ACOUSTIC EXCITATION OF NUCLEAR SPIN RESONANCE IN SINGLE-CRYSTAL METALLIC TANTALUM*

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It is well known[1] that the interaction between the nuclear electric quadrupole moment and the gradient of inhomogeneous crystalline electric fields provides an effective means for line broadening and relaxation effects in the nuclear magnetic resonance spectra of many solids. The relaxation process is one in which thermal vibrations of the lattice cause a modulation of the gradient of the crystalline electric fields, and this modulation induces transitions among the nuclear magnetic energy levels through the quadrupolar interaction.

Kastler[2] and Al'tshuler[3] proposed that one should be able to drive this relaxation process in reverse, i.e., to detect a resonant absorption of coherent acoustic energy from an external source by the nuclear magnetic spin system. This effect was first observed indirectly in the acoustic saturation experiments of Proctor and Tantilla[4,5] on Cl^{35} nuclei in $NaClO_3$, wherein acoustic energy of the proper frequency was introduced into the sample, and its saturation effect on the conventional nuclear magnetic resonance signal was observed. Other investigators have used the acoustic saturation method to study the alkali halides.[6-9]

The first direct observation of the resonant absorption of acoustic energy by nuclear spins was done by Bolef and Menes in indium antimonide[10] and in a series of alkali halides.[11-13] Using the direct method, they observed a resonant increase in the attenuation of a standing sound wave in the sample when the acoustic frequency and the magnetic field were set properly for nuclear spin resonance. This direct method, developed by Bolef and Menes, is very analogous to conventional nuclear spin resonance techniques wherein a resonant absorption of rf electromagnetic radiation is detected.

We have observed, using the method of direct ultrasonic excitation, the Ta^{181} nuclear spin resonance in single-crystalline metallic tantalum.[14] For work in bulk single-crystal metals, the advantages of the acoustic excitation over conventional nuclear resonance techniques are obvious since one is not hampered by electrical skin-depth problems. The sound waves, as opposed to rf electromagnetic radiation, are capable of penetrating into the interior of the metal and interact with the bulk of the nuclei. The interaction between the externally generated acoustic wave and the nuclei is via the torque exerted on the nuclear electric quadrupole moment by inhomogeneous electric fields created by the passage of the wave. The direct ultrasonic excitation method of studying nuclear spin resonance in single-crystal metals should be extremely useful in many materials whose constituent nuclei possess reasonably sized quadrupole moments.

The quadrupolar selection rules permit the acoustic excitation of both $\Delta m = \pm 1$ and $\Delta m = \pm 2$ transitions among the nuclear magnetic energy levels, where m is the magnetic quantum number of the nucleus. An energy-level diagram of the nuclear ground-state multiplet of $Ta^{181}(I = \frac{7}{2})$, with the allowed acoustically induced transitions indicated, is shown in Fig. 1. These transitions may be induced by the pas-

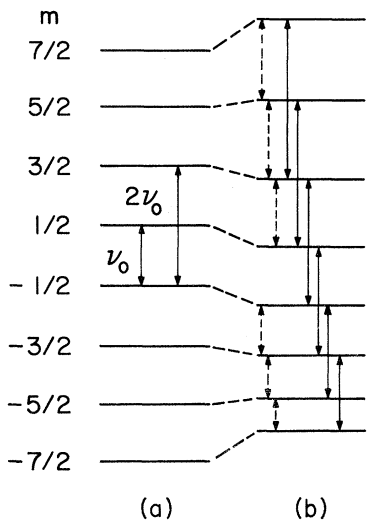

FIG. 1. High-field energy levels for nuclear spin $I = \frac{7}{2}$. (a) Linear Zeeman effect. No static quadrupole interaction. This situation persists for nuclei occupying sites with cubic symmetry as in perfect tantalum. (b) Effect of static quadrupole interaction to first order, e.g., for nuclei in the vicinity of lattice strains. ν_0 is the Larmor frequency. The dotted lines indicate the allowed $\Delta m = \pm 1$ transitions and the heavy lines the $\Delta m = \pm 2$ transitions.

