

We have recently shown, however, that a ferromagnet is not required for spin pumping, since applied radiofrequency (rf) magnetic fields can produce spin accumulation in paramagnetic materials [7]. In this case, the paramagnetic medium absorbs energy from the external microwave magnetic field to produce the nonequilibrium spin accumulation. In this Letter we describe a new effect in which the energy flow can be reversed: by combining an external spin-injection source with microwave field spin pumping in a paramagnetic medium, the system can be driven from one that absorbs microwave energy to one that emits microwave energy. When the medium is placed within a resonant circuit, it is possible to obtain microwave amplification by stimulated emission of radiation (maser) action driven by spin injection.

The operation of a maser device requires the generation of population inversion. In a two-level paramagnetic maser system, both excitation and stimulated emission occur from the same pair of levels [8]. Population inversion can be produced by separating the processes of excitation and emission in time, for instance by pulsed excitation. In multilevel systems the levels used for excitation and emission can be separated, and continuous operation is then possible [8]. For example, a proton nuclear spin maser was realized using the coupling between the nuclear and electron spins, and exciting the latter with microwaves [9], or by optically pumping nuclear transitions in ³He [10]. The

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continuing progress in the field of spintronics in both technological advances as well as the understanding of fundamental processes, offers a new possibility in which a spin current injected into the paramagnetic medium can act as a continuous source of population inversion. As we will show, this allows for a straightforward description of maser operation with semiclassical Bloch-type equations written in terms of spin accumulation. The proposed spin injection-driven maser, at least in its present form, is not a practical design for use in applications, but we show that it is feasible to fabricate and study one with the current level of technology, as demonstrated in, for instance, spin current switching experiments [11] and microwave pulsed field-induced ferromagnetic switching [12].

The principle behind the device is illustrated in Fig. 1. In Fig. 1(a), part of the rf power incident on a paramagnetic medium is absorbed in the process of creating spin accumulation in the system. Power is lost to internal dissipation due to spin relaxation and to an external "spin sink." In a real device this could be a ferromagnetic electrode used to probe the spin accumulation in the paramagnetic region. However, the role of the spin sink can be reversed by driving a spin-polarized current into the paramagnet from an externally powered spin source, as shown in Fig. 1(b).



FIG. 1. Power flow in and out of a paramagnetic medium to illustrate the principle behind the spin-injection maser. (a) Part of the input rf power is absorbed to produce precessing spin accumulation in the medium; power is lost to internal dissipation due to spin relaxation processes, and to a spin sink. (b) When additional spins are provided by an externally powered spin-injection source, it is possible for the system to exhibit gain, resulting in net emission of rf power.

As we will demonstrate below, this allows the medium to exhibit rf power gain. Figure 2 shows schematically a bilayer structure for injecting spins electrically: a ferromagnetic layer is deposited on top of the paramagnetic layer (separated by a thin aluminum oxide tunnel barrier), and the layers are connected to opposite terminals of a voltage source so that an electrical current is driven across the interface between the layers. This will cause a spin current I_s to flow into the paramagnetic layer, whose thickness is assumed to be much less than its spin relaxation length so that a uniform spin accumulation μ_s will build up in the layer. The role of the ferromagnet in Fig. 2 is purely as a source of spin current. The dynamics of the magnetization or spin waves in the ferromagnet are not relevant to our description. We will first show how the injected spin current modifies the solutions for the spin-pumped spin accumulation in the medium, and then analyze the response of the system within an electrical circuit model.

In a previous publication [7], we have described the dynamics of the spin accumulation $\vec{\mu} = (\mu_x, \mu_y, \mu_z)$ in response to a time-dependent magnetic field $\vec{B}(t)$. We now extend this description to include interaction with an external spin source:

$$\frac{d}{dt}\vec{\mu}(t) = -\hbar \frac{d}{dt}\vec{\omega}_B(t) - \frac{\vec{\mu}(t)}{\tau} + \vec{\omega}_B(t) \times \vec{\mu}(t) + \vec{I}_s(t), \quad (1)$$

where $I_s(t)$ is the injected spin current (in units of power, corresponding to a rate of injected spin accumulation), $\vec{\omega}_B$ is the Larmor frequency $\hbar \vec{\omega}_B = g \mu_B \vec{B} \ (g \simeq 2$ is the electron g factor), and τ is the spin relaxation time. Equation (1) describes the time evolution of the spin accumulation in response to four physical processes (the four terms on the right-hand side of the equation): spin pumping, spin relaxation, spin precession, and spin injection. The spin pumping term describes the rate at which the time-dependent magnetic field, via the Zeeman energy, pushes spins aligned with the magnetic field below the Fermi level, and antialigned spins above the Fermi level,



