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Citation: Appl. Phys. Lett. **79**, 1694 (2001); doi: 10.1063/1.1401790
View online: http://dx.doi.org/10.1063/1.1401790
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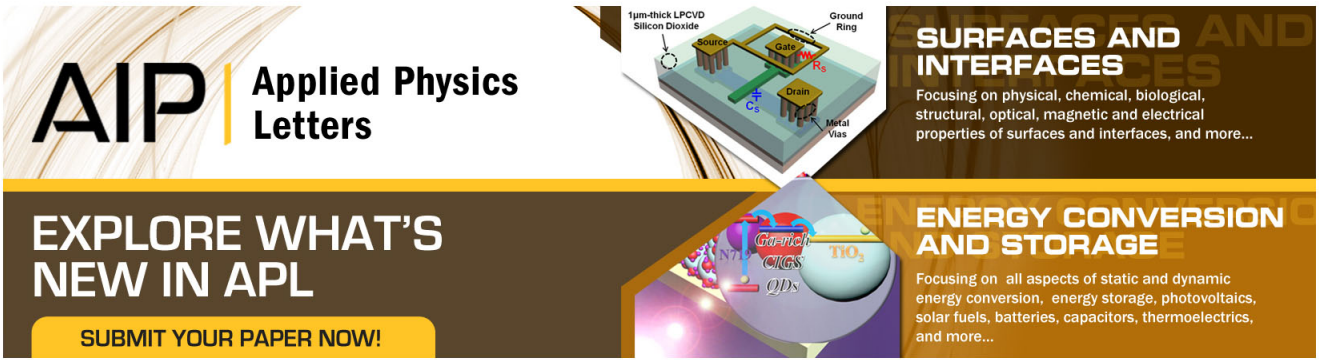

Applications of photoinduced electron spin polarization at room temperature to microwave technology

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(Received 28 March 2001; accepted for publication 13 July 2001)

We present a method for controlling the bulk permeability via spin polarization generated by light excitation. This process involves the magnetic interaction of photoexcited triplets with stable radicals in liquid solution. The resulting non-Boltzmann spin population of the stable radical is reflected by a significant change in the permeability of the chemical system. We demonstrate how these light-driven changes result in conspicuous changes in the reflection coefficient (amplitude and phase) of a microwave cavity in which the active chemical system is placed. This effect can lead the way to ultralow noise microwave amplifiers and low-loss microwave phase shifters, operating at room temperature with very low spin temperature (<16 K). Moreover, the nonlinear character of the phenomenon can be utilized for devices, which protect sensitive instrumentation from a strong destructive microwave pulse. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1401790]

Controlling the magnetic permeability in paramagnetic materials has been recognized as an important feature in microwave technology.[1–3] The general concept is to exploit changes in the electron spin population of the magnetic Zeeman levels, thus allowing us to amplify the electromagnetic radiation in the bulk material. These early studies resulted in the three-level solid-state maser (microwave amplification by stimulated emission of radiation) amplifier based upon the paramagnetic properties of the electron.[4,5] The main advantage of using the maser is typified by its extremely low-noise figure as compared to conventional microwave amplifiers, e.g., vacuum tubes and GaAs field effect transistors. For example, maser technology was used in the discovery of the 3.5 K background blackbody radiation of the universe.[6] Even today, the most advanced state-of-the-art semiconductor microwave amplifiers, e.g., high electron mobility transistors, cooled to cryogenic temperatures can achieve noise temperature of ~6 K, while the maser achieves noise temperature of ~3 K at ~9 GHz.[7] In general, the gap between the maser noise performance relative to cryogenic semiconductor amplifiers increases with microwave frequency.

To achieve amplification in a paramagnetic-based maser, one must achieve inverted spin population, which corresponds to a negative spin temperature. This is done in conventional masers by microwave pumping, but with the restriction of operating at very low temperatures ($T<2$ K). This restriction, which precluded the wide use of masers, can be accounted for by two reasons. First, by microwave pumping high population inversion can be achieved, only if $k_B T \ll h\nu$. The second reason is that the active materials in the microwave pumped masers[8] exhibit a very steep dependence of the spin-lattice relaxation (SLR) time upon temperature. Thus, at high temperatures, the fast relaxation cannot allow for efficient pumping of the magnetic levels. Pumping the levels by optical excitation can overcome the first difficulty of very low temperature.[8–11] Nevertheless, the second constraint of short SLR time still limits the maximum temperature of operation to ~10 K.

