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Nuclear Acoustic Resonance of Liquid He^3 on a Metal Surface

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The nuclear acoustic resonance of He^3 spins has been excited and detected via Rayleigh waves on a metal surface. The signal is many orders of magnitude larger than the nuclear acoustic resonance signal detected in the bulk of solids. Its magnitude is very sensitive to surface damage. The effect has been observed with liquid He^3 in contact with gallium, aluminum, and gold.

Nuclear acoustic resonance (NAR) has been observed in solids. The resonance is excited either via the quadrupolar coupling[1] or via the dipolar one.[2,3] To my knowledge, NAR has not been observed in liquids. In this paper, I report the observation of the NAR of liquid He^3 in contact with a metal surface. The NAR is excited by forcing the solid surface to vibrate during a short time at a frequency and magnetic field which correspond to the resonance condition for the He^3 spins ($\Delta m = 1$ transition). The detection is made by recording the free decay of the surface vibration following the acoustic pulse. The experiment is similar to Hahn's experiment with radiofrequency waves.[4]

The observation conditions of this new effect are as follows: The sample is a metallic single crystal which has two faces flat and parallel. The area of these faces is about 1 cm^2 and the distance between them is 1 cm. The sample is in a liquid He^3 bath and has an X-cut quartz transducer bonded to one of its flat faces. The transducer diameter is 0.5 cm. An rf pulse of 1 μsec duration is applied to the transducer which converts it into a longitudinal acoustic wave train which propagates in the bulk and on the sample surface.[5] In the bulk, the wave is reflected successively on the two parallel sample faces and gives rise to acoustic echoes on the quartz transducer which are converted into rf pulses, then amplified and demodulated.

On the other hand, a Rayleigh wave propagates on the sample surface. Nevertheless, this wave does not return toward the transducer because the wave front (that one can consider circular in the simplest case of an isotropic medium) does not reach the different points of the sample edge (which is rectangular) at the same time. It is important to note at this stage that, except for the special case considered later, the vibrations of the quartz transducer are damped as soon as the rf pulse is removed (i.e., less than 0.5 μsec). As a result, the quartz vibrates only when an echo from the bulk reaches the sample surface. When the magnetic field is set up (normal to the sample faces in the present experiments), the echo amplitude varies because of all the magnetoacoustic effects. Nevertheless, one does not observe any change in the damping of the quartz transducer.

When the NAR condition is fulfilled for the He^3 spins (i.e., $\omega = \gamma H$, where ω is the acoustic angular frequency, γ the gyromagnetic factor, and H the magnetic field), one observes on the quartz transducer an acoustic free decay (AFD) following the acoustic pulse (i.e., the quartz does not stop vibrating as soon as the acoustic pulse is removed). The AFD is exponential (Fig. 1) and is quite similar to the nuclear induction signal induced in a coil following an rf pulse.[4] Furthermore, the AFD interfers with the echoes from the bulk. So, when the magnetic field is swept around the NAR, beats are observed.

Figure 2 shows the beat patterns obtained for the successive echoes. From the amplitude variation of the first echo (Fig. 2), one can see that the effect is very large. It gives an apparent attenuation variation of the order of 3×10^{-1} cm^{-1}. This value is to be compared with the usual attenuation vibration due to the NAR in the bulk which is of the order of 10^{-6} cm^{-1}.[1,3] Figure 2 shows also that the period δH_n of the beat pattern

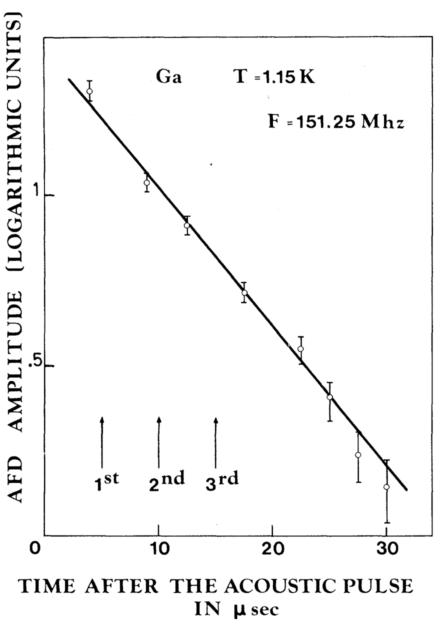

FIG. 1. Amplitude of the AFD at the resonance as a function of the time after the acoustic pulse. The time duration of the pulse is 1 μsec. The arrows indicate the arrival times of the first three echoes from the bulk.

