Detection of superradiant emission from a system of nuclear magnetic moments

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Spontaneous formation of a Dicke superradiance state was observed in a system of inversely polarized proton spins with a "frozen" polarization. Superradiance was observed when the Larmor frequency coincided with the resonance frequency of a passive oscillatory circuit. The initial reason for the appearance of superradiance was incoherent maser radiation emitted by spins. At ultralow temperatures the frequency of a superradiant oscillator was tunable from several hundreds of kilohertz to hundreds of megahertz. The dependence of incoherent and coherent emission on the initial polarization was studied. Depending on the occupancy factor and on the polarization of the nuclei, one could expect an rf analog of an optical superradiant laser in the case of weakly amplifying and strongly amplifying active media. In the latter case, superradiant reversal of the negative polarization was observed.

In 1953, Dicke[1] extended theoretically the idea of the Hahn experiment[2] on the formation of coherent states in a system of nuclear spins to microwave spectroscopy and optics. The coherence was established by irradiation of matter with an rf field. Another method for spontaneous time correlation of spatially separated radiators (superradiant SR) was achieved in the optical and infrared ranges.[1] Superradiance is a nonlinear process with an output intensity proportional to the square of the number of radiators and to the maximum possible rate of "depletion" of the populations. Recent developments in the technique of dynamic polarization of nuclei in condensed media at meter wavelengths and ultralow temperatures could be used to achieve values of the Boltzmann factor $\mu H/kT$ (μ is the magnetic moment of a nucleus) like those in microwave spectroscopy at liquid helium temperatures. Naturally, we are then faced with the fundamental question of the possibility of spontaneous appearance of SR of magnetic dipoles in a state of thermodynamic equilibrium characterized by a negative spin temperature. In contrast to the optical and infrared ranges, the probability of spontaneous emission of rf radiation is negligible, so that spontaneous emission cannot be the cause of SR. For this reason, in contrast to the optical case, we cannot generate coherent radiation at radio frequencies without a resonator. Nevertheless, at meter wavelengths one can induce not only spontaneous SR (without the application of a $\pi/2$ pulse), but also to produce oscillations differing from that in an SR laser only by the process of formation of a population inversion. In fact, we shall show that in the case of a strong coupling of nuclear spins to a passive resonant circuit, an incoherent maser oscillation acting as spontaneous emission in optics can develop a sufficient field amplitude in a circuit for the onset of time-correlated rotation of the total magnetization of the spin system.

The feasibility of SR reversal of the polarization was demonstrated by Bloembergen and Pound.[3] Faĭn[4] proposed the name kogetron for a new coherent source of radiowaves. An attempt to induce SR in a system of aluminum nuclei in ruby (Al_2O_3) was reported in 1977 by Bösiger *et al.*[5] However, these authors[5] were only able to observe incoherent maser action which in the case of weakly amplifying media and weak polarization of nuclei was of intensity much higher than the intensity of the coherent SR process, so that it was impossible to observe the latter. We shall show that an increase in the proton polarization above the maser threshold gives rise to a coherent SR process. We shall report an investigation of the dependence of the maser action and SR on the degree of polarization of nuclei.

EXPERIMENTAL ARRANGEMENTS DESIGNED TO DETECT RADIOFREQUENCY SUPERRADIANCE

Our active substance was an insulator with a high proton concentration, propanediol ($C_3H_8O_2$), containing a paramagnetic Cr^{+V} impurity. The substance in the form of cooled spheres 1 mm in diameter was poured into a chamber where ^3He was dissolved in ^4He and magnetized by a static field of $H_0 = 26.4$ kG. The fill factor was 0.6 and the spin concentration was $n = 3.8\times10^{22}$ cm^{-3}. Experiments were carried out using two cryogenic systems. In the first the same two-turn coil inside a microwave resonator, filled completely with a sample of 12 cm^3 volume, was used to measure the polarization and to generate SR.