FIG. 2. Schematic of an electrical spin-injection source for the paramagnetic medium. An electrical current is driven across the interface between a ferromagnetic layer and the paramagnetic layer (separated by an oxide tunnel barrier), which causes a spin current I_s to flow into the paramagnet. It is assumed that the static field B_z is sufficiently strong to saturate the magnetization M of the ferromagnetic layer normal to the film plane.

behaving essentially as a source of locally injected, timedependent spin currents [7]. In a uniform, paramagnetic system, we previously found that for a magnetic field rotating with angular frequency ω in the x-y plane, a constant spin accumulation is generated in the z direction. Equation (1) [with $\vec{I}_s(t) = 0$] may be connected to electron spin resonance theory by observing that the timedependent magnetization is given by $\vec{m}(t) =$ $\frac{1}{4}gN_F\mu_B[\vec{\mu}(t) + \hbar\vec{\omega}_B(t)]$, where N_F is the total electron density of states at the Fermi level. The first term $\vec{\mu}(t)$ in the expression for $\vec{m}(t)$ describes the nonequilibrium magnetization, and the second term describes the equilibrium magnetization due to the field-induced paramagnetism [9,13].

The system in Fig. 2 is now spin pumped by applying a magnetic field B_{xy} that rotates in the *x*-*y* plane at microwave-frequency ω , and a static field B_z is applied in the *z* direction; ω_{xy} and ω_z are the Larmor frequencies associated with these fields. A constant spin current $\vec{I}_s(t) = I_s \hat{z} = \mu_s \tau^{-1} \hat{z}$ is injected into the system, where μ_s is the spin accumulation that would be present in the paramagnet in the absence of the applied field, and is collinear with B_z . The sign of μ_s depends on the orientation of the magnetization (usually fixed by B_z) and the direction of charge current flow across the interface. The spin accumulation is found by solving Eq. (1) after transforming to a rotating reference frame in which the in-plane magnetic field is static [7]. The solutions are

$$\mu_{\parallel} = \frac{(\omega_z - \omega)\tau(\omega_{xy}\tau)}{1 + (\omega_{xy}\tau)^2 + (\omega_z - \omega)^2\tau^2} (\hbar\omega + \mu_s), \quad (2)$$

$$\mu_{\perp} = -\frac{(\omega_{xy}\tau)}{1 + (\omega_{xy}\tau)^2 + (\omega_z - \omega)^2\tau^2}(\hbar\omega + \mu_s), \quad (3)$$

$$\mu_{z} = \mu_{s} - \frac{(\omega_{xy}\tau)^{2}}{1 + (\omega_{xy}\tau)^{2} + (\omega_{z} - \omega)^{2}\tau^{2}}(\hbar\omega + \mu_{s}), \quad (4)$$



FIG. 3. A vector diagram in the rotating reference frame showing the field (white arrow) and the in-plane spin accumulation (black arrow), for (a) $\mu_s = 0$, (b) $\mu_s = -\hbar\omega$, and (c) $\mu_s = -2\hbar\omega$. The μ_{\parallel} axis is the spin accumulation in phase with the field, while the μ_{\perp} axis is the spin accumulation out of phase with the field. The half-plane $\mu_{\perp} < 0$ represents in-plane μ that lags the field (absorption), the half-plane $\mu_{\perp} > 0$ represents in-plane μ that leads the field (emission).

