PHONON-PHOTON DOUBLE-QUANTUM TRANSITIONS AS A DETECTOR OF MICROWAVE ULTRASONICS

N. S. Shiren
General Electric Research Laboratory, Schenectady, New York
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We have observed double-quantum transitions, in which a microwave photon and an ultrasonic phonon, at different frequencies, are simultaneously absorbed. Orton, Auzins, and Wertz[1] have previously reported similar observations in which photons at two different frequencies were absorbed in a transition between the $S_z = -1$ and $S_z = +1$ levels of Ni^{2+} in MgO at 77°K. Our observations were made at 2°K, on the spin systems of Ni^{2+} and Fe^{2+} in MgO. We are able to confirm the double-quantum nature of the narrow line observed in the spectrum of the latter,[2] as predicted by Orton et al.[1] Further results on phonon-photon double-quantum transitions, and on the interaction of microwave ultrasonics with the two aforementioned ions, will be reported more fully. However, it is the purpose of this Letter to discuss the use of these transitions in the detection of ultrasonic radiation at microwave frequencies, and to report an experimental demonstration of the feasibility of the method.

Detection of microwave ultrasonics by the concomitant saturation of electron spin resonance transitions has been suggested,[3] and its feasibility has been demonstrated under c.w. conditions.[4] As a detector of microsecond pulses, however, the saturation method fails because of the long spin-lattice relaxation times which are characteristic of paramagnetic impurities in solids.

Pulse detection of microwave ultrasonics is presently effected by the inverse of the piezoelectric effect used for ultrasonic generation.[5,6] Because of the small power conversion ($\sim 10^{-4}$ at X band with a cavity $Q = 10^3$) the sensitivity is low. The principal disadvantage, however, is that the output is not always a measure of acoustic intensity, because it is determined by the acoustic wave amplitude, integrated over the face of the piezoelectric crystal. The detected pulse amplitude is consequently extremely sensitive to any deviation of this face from being a constant-phase surface of the wave. Such effects lead to nonexponential decays in pulse echo experiments, and have been discussed in detail by Jacobsen.[5]

Referring to Fig. 1, the double-quantum detection method consists in the measurement of

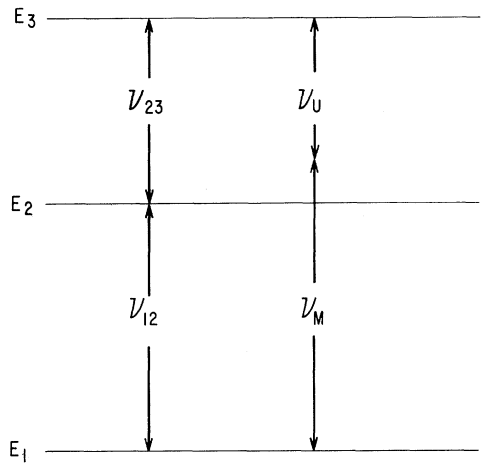

FIG. 1. Energy level scheme showing a possible double-quantum transition for a phonon at frequency ν_U and a photon at ν_M.

microwave power, at frequency ν_M, which is absorbed in double-quantum transitions, at frequency ν_{13}, when ultrasonic power is applied at frequency $\nu_U = \nu_{13} - \nu_M$. Used as a pulse detector, the energy level system is allowed to assume its steady state under the influence of the c.w. microwaves. There is then an increased microwave absorption concurrent with the ultrasonic pulse. The method is capable of detecting very fast pulses because the rise time for absorption is of order T_2, rather than the much larger time, of order T_1, associated with partial saturation at low power levels. The decay time is also $\sim T_2$ since, in the absence of double-quantum transitions, the return of the 1-3 population difference to equilibrium is not observed. Since the double-quantum transition probability is proportional to the product of the two energy densities, the microwave power absorbed is a measure of the acoustic intensity. By using an energy level scheme in which $\nu_{12} > \nu_{23}$, and therefore, for a given transition probability, $\nu_M > \nu_U$, conversion "gain" may be realized, in the sense that the rf power absorbed is greater than the ultrasonic in the ratio ν_M/ν_U.[7]

We have demonstrated double-quantum pulse detection using Fe^{2+} in MgO. As a detector of

ultrasonic energy the ground-state Zeeman transitions of this ion are particularly favorable because of the large spin-phonon coupling. Comparative measurements in the same crystal show that the spin-phonon interaction is $\sim 4\times 10^3$ larger than that for Mn^{2+}, and ~ 30 larger than for Ni^{2+}. The effective spin is[2] $S=1$ and, as in Ni^{2+},[1] the $S_z=\pm 1$ levels are shifted by local departures from cubic symmetry, leading to an inhomogeneously broadened line, about 400 gauss wide (peak to peak derivative), for the $\Delta M=1$ transitions. The $\Delta M=2$ transition remains sharp, however (6 gauss in our crystals). For a fixed value of ν_M it should therefore be possible to find magnetic field values such that equivalent double-quantum transitions will occur over a range $\nu_U=\nu_M\pm 1000$ Mc/sec, and we have observed them with $\nu_U=\nu_M\pm 300$ Mc/sec. Used as a detector, the need for tunable receiving and/or generating circuits is therefore eliminated.

