Quantum rf emission in the spectrum of electron spin-spin interactions of a solid

É. V. Avagyan, V. A. Atsarkin, and V. V. Demidov
Institute of Radio Engineering and Electronics, Academy of Sciences of the USSR

(Submitted 23 December 1986)
Pis'ma Zh. Eksp. Teor. Fiz. **45**, No. 3, 157–160 (10 February 1987)

Quantum emission in the rf range has been achieved for the first time through the induced emission of energy from the electron spin-spin reservoir of a solid paramagnet in a zero magnetic field.

The existing quantum oscillators and amplifiers in the rf range (masers)[1] operate by virtue of an induced emission from a system of discrete energy levels separated by an interval $\Delta E = \hbar\Omega$, where Ω is the frequency of the signal which is excited or amplified. In particular, it is assumed that the operation of a maser in the standard three-level model (Fig. 1a) is possible only under the condition $\Omega \gg \omega_L = \gamma H_L$, where H_L is the rms local field, and γ is the gyromagnetic ratio. This assumption rules out a simultaneous saturation of the 1–3 and 1–2 transitions by the pump field. This condition restricts the concentration of active paramagnetic centers which can be attained to a level $\sim 3 \times 10^{-4}$, when Ω is in the microwave range.

On the other hand, we know that in a solid the saturation of an ESR line with some frequency offset $\Delta = \omega - \omega_0$ from the center of the line, ω_0, causes a sharp change in the temperature (T_{SS}) of the electron spin-spin reservoir. At $\Delta > 0$, a population inversion arises[2,3] ($T_{SS} < 0$; Fig. 1b) over the entire continuous spectrum of the reservoir, with a width $\sim \omega_L$. However, it is usually not possible to exploit this inversion to achieve quantum emission at frequencies $\Omega \sim \omega_L$, since in magnetic fields $H_0 \gg H_L$ induced transitions within the spectrum of the electron spin-spin reservoir are

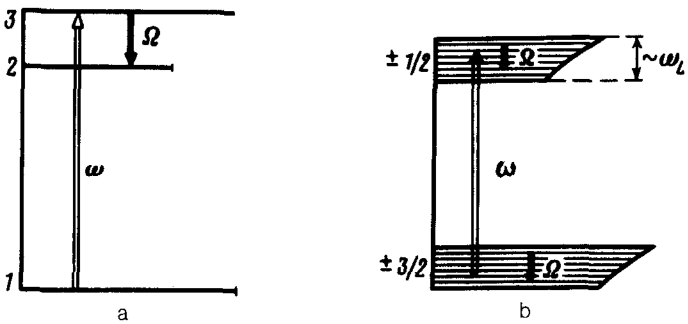

FIG. 1. Energy diagrams of the production of a population inversion. a—In a three-level system; b—in the quasicontinuous spectrum of an electron spin-spin reservoir. The length of the horizontal line segments is proportional to the population of the corresponding level.

 0021-3640/87/030195-04$01.00

forbidden by the selection rules for the secular part $\hat{\mathscr{H}}_d^0$ of magnetic dipole-dipole interactions. This difficulty can be circumvented by using paramagnetic ions with an effective spin $S > 1/2$ at $H_0 = 0$. For example, the energy spectrum of an ion with $S = 3/2$ in a zero magnetic field consists of two degenerate Kramers doublets with $\pm 1/2$ and $\pm 3/2$, separated by an "initial" stark splitting $\hbar\omega_0$ in the crystal field. The Hamiltonian $\hat{\mathscr{H}}_d^0$ now includes terms which allow a change of ± 1 in the magnetic quantum number, so that transitions induced by the rf field are allowed in the spectrum of the electron spin-spin reservoir (Fig. 1b).

We have observed a quantum generation due to an induced emission in the spectrum of an electron spin-spin reservoir in crystals of rutile (TiO_2) containing $n = 6 \times 10^{19}$ cm^{-3} paramagnetic Cr^{3+} ions ($S = 3/2$) at $H_0 = 0$ and at a temperature $T_0 = 1.7$ K. The sample, with a volume $V_0 = 0.4$ cm^3, is placed in the inductance coil of a high-Q rf circuit which is tuned to the frequency $\Omega/2\pi \sim 10^7$ Hz. It is irradiated with a microwave pump field at a frequency $\omega = \omega_0 + \Delta$, where $\omega_0/2\pi = 42.8$ GHz is the initial splitting of the Cr^{3+} levels in rutile.[4] The frequency offset $\Delta \simeq 400$–600 MHz maximizes the steady-state inversion in the spectrum of the reservoir,[4] $E = T_0/T_{SS} \approx -40$, at a pump power of about 0.1 W. The generation occurs only if the quality factor of the rf circuit, Q, exceeds a certain critical Q_c. This value is about 750 at the optimum frequency, $\Omega/2\pi = 18.5$ MHz, and increases slightly at the edges of the range studied, 8–30 MHz. Figure 2 is an experimental oscilloscope trace. We see a delay in the beginning of the generation due to the time required for the inversion to rise to the critical value. Then we enter a steady state. Finally, the oscillations decay after the pump pulse ends. The maximum generation power is about 0.2 mW.