A high-Q coaxial cable, representing a part of an oscillatory circuit, was used to couple the coil to a system for measuring the nuclear polarization with a Q meter. In this system we observed SR at a frequency of 65 MHz in a circuit comprising the coil, surrounded by the microwave resonator, and the cooled part of the coaxial cable with the Q meters disconnected. In this experimental geometry we were able to achieve SR reversal of the sign of the polarization. All the other experimental results reported below were obtained using the second system. In this case, analysis of the experimental results was simplified by observing SR in a resonant circuit composed of a 10-turn coil with an area of 2.7 cm^2 and a 29-pF capacitor. The resonant frequency of the circuit was 21.5 MHz in the cooled state and the Q factor was $Q = 580$. A Teflon chamber for the dissolution of ^3He in ^4He with a sample of 0.5 cm^3 volume was placed inside the coil. After cooling down to $T = 0.3$ K the resonator was supplied with microwave power at a frequency of 120 MHz higher than the central frequency of the ESR line of the Cr^{+V} ion (73 GHz). In the case of nonresonant irradiation of the ESR line, "dynamic cooling" took place,[6] the result of which was a high negative polarization of nuclear spins. Since the dissolution chambers were constructed differently, the maximum polarization of the nuclei in the first system was $P = -0.9$,

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whereas in the second system it was $P = -0.7$. At the end of the polarization process the microwave source was switched off and the temperature of the dissolution chamber was lowered to < 50 mK. Cooling slowed down greatly the process of nuclear spin–lattice relaxation and, consequently, the nuclear and electron spin systems became practically uncoupled. By way of example, we should point out that characteristic longitudinal spin–relaxation time of nuclei considered as a function of the field H_0 (5–26.4 kG) ranged from 50 to 500 h. We observed SR when the Larmor frequency of the nuclei coincided with the resonance frequency of the circuit when H_0 was varied adiabatically slowly. The "frozen-in" magnetization was retained up to the appearance of the maser effect or SR, and for the spin $I = 1/2$ when $P = 0.6$ it amounted to

$$M = {}^1/_2 n\gamma hP = 0.32 \text{ G}, \tag{1}$$

where $\gamma = 4.257 \times 10^4$ Hz/Oe is the gyromagnetic ratio for protons and h is the Planck constant. A static nuclear susceptibility of the sample was $\chi_0 = M/H_0 = 1.2 \times 10^{-5}$ ($H_0 = 26.4$ kG). In the case of the Gaussian line profile the maximum hf nuclear susceptibility $\chi''(\nu_{\max})$ was (for $P = 0.6$)

$$\chi''(\nu_{max}) = 0.47\pi\chi_0\nu_{max}/\gamma\Delta H = 0.47\pi M/\Delta H = 3.6\cdot 10^{-2}, \tag{2}$$

where $\Delta H = 13$ Oe is the width of the NMR line at midamplitude and $\nu_{\max} = \gamma H_0$. The appearance of an hf voltage in the circuit resulted in generation of a starting pulse by a special triggering device; this pulse switched on an analyzer which operated as a digital oscilloscope. The analyzer recorded the shape of the video signal representing the envelope of the rf pulse profile.

EXPERIMENTAL RESULTS

When the Larmor frequency of the nuclei coincided with the resonance frequency of the circuit, the initial polarization $P = -0.85$ was reversed spontaneously. Depending on the rate of passage across the resonance of the circuit, the final polarization had values between 0.12 and 0.5. This represented SR reversal of the magnetization under the influence of a high-power rf pulse generated in a passive circuit (possibly in the walls of the microwave resonator) by the nuclear spins. This pulse could be observed directly on the screen of an oscilloscope. The most probable (under statistical equilibrium conditions) initial value of the cooperative number r (Ref. 1) was

$$r = |\bar{m}| = {}^1/_2|n_+ - n_-| = {}^1/_2 n|P|, \tag{3}$$

where n_+ and n_- are the populations of the sublevels $+1/2$ and $-1/2$. In the case of polarization reversal the cooperative number changed in accordance with Eq. (3), indicating directly the presence also of incoherent radiation. The maximum intensity I proportional to the square of the voltage across the circuit in the Dicke state (r, $m = 0$) should include a term

$$I \propto (r+m)(r-m+1) \propto n^2P^2, \tag{4}$$

proportional to the square of the polarization P. An investigation of the dependence of the maser effect and SR on P was carried out using the second system. Such experiments established that in the range $-(0.39\text{–}0.45) < P < -0.09$, where the lower limit depended on the rate of change of the field, only one maser pulse with a characteristic wide pulse profile was observed, lasting up to 0.3 s. The leading edge of this pulse transformed into a separate steep-ended SR pulse as the modulus of the polarization increased (Fig. 1). Using the initial value $P = -0.09$ at which the maser effect was observed, we could calculate the fill factor η of the coil by the sample:

