Giant lasing effect in magnetic nanoconductors

A. Kadigrobov\textsuperscript{1,2}, Z. Ivanov and T. Claeson\textsuperscript{1}

\textsuperscript{1}Department of Microelectronics and Nanoscience, Chalmers University of Technology, SE-412 96 Göteborg, Sweden

R. I. Shekhter\textsuperscript{2}, and M. Jonson\textsuperscript{2}

\textsuperscript{2}Department of Applied Physics, Chalmers University of Technology and Göteborg University, SE-412 96 Göteborg, Sweden

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We propose a new principle for a compact solid-state laser in the 1-100 THz regime. This is a frequency range where attempts to fabricate small size lasers up till now have met severe technical problems. The proposed laser is based on a new mechanism for creating spin-flip processes in ferromagnetic conductors. The mechanism is due to the interaction of light with conduction electrons; the interaction strength, being proportional to the large exchange energy, exceeds the Zeeman interaction by orders of magnitude. On the basis of this interaction, a giant lasing effect is predicted in a system where a population inversion has been created by tunneling injection of spin-polarized electrons from one ferromagnetic conductor to another — the magnetization of the two ferromagnets having different orientations. Using experimental data for ferromagnetic manganese perovskites with nearly 100\% spin polarization we show the laser frequency to be in the range 1-100 THz. The optical gain is estimated to be of order 10\(^7\) cm\(^{-1}\), which exceeds the gain of conventional semiconductor lasers by 3 or 4 orders of magnitude. A relevant experimental study is proposed and discussed.

The ability to synthesize magnetically ordered, layered conductors with nearly 100\% spin-polarization of the conducting electrons \cite{1}, has opened up a new field in solid state physics, the field of “spintronics” \cite{2,3}. Spin-dependent tunneling of electrons is one phenomenon that has already found commercial applications based on the resulting “giant” magnetoresistance of certain layered structures \cite{2,3}. Other applications are bound to follow. The possibility to control, by means of a bias voltage, not only the energy but also the spin of electrons injected into a magnetic conductor makes it feasible to investigate the properties of highly excited spin-polarized electrons. An example of such a system is presented in Fig.1, where the hatched region corresponds to an equilibrium distribution of (spin-up) electrons in a spin-polarized conductor. The dotted area marks a non-equilibrium distribution of “hot” (spin-down) electrons. Relaxation of the spin-down electrons to an equilibrium configuration requires spin-flip processes and is therefore completely blocked if such processes are not allowed. In the presence of such a “spin lock” against relaxation, highly excited states in the material may have a long lifetime, which may in turn determine novel “spintronics” effects in spin-polarized conductors. The objective of this Letter is to demonstrate how electromagnetic radiation may remove the spin-lock effect and to demonstrate some important consequences of this effect for spintronics. We will show that the radiation makes the blockade of relaxation weaker through its coupling to the exchange interaction in magnetically ordered conductors. This comes about via the dependence of the exchange constant on the momenta of the conduction electrons. As a result a lasing effect is shown to occur in systems where an inverted electron population has been created by the tunneling injection of spin-polarized electrons from one ferromagnetic conductor to another (the orientation of the magnetization being different in the two ferromagnets).

An example of such a system is presented in Fig.3. Our estimations show that a laser with an optical gain that exceeds the gain of conventional semiconductor lasers by three or four orders of magnitude can be built and argue that laser action can be achieved provided care is taken to design the system so that the lasing region is not too much heated. The frequency of these lasers can be in a wide range that includes the interval 1 - 100 THz.

\begin{equation}
\hat{H}_0 = \hat{\sigma}_0 \frac{\hat{p}^2}{2m^*} - \hat{\sigma} \mathbf{1},
\end{equation}

where \(m^*\) is the effective mass, \(\hat{p}\) is the momentum operator, \(\hat{\sigma}\) are Pauli matrices, \(\sigma_0\) is the 2 \times 2 unity matrix,

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{fig1}
\caption{Schematic representation of the band structure of a magnetically ordered conductor. The hatched region corresponds to an equilibrium distribution of spin-up electrons in the lower band, while the dotted region indicates a non-equilibrium distribution of "hot" spin-down electrons in the upper band. Arrows show electron spin directions in the bands.}
\end{figure}
and \( J \) is the exchange energy. According to Eq.(1) the dispersion law for electrons with spins pointing up/down is

\[
E_{\uparrow}(p) = \frac{p^2}{2m^*} + I \tag{2}
\]