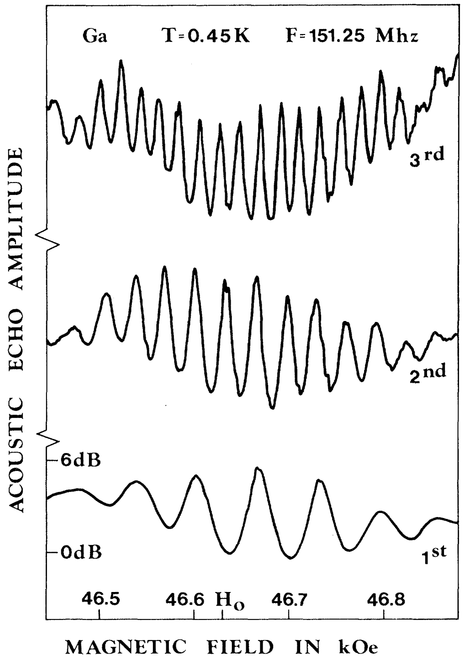

FIG. 2. Amplitude variation of the successive echoes when the magnetic field is swept around the NAR. H_0 $=46.63$ kOe is the value corresponding to the resonance of the He^3 spins for $F=151.25$ MHz.

for the nth echo is of the form $\delta H_1/n$, where δH_1 is the period for the first echo.

One can explain this experimental result and calculate the value of the period δH_n by supposing that the frequency of the AFD is the Larmor frequency of the He^3 spins (as in the case of the nuclear induction signal in a coil, following an rf pulse). The superposition of the AFD and the nth echo on the transducer can be written

$$A = A_1 \cos(\omega_0 t) + A_2[Y(t - 2nL/V_b) - Y(t - \tau - 2nL/V_b)]\cos[\omega(t - 2nL/V_b)], \tag{1}$$

where $\omega_0 = \gamma H$ is the angular Larmor frequency of the spins, ω the angular frequency of the acoustic pulse, Y the Heaviside function, τ the pulse duration ($\simeq 1$ μsec in this case), L the thickness of the sample ($=1$ cm), and V_b the sound velocity in the metal. In the time interval $2nL/V_b \leq t \leq 2nL/V_b + \tau$ (which is the arrival time of the nth echo on the transducer) Eq. (1) can be put in the form

$$A = (A_2 - A_1)\cos[\omega(t - 2nL/V_b)] + 2A_1 \cos[\tfrac{1}{2}(\omega_0 - \omega)t + nL/V_b]\cos[\tfrac{1}{2}(\omega_0 + \omega)t - nL/V_b]. \tag{2}$$

For $A_1 \ll A_2$, the first term is the unperturbed echo. The second term is an oscillating signal, the frequency of which is $\frac{1}{2}(\omega_0 + \omega) \cong \omega$. Its amplitude $2A_1 \cos[\frac{1}{2}(\omega_0 - \omega)t + nL/V_b]$ depends on time and magnetic field (via ω_0). The time dependence is negligible during the time interval $2nL/V_b \leq t \leq 2nL/V_b + \tau$ since the condition $(\omega_0 - \omega)\tau \ll 2\pi$ is experimentally satisfied when the magnetic field value is near the resonance one. Hence, the amplitude of the second term of Eq. (2) can be written

$$2A_1 \cos[(\gamma H - \omega)nL/V_b + nL/V_b]. \tag{3}$$

This expression depends on the magnetic field and the echo number. A simple calculation gives for the modulation period

$$\delta H_n = \pi V_b/\gamma L n. \tag{4}$$

In the case of gallium, one obtains from Eq. (4) a value $\delta H_1 = 63.2$ Oe (the sample is a single crystal having its faces normal to the $\vec{b}$ axis and the sound velocity in this direction is 4.1×10^5 cm sec^{-1}). The experimental value is $\delta H_1 = 63$ ± 6 Oe (Fig. 2) and is in good agreement with the calculated one.