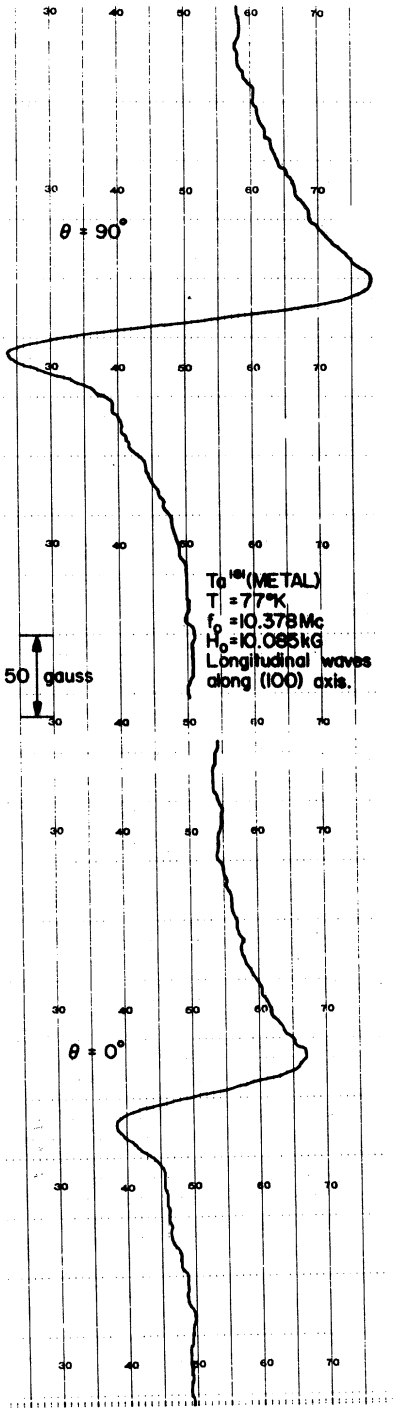

FIG. 2. Typical $\Delta m = 2$ acoustically excited nuclear spin resonances. θ is the angle between the magnetic field and the direction in the sample along which the sound wave is set up. f_0 and H_0 are the acoustic frequency and magnetic field at resonance, respectively. f_0 is equal to ν_0 for $\Delta m = 1$ transitions and to $2\nu_0$ for $\Delta m = 2$ transitions. Signal-to-noise ratios as high as 70 have been observed. The particular curve is for Ta^{181} (metal), $T = 77°K$, $f_0 = 10.378$ Mc/sec, $H_0 = 10.085$ kG, with longitudinal waves along the (100) axis.

sage of either longitudinal or transverse acoustic waves through the sample.

In our experiments both longitudinal and transverse waves were successfully employed to detect the nuclear spin resonance. The samples were in the form of cylinders about 0.5 cm in diameter and 1.0 cm long, the cylinder axis being accurately oriented along the desired crystalline direction using x rays. The end faces of the cylindrical samples were polished optically flat and parallel to facilitate the generation of either longitudinal or transverse standing sound waves using quartz piezoelectric transducers. Experiments were done on samples oriented along both the [100] and [111] crystalline axes. Nuclear spin resonances were detected where $\Delta m = 1$ and $\Delta m = 2$. Figure 2 shows two typical $\Delta m = 2$ resonances excited and detected using a longitudinal wave along the [100] crystalline axis. The traces are the derivitives of the actual absorption lines due to the use of 37-cps magnetic-field modulation and synchronous detection at the modulation frequency. The acoustic frequency was kept constant while the magnetic field was swept slowly through the resonance. The intensity of the absorption line should depend strongly on θ, the angle between the magnetic field and the direction along which the standing sound wave is set up. For longitudinal waves set up along the [100] axis of a cubic material, the intensity of the $\Delta m = 2$ absorption line should vary as $\sin^4\theta$.[15] Another factor which enters into the expression for the intensity is the line-shape factor $g(\nu)$ which may also be dependent on θ. The angular dependence of $g(\nu)$ arises, in some cases,[16] from the preferential orientation of dislocations along certain crystalline axes. The angular dependence of $g(\nu)$ will then detract from the strict $\sin^4\theta$ dependence mentioned above. However, in our experiments we observe, for both longitudinal and transverse waves, a departure from the expected angular dependence which cannot be blamed on an anisotropic $g(\nu)$. The reason for this disagreement is thought to be predominantly due to acoustic-mode conversion[17] in the sample arising because of the relatively small ratio of sample diameter to sound-wave length. For example, in the case of a longitudinal standing wave set up along the axis of the cylindrical sample there can be, in addition to axial motion, a considerable amount of radial motion. Support for this conclusion is the fact that when care was taken to polish the transducer electrode

optically flat, a more reasonable dependence of the absorption intensity on θ was observed, as compared to the almost complete isotropy previously observed. If the transducer electrode was not flat, this would be expected to contribute to acoustic-mode conversion. Further evidence is the fact that the observed angular dependence of the absorption intensity for both longitudinal- and transverse-wave excitation can be understood if one assumes a mixture of modes present. One way to diminish the mode mixing is to use larger diameter samples and operate at higher acoustic frequencies. In experiments done on I^{127} in KI by us and others,[12] using more favorable sample geometry, the deviation from the expected angular dependence can be explained in terms of an anisotropic $g(\nu)$. Experiments using larger diameter tantalum samples, as well as experiments on other metals, temperature dependence of the resonance, effects of sample purity, etc., are in progress. A more detailed experimental and theoretical description of this work will be published later.

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EFFECT OF OPEN ORBITS ON HELICON AND ALFVÉN-WAVE PROPAGATION IN SOLID-STATE PLASMAS

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Since the original suggestion of Konstantinov and Perel[1] and Aigrain[2] that certain low-frequency electromagnetic (helicon) waves can be propagated with little attenuation in noncompensated solid-state plasmas, and of Buchsbaum and Galt[3] that magnetohydrodynamic waves can propagate in compensated solids, there has been great activity in this area by both experimentalists and theorists.[4] The work to date has dealt with materials under conditions such that only closed cyclotron orbits were present. There is then a close analogy between the characteristics of helicon and Alfvén waves in solids, and the corresponding waves in gaseous plasmas. In this Letter we discuss a phenomenon which is peculiar to solids, namely, the effect of open orbits on helicon and Alfvén-wave propagation. We will show that the presence of a single band of open orbits causes strong damping of the helicon wave in an uncompensated material and, in extreme circumstances, can convert the helicon wave into a damped Alfvén wave. This effect has been seen by Grimes.[5] The additional damping is easily observable since it is a strong function of magnetic-field orientation. It falls rapidly to zero as the field is turned away from directions which give rise to open orbits. In uncompensated metals, the