susceptibility of a random dilute ferroelectric system. Because of the generalized nature of the treatment, it is expected that the same theory should describe the paramagnetic susceptibility of a random dilute paramagnetic system[7] such as $La_{3-x}Gd_xIn$ (for sufficiently small values of x). The underlying basis of the theory is that the spin correlation between widely separated spin impurities (or the correlation between widely separated electric dipoles in the ferroelectric case) is broken up by randomly positioned impurities between them. There are small strongly correlated clusters of spins which are only weakly coupled to other clusters. Klein[4] has considered in detail only the case where the forces between the impurities are limited to dipole-dipole interactions, for which he obtains a temperature maximum in the susceptibility and the prediction that $T_{\max}$ is proportional to the concentration of impurities, but that the height of the peak is independent of the concentration and the susceptibility is smaller than the free-ion susceptibility at all temperatures.

It is conceivable that the strong concentration dependence of $T_{\max}$ and the gigantic magnetic susceptibilities of the $La_{3-x}Gd_xIn$ system can be brought into agreement with the Marshall-Brout-Klein theory if strong short-range ferromagnetic forces are included in the theory. On the other hand, it may be that the theory is inapplicable to systems with impurity concentrations as high as those reported here.

It is a pleasure to thank M. W. Klein for stimulating discussions.

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*Work supported by the National Science Foundation.

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MULTIPLE SPIN ECHOES AND SPIN LOCKING IN SOLIDS

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(Received 16 May 1966)

It is commonly believed that spin-echo techniques[1] succeed in recovering transverse spin magnetization $\gamma\hbar\langle I_x\rangle$ only to the extent that it has been lost through inhomogeneous broadening. We were, therefore, surprised to observe long trains of echoes in a number of homogeneously broadened solids when pulse sequences timed according to the Carr-Purcell prescription[2] were applied. (An initial 90° pulse is followed after a time τ by a train of pulses of repetition period 2τ.) An example is shown in Fig. 1. The experiments were all performed near room temperature with a pulse spectrometer which provided a rotating field $H_1 \approx 50$ Oe at 30 MHz and recovered from pulse overloads in ~5 μsec.

The behavior of this effect can be described as follows: (1) The phase of the rf carrier in the first pulse must differ by ~90° from that during the rest of the experiment.[3] (2) The effect occurs only when $\tau \lesssim T_2$. The echo en-

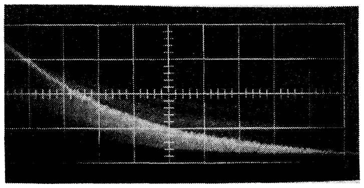

FIG. 1. Train of ^{19}F echoes in powdered $Co(NH_3)_6(BF_4)_3$. Sweep speed: 1 msec/cm. $\tau = 10$ μsec. For this substance $T_2 \approx 45$ μsec.

velope then decays exponentially (after an initial transient) with a time constant $T_2^\dagger$ which becomes longer as τ/T_2 is reduced. (3) The effect is maximized when all the pulses are 90° pulses, but for small τ/T_2 this requirement becomes less strict. (4) A plot of $T_2^\dagger$ vs τ extrapolates for $\tau \to 0$ to $T_{1\rho}$, the longitudinal relaxation time in the rotating frame.[4] (5) The number of echoes obtainable can be very large. In K_2SiF_6 (^{19}F resonance) we observe $T_2^\dagger = 0.12$ sec and $\tau = 10$ μsec, corresponding to a decay of less than 0.02% per echo.

We believe that this phenomenon can be understood from either of two points of view:

(1) For the first few echoes it is convenient to calculate explicitly the development of the density matrix, treating the (90°) pulses as pure rotations and computing the development between pulses by perturbation theory. This procedure is an extension of that used by Powles and Strange,[5] who have observed and explained single 90°-90° echoes in solids. They find at the first echo maximum that the leading term in $\langle I_x\rangle_1 - \langle I_x\rangle_0$ is of order τ^4. Quantitative estimates for our experimental conditions suggest on this basis a loss of 5-10% per echo. We have carried the calculation to the second echo maximum and find that the fourth-order decrement is recovered, and that $\langle I_x\rangle_2 - \langle I_x\rangle_0$ is of order τ^6. We do not know whether a further cancellation occurs on the fourth echo, but it is clear that a multiple-pulse experiment preserves $\langle I_x\rangle$ far longer than would be guessed by extending the results of Powles and Strange through a Stosszahlansatz.

(2) In the quasisteady state after the first few pulses, the pulse train can be regarded as a carrier of amplitude $\bar{H}_1 = H_1(t_w/2\tau)$, where t_w is the pulse width, together with a set of sidebands spaced in angular frequency by π/τ. An examination of the effective spin Hamiltonian shows that as τ/T_2 becomes small, the sidebands are nonsecular and can be ignored. One then has the conditions for "spin locking"[6] in the field $\bar{H}_1$. $\langle I_x\rangle$ decays in a time $T_{1\rho}$. The decay is no longer critically dependent on t_w.

The possibility of "continuous" observation of $\langle I_x\rangle$ during spin locking appears potentially valuable in the detection of double resonance in the rotating frame,[7] in that the resonance condition for the secondary spins can, in principle, be detected (if the relevant cross-relaxation time is sufficiently short) without producing a new spin lock for each trial. The case of intermediate τ/T_2 is also intrinsically interesting as a means for the detailed study of spin interactions. Related phenomena are expected in liquids as well as solids: One must be clearly aware that multiple-pulse experiments are qualitatively different from one- and two-pulse experiments, and each requires a detailed consideration of the relevant spin Hamiltonian.

A detailed account and analysis of these experiments and others in progress will be published elsewhere.

We thank Professor E. L. Hahn for his comments.

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