Superfluorescent Transients of an Inhomogenously Broadened Q-Switched Nuclear-Magnetic-Resonance System

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Strongly inverted ^{19}F spins have been used to investigate the transient response of a low-Q CaF_2 NMR laser after Q switching. The experiments are in fair agreement with the numerical results from a mean-field approach to superfluorescence. The model is based on a set of Bloch-type dynamic order-parameter equations for a distribution of spin packets. It takes into account such well known phenomena as spin fanning, hole burning, and spectral diffusion and, therefore, links basic concepts of nonlinear optics with conventional NMR in solids.

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The spontaneous formation of well-organized spatial, temporal, or spatio-temporal structures out of random states is the central theme of the cross-disciplinary field which is called synergetics.[1] Lasers, in particular NMR lasers,[2] have been used over the past few years to demonstrate various macroscopic features of the dynamics of temporal pattern formation by means of cooperative self-organization in nonlinear many-body systems. For example, a laser-type ^{27}Al nuclear spin system of a single ruby crystal has been useful in studying[3] superfluorescence, self-pulsing, hopping-mode patterns, bistability, frequency-doubling bifurcations, paths to chaotic behavior, and first- and second-order–type phase transitions in systems far from equilibrium. A reasonable description of the experimental observations has been obtained from a model based on Bloch-type order-parameter differential equations for a fictitious spin-$\frac{1}{2}$ system in a low-Q resonant structure. The bold assumption of a homogeneously broadened NMR system has been verified surprisingly well, at least for low excitation levels.

To extend these investigations to true spin-$\frac{1}{2}$ systems and to higher excitations, we found that CaF_2, doped with Gd^{3+}, has appropriate physical properties (high spin-$\frac{1}{2}$ density of ^{19}F, high spin inversion, convenient relaxation and pumping times) for laser experiments. It is the purpose of this Letter to report on novel observations with a CaF_2 NMR laser[4] which reflects general features of arbitrary laser-type devices where inhomogeneous broadening is of importance.

The required spin inversion of ^{19}F above the laser threshold is obtained by the method of dynamic nuclear polarization (DNP). At the temperature of liquid He, a single crystal of 1.9 cm^3 CaF_2:Gd^{3+} is irradiated at a selected ESR frequency of Gd^{3+} in a magnetic field of about 1.1 T. The pumping time is $\tau_p = 1200$ s and the spin-lattice relaxation time is $T_1 = 760$ s. The polarization attained is of the order of 10%. The laser resonator with a $Q = 190$ consists of a coil wound on the crystal, an external tuning capacitor, and an electronically controlled tuning device, acting effectively as a Q switch. The filling factor η is approximately 0.1. After triggering the switch, the LC circuit initiates a well-known delayed giant pulse for a duration of typically 50 to 100 μs. After the superfluorescent pulse has died out, the system remains quiescent for a period of several milliseconds. Then the system recovers and puts out a sequence of weaker pulses which evolves towards a state of steady laser activity if DNP pumping is continued.

Three observations are remarkable: firstly, the pronounced structure of the giant pulse (Fig. 1); secondly, the unexpected appearance of follower pulses (Fig. 2) after a recovery time t_{12} of some milliseconds (Fig. 3); and thirdly, the fact that the shape of the overall superfluorescent transient is unaffected by the DNP pump after the Q switch has been activated. The follower pulses are also present when the pump is turned off after triggering the laser. In contrast, the usual Bloch-type laser equations for homogeneous laser systems for the low-Q case predict a hyperbolic secant profile for the giant pulse and a recovery time t_{12} of the order of τ_p, if the pump is kept on.

Several models have been invoked to account for the unforeseen effects. We arrived at the conclusion that the source of the pronounced structural features of the observed transients is the interplay of inhomogeneous and homogeneous spin-spin interactions between Gd^{3+}-^{19}F and ^{19}F-^{19}F spins, respectively. Both interactions contribute to the dephasing of nuclear spins and thus to the dissipation of spin order. There are interactions leading to an irreversible loss of phase memory. They are expressed by the transverse relaxation time T_2. In addition, there exist static line-broadening effects due to Gd^{3+} which cause a reversible fanning of ^{19}F spins which may be reversed, as in conventional echo experiments,[5,6] yielding constructive interference patterns. Operationally, spin fanning may be characterized by a dephasing time T^{2+}. Therefore one may argue that this fanning is the cause of the marked interference pattern within the giant pulse, whereas the recovery time and the follower pulses reflect the response to hole-burning effects[7] during the giant pulse and its healing through spin diffusion[8] during the recovery time.