We will deal with a system, schematically presented in Fig.3, in which two potential barriers divide the magnetic conductor into three parts; the magnetization of two adjacent magnetic conductors (regions A and B) are pointing in opposite directions. A bias voltage \( V \) is applied between regions A and C. We assume that the spin relaxation time \( \nu_0^{-1} \) [4], the time of energy relaxation without changing the electron spin direction \( t_E \) and the time of electron tunneling \( t_{tun} \) obey the inequality \( t_E \ll t_{tun} \ll \nu_0^{-1} \). In the absence of spin-flip processes, the energy relaxation of injected spin polarized electrons creates a non-equilibrium state in which equilibrium is established only within each group of electrons with a fixed spin polarization. Therefore, in region B electrons in the spin-up and spin-down energy bands are in local equilibria corresponding to the different chemical potentials \( \mu_{\uparrow} \) and \( \mu_{\downarrow} \), respectively, while the system as a whole is far from equilibrium.

According to Eq.(2) the energy conservation law for vertical transition of electrons with emission of photons of frequency \( \omega \) does not depend on the electron momentum:

\[
E_{\uparrow}(p) - E_{\downarrow}(p) - \hbar \omega = 2I - \hbar \omega \tag{3}
\]

It follows that for \( \omega = 2I/\hbar \) all "hot" electrons are in resonance with the electromagnetic field, and hence stimulated emission of light due to transitions of electrons from filled states of the upper band to the empty states of the lower band is possible for all electrons in the energy range \( \mu_{\uparrow} - \mu_{\downarrow} \). As seen in Fig.3, under the neutrality condition \( N_{\uparrow} + N_{\downarrow} = N_{\uparrow0} \) (\( N_{\uparrow} \) and \( N_{\downarrow} \) are the densities of spin-up and spin-down electrons, \( N_{\uparrow0} \) is the equilibrium electron density) the population inversion needed for lasing requires a large enough bias voltage, \( V > (2I - \mu_0)/e \).

The conventional Zeeman term \( \hat{H}_Z = g\mu_B \hat{H}_\sigma \) describing interaction between the (hot) electrons and an electromagnetic field does provide a mechanism for stimulated radiative transitions between the energy bands containing electrons with opposite spin directions. However, it is relatively small in magnitude and it is not the most important mechanism. For ferromagnets, we would like to suggest a much more effective mechanism of interaction between light and conduction electron spins. This mechanism is based on the dependence of the exchange energy (1) on the momentum \( \mathbf{p} \) of the conduction electron. The momentum dependence has to do with the overlap of the wavefunctions of the conduction electron and the magnetic sub-system (see, e.g., [5]). It is determined by the value of \( p a/\hbar \), where \( a \) is the characteristic size of the orbital (that is \( a \) is an atomic-scale length).

This is why it varies with the momentum of the conduction electron. In the absence of an electromagnetic field the Hamiltonian which describes this situation can be written as

\[
e(\hat{p}) = \sigma_0 \frac{p^2}{2m^*} - \hat{\sigma} \mathbf{I} \hat{p} \tag{4}
\]

In the presence of an electromagnetic field with vector potential \( \mathbf{A} \) the momentum operator \( \hat{p} \) in Eq.(4) must be changed to \( \hat{p} - (e/c)\mathbf{A} \). Expanding in powers of \( (e/c)\mathbf{A} \) one gets an effective Hamiltonian \( \hat{H}_{eff} = \epsilon(\hat{p}) + \hat{H}_{eff}^{(1)} \) where the perturbation Hamiltonian [6] is

\[
\hat{H}_{eff}^{(1)} = -\frac{e}{2c} \sigma \left( A_i \frac{\partial}{\partial p_i} + \frac{\partial}{\partial p_i} A_i \right)_{\mathbf{p} = \hat{p}} \tag{5}
\]

In Eq.(5) we have omitted terms that do not flip spins; summation over double indices is implied: \( a_i b_i = ab \).