where μ_{\parallel} and μ_{\perp} refer to the components of the spin accumulation that rotate in phase (dispersive component) and 90° out of phase (absorptive or emissive component) with the field B_{xy} , respectively. In our previous work we focused on μ_z , which can be measured electrically in a dc transport experiment by using ferromagnetic detection electrodes. Here, with the addition of the injected spin current, we instead focus on μ_{\parallel} and μ_{\perp} . In Fig. 3 we illustrate three cases for specific values of μ_s . When $\mu_s =$ 0, μ_{\perp} is always negative. This situation is shown in Fig. 3(a), where the black vector is the in-plane spin accumulation in the rotating reference frame. The vector will always lie in the half-plane $\mu_{\perp} < 0$, which represents energy absorption from the field. When $\mu_s = -\hbar\omega$, we have that $\mu_{\parallel} = \mu_{\perp} = 0$ and $\mu_z = \mu_s$, thus giving the remarkable result that the spin pumping effect can be turned off with an injected spin current. This is shown in Fig. 3(b), where the in-plane spin accumulation vector has disappeared. When $\mu_s < -\hbar\omega$, both μ_{\parallel} and μ_{\perp} change sign as shown in Fig. 3(c). The spin accumulation vector now lies in the half-plane $\mu_{\perp} > 0$, which represents energy emission to the field. This corresponds to stimulated emission resulting in the medium exhibiting gain.

In other words: the injected spins, originally oriented along the z direction, will precess around the field B_{xy} . Depending on their initial polarization in the z direction, they will give either a positive or negative contribution to μ_{\perp} , and depending on the sign of ω , they will either enhance the damping or generate gain.

Using Eqs. (2)-(4) we find the power per unit volume in the paramagnet dissipated due to spin relaxation:

$$P_{\rm diss} = \frac{N_F}{4\tau} |\vec{\mu}|^2 = \frac{1}{4\tau} N_F [\mu_s^2 - \omega_{xy} \tau (\hbar \omega - \mu_s) \mu_{\perp}].$$
(5)

The net power extracted from or supplied to the field is determined by the time-averaged value of $\vec{m} \cdot \frac{d}{dt} \vec{B}$ [14]:

$$P_{\text{field}} = -\frac{1}{4\tau} N_F \omega_{xy} \tau \mu_{\perp} \hbar \omega.$$
 (6)

This power can be positive or negative, as discussed above. The power supplied from the external spin source is

$$P_{\text{ext}} = \frac{1}{4} I_s \mu_z N_F = \frac{1}{4\tau} N_F (\mu_s^2 + \omega_{xy} \tau \mu_s \mu_\perp) \qquad (7)$$

 $P_{\text{ext}} + P_{\text{field}} = P_{\text{diss}}$, as required by conservation of energy [15].

While it is possible to generate locally rotating rf magnetic fields in a device, it is generally more practical to use a linear rf driving field in resonance with ω_z , similar to conventional electron spin resonance experiments. A linear field can be decomposed into two counterrotating magnetic fields of half the magnitude. For positive ω_z , only the field rotating counterclockwise has the right sense for the resonant condition $\omega = \omega_z$ to hold, and thus to contribute significantly to the response. We have checked this explicitly for the relevant range of parameters with exact, numerical solutions of Eq. (1). To simplify the expressions that follow, we will therefore assume the resonant condition with the convention of positive ω , and further assume that $(\omega_{xy}\tau)^2 \ll 1$, which is typically the case experimentally. We then have $\mu_{\parallel} \simeq 0$ and $\mu_{\perp} \simeq -\omega_{xy}\tau(\hbar\omega + \mu_s)$, and the in-plane magnetization components m_{\parallel} and m_{\perp} are now both linear in ω_{xy} . We define the complex magnetic susceptibility $\chi = \chi_{\parallel} + i\chi_{\perp}$ for $\mu_0(m_{\parallel} + im_{\perp}) = \chi B_{xy}$, and find the components to be

$$\chi_{\parallel} = \frac{1}{4} g N_F \mu_B^2 \mu_0 \equiv \chi_0, \tag{8}$$

$$\chi_{\perp} = -\chi_0 \frac{\tau}{\hbar} (\hbar \omega + \mu_s), \qquad (9)$$

where χ_0 is a constant determined by the material parameters g and N_F .

