Acoustic Properties of Solid ^{3}He at Low Temperatures

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We have measured the velocity and attenuation of longitudinal sound at 10 MHz in solid ^{3}He as a function of temperature from the melting temperature to as low as 12 mK. The measurements were made at molar volumes between 23.2 and 24.6 cm^3 and at two different concentrations of ^{4}He impurities. In the sample containing less than 5 ppm of ^{4}He the velocity varies as $1/T$ at low temperatures and the attenuation varies exponentially with temperature. The sample containing 158 ppm ^{4}He reveals very different behavior.

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The properties of solid ^{3}He have been of special interest because it is the most "quantum" solid in nature. The influence of the quantum exchange has been measured on NMR relaxation, magnetic susceptibility, pressure at constant volume, and specific heat. At low temperatures the exchange energy should also be expected to dominate the compressibility and therefore the sound velocity.[1,2] We have observed the temperature dependence associated with this effect for the first time. We have also found that the effect is masked by other phenomena in a sample containing 158 ppm of ^{4}He.

Another manifestation of the quantum nature of this solid is the presence of a variety of unique excitations such as vacancy and impurity waves. These could attenuate acoustic waves. At high temperatures vacancies are thermally activated with an activation energy between 2 and 10 K, depending on the molar volume.[3,4] However, in this temperature range the acoustic attenuation is dominated by dislocations. We have found that the attenuation, below about 300 mK, varies exponentially with $1/T$ but the activation energy is an order of magnitude smaller than that expected for vacancies. Thus some other excitations appear to be responsible.

A third phenomenon which could be expected to influence sound propagation is the isotopic phase separation of mixtures of ^{4}He in ^{3}He.[5] Previous work has suggested that the phase separation alters the acoustic properties of the crystal by creating dislocations.[2] We have observed this phenomenon at lower temperatures than previous workers and have found that the phase separation takes place gradually as a function of temperature. Therefore it does not appear to proceed by nucleation. It is this phase separation and its generation of dislocations which masks the influence of the exchange on the sound velocity in impure crystals.

The ^{3}He crystals for this experiment were grown at constant pressure from a chemically polished cold finger at rates between 0.2 and 0.4 mm/min. They were then annealed within 50 mK of the melting temperature for as long as 20 h. Even with the best crystals the stability and reproducibility of the acoustic data could be degraded by rapid changes of temperature, excessive mechanical vibrations, or sudden changes of pressure in the room (opening and closing of the door). Temperature was measured using cerium magnesium nitrate and the calibration was checked on each cooldown against three superconducting fixed-point thermometers at 0.273, 0.137, and 0.096 K. A transmitter and a receiver transducer separated by 4.27 mm were used to measure changes of velocity and amplitude of acoustic pulses transmitted through the crystals. The pulses were always well below the amplitudes at which other workers have observed amplitude-dependent behavior.[6]

The temperature dependence of the amplitude, $A(T)$, of the transmitted sound in the pure samples (<5 ppm ^{4}He) is shown in Fig. 1 for a crystal at volume 24.6 cm^3/mole. It shows that $\ln\{[A(0)-A(T)]/A(0)\}=B-\phi/T$. In terms of an attenuation coefficient, $\beta=\beta_0+\beta(T)$, consisting of a temperature-independent part, β_0, and a temperature-dependent part, $\beta(T)$, which vanishes at $T=0$, the amplitude at distance L from the transmitter will be $A(T)=A_0\exp(-\beta L)=A(0)\exp[-\beta(T)L]$ and $\ln[A(0)/A(T)]=\beta(T)L$. Observed values of $A(0)/A(T)$ are always between 1.08 and 1.00 so that