![Fig. 2. Schematic illustration of how electrons are injected into the active region for the case that the adjacent ferromagnets have different magnetization directions (shown with long arrows; electron spins, \( \sigma \), are shown with short arrows with black dots at the end). An electron from the left-side ferromagnet with its spin parallel to the magnetization direction passes through a sharp boundary (shown as a vertical thick line) between the ferromagnets without changing spin direction. In the right-side ferromagnet it emerges with its spin in a superposition of the spin-up and spin-down states. The classical precession of the spin is indicated by a dotted ellipse. If the injected electrons are prepared in such a way that their spins are not parallel to the magnetization in the active region B (see Fig.3), the Hamiltonian (5) produces spin-flips and hence stimulates the needed radiative transitions of hot electrons in the upper band to the lower energy band. This process is illustrated in Fig.2, where an electron (with its spin parallel to the magnetization) is impinging on the boundary from the left, passes through the boundary and is scattered into a quantum superposition of spin-up and spin-down states in the active region to the right of the boundary. The wave function of the electron in the active region B is

\[
\Psi(\mathbf{r}) = e^{i\mathbf{p}_r \cdot \mathbf{r}} \sum_{k=1}^{2} \alpha_k \left( I_x/(I_z + (-1)^k I) \right) e^{i p_k x} \tag{6}
\]

where the \( x \)-axis is perpendicular and the \( y \)- and \( z \)-axes are parallel to the boundary; the projections of the
electron momentum and the coordinate on the boundary are \( p_\perp = (0, p_y, p_z) \) and \( r_\perp = (0, y, z) \), while \( p_{1,2} = \sqrt{2m (E + \pm I)} - p_\perp^2 \) and \( I_\pm = I_x + iI_y \); coefficients \( a_{1,2}(E) \) are found by matching the wave functions of the electron in the active region (the right-hand side of Fig. 2) and in the injecting region (left-hand side of Fig. 2) at the boundary (we do not present their explicit form here).

Using Eq.(6) as the initial proper state \( \Psi_f(r) \) belonging to the initial energy \( E_f \) and the final state \( \Psi_f(r) \) belonging to the final energy \( E_f \) one sees that matrix element \( \langle \Psi_f(r) | \hat{H}_{\text{eff}}^{(1)} | \Psi_i(r) \rangle \) (the probability amplitude for a radiative electronic transition between the unperturbed energy bands) is not zero if \( p_1(E_i) = p_2(E_f) \). From here it follows that the difference between the initial and the final energies should be \( E_i(p) - E_f(p) = 2I(p) \).

![Fig. 3. Schematic illustration of a population inversion in a magnetic conductor (region B). A bias voltage \( V \) is applied between magnetic conductors (A and C) which have opposite directions of their magnetizations (the thick vertical lines represent potential barriers). Arrows show the electron spin directions in the electron energy bands; \( \mu_0 \) is the equilibrium magnetic chemical potential (in the absence of bias). Population inversion requires the bias voltage to be \( V > (2I - \mu_0)/\epsilon \). The insertion shows a possible realization of a structure in which two magnetic conductors with opposite magnetization directions are coupled through a microbridge (B).](image)

Taking such wave-functions as the initial and the final states and performing standard calculations \([9]\) using Eq.(5) and Fermi’s golden rule one gets the stimulated transition rate per unit volume of active material as

\[
R_{st} = \frac{32\pi e^2 \mu}{n^2} \left| I \right|^2 \frac{\hbar \nu_s}{\omega} \left( \frac{b_1 N_1^2 - b_2 N_2^2}{2(\hbar \omega - I)^2 + (\hbar \nu_s)^2} \right) N_p \]

\[
\times \left\{ \left( \epsilon_d \cdot \epsilon_a \right)^2 + \left( \epsilon_d \cdot \epsilon_i \right)^2 \right\} \]

(7)

Here \( \mu \) and \( n \) are the magnetic permeability and the refractive index of the medium, respectively, \( N_p \) is the photon density, \( \omega \) is the photon frequency, the constants \( b_1 \) and \( b_2 \) are of order unity, the unit vectors \( \epsilon_a \) and \( \epsilon_i \) are directed along the magnetizations in the active (right-hand side of Fig.2) and injection (left-hand side of Fig.2) regions, respectively, while \( \epsilon_d \) is parallel to the vector \( I' \equiv \epsilon \partial I/\partial p_i \), where \( \epsilon \) is the unit polarization vector in the direction of the vector potential \( A \).