It is noteworthy that a radio-frequency amplifier, based upon nuclear polarization at room temperature was suggested in the past.[12] However, this approach is restricted to low radio frequency (<50 MHz and magnetic fields of ~10–20 kG), where existing solid-state electronics provides better noise performances.

The population difference ΔN between the spin levels is directly related to the macroscopic magnetic permeability of the active material, $\mu=1+4\pi\kappa$, where the volume magnetic susceptibility, $\kappa=\kappa'-i\kappa''$, is expressed by[13]

$$\kappa''(\nu-\nu_0)=\Delta N/8\cdot\hbar\gamma^2\cdot(1.75)\cdot f''(\nu-\nu_0), \tag{1}$$

where γ is the electron gyromagnetic ratio (1.76×10^7 $G^{-1}s^{-1}$) and $f''(\nu-\nu_0)$ is the normalized absorption/emission line shape function (usually a Lorentzian)[14] with a maximum at ν_0. The real part of the volume magnetic susceptibility κ', which is related to the signal phase, can be obtained by replacing the line shape function $f''(\nu-\nu_0)$ with the function $f'(\nu-\nu_0)$.[13]

We present here an approach to change μ via photoexcitation, by controlling ΔN [Eq. (1)]. This effect is also known as electron spin polarization (ESP), generated in photoexcited chemical systems.[15] By optimizing the parameters affecting the ESP value, we have achieved changes in μ, which are large enough to produce significant changes in the microwave power reflected from the cavity loaded with the active material. Thus, by controlling μ, two necessary conditions for possible microwave devices can be materialized, namely: (a) achieving maser action, i.e., amplification of a microwave signal with very low noise; and (b) controlling the phase of a microwave signal (low-loss phase shifter). The latter condition was accomplished and can be implemented on a prototype device for protecting sensitive receivers from strong pulses (see below).

In order to generate high ESP, we have utilized a unique process based on the interaction of photoexcited triplets with stable radicals in solution.[16,17] This interaction is powered by the spin exchange and the triplet's zero-field splitting (ZFS)

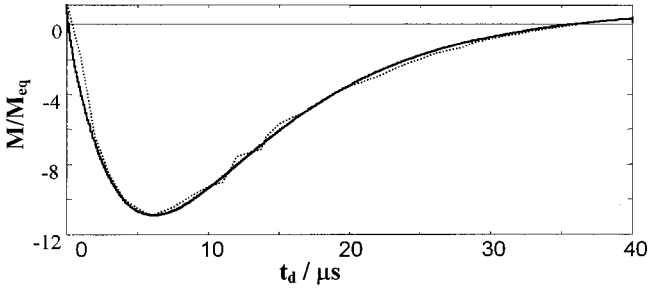

FIG. 1. Experimental (dotted) and calculated (solid) magnetization as a function of the delay time t_d after the laser pulse. Notice the change in magnetization from positive values (thermal equilibrium in absorption) into negative ones (spin polarized in emission). The chemical system consists of the free radical trityl-(tris-(8-carboxyl-2,2,6,6-tetrakis-methylbenzo[1,2d:4,5-d′]bis(1,3)dithiole)methyl), with a linewidth of ~1 MHz and the triplet of etioporphyrin in a mixture of 1-chloronaphthalene:paraffin oil (20:80) with a viscosity of 10 cP. Further experimental and theoretical details are given in Ref. 19.

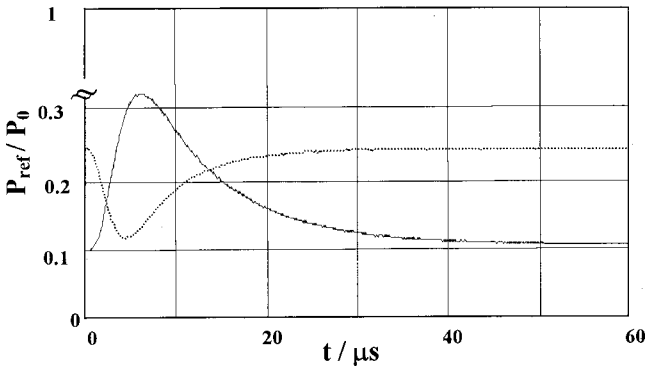

FIG. 3. (Solid line) Time dependence of the reflection coefficient, measured for the system of trityl-etioporphyrin (conditions as in Fig. 2) in a single-ring dielectric cavity with a hole in it; (Dotted line) Same experiment, but employing a double stacked dielectric resonator with different coupling parameters and a smaller filling factor.

parameters, leading to high, controllable, and reversible ESP generated in the radical.[16,18,19] The radical's polarization depends upon the triplet and radical properties, solvent viscosity, and temperature, which can be optimized to obtain high values of ESP.[18–20] The optimal parameters, as predicted by theory, are 300–500 G for the triplet ZFS parameter, solvent viscosity of 20–100 cP, and radical linewidth as small as possible. Figure 1 presents typical experimental results, confirmed theoretically, of the radical's magnetization as a function of the delay time after the laser pulse.

