Radio-frequency superradiance in a resonator

Yu. F. Kiselev and É. I. Aliskenderov
United Institute of Nuclear Research

(Submitted 20 November 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **51**, No. 1, 14–16 (10 January 1990)

Modified Bloch equations are derived with allowance for the phase-frequency characteristic of the oscillatory circuit. It is shown that nonlinear processes of radio-frequency super-radiation (SR) and maser oscillation (MO) as well as the oscillating frequency are closely related to the phase characteristic of the circuit; this makes it possible in an experiment to separate the SR effect from the MO effect.

Kiselev *et al.*[1] reported the detection of the phenomenon of radio-frequency superradiation (SR), and this was confirmed by independent studies.[2] In the experiments of Ref. 1, the high nuclear polarization of the proton spins of propanediol, obtained by the method of dynamic cooling, was frozen by lowering the lattice temperature to 50 mK. Then the Larmor frequency ω_0 of the spin system inversely polarized with respect to the external magnetizing field H_0 was matched with the resonance frequency of a passive radio-frequency circuit by changing H_0, whereupon, depending on the magnitude of the initial polarization, either a long pulse of maser oscillation (MO) or SR and MO pulses separated by a time interval were observed. A more detailed analysis of the data of Ref. 1 shows that as the absolute value of the initial polarization increases, the MO pulse width increases (Fig. 1a and 1b), and the SR pulse width decreases. As follows from Bloom's calculations,[3] the Bloch equations do not even give a qualitative explanation for the widening of the MO pulse width as the absolute value of the initial polarization increases. A characteristic feature of the experiments of Ref. 1 is the approximate equality of NMR line width and circuit passband (~40 kHz). The disagreement with experiment is due to the fact that the Bloch equations do not allow for the complex nature of the radio-frequency circuit impedance, when the radiation frequency is not equal to the circuit tuning frequency. Using the notation of Ref. 3, we obtain modified Bloch equations which take into account the phase-frequency characteristic of the circuit, and which provide at least a qualitative explanation of the observed effect. If H_0 is directed along the z axis, and θ is the angle between z and the magnetization vector $\overline{M}$, then its components are $M_x = M \sin\theta \cos\phi$; $M_y = -M \sin\theta \sin\phi$; $M_z = M\cos\theta$, where ϕ is the angle between the magnetization component in the x, y plane and the x axis. The components of the magnetic field, with allowance for the phase shift φ between the voltage and the current in the circuit, are

$$H_x = H_1 \cos\omega t + H_r \sin(\phi - \varphi), \qquad H_y = -H_1 \sin\omega t + H_r \cos(\phi - \varphi),$$
$$H_z = H_0, \qquad \varphi = \operatorname{arctg}[(\omega_1 L - 1/\omega_1 C)/R], \tag{1}$$

where ω_1 is the frequency of the current in the circuit, and L, C and R are the circuit

© 1990 American Institute of Physics

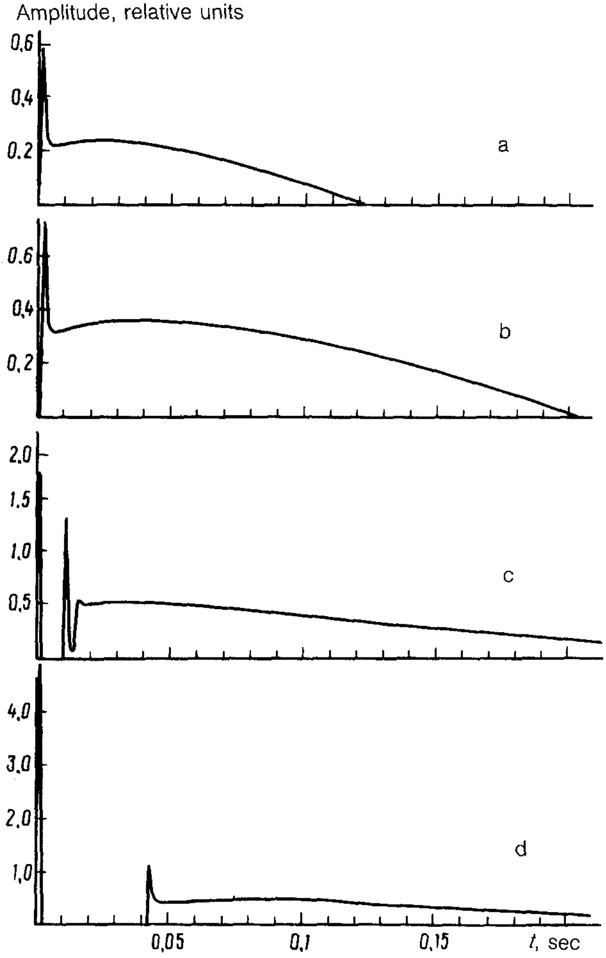

FIG. 1. Amplitude of MO and SR pulses plotted as a function of the initial polarization of protons; $P = -0.18$, -0.34, -0.49, and -0.50, respectively, for (a), (b), (c), and (d), respectively. The MO pulse width in (a) and (b) is proportional to the polarization, in accordance with Eq. (4). Rate of change $H_0 = 100$ G/sec.