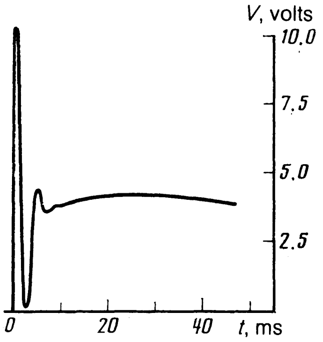

FIG. 1. Leading edge of a detected maser effect voltage used in separation of an SR pulse (frequency 21.5 MHz, $|P| = 0.45$, $dH_0/dt = 100$ Oe/s).

$$\eta = [4\pi Q\chi''(\nu_{max})]^{-1} = 0.025. \tag{5}$$

The value of η was approximately eight times less than the geometric fill factor of the coil. Since $\chi''(\nu_{\max}) = \pi\gamma\chi_0 H_0 T_2$ (T_2 is the spin–spin relaxation time), Eq. (5) was identical with the condition for the excitation of a spin oscillator:

$$\eta \geqslant [(2\pi)^2\gamma QMT_2]^{-1}.$$

Further increase in $|P|$ caused the SR intensity to rise rapidly and the onset of the maser effect to shift away from the SR pulse on the time axis (Fig. 2). Modulation of the field H_0 by a field of acoustic frequency (50 Hz) caused amplitude modulation of the maser pulse at the same frequency (Fig. 3). An increase in the depth of modulation could be used to interrupt and start again the maser action repeatedly. Consequently, a "long" pulse (Figs. 2 and 3) did indeed represent incoherent maser radiation (as demonstrated also by the shape of its pulse). A calculation of the duration of the maser pulse, in the case of a known rate of field scanning, indicated that in the course of the maser action the spin system became "locked" to the resonance frequency of the circuit.[3] Consequently, the pulse duration could be several times longer than the time taken by the field to cross the NMR line.

In contrast to the maser signal, the SR pulse was related

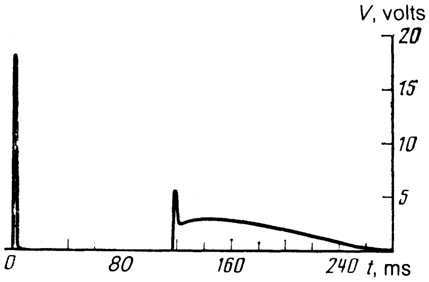

FIG. 2. General appearance of SR and maser effect pulses. Here, V is the amplitude of an hf voltage across the circuit.

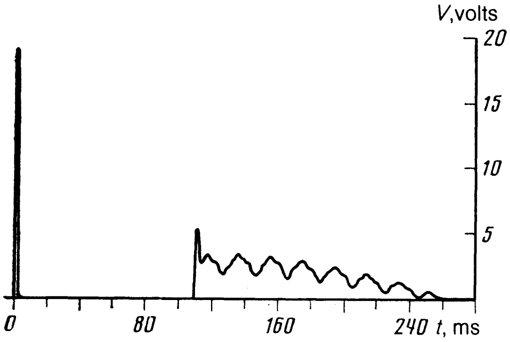

FIG. 3. Same pulses as in Fig. 2, but under conditions of amplitude modulation of the field H_0 at a frequency 50 Hz.

to the time-correlated rotation of the magnetization which reversed the central part of $\chi''(\nu)$. Since during our experiments the field H_0 varied continuously, the maser effect was observed in the unreversed wing of $\chi''(\nu)$ and was delayed in time relative to the SR pulse. Figure 4 shows in extended form the voltage across the coil due to an SR pulse, which in accordance with the theory of Ref. 7 obeyed a dependence of the type

$$V \propto \operatorname{sech}[(t-t_0)/\tau],$$

where t_0 is the delay time and τ is the time representing the width of a pulse. Knowing the fill factor, we could estimate the time constant of radiative damping $\tau_R = (4\pi^2 \times \eta M Q \gamma)^{-1}$, amounting to 1 μs for $|P| = 0.6$.