For a theoretical description of the effect we work from the Provotorov equations,[2] supplementing them with terms to reflect the interaction of the electron spin-spin reservoir with the rf field $h\exp(i\Omega t)$ and also with an equation for the energy of this field, $\epsilon_\Omega = h^2 V_0/8\pi\eta$ (the coefficient η gives the filling of the circuit with the material):

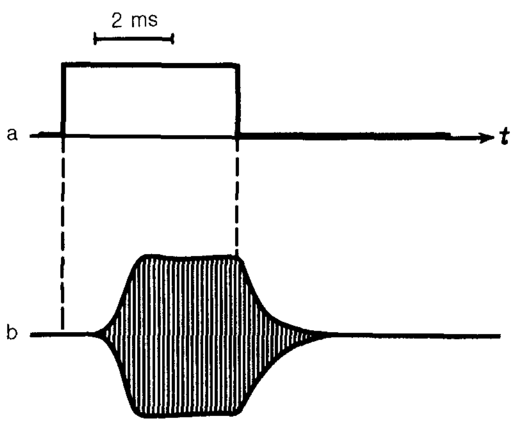

FIG. 2. Quantum generation on levels of the electron spin-spin reservoir in rutile. a—The microwave pump pulse; b—the generation signal at 18.5 MHz.

$$\begin{cases} \dfrac{d}{dt}\beta_S = -2W_\omega(\beta_S + \beta_{SS}\Delta/\omega_0) - \tau_{SL}^{-1}(\beta_S - \beta_0) \\ \dfrac{d}{dt}\beta_{SS} = -2W_\omega\left(\dfrac{\Delta^2}{\omega_L^2}\right)(\beta_S\omega_0/\Delta + \beta_{SS}) - \tau_{SSL}^{-1}(\beta_{SS} - \beta_0) - 2W_\Omega\left(\dfrac{\Omega^2}{\omega_I^2}\right)\beta_{SS}, \\ \dfrac{d}{dt}\epsilon_\Omega = -(\hbar^2/2k)W_\Omega n V_0\Omega^2\beta_{SS} - \tau_\Omega^{-1}\epsilon_\Omega \end{cases} \quad (1)$$

where $\beta_S^{-1} = T_S$, $\beta_{SS}^{-1} = T_{SS}$, and $\beta_0^{-1} = T_0$ are the temperatures of the Stark subsystem, the electron spin-spin reservoir, and the lattice; W_ω and W_Ω are the probabilities for the induced transitions at the frequencies ω and Ω; τ_{SL} and τ_{SSL} are the spin-lattice relaxation times of the Stark subsystem and of the reservoir; and $\tau_\Omega = Q/\Omega$ is the decay time of the oscillations in the rf circuit. We are ignoring the slight spontaneous emission with the temperature T_0. Similar equations were derived in Ref. 5 in an analysis of the maser effect at the wing of an ESR line during pumping not strictly at resonance.[6]

The probability for an induced transition in the spectrum of the reservoir is

$$W_\Omega = \frac{\pi}{2}\hbar^{-2}|\mu_\Omega|^2 G(\Omega)h^2, \quad (2)$$

where μ_Ω is the matrix element, and $G(\Omega)$ is the form factor, both of which are determined by the Hamiltonian $\hat{\mathscr{H}}_d^0$ at $H_0 = 0$. It is a difficult matter to calculate these parameters, so it is more convenient to study system (1) by introducing $\chi''(\Omega)$: the imaginary part of the magnetic susceptibility at the frequency Ω. This quantity is related to the absorption of rf energy in the material:

$$\left(\frac{\partial}{\partial t}\epsilon_\Omega\right)_{\mathrm{ESSR}} = -\frac{1}{2}\Omega\chi''(\Omega)V_0 h^2 . \quad (3)$$

It is furthermore proportional to[4] β_{SS}. Restricting the analysis to the case of a pronounced saturation at the pump frequency[3] ($W_\omega\tau_{SL} \gg 1$, $-\beta_S \to \Delta\beta_{SS}/\omega_0$), and using (3), we find

$$\begin{cases} \dfrac{d}{dt}\beta_{SS} = -(\tau_{SL}^*)^{-1}(\beta_{SS} - \beta_{SS}^\infty) - \dfrac{2k}{\hbar^2}\dfrac{\Omega}{(\Delta^2 + \omega_L^2)n}\chi''(\Omega)h^2 \\ \dfrac{d}{dt}(h^2) = -4\pi\Omega\eta\chi''(\Omega)h^2 - \tau_\Omega^{-1}h^2 \end{cases} \quad (4)$$

where $\beta_{SS}^\infty = E^\infty\beta_0 = -\omega_0\Delta\beta_0/(\Delta^2 + a\omega_L^2)$ is the steady-state value of β_{SS} at strong microwave saturation, but without allowance for the effect of the generation field; $\tau_{SL}^* = \tau_{SL}(\Delta^2 + \omega_L^2)/(\Delta^2 + a\omega_L^2)$; and $a = \tau_{SL}/\tau_{SSL}$.