The experimental geometry was similar to that previously reported.[4] A 1.75-cm long x-cut quartz crystal transducer was bonded to an MgO crystal 1.4 cm in length along a cubic axis. Both crystals were 3 mm in diameter. The end faces of the quartz and the bonding face of the MgO were optically polished. The back end of the latter was rough cut. The MgO crystal extended across the short dimension of a full-wave rectangular cavity resonator tuned to 9.2 kMc/sec, and the bonded face of the crystal lay just inside the cavity. The back end extended outside the opposite cavity wall. 0.5-μsec ultrasonic pulses were generated at the opposite end of the quartz by standard techniques.[5] Video detection of the microwave absorption was effected by a pair of MA408A crystals.

The observed double-quantum pulses are shown on the lower trace of Fig. 2, for a 50-watt ultrasonic drive pulse at 9.45 kMc/sec, and 30-mw c.w. microwaves at 9.21 kMc/sec. The upper trace is the piezoelectrically detected output at the generating end of the quartz. The transmitter pulse is observed first, followed by echoes in the quartz. No echoes from the back end of the MgO are seen, due to the rough surface and strong absorption by Fe^{2+} $\Delta M=1$ transitions. The double-quantum pulses appear halfway (measured at the start of the rise) between the echoes in the upper trace, since they are observed at the time of transmission through the bond. Their rise and decay times are determined by the ultrasonic pulse length, i.e., by the time taken for all the energy to be within the

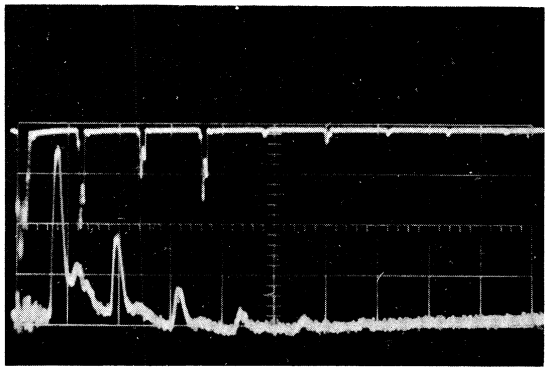

FIG. 2. Lower trace: double-quantum signal after crystal video detection, for ultrasonic frequency 240 Mc/sec higher than the rf. Upper trace: piezoelectric signal after superheterodyne detection. The time between successive echoes is 6.2 μsec. The initial wiggles in the double-quantum output, which appear simultaneously with the direct magnetron pulse in the upper trace, are due to electrical pickup from the pulser. The shape of the piezoelectric echoes is due to double pulsing and frequency modulation of the magnetron.

cavity. The total observed double-quantum pulse width is thus the sum of this rise time and the transmission time through the cavity. The two smaller pulses following each large one are consecutive reflections from the back and front faces of the MgO. They are observed despite the loss of a coherent wave front (by reflection at the rough surface) which partially destroys these echoes in the piezoelectric detector. Also, the nonexponential decay of the echo pattern in the upper trace, which is caused by a similar effect mentioned earlier, is not observed in the double-quantum echo pattern which does decay exponentially. The background level is believed to be an enhanced rf absorption by those spins for which $\nu_M=\nu_{12}$ and $\nu_U=\nu_{23}$, due to partial saturation of the 2-3 $\Delta M=1$ transition by the ultrasonics. Its decay time, which should be $\sim T_1$, is shortened by the amplifier response.

It will be noted that the detection sensitivity is lower than for the piezoelectric output with superheterodyne post detection. However, it can be improved by using higher rf measurement power, better video amplification (noise in the lower trace is all amplifier noise), and particularly by using higher Fe^{2+} concentrations. In our crystals the concentration is $\sim 0.02\%$, and the effective concentration for the narrow double-quantum transition is about 1% of this. The crystal length

should be smaller than the one we used in order to obtain the best ratio of pulse amplitude to total energy absorption. The optimum length is one just long enough to completely contain the ultrasonic pulse so that the output reaches its full amplitude. Results on optimization of the detector, together with a detailed analysis, will be reported at a later date.