To substantiate these ideas we have developed a theoretical model which is numerically tractable and which is based on the concept of spin packets P_i. To each packet we assign a local field $B_{0i} = \omega_i/\gamma$ and an occupation number N_i in accordance with an assumed Gaussian distribution $g(\omega_i)$, where ω_i is the NMR frequency of P_i with all packets having the same transverse relaxation T_2, independently of the strength of the laser field. Thus we neglect Redfield-type ordering effects in the rotating frame.[9] We further assume that spectral spin diffusion leads to a cross relaxation among the spin packets which we describe by the time constant T_p, a model-dependent fit parameter. We treat the radiation field in the mean-field (MF) approximation. Therefore it is the MF value of the self-induced field which acts back onto the spins of the different packets.

In the rotating-frame approximation the ith spin-packet nuclear magnetization then follows Bloch-type dynamic equations:

$$\dot{M}_{ui} = M_{vi}\,\Delta\omega_i - \gamma M_{zi} B_v^{\,i} - M_{ui}/T_2\,, \tag{1}$$

$$\dot{M}_{vi} = \gamma M_{zi} B_u^{\,i} - M_{ui}\,\Delta\omega_i - M_{vi}/T_2\,, \tag{2}$$

$$\dot{M}_{zi} = \gamma M_{ui} B_v^{\,i} - \gamma M_{vi} B_u^{\,i} - \frac{1}{T_p}\left(M_{zi} - \frac{c_i}{c_{i+1}} M_{z\,i+1}\right) - \frac{1}{T_p}\left(M_{zi} - \frac{c_i}{c_{i-1}} M_{z\,i-1}\right)\,. \tag{3}$$

The total nuclear magnetization is given by $M_u = \sum_i M_{ui}$, $M_v = \sum_i M_{vi}$, and $M_z = \sum_i M_{zi}$.

Since we are interested in the transient time regime only, we have dropped terms which express the spin-lattice relaxation (T_1) and the DNP pump (τ_p). M_{ui}, M_{vi}, and M_{zi} are the two transverse and the longitudinal components of nuclear magnetization of P_i in the frame rotating with the center frequency ω_0 of $g(\omega)$. We have set $\Delta\omega_i = \omega_i - \omega_0$, $\omega_c = (LC)^{-1/2}$, and $c_i = M_{zi}(t=0)/M_z(t=0)$ which is a measure of the longitudinal Zeeman order of the ith packet relative to $M_z(t=0)$. $B_u^{\,i}$ and $B_v^{\,i}$ are the two orthogonal components of the rotating part of the radiation field in the (u,v,z) frame. For our low-Q case the ringing time is $\tau_R = 1.3\ \mu s$, which is short compared with T_2 and T_p. Hence, the radiation field may be eliminated adiabatically from the order-parameter equations in accordance with

$$B_u^{\,i} = 0.5\mu_0\eta Q\left[D\left(\textstyle\sum_i M_{ui}\right) - E\left(\textstyle\sum_i M_{vi}\right)\right]\,, \tag{4}$$

$$B_v^{\,i} = 0.5\mu_0\eta Q\left[E\left(\textstyle\sum_i M_{ui}\right) + D\left(\textstyle\sum_i M_{vi}\right)\right]\,, \tag{5}$$

$$D = \frac{(\omega_c^{\,2} - \omega_0^{\,2})Q/\omega_0^{\,2}}{1 + (\omega_c^{\,2} - \omega_0^{\,2})^2 Q^2/\omega_0^{\,4}}\,, \tag{6}$$

$$E = \frac{1}{1 + (\omega_c^{\,2} - \omega_0^{\,2})^2 Q^2/\omega_0^{\,4}}\,. \tag{7}$$

The laser output is proportional to the rotating component of the nuclear magnetization and thus to $M_T = (M_u^{\,2} + M_v^{\,2})^{1/2}$.