In order to analyze the performance of our system within a device, we model the system as the resistor-inductorcapacitor electrical network (RLC) circuit shown in Fig. 4. We follow a standard treatment for spin resonance theory [9,16]. The inductor coil is used to both excite and detect the magnetization induced in the medium. The inductance is modified by χ as $L = L_0(1 + \eta \chi)$, where η is the filling factor of the medium in the coil [17]. The total impedance of the circuit is then given by

$$Z = R_0 - \omega L_0 \eta \chi_{\perp} + i \omega L_0 (1 + \eta \chi_0) + \frac{1}{i \omega C_0}.$$
 (10)

The effect of the magnetic susceptibility on the inductance thus appears as an additional resistance and inductance, indicated as R' and L' in the equivalent circuit of Fig. 4(b). The resistance $R' = -\omega L_0 \eta \chi_{\perp}$ is positive in the absence of spin injection ($\mu_s = 0$), resulting in increased damping in the circuit. However, with $\mu_s < -\hbar\omega$, R' becomes negative and reduces the damping, which can be measured as a change in the transmitted power in a suitably designed waveguide structure [18]. Self-sustaining, maser oscillations [19] are obtained when the magnitude of the negative



FIG. 4. (a) Resonant RLC circuit for the spin-injection maser connected to a rf voltage source. The paramagnetic medium is enclosed within the inductor coil (the bare coil has inductance L_0). The modification of the inductance by the complex magnetic susceptibility of the paramagnet results in the equivalent circuit (b), with an additional resistance R' and inductance L'.

R' is larger than R_0 , leading to negative effective resistance in the circuit. We can write the condition $R_0 + R' < 0$ in terms of the quality factor $Q = \frac{\omega L_0}{R_0}$ of the unloaded circuit:

$$Q > \left(-\eta \chi_0 \frac{\tau}{\hbar} (\hbar \omega + \mu_s)\right)^{-1}.$$
 (11)

Using typical parameters for Al with τ of order 10^{-10} s at room temperature, the product $\eta \chi_0 \frac{\tau}{h} \simeq 1/\text{eV}$. When $|\mu_s| \gg \hbar \omega$, then Eq. (1) reduces to $Q > 1/|\mu_s(eV)|$. To find a value for Q we then only need to estimate μ_s . Spin relaxation in a confined medium can be described as an effective resistance between parallel spin up and spin down channels. For injection into an Al metal island (500 \times $500 \times 50 \text{ nm}^3$), a spin resistance of 10 Ω was found experimentally [20]. Scaling to a larger medium $1 \times 1 \times$ $0.2 \ \mu m^3$, the spin resistance is reduced to about 1 Ω . Assuming a ferromagnet polarization of 50% and a tunnel barrier resistance of 1 k Ω (about the lowest value that can be obtained technologically) that limits the charge current to 10 mA, a spin accumulation voltage μ_s of order 5 meV can be achieved (this is indeed much greater than $\hbar\omega$ for microwave frequencies up to tens of GHz). This gives a minimum Q of 200 [21]. Although this is relatively large, it should be possible to fabricate such an on-chip resonator, especially in a proof-of-principle experiment in which superconducting strip lines are used to construct the resonant circuit [22].

To estimate the typical microwave power which can be emitted by this device, we assume that the amplitude of the oscillating B_{xy} is limited by the condition $\omega_{xy}\tau \leq 1$, at which point the stimulated emission becomes less efficient [Eqs. (3) and (6)]. From Eq. (6) we then find that a typical power is 1 μ W (note, however, that this scales with the device dimensions). The total power supplied by the voltage source is of order 100 mW. The main reason for this low efficiency is the mismatch between the 1 k Ω resistance of the tunnel barrier and the typical 1 Ω spin resistance of the system. This is purely a technological problem related to the limitations of using aluminum oxide tunnel barriers.

In conclusion, we have shown that by combining an external spin injection source with microwave field spin pumping on a paramagnetic medium, it can be made to exhibit microwave energy gain for sufficiently large injected spin currents. This effect does not depend on the details of the paramagnet or the spin-injection source. While we have used a simple metallic multilayer system to illustrate our proposal for a spin-injection maser, our description can be readily applied to other systems, such as semiconductors, and other sources of spin injection, such as optical pumping.

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