The advent of an intensity (rather than integrated amplitude) detector will facilitate experiments which have not heretofore been possible. As an example, it will allow observation of incoherent acoustic energy, important in the study of boundary scattering. Absolute ultrasonic energy measurements are also possible, since the microwave power absorbed may be calibrated if the transition probability and numbers of spins are known.

It is a pleasure to acknowledge discussions on various aspects of this work with Dr. T. G. Castner, Dr. E. H. Jacobsen, Dr. E. B. Tucker, and Dr. G. D. Watkins. T. G. Kazyaka aided in all the experimental work.

[1]J. W. Orton, P. Auzins, and J. E. Wertz, Phys. Rev. Letters 4, 128 (1960).

[2]W. Low, Phys. Rev. 118, 1130 (1960).

[3]C. Kittel, Phys. Rev. Letters 1, 5 (1958).

[4]E. H. Jacobsen, N. S. Shiren, and E. B. Tucker, Phys. Rev. Letters 3, 81 (1959).

[5]E. H. Jacobsen, Quantum Electronics (Columbia University Press, New York, 1960), pp. 468-482.

[6]H. E. Bömmel and W. Dransfeld, Phys. Rev. 117, 1245 (1960).

[7]E. H. Jacobsen has pointed out that the inverse, in a properly chosen energy level scheme such that $\nu_U >> \nu_M$, allows detection of very high frequency phonons by low-frequency rf.

SURFACE STATES ON CLEAVED SILICON*

D. R. Palmer, S. Roy Morrison, and C. E. Dauenbaugh
Honeywell Research Center, Hopkins, Minnesota
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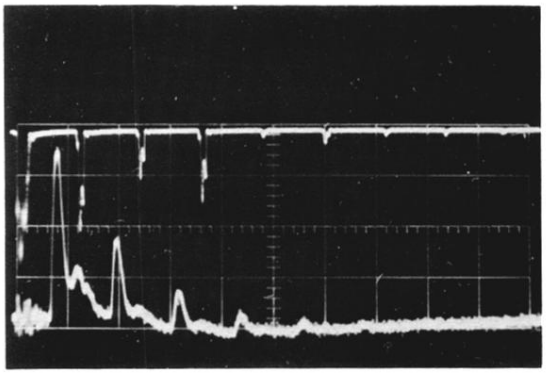
Measurements of surface band structure on silicon (111) surfaces cleaved in vacuum (10^{-7} to 10^{-10} mm) have been made using both the field-effect approach[1,2] and the channel method of Statz and his co-workers.[3]

In the field-effect measurements the portion cleaved off the sample was used as the field plate, and measurements were taken with fields up to 10^4 volts/cm. The field-effect measurements show a slightly n-type surface on both p- and n-type bulk material, of field-effect mobility of about 50 cm^2/v sec. The surface is extremely insensitive to oxygen admission, with up to atmospheric oxygen pressure producing negligible change in field effect, and no sudden changes of conductance with increasing pressure. The latter measurement is unreliable over long periods of time due to temperature drifts. Because of this insensitivity to ambient, the measurement cannot distinguish between a strongly n-type surface with a high density of states or a surface which is almost intrinsic.

To discriminate between the above techniques, the channel method of Statz was employed. Samples were made by diffusing phosphorus into 80-ohm-cm p-type silicon to form *npn* units and by diffusing boron into 20-ohm-cm n-type silicon to form *pnp* units. The "base" width was 0.04 inch. By cleaving so as to leave the junctions intersecting a cleaved surface, we were able to make measurements of the channel conductance, if any, caused by the cleaved surface. In neither the *npn* nor the *pnp* case was the conductivity of the cleaved surface greater than 10^{-2} μmho/square at 1.5 volts bias, indicating there was no channel (inversion layer) present. This can be compared, for example, with a conductance of about 90 μmho/square as measured by the Statz technique on a cleaved surface of 5-ohm-cm n-type germanium (*pnp* configuration).

Thus we conclude that the dominant surface states are near the center of the gap with a freshly cleaved silicon surface, if we assume the surface state structure independent of the bulk type.

This result is in disagreement with work on ion-bombarded surfaces, where Law[4] has found a p-type accumulation layer on p-type silicon after bombardment and annealing and has found that the surface becomes n-type after bombardment with 1000-volt argon ions. He has also stated without reference that earlier work showed



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