In a time interval short compared with T_p cross relaxation is ineffective. Thus the giant pulse is affected only by spin fanning which leads to consecutive phases of constructive interfer-

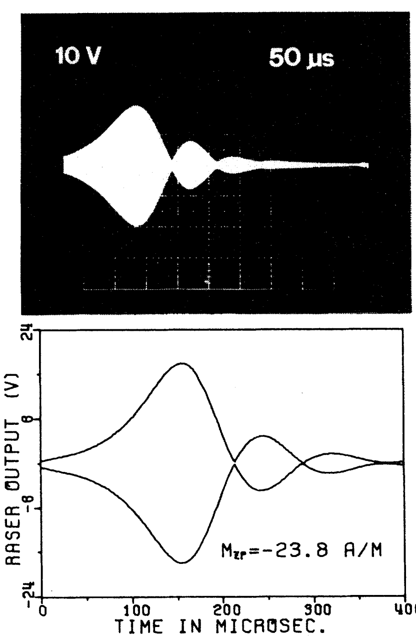

FIG. 1. Measured (above) and numerically computed (below) structure of the superfluorescent pulse for the well-tuned NMR laser. The number of spin packets was set equal to 15.

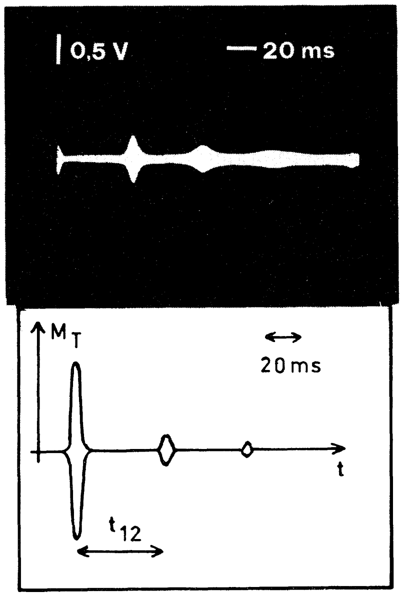

FIG. 2. Experimental (above) and computed (below) behavior of the start and the follower pulses.

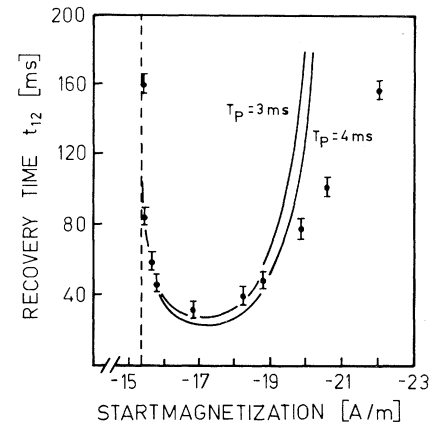

FIG. 3. Experimental and computed recovery time t_{12} between the start and the first follower pulse as functions of the pump magnetization.

ence. Figure 1 shows the numerical solution together with the corresponding experimental pulse for a well-tuned NMR laser, $\omega_c = \omega_0$. Detuning the laser leads to a less pronounced output structure without the zero crossings. For all cases the agreement is gratifying. The numerical solutions $M_{zi}(t)$ show further that the giant pulse ends in a state with a burned hole in the $M_{zi}(t)$ distribution. Superfluorescence therefore leads to self-burning of a hole in the distribution of the longitudinal packet magnetization. Spin diffusion then heals the hole during the recovery time which is a function of its width and depth, and as such, of the initial spin inversion. During the healing process the spin system may be brought above the laser threshold where it becomes ready for a second superfluorescent pulse which is then followed by a third and so on (Fig. 2). Figure 3 compares the experimentally measured recovery time t_{12} between the first and the second pulses and the numerical predictions from Eqs. (1)–(3). For weak hole self-burning, a fit with a cross relaxation T_p of some millseconds is in fair agreement with estimates from spin-diffusion considerations.

We have also performed conventional NMR experiments in order to provide additional support for the proposed model. From free-induction-decay, spin-echo, and external hole-burning measurements, we find an overall reasonable agreement between the theoretical model and the experimental data. Observed deviations might be due to the spatial inhomogeneity of the laser field, the coupling of the laser active bulk spins to the frequency-shifted near neighbors of a paramagnetic impurity ion, or the radiation-field-dependent spin-spin relaxation in the rotating frame. However, estimates show that neither process is strong enough to account quantitatively for the observed facts.