Eq.(7) was derived under the assumption that \( |I'|p_F \nu_s < h \nu_s \) \((p_F \nu_s \) is the Fermi momentum of electrons in the upper band\), that is the additional dispersion caused by the dependence of \( I \) on the electron momentum (see Eq.(4)) is smaller than the broadening of the electron energy due to spin-flip processes. In the opposite limiting case \( |I'|p_F \gg h \nu_s \) one has

\[
R_{st} = \frac{32\pi e^2 \mu}{n^2} \left| I \right|^2 \frac{\hbar \nu_s}{\omega} \left( \frac{b_1 N_1^2 - b_2 N_2^2}{2(\hbar \omega - I)^2 + (\hbar \nu_s)^2} \right) N_p \]

\[
\times \left\{ \left( \epsilon_d \cdot \epsilon_a \right)^2 + \left( \epsilon_d \cdot \epsilon_i \right)^2 \right\} \]

(8)

where the constants \( b_{3,4} \sim 1 \).

One of the necessary conditions for the lasing effect to be realized is (see, e.g., \([9]\)):

\[
R_{st} = \nu_p N_p ,
\]

(9)

where \( \nu_p \) is the photon relaxation rate. We consider the case that damping of electromagnetic waves is mainly due to absorption by free charge carriers, the frequency of the photon relaxation being \( \nu_p = 2k\omega/n \) (see \([10]\)). For estimating the parameters of the problem we use standard formulae for the refractive index \( n \) and the absorption coefficient \( k \) for metals subject to electromagnetic fields (see, e.g., \([11]\)). One finds that

\[
\nu_p = \frac{\mu}{n^2} \frac{4\pi \sigma(0)}{1 + (\omega t_0)^2} .
\]

(10)

where \( \sigma_0 \) is the static conductivity of the conductor, \( t_0 \) is the transport electron relaxation time.

Using Eq.(7) and Eq.(10) one can rewrite Eq.(9) as

\[
\frac{(N_1 - N_2)}{N_1 + N_2} \approx \frac{\hbar \nu_s p_0}{\nu_p} \frac{1}{2I} \frac{1}{h \nu_s} \frac{1}{1 + (\omega t_0)^2} \]

(11)

where \( p_0 = \hbar/a \sim 10^{-19} \text{erg sec/cm} \).

It seems that for achieving the lasing effect the most favorable materials are ferromagnetic manganese perovskites with nearly 100% spin polarization of the conduction electrons (\([1,12,13]\)). The high degree of polarization of the carriers permits the creation of a population inversion of the energy bands in the active region B (see Fig.3). Here and below we use experimental values of the needed parameters: the mean free path \( l_0 = 1.4 \times 10^{-7} \) cm, the Fermi velocity \( v_F \approx 10^8 \text{ cm/s} \), \( t_0 \approx 10^{-15} \text{ s} \) and \( \nu_p = 0.3m \), where \( m \) is the free electron mass, the number of carriers \( \approx 3.4 \times 10^{21} \), the resistivity \( \rho \approx 10^{-4} \Omega \cdot \text{cm} \) (\([14]\)). Inserting these values into Eq.(11) one finds the lasing condition to be

\[
\frac{N_1 - N_2}{N_1 + N_2} \approx \frac{5 \hbar \nu_s}{I}
\]
For the case $|I|p_{F\uparrow} \gg h\nu_s$ (see Eq.(8)), the lasing condition Eq.(9) is

$$\frac{N_{\uparrow}^{2/3} - N_{\downarrow}^{2/3}}{N_{\uparrow} + N_{\downarrow}} \approx 0.5 \times 10^{-7} \text{ cm}$$

From these equations it follows that the lasing condition $R_{st} = \nu_s N_p$ is easily satisfied since one needs $h\nu_s/I$ to be less than $10^{-1}$ while the theoretical estimate of the spin relaxation rate $\nu_s$ gives the value $10^{-2}$ for this ratio. Estimations based on the above experimental values of the parameters show the optical gain to be $g_{opt} = (n/c)R_{st} \sim 10^7 \text{ cm}^{-1}$ and the threshold current density to be $j_{th} = e\nu_s N_{\uparrow} \sim 10^7 \div 10^8 \text{ A/cm}^2$ if the length of the active region is $l = 10^{-5} \text{ cm}$. Estimations for $N_e \sim 10^{18} \text{ cm}^{-3}$ shows the optical gain and the threshold current to be $g_{opt} \sim 10^5 \div 10^4 \text{ A/cm}^{-1}$ and $j_{th} \sim 10^5 \text{ A/cm}^2$.