We demonstrate here a preliminary device (Fig. 2), operating at room temperature, based on a chemical system which upon photoexcitation exhibits ESP. This prototype enables us to significantly change the amplitude and phase of the loaded cavity's reflection coefficient P_{ref} due to photoinduced permeability changes in the chemical system (Fig. 3). As of now, we have achieved a maximum change of about 25% in the reflected wave, relative to the impinging radiation amplitude.

We discuss the experimental results of Fig. 3 through the relation between P_{ref} and μ. For a nearly critically coupled cavity, and within the first-order approximation of small changes in the reflected power ΔP_{ref}, one obtains[21]

$$\frac{\Delta P_{\mathrm{ref}}}{P_0} = \pm i 0.193 \cdot 4\pi\kappa\eta Q_c = \pm 0.193 \cdot 4\pi\eta Q_c |\kappa| e^{i\varphi}, \tag{2}$$

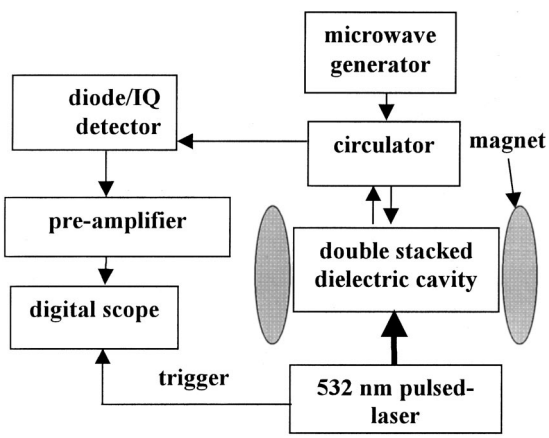

FIG. 2. Schematic presentation of the microwave system, which measures the time-resolved reflected power P_{ref} from the microwave cavity.

where ΔP_{ref} is a complex number whose phase is related to the cavity's coupling properties and the radical's ESP mode (absorption or emission), P_0 is the incident microwave power on the cavity, η is the filling factor of the illuminated sample,[21] $\varphi = \arctan(\kappa'/\kappa'')$, and the quality factor of the cavity Q_c already includes the dielectric losses of the sample, but does not include the losses due to power delivered to the external load. At resonance we relate only to κ'', which is $\sim 5\times10^{-4}$ [calculated by Eq. (1) with $\Delta N \sim 1.2\times10^{16}$ spin/cc (Fig. 1),[19] and the radical linewidth of ~1 MHz]. Finally, the filling factor η of the illuminated part of the two cavities employed (double stacked dielectric resonator[22] and a single dielectric ring resonator) was calculated to be ~0.15 and 0.25, respectively,[23] and the loaded Q_c was measured to be 1000 for both resonators. Inserting these values in Eq. (2), we obtain a predicted noticeable change of $\Delta P_{\mathrm{ref}}/P_0 = 0.18$ and 0.3 for the two cavities at room temperature, which fits well with our experimental findings shown in Fig. 3. Thus, the current microwave system (Fig. 2) allows us to view changes in $\Delta P_{\mathrm{ref}}/P_0$, due to changes in photoexcitation intensity, which vary between 0 and 0.25 and may extend over several hundreds of microseconds after the laser pulse. In addition to the amplitude change of the reflection coefficient a phase change of $\sim 30^0$ was measured using in-quadrature phase detector.