Summarizing, one may say that the phenomenon of spin fanning and self-burning of holes is of rather general nature which should be found also in other superfluorescent systems such as optical lasers and masers. Laser transients offer a new possibility to investigate the spin-packet line shape and the spectral diffusion of inhomogeneously broadened resonances in a single-shot experiment. This method may then be applied to systems, for example, which have recently been investigated in the realm of ESR.[10] The remarkable structure of superfluorescent transients may also be considered as the manifestation of self-organization among slightly different individuals of a nonlinear many-body system and, therefore, such transients are a further illustrative example of synergetic behavior.

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[1]See *Springer Series in Synergetics*, edited by H. Haken (Springer-Verlag, Berlin, 1978–1982), Vols. 1–18.

[2]P. Bösiger, E. Brun, and D. Meier, Phys. Rev. Lett. 38, 602 (1977).

[3]P. Bösiger, E. Brun, and D. Meier, Phys. Rev. A 18, 671 (1978); P. Bösiger *et al.*, Phys. Rev. A 20, 1073 (1979); P. Bösiger *et al.*, in *Dynamics of Synergetic Systems*, edited by H. Haken, Springer Series in Synergetics Vol. 6 (Springer-Verlag, Berlin, 1980), p. 48. P. Bösiger *et al.*, in *Optical Bistability*, edited by C. W. Bowden (Plenum, New York, 1981), p. 173; D. Meier, R. Holzner, B. Derighetti, and E. Brun, in *Evolution of Order and Chaos*, edited by H. Haken, Springer Series in Synergetics Vol. 17 (Springer-Verlag, Berlin, 1982), p. 146.

[4]E. Brun, B. Derighetti, H. Marxer, and F. Waldner, in *Bulletin of Magnetic Resonance*, edited by J. Smith and W. H. Wisman (Franklin Institute Press, Philadelphia, 1980).

[5]E. L. Hahn, Phys. Rev. 80, 580 (1950).

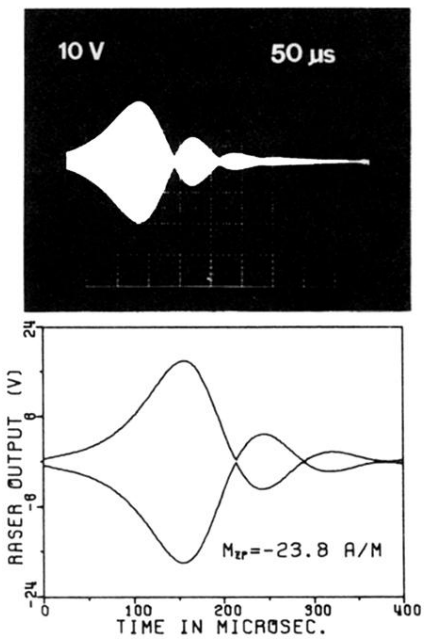
[6]M. Kunitomo and T. Hashi, Phys. Lett. 81A, 299 (1981).

[7]N. Bloembergen, thesis, University of Leiden, 1948 (unpublished).

[8]G. W. Leppelmeier and J. Jeener, Phys. Rev. 175, 489 (1968).

[9]A. G. Redfield, Phys. Rev. 98, 1787 (1955).

[10]R. Boscaino and F. M. Gelardi, J. Magn. Reson. 49, 476 (1982), and J. Phys. C 15, 6245 (1982).



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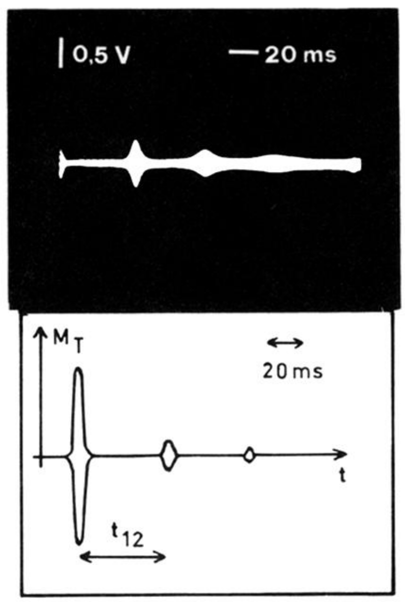

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