I have looked for the experimental conditions required for the observation of the AFD. At first, the sample in the He^3 bath must be a metal. To prove this assertion, I have done an experiment with a nondegenerate semiconductor (germanium doped with 10^{17} indium/cm^3) and have found that the AFD does not exist.[6] I have recorded the effect in gallium, aluminum, and gold. The experiment with the gold was done to prove that there is no fundamental connection between the AFD effect and the existence of an oxide layer on the metal surface. The magnitude of the effect is very sensitive to the quality of the metal surface which holds the quartz transducer. When this surface is damaged (defect size $\gtrsim$ acoustic wavelength), the AFD is strongly reduced. Furthermore, the AFD is 1 order of magnitude larger in gallium than in aluminum and gold. This fact can be explained by taking into account the preparation difference between the samples.

For aluminum[7] and gold, the single crystals were spark cut, mechanically polished, and then annealed. Unlike these, the gallium was crystallized in a mold which gave directly a flat surface.[8] Thus the gallium surface was certainly less strained than the aluminum and gold ones.

I have also checked that there must be liquid He^3 on the sample face which holds the transducer. To do this, I have pumped on the He^3 bath (at constant temperature) and observed the disappearance of the AFD as soon as the liquid level was below the metal face. The frequency and temperature ranges of the effect are as follows: The ultrasonic frequency must be $\gtrsim 100$ MHz and the temperature $\lesssim 1.7$ K in order that the AFD be observed.[6]

One can explain qualitatively the AFD effect by taking into account the free Larmor precession of the He^3 spins and the magnetic field associated with the acoustic surface wave.

Following the acoustic pulse, a Rayleigh wave propagates on the metal surface. The wave produces a lattice distortion and the electric field associated with this strain is screened by the conduction electrons. Thus an electronic current and a magnetic field are created. The magnetic field induces transitions in the spin system in contact with the metal. Quinn and Ying[2] considered a similar spin-phonon coupling in the bulk to deal with the NAR in metals.

Consider a surface element dS in contact with the liquid. When it is irradiated with an acoustic pulse near the resonance, the phonons are absorbed by the nuclear magnetic moments in the vicinity of dS. Following the pulse these moments return part of the absorbed acoustic energy to dS during their free Larmor precession. At this stage, there is a complete analogy between the AFD effect and Hahn's experiment.[4] However, in the case presented here, it is necessary to deal with the propagation of the acoustic wave, since the wavelength is much smaller than the characteristic lengths of the sample surface.

The phonons received by dS from the nuclear moments can propagate in any direction. Thus a wave of the form $e^{-t/T}\cos[\omega(t-2r/V_R)]$ reaches the transducer (T is the characteristic relaxation time of the effect, V_R the surface wave velocity, and r the distance between dS and the origin). Integrating over all the free surface, one can predict on the transducer an AFD of the form $e^{-t/T}\cos(\omega t+\varphi)$. In the case of Fig. 1, T is found to be 11 μsec. This value is much smaller than the spin-spin relaxation time T_2 in liquid He^3 which is about 3 sec.[9] It is not surprising to find T much smaller than T_2 since in a quantitative theory of the AFD effect it would be necessary to take into account the effect of the metal surface upon the relaxation processes and the spin diffusion in liquid He^3. Lastly, from papers on nuclear magnetic resonance in adsorbed films of He^3,[10] it appears that a decay time of 11 μsec is not unusual near a surface.