If the spin-spin relaxation time T_2 was estimated from the width of the Gaussian line profile of protons in propanediol, amounting to 13 Oe, the value of this relaxation time was found to be

$$T_2=(\pi^{-1}\ln 2)^{1/2}/\gamma\Delta H=8.5\ \mu\text{s}.$$

The time constant of the oscillatory circuit was $\tau_c = 2Q/2\pi\nu \approx 9\ \mu$s. In an additional experiment characterized by $Q = 130$ and $\tau_c = 2\ \mu$s, the initial value of $|P|$ at which the maser effect was observed increased inversely proportionally to Q. Nevertheless, neither the condition $\tau_R < T_2 \approx \tau_c$ nor $\tau_c < \tau_R < T_2$ ensured reversal of the polarization in a sample of 0.5 cm^3 volume. Consequently, inhomogeneous broadening of the NMR line appeared at ultralow temperatures. Successful reversal of the polarization in a sample of 12 cm^3 volume was attributed to an increase in the intensity of the radiation emitted by this large-volume sample. The attainment of reversal in practice indicated that it should be possible to generate rf SR in strongly amplifying media.

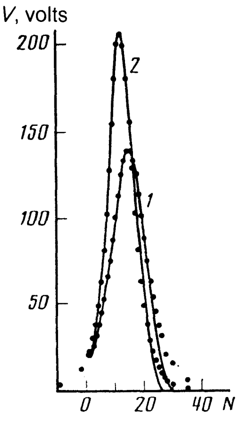

FIG. 4. Experimental (continuous curve) and theoretical (points) profiles of the detected voltage due to an SR pulse in an oscillatory circuit in the case when $|P| = 0.52$ (1) and 0.57 (2); N is the number of analyzer channels (20 μs/channel).

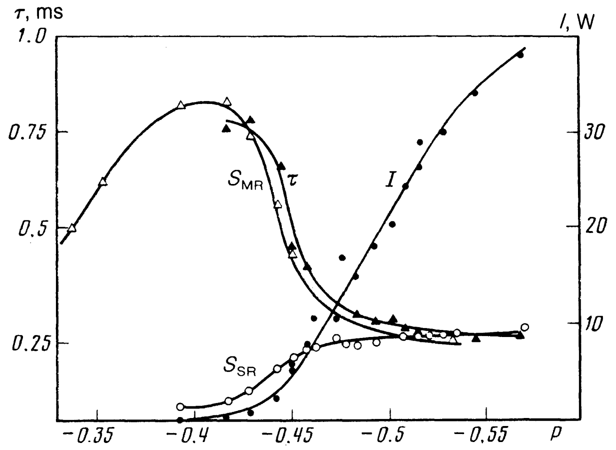

FIG. 5. Areas under the maser radiation and SR pulses (arbitrary units), the maximum intensity I, and the duration of the SR pulse τ plotted as a function of the dependence of the initial polarization of the nuclei.

Figure 5 shows the dependences of the areas under the maser radiation pulse (S_{MR}) and under the SR pulse (S_{SR}), of the maximum intensity I, and of the width of the SR pulse τ on the polarization when the scanning rate was 200 Oe/s. Clearly, the appearance of SR resulted in redistribution of the energy of the conservative spin system in favor of coherent radiation, which reduced strongly the value of τ. In accordance with Eq. (4), the maximum intensity depended quadratically on P during the initial part of the characteristic. On the whole, the dependence was analogous to the case of SR in a weakly amplifying system considered at optical frequencies in Ref. 7. Under similar conditions in the optical case there was no reversal of the populations, which was due to partial cooperation of an effective number of radiators (by analogy with inhomogeneous broadening in the case discussed in the present paper). It should also be pointed out that the width of the SR pulse increased as the rate of scanning of the field was reduced. This dependence was related either to inhomogeneous broadening of $\chi''(\nu)$, which could appear because of the static field of highly polarized electron spins, or due to the dependence of the boundary value of the transverse magnetization at which SR began on the delay time t_0. The observation method, which could not give the value of t_0 prevented us from identifying unambiguously the cause of such behavior.

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