We predict an extremely large optical gain in systems with a high density of charge carriers. The price to be paid for the gain exceeding what can be achieved in semiconductors by 3 or 4 orders of magnitude is the high currents needed for an effective tunneling pumping of the system. The current value $j = 10^8 \div 10^9 \text{ A/cm}^2$ seems to be very large for homogeneous bulk metals because of the accompanying Joule heating. Special measures are needed to avoid heating the active, lasing region. One solution to that problem is to arrange for the current injection to be inhomogeneous in space. This can be achieved if the magnetic electrodes are electrically connected through a point contact. The spreading out of the current far from the narrow point contact provides for an efficient dissipation of heat [15]. A current density $j \sim 10^8 \text{ A/cm}^2$ can be reached without significant heating of the contact region. On the other hand, the extremely large optical gain $g_{opt} \sim 10^7 \text{ cm}^{-1}$ means that it is enough to have a small volume of active lasing region. Such a structure can be prepared on the basis of the technique suggested in Ref. [16] for fabrication of biepitaxial films of $La_{0.75}Sr_{0.3}MnO_3$ with $45^\circ$ in-plane rotated domains.

In summary we have proposed a new principle for a compact solid-state laser working in the 1-100 THz regime. The proposed laser is based on a new mechanism for creating spin-flip processes in ferromagnetic conductors. The mechanism is due to the interaction of light with conduction electrons; the interaction strength, being proportional to the large exchange energy, exceeds the Zeeman interaction by orders of magnitude. On the basis of this interaction, a giant lasing effect was predicted for systems where a population inversion can be created by tunneling injection of spin-polarized electrons from one ferromagnetic conductor to another — the magnetization of the two ferromagnets having different orientations. Using experimental data for ferromagnetic manganese perovskites with nearly 100% spin polarization we show the laser frequency to be in the range 1-100 THz. The optical gain is estimated to be of order $10^7 \text{ cm}^{-1}$, which exceeds the gain of conventional semiconductor lasers by 3 or 4 orders of magnitude. An experimental study based on a point contact geometry to avoid heating by the necessarily large injection currents was proposed and discussed.

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[1] ”Collosal Magnetoresistive Oxides” edited by Y. Tokura (Gordon and Breach Science Publishers, Amsterdam, 2000).
[2] G.A. Prinz, Science, 282, 1660-1663 (1998).
[3] S.A. Wolf et al., Science 294, 1488 (2001).
[4] The main mechanism for relaxation of hot electrons is the spin-flip process caused by emission of spin-waves.
[5] H.J. Zeiger and G.W. Pratt, ”Magnetic Interactions In Solids”, Clarendon Press, Oxford, 1973.
[6] The Hamiltonian Eq.(5) gives an effective ”spin-orbit” interaction in magnetic films which are usually under a considerable strain caused by the substrate and grain boundaries (see, e.g., [7,8] and references therein). If $pa/h \ll 1$, for bulk materials $\partial I/\partial p_i = 0$, one should take the next term in the expansion, that is change $\partial I/\partial p_i$ to $p_i \partial^2 I/\partial p_i \partial p_i$ in Eq.(5).
[7] Yan Wu, Y. Suzuki, U. Rudiger, J Yu, and A.D. Kent, Appl. Phys. Lett. 74, 2295 (1999).
[8] Yeong-Ah Soh et al., J. Appl. Phys. 91, 7742 (2002).
[9] G.P. Agrawal and N.K. Dutta ”Long-wavelength semiconductor lasers”, Van Nostrand Reinhold Company, New York, 1986;
[10] F.J. Blatt ”Physics of electronic conduction in solids”, McGraw-Hill Book Company, 1968.
[11] J.M. Ziman ”Principles of the theory of solids”, Cambridge, 1964.
[12] J.-H. Park et al., Nature, 392, 794 (1998).
[13] Y.-D. Chuang et al., Science, 292, 150 (2001).
[14] The values are taken for a compound similar to the perovskites manganites found to be nearly 100% spin-polarized, see [13].
[15] A.V. Khotkevich, I.K. Yanson ”Atlas of point contact spectra of electron-phonon interactions in metals”, translation by R.C. Reinertson, Kluwer Academic Publishers, Boston/Dordrecht/London, 1995.
[16] R. Mathieu, P. Svedlindh, R.A. Chakalov, and Z.G. Ivanov, Phys. Rev. B, 62, 3333 (2000).