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[6] The observation is in fact limited by the sensitivity of the apparatus which can detect on the acoustic echoes an amplitude variation $\gtrsim 10^{-2}$.

[7] The author thanks Dr. G. Revel of Centre d'Etude de Chimie Metallurgique de Vitry for supplying a high-purity aluminum single crystal.

[8] The author is indebted to P. de la Breteque of Alusuisse for the supply of pure gallium, and thanks C. Alquie (Ecole de Physique et Chimie, Paris) who showed

him the technique for making oriented gallium crystals in a mold.

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Short-Wavelength Collective Excitations in Liquid Rubidium Observed by Coherent Neutron Scattering[*]

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The short-wavelength collective excitations in liquid rubidium at 320 K have been studied by coherent neutron scattering. For values of $\kappa = 2\pi/\lambda$ up to $1.0\ \mathrm{\AA}^{-1}$, clear evidence of propagating modes was found from the shape of the scattering function $S(\kappa,\omega)$ at constant values of κ. This result shows that the existence of such modes does not depend upon either quantum effects or low thermal population of the modes.

The existence of propagating collective modes of finite wavelengths in simple liquids is a subject of intense, continuing interest. Recent neutron-scattering studies[1] of liquid parahydrogen near its freezing point established the existence of such modes for values of $\kappa = 2\pi/\lambda$ as large as $3.1\ \mathrm{\AA}^{-1}$, and suggested the existence of transverse excitations. Since the first peak in the structure factor $S(\kappa)$ occurs at $\kappa = 2.0\ \mathrm{\AA}^{-1}$, this implies that the wavelengths of these modes are smaller than typical interatomic spacings. Earlier measurements[2] on liquid lead near the freezing point were interpreted as evidence for the existence of both longitudinal and transverse modes with wavelengths comparable to two interatomic spacings. Recent molecular-dynamics studies[3] of a Lennard-Jones fluid with parameters chosen to simulate liquid argon show evidence of such excitations, but only for κ's smaller than $0.3\ \mathrm{\AA}^{-1}$, implying wavelengths greater than 5–7 interatomic spacings. Neutron-scattering studies[4] on liquid argon at 85.2 K show no evidence of peaks for values of $\kappa \geq 1.0\ \mathrm{\AA}^{-1}$. In this Letter we present neutron-scattering results for liquid Rb at 320 K which show evidence of "longitudinal" modes for $\kappa \leq 1.0\ \mathrm{\AA}^{-1}$, but no evidence of "transverse" modes.

The lattice dynamics of Rb have been extensively studied,[5] and the results have been used to derive a volume-dependent effective two-body potential.[6] Molecular-dynamics calculations[7] with this potential have been compared in detail to neutron-scattering results[8] in the range $1.25 < \kappa < 5.50\ \mathrm{\AA}^{-1}$ and good agreement found. Further computations[9] at wave vectors $\kappa < 1.0\ \mathrm{\AA}^{-1}$ predicted the existence of well-defined propagating modes in the spectrum of density fluctuations. Since the ratio of coherent[10] to incoherent[11] neutron-scattering cross sections is high ($\sigma_{\mathrm{coh}}/_{\mathrm{inc}} \geq 1800$), liquid rubidium is an excellent choice for experimental study.

The experiments were performed at the thermal-neutron time-of-flight spectrometer[12] at the Argonne National Laboratory CP-5 reactor. The incident energy was 33.0 meV, with an overall energy resolution of approximately 1.0 meV (full width at half-maximum). As has been discussed in Ref. 2, large incident energies are required in order to make measurements at small momentum transfers over a useful range of energy transfers. Using the computer code MSCAT,[13] the sample container was designed to maximize the ratio of first scattering to multiple scattering, while maintaining a large first-scattering probability. The final sample container consisted of 32 slabs of liquid, each 2.54 cm wide by 0.22 cm high by 10.16 cm long, separated from each other by horizontal boron-nitride spacers 0.10 cm thick. Even with this large a sample ($182\ \mathrm{cm}^3$ of Rb) 32 days of running time were required. The alumi-