The threshold value for maser action requires that the amplification due to κ'' should be larger than the dielectric losses, implying that[13]

$$\kappa'' > (4\pi\eta Q_L)^{-1} \tag{3}$$

and $1/Q_L = 1/Q_c + 1/Q_{\mathrm{ex}}$, where Q_{ex} corresponds to additional losses of power delivered to the external load.[13] Further improvements in the filling factor of the cavity and of the chemical system may close the gap between the current performance and the required one for achieving this threshold (a factor of ~4). Realization of such a maser can lead to new types of microwave amplifiers, operating at room temperature and still maintaining a low noise figure. The noise temperature T_m for such a device can be approximated by the expression[3,9,10]

$$T_m = \frac{1}{1-\lambda}[\lambda T_\lambda + |T_s|], \tag{4}$$

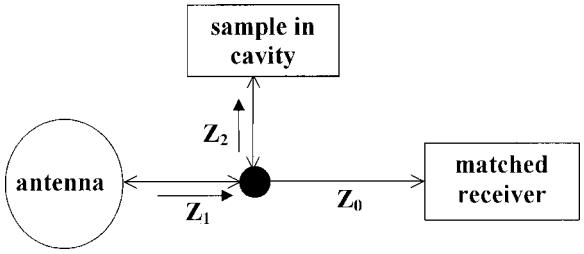

FIG. 4. Schematics of an ELD.

where λ represents the losses in the transmission line from the source to the cavity, T_λ is the ambient temperature of the transmission line, and T_s is the spin temperature, which depends on the deviation from thermal equilibrium of the spin levels and can be calculated using the equation[24] $|T_s| = |\hbar \gamma H_0 / k_B \ln B|$, where $B=(1-\Delta N/N)/(1+\Delta N/N)$, and H_0 is the magnetic field. With the chemical systems used in our experiments we have obtained $\Delta N/N \sim -0.014$, which corresponds to $T_s \sim 15.5$ K. Inspection of Eq. (4) shows that the noise temperature can be much smaller than the ambient temperature, i.e., when $\lambda \rightarrow 0$.[9–11] In practical designs, λ can be as low as 0.01–0.055, which gives a noise temperature T_m that is very close to that of the spin temperature T_s.[3,9,11,25] Noise temperature below the ambient temperature is also common in conventional solid-state electronic amplifiers.[25]

Combining the nonlinear properties of the spin system and the ability to control the cavity's reflection coefficient enabled us to construct an electromagnetic limiting device (ELD) (Fig. 4). This application should protect sensitive receivers, which may be exposed to strong microwave pulses. The advantage of utilizing the ESP phenomenon in such a device is the low losses of the system, while other protective devices add substantial noise and losses to the system. Thus, a spin system subjected to high intensity of microwave radiation is saturated, implying that $\Delta N \rightarrow 0$ and $\mu \rightarrow 1$. The signal from the antenna is reflected at the waveguide junction with reflection coefficient amplitude: $\Gamma=(Z_1-Z_0)/(Z_1+Z_0)$, where Z_0 is the waveguide's impedance, Z_1 is the impedance at the junction (obeying $1/Z_1=1/Z_0+1/Z_2$), and Z_2 is the impedance at the entrance of the stub leading to the cavity. Without laser excitation, the system is designed such that $Z_2 \ll Z_0$, i.e., $Z_1 \sim Z_2 \ll Z_0$, implying that most of the power is reflected back from the junction. When the chemical system is photoexcited, the bulk's permeability is changed such that $Z_2 \gg Z_0$. Under this condition, the microwave signal is channeled directly from the antenna to the receiver. If the antenna is subjected to a high intensity signal, it will saturate the spin system in the cavity, and the non-Boltzmann magnetization, generated by light excitation, will be destroyed rapidly (resulting in $Z_2 \ll Z_0$, which is equivalent to the case with no laser light). Therefore, destructive signals will not be able to penetrate the junction and will be reflected.

The short lifetime of the photophysical processes described (up to 300 μs), allows operating the devices only with synchronized microwave pulses. In principle, these devices can be operated in a continuous wave microwave excitation mode, using high-repetition laser pulses. In our system, this will require a laser power of ~10 W=1 mJ/pulse $\times$1/100 μs to maintain steady state magnetization. This value is too large for the sample to sustain heating effects and photodegradation. Thus, it can be concluded that cw operation requires further improvement of the device constituents (e.g., cavity, active chemical material, and laser performance).

To summarize, this work demonstrates the applicative aspects of ESP, generated by a photophysical process at room temperature.

This work was partially supported by the Israel Ministry of Science and Ministry of Defence. The Farkas Research Center is supported by the Bundesministerium für die Forschung und Technologie and the Minerva Gesellschaft für Forchung GmbH, Germany. This work is in partial fulfillment of the requirements for a Ph.D. degree (A.B.) at the Hebrew University of Jerusalem. The authors are grateful to Dr. Lars-G. Wistrand from Nycomed Innovations in Sweden for providing them with the trityl radical. Valuable discussions with Dr. A. Berg of the Hebrew University are highly appreciated.

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