inductance, capacitance, and resistance, respectively. It is assumed that the external radio-frequency field $2H_1 \cos \omega t$ is directed along the x axis, and H_r is the field component produced by the rotating magnetization $M \sin \theta$. Since H_r is proportional to the absolute value of the current in the circuit, we can write

$$H_r = kM \cos\varphi \sin\theta, \qquad (2)$$

where the constant k should be determined at the circuit resonance frequency for $\varphi = 0$. Introducing the new variables[3] $u = M_x \cos \omega t - M_y \sin \omega t = M \sin \theta \cos(\omega t - \phi)$, $v = -M_x \sin \omega t - M_y \cos \omega t = -M \sin \theta \sin(\omega t - \phi)$, and using Eqs. (1) and (2), we obtain a modified system of Block equations for the complex impedance of the radio-frequency circuit

$$\frac{du}{d\tau}+\beta u+\delta v=-rM_z[u\cos\varphi+v\sin\varphi],$$

$$\frac{dv}{d\tau}+\beta v-\delta u+M_z=-rM_z[v\cos\varphi-u\sin\varphi],\tag{3}$$

$$\frac{dM_z}{d\tau}+\alpha M_z-v=\alpha M_0+r[u^2+v^2]\cos\varphi,$$

where $\tau=\gamma H_1 t$, $\alpha=1/\gamma H_1 T_1$, $\beta=1/\gamma H_1 T_2$, $\delta=(\omega_0-\omega)/\gamma H_1$, $\omega_0=\gamma H_0$, and $r=k\cos\varphi/H_1$. For $\varphi=0$, the system of equations (3) is identical to the equations of Ref. 3. We thus have $k=2\pi\eta Q$, where η is the duty factor, and Q is the circuit quality factor. The terms with r in Eqs. (3) now depend on the circuit phase-frequency characteristic via angle φ, resulting in qualitatively new effects. It follows from Eqs. (3) that the oscillating frequency of the spin system, which is strongly coupled to the passive resonance circuit when $\varphi\neq0$,is not equal to the Larmor frequency ω_0. When $H_1=0$ we have

$$\omega_1=\frac{d\phi}{dt}=\omega_0+\gamma kM_z\sin\varphi\cos\varphi,\tag{4}$$

A shift of the oscillating frequency also means that the duration of the emission depends on M_z (Fig. 1a and 1b). In our case, the frequency shift $\omega_0/2\pi=2.1\times10^7$ Hz amounts to 10^5 Hz. We recall that $H_0(t)$ changes at a constant rate during the experiment. Modulation of H_0 leads to a deviation φ and gives rise to amplitude modulation, which was observed in Ref, 1. We note that a shift in oscillating frequency was predicted some time ago,[4] but this effect is very slight for liquids, and we do not know whether this shift had been detected earlier by anyone. As the initial polarization increases further, a partial reversal of magnetization M_z takes place (Fig. 1c and 1d), so that the lasing is temporarily interrupted. The nonreversed part of the NMR line then excites MO again (long pulses in Fig. 1c and 1d). Note, in particular, that in all of the experiments discussed, as a result of the action of the spin diffusion mechanism, the final nuclear polarization decreases in absolute value, but it remains a negative quantity. Thus, allowing for the circuit phase characteristic in Eqs. (3) makes it possible to substantiate the method of separation of SR and MO in terms of the dependence of the duration of the emission on the magnitude of the nuclear polarization, even if the magnetization does not change sign. As noted in Ref. 1, other methods include the reversal of total magnetization in the case of superradiant lasing, and the possibility of amplitude modulation in the case of MO. Hence, as stated in Ref. 2, a change in the sign of magnetization is by no means a necessary condition for identifying SR.

Equations (3) imply that there is a change in the condition for MO, which is a necessary condition[5] SR

$$T_2^{-1}=2\pi\eta\gamma Q\,|M_z|\cos^2\varphi,\quad M_z<0.\tag{5}$$

This equation signifies that emission cannot occur outside the circuit passband. As shown by the calculation of Eqs. (3), the time of which the emissions occurs depends essentially on the angle φ, a situation which cannot be inferred from the standard Bloch equations.

Thus, in the nonlinear processes of SR and MO, a real passive circuit shifts the oscillating frequency relative to the Larmor spin frequency and affects the condition of self-excitation of MO and SR. The modified equations (3) can be used in important practical applications in connection with the possibility of obtaining high polarization in liquids at room temperatures by the method of chemical polarization of nuclei.[6]

We are very grateful to N. Blombergen for interest in this work[1] on the detection of radio-frequency SR, to N. N. Bogolyubov for support, to A. S. Shumovskii for helpful discussions, and to A. F. Prudkoglyad for allowing us to use the results of a joint experiment.

[1] Yu. F. Kiselev *et al.*, Zh. Eksp. Teor. Fiz. **94**, 344 (1988) [Sov. Phys. JETP **67**, 1931 (1988)]; Mod. Phys. Lett. **B 1**, 409 (1988).

[2] N. A. Bazhanov *et al.*, Fiz. Tverd. Tela **31**, 206 (1989) [Sov. Phys. Solid State **31**, 291 (1989)].

[3] S. J. Bloom, Appl. Phys. **28**, 800 (1957).

[4] F. I.Skripov and E. D. Al'tman, Izv. Vyssh. Uchebn. Zaved. Radiofiz. **5**, 104 (1962).

[5] Yu. F. Kiselev *etal.*, Preprint R17-89-441, JINR, 1989.

[6] A. L. Buchachenko *et al.*, *Magnetic and Spin Effects in Chemical Reactions* (Nauka, Moscow, 1978), Ch. 2.

Translated by A. Peiperl