ens (Ref. 8).

¹⁰W. Webb, Phys. Rev. <u>55</u>, 297 (1939).

¹¹D. F. Simons and M. B. Salamon, Phys. Rev. Lett. 26, 750 (1971).

⁻¹²The resistivity was computed by numerical integration using the values of $(d\rho/dT)/\rho_{25}$ °C (measured on a sample with the same T_c) of Ref. 11 and using the data of Ref. 10 to obtain $\rho(T_c) = 14.31 \ \mu\Omega$ cm and ρ_{25} °C = 4.51 $\mu\Omega$ cm.

¹³Summaries of results are given in M. Hansen, *Constitution of Binary Alloys* (McGraw-Hill, New York, 1958), and in J. E. Ashman, Ph.D. thesis, Univ. of Illinois, 1970 (unpublished).

¹⁴Smithsonian Physical Tables, edited by W. E. Forsythe (Smithsonian Institute, Washington, D. C., 1959); and *Reference Tables for Thermocouples*, edited by H. Shenker *et al.*, U. S. National Bureau of Standards, Circular No. 561(U. S. GPO, Washington, D. C., 1955).

¹⁵Landolt-Börnstein: Zahlenwerte und Functionen, edited by K.-H. Hellwege and A. M. Hellwege (Springer, Berlin, 1959), Vol. II, Part 6. ¹⁶P. D. Mercia and L. W. Schad, Nat. Bur. Stand. (U. S.), Bull. 14, 571 (1917).

¹⁷C. Herring, in *Magnetism*, edited by G. Rado and H. Suhl (Academic, New York, 1966), Vol. IV, p. 129; E. C. Stoner, Proc. Leeds Phil. Soc. <u>2</u>, 149 (1930). ¹⁸It is possible that the similar disparity observed between dQ/dT and C_p in Ni is not an "experimental artifact" as was suggested by Tang, Craig, and Kitch-

¹⁹This noise reflects the smallness of the thermopower anomaly in β -brass, which, within the framework of the present model, is traceable to the smallness of the resistivity anomaly.

²⁰It would seem fair to point out that neutron-scattering experiments have been unable to resolve correlation functions in the immediate vicinity of T_c to better precision than that obtained in the present experiment or from the measurement of only the resistivity anomaly [see, e.g., J. Als-Nielsen, Phys. Rev. <u>185</u>, 664 (1969)].

Boson Echoes: A New Tool to Study Phonon Interactions

J. Joffrin and A. Levelut

Laboratoire d'Ultrasons,* Université Paris VI, 75230 Paris Cedex 05, France (Received 5 September 1972)

The phenomenon of boson echoes, which presents some similarities with the spin-echo phenomenon, is explained in terms of phonons. In particular, the nature of the two relaxation times T_1 and T_2 is explored, and they are ascribed to anharmonic and impurity scattering processes, respectively.

In a recent paper¹ we have reported on preliminary experiments on boson echoes, a phenomenon presenting some analogy with spin echoes: A crystal, placed in a cavity, receives two short pulses of hyperfrequency electric field at times 0 and τ ; at time 2τ , it reradiates a signal (an echo) at the same frequency. Actually, the effect is strongly different from spin echoes in the following respects: (a) The echo intensity is not a periodic function of the power or length of the pulses; (b) it occurs at any frequency without adjusting a biasing field; (c) its width Δt is determined by the widths of the exciting pulses; and (d) only crystals without a center of symmetry are able to produce this effect. These echoes have been explained as boson echoes.¹

In this Letter we report on further experiments with two cavities and with two or three pulses (Fig. 1) which enable us to attribute the echoes to the crystal phonon modes and to obtain some insight on their relaxation times. In particular, we may distinguish between the coherence relaxation time T_2 and the relaxation time T_1 of phonon



FIG. 1. Schematic of an echo experiment. (a) Twopulse sequence; (b) three-pulse sequence. Only echoes at times 2τ and $T + \tau$ are represented, with the relaxation times corresponding to each period. The operators a and a^{\dagger} correspond to dipolar echoes; a^2 and $a^{\dagger 2}$ to quadrupolar echoes.