In the steady state ($d/dt = 0$) system (4) has two solutions: the trivial solution ($h_{\mathrm{st}} = 0$, $\beta_{SS}^{\mathrm{st}} = \beta_{SS}^\infty$) and the solution corresponding to generation at the frequency Ω:

$$\begin{cases} h_{\rm st}^2 = -\dfrac{\hbar^2\beta_0(\Delta^2 + a\omega_L^2)n}{2k\tau_{SL}\Omega}\left(\dfrac{1}{\chi_0''(\Omega)} + 4\pi\eta Q E^\infty\right) \\ \beta_{SS}^{\rm st} = -\beta_0/4\pi\eta\chi_0''(\Omega)Q\ , \end{cases} \tag{5}$$

where $\chi_0''(\Omega)$ is the equilibrium value of $\chi''(\Omega)$ at the temperature T_0. Solution (5) obviously exists only if $E^\infty < 0$, and for it we have the threshold condition

$$Q^{-1} < Q_c^{-1} = 4\pi\eta\,|E^\infty|\,\chi_0''(\Omega), \tag{6}$$

which is the standard condition for paramagnetic masers.[1] The amplitude of the generation is limited in the steady state by a partial saturation of the reservoir by the generation field. From (5) and (6) we find $|\beta_{SS}^{\rm st}| < |\beta_{SS}^\infty|$.

Assuming $[\chi_0''(\Omega)]_{\max} \simeq \chi_0/2$, where χ_0 is the static susceptibility, we find $Q_c \sim 10^2$ from (6) for our sample. This value is noticeably lower than the experimental value. The apparent reasons for the discrepancy are the incomplete microwave saturation and the roughness of our estimate of $\chi_0''(\Omega)$. The steady-state rf power dissipated in the circuit is

$$P = \epsilon_\Omega/\tau_\Omega = \frac{\hbar^2 n V_0\omega_0\Delta}{4kT_0\tau_{SL}}\left(1 - \frac{Q_c}{Q}\right) \tag{7}$$

Its maximum value, calculated from (7) at the optimum frequency offset[3] $\Delta = \omega_L\sqrt{a}$, at $Q \gg Q_c$, and with the parameter values $\gamma_{SL} = 8$ ms, $\omega_L/2\pi = 200$ MHz, and[4] $a = 8$ for the material, is 3×10^{-4} W, in fair agreement with experiment.

In summary, a direct conversion of the collective energy of spin-spin interactions of a solid paramagnet into coherent electromagnetic waves has been achieved for the first time. In contrast with other quantum generators for the rf range,[7,8] it is possible in this case to continuously tune the frequency Ω over the entire continuous spectrum of the electron spin-spin reservoir, which ranges essentially from 0 to 10^8–10^9 Hz.

[1] G. Trope, Quantum Amplifiers and Generators (Russ. trans., IL, Moscow, 1961).

[2] B. N. Provotorov, Zh. Eksp. Teor. Fiz. **41**, 1582 (1961) [Sov. Phys. JETP **15**, 1098 (1961)].

[3] V. A. Atsarkin and M. I. Rodak, Usp. Fiz. Nauk **107**, 3 (1972) [Sov. Phys. Usp. **15**, 251 (1972)].

[4] É. V. Avagran, V. A. Atsarkin, and G. A. Vasneva, Zh. Eksp. Teor. Fiz. **85**, 1790 (1983) [Sov. Phys. JETP **58**, 1041 (1983)].

[5] N. M. Aminov and B. I. Kochelaev, Pis'ma Zh. Eksp. Teor. Fiz. **15**, 224 (1972) [JETP Lett. **15**, 156 (1972)].

[6] R. M. Valishev, B. I. Kochelaev, and A. Kh. Khasanov, Fiz. Tverd. Tela (Leningrad) **16**, 3726 (1974) [Sov. Phys. Solid State **16**, 2424 (1974)].

[7] R. Bösiger, E. Brun, and D. Meier, Phys. Rev. A **18**, 671 (1978).

[8] A. L. Buchachenko and V. L. Berdinskiĭ, Vestn. Akad. Nauk SSSR No. 1, 91 (1981).

Translated by Dave Parsons