FIG. 2. Schematic of the experiment. The coherent phonons are generated where there is a gradient of the piezoelectric constant (at the surface which is represented by a thick line). Their evolution is reversed in the region irradiated by the electrical field (crosshatched part of the crystal).

population excited by the pulses. Some consequences of that distinction are emphasized because it seems that this type of experiment may become a new tool for solid-state physics.

The first point to be elucidated is the nature of the bosons. In our previous experiments,¹ made with two pulses and a single cavity, an echo was obtained independent of the interval of time τ between the two pulses. This result does not allow one to decide whether the echoes are due to macroscopic vibration modes of the polarization or to phonons.

We have performed an experiment in the new configuration represented in Fig. 2. It was done at 4.2°K and 9 GHz with a CdS single crystal. The first pulse generates the coherent state in cavity 1; the second pulse, which reverses the time, is applied in cavity 2; the echo is detected in cavity 1. As long as τ is shorter than l/v_L (where l is the crystal length and v_L the sound velocity of longitudinal waves), no echo is observed. When τ approaches this value, the echo grows suddenly and reaches a sharp maximum, just at time 2τ = $2 l/v_L$. For τ larger than l/v_L , the echo is still present and its intensity decreases with a characteristic time roughly equal to the time measured in the previous experiments with only one cavity. For this experiment the sample is depolished so that no hypersonic signal corresponding to a round trip of the first pulse in the crystal is detected at time $2l/v_L$. This proves that the bosons are acoustical phonons.² The so-called time reversal is actually a wave-vector reversal produced by a parametric coupling between a hyperfrequency photon and two phonons with opposite wave vectors and the same frequency. Such an interaction, which is governed by a fifth-rank

tensor, is allowed only in crystals without a center of symmetry.

The second point to be discussed is the problem of the relaxation times. For spin echoes, two different times T_2 and T_1 are defined: The former is connected with the relaxation of the observables S_+ and S_- which do not commute with the Zeeman Hamiltonian, while the latter governs the relaxation of S_Z . Similarly, for phonon ehcoes, T_1 corresponds to the relaxation of the population $N=a^{\dagger}a$, while the nondiagonal quantities such as aand a^{\dagger} (or a^2 and $a^{\dagger 2}$) relax with a time constant T_2 .

The phonons which compose here the active population are determined by the frequency and width of the pulses. Their wave vectors and polarizations depend on the sample shape, the electric-field homogeneity in the cavity, and the crystal coupling tensor.

Experimentally, T_2 is measured by the decrease of the echo intensity obtained from a series of two pulse sequences in which τ is varied [Fig. 1(a)]. Crystal impurities reduce its value because, by scattering, they destroy the phonon population coherence. Time reversal is produced by the second pulse at time τ : The incoming phonons with wave vector q are amplified and, simultaneously, phonons with wave vector -q are generated. The latter travel backwards during the time interval $(\tau, 2\tau)$ along the same path as the incoming phonons traveled during the interval $(0, \tau)$. Only the phonons which have not been scattered by impurities restore the coherence at time 2τ . T_2 is then a measure of the quality of the sample independent of its surface quality and shape and is not very different from the relaxation time deduced from the ultrasonic attenuation experiments at the same frequency which, we know, is also governed by impurities. It is typically ~10 μ sec at 4°K and 9 GHz in all the crystals studied so far.

On the other hand, T_1 is experimentally measured by the decrease of the $(T + \tau)$ -echo intensity in three-pulse sequences when the time interval (τ, T) is changed [Fig. 1(b)]. T_1 is the relaxation time of the quantity associated with the operator $N + \frac{1}{2}$, or, equivalently, with the phonon energy density. The lifetime of this observable is no longer governed by the impurities but only by the inelastic scattering of the excited phonons. This process is very ineffective at low temperature and depends mainly on the anharmonicity of the crystal; $T_1 \approx 0.1 \sec$ for CdS at 9 GHz and 4°K and is in good agreement with standard theoreti-

cal values.³

On the contrary, ultrasonic attenuation experiments at low temperatures never succeed in reaching that relaxation time because of the stronger impurity effect. It is here that the usefulness of the method of boson echoes enhances the possibility of studying inelastic scattering in the low-temperature regime.

In this Letter, some simplifications have been made. In fact, there are two kinds of echoes: (i) Those for which the radiated electromagnetic field is proportional to ϵ^2 (where ϵ is the strain). We call them quadrupolar elastic echoes; detailed calculations show that all the echoes (at times 2τ , 2T, $2T-\tau$, and $T+\tau$) have a quadrupolar contribution. (ii) The others for which the radiated electromagnetic field is proportional to ϵ . We call these dipolar elastic echoes; they give no contribution to the echo at $T+\tau$.

Besides these results, other effects take place at high power levels of the pulses. Saturation of the echoes appears first, and when power is further increased, nonlinear interactions between the phonons themselves generate new echoes whose origin has been explained by Herrmann, Kaplan, and Hill⁴ in the case of magnetoelastic modes.

In conclusion we must emphasize first the simplicity of this method, especially when it is conpared with the ultrasonic method which requires a careful preparation of the samples; however, it depends on the absence of a center of symmetry for the crystal studied. Second, the fact that T_1 may be as large as 0.1 sec at 4°K indicates that the phonon population has a Q factor of the order of 10⁹. Thus the phonon echoes may become a very powerful tool for studying weak inelastic interactions in crystals.

Detailed calculations and more information on the numerical results will be published elsewhere.

*Associated with the Centre National de la Recherche Scientifique.

¹Ch. Frenois, J. Joffrin, A. Levelut, and S. Ziolkiewicz, Solid State Commun. 11, 327 (1972).

²Another argument supporting the idea of phonon echoes is the close analogy between the results of N. N. Krainik, S. N. Popov, and I. E. Mylnikova, [J. Phys. (Paris), Colloq. <u>33</u>, C2-179 (1972)] on the measurement of T_2 at low frequency in a wide temperature range in SbSI, and the curve obtained by E. F. Steigmeier and W. J. Merz [Helv. Phys. Acta <u>41</u>, 1206 (1968)] in the same material for the thermal conductivity, expecially in the critical region.

³P. C. Kwok, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1967), Vol. 20, p. 213.

⁴G. F. Herrmann, D. E. Kaplan, and R. M. Hill, Phys. Rev. 181, 829 (1969).

Giant Electron Spin-Resonance Transmission in Cu Ion Implanted with Mn⁺

P. Monod, H. Hurdequint, A. Janossy,* J. Obert, and J. Chaumont

Laboratoire de Physique des Solides, Institut de Physique Nucléaire.

et Laboratoire de Spectrométrie de Masse, Université de Paris-Sud, 91 Orsay, France (Received 7 August 1972)

We observed for the first time the transmission of electron spin resonance through a thin copper foil implanted with Mn on either one or both sides at a dose of about 10^{15} ion/ cm². We found an increase of intensity over that of the pure metal, at 1.5° K, by a factor of 50 for one-sided-implanted samples and 2500 for two-sided-implanted samples. This enhancement decreases with increasing temperature, while the g factor remains very close to that of pure copper metal.

We wish to present here a set of new observations concerning the transmission of conductionelectron spin resonance (CESR) through a Mnimplanted thin slab of a single crystal of copper. Although we do not at the present time have a satisfactory theoretical or phenomenological model to account for all the features of the observed resonances, we believe that our results are sufficiently striking to be presented as such

without detailed analysis of their microscopic meaning.

Conduction-electron spin resonance in dilute alloys so far has been studied in situations where the concentration of the alloy was homogeneous over the whole sample. In that case it is possible to analyze the results in terms of the same formalism as for a pure metal^{1, 2} by properly rewriting the ESR parameters in terms of impurity