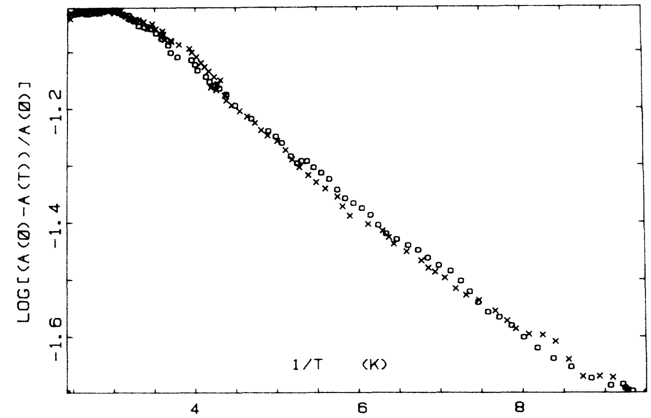

FIG. 1. $\log_{10}\{[A(0)-A(T)]/A(0)\}$ as a function of $1/T$. $A(0)$ is the low-temperature limiting value of the amplitude and $A(T)$ is the amplitude at temperature T. Crosses are data during cooldown and circles are data during warmup. The volume is 24.6 cm^3/mole.

$\ln[A(0)/A(T)] \sim A(0)/A(T) - 1$. Thus

$$\ln\ln[A(0)/A(T)] \sim \ln\{[A(0)-A(T)]/A(T)\} = \ln L + \ln\beta(T)$$

and our results require that $\beta(T) = \exp(-\phi/T)$. From this we conclude that the attenuation involves thermally activated excitations and we find an activation energy $\phi \sim 0.25$ K.

The velocity in the pure samples increases with decreasing temperature at high temperatures, reaches a maximum at about 100 mK, and then decreases as $[c_0 - c(T)]/c_0 = \alpha/T$. This is shown in Fig. 2 for a crystal grown at a volume of 24.1 cm^3/mole. The melting curve, below its minimum, does not reach this volume so that data in the solid can be obtained to the lowest obtainable temperatures. For this reason the data shown extend over a wider temperature range than for any of the other crystals. It also reveals another phenomenon which could obscure the $1/T$ dependence. The measurements shown were made over a longer period of time (4 days) and subjected to more mechanical disturbances than other crystals. In addition, the temperature was not varied monotonically in time. As a consequence the amplitude was not constant at low temperatures and there was some correlation between these changes in amplitude and the velocity. What appear to be hysteresis loops near the center of the figure reflect corresponding loops in the amplitude. Their sizes as they appear in the plot have been reduced by about a factor of 2 from the raw data by removal of the amplitude dependence. To do so, the amplitude was fitted to the velocity for portions of the data where the velocity and amplitude changed proportionately as a function of time while the temperature remained almost constant. The amplitude time series was then multiplied by the factor determined from the fit and subtracted from the velocity time series for the entire run. The only effect of this subtraction was to reduce the size of the loops. The physical mechanism for the proportional change of amplitude and velocity is probably the generation of crystal defects by thermal or mechanical stresses. This proportional variation of velocity and amplitude was observed consistently in impure crystals as described below.

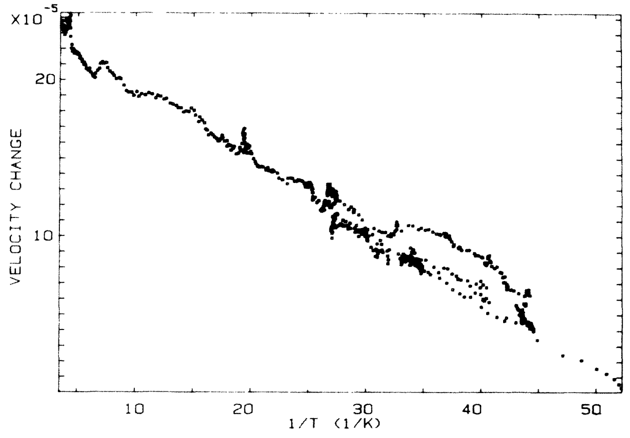

FIG. 2. Fractional change in velocity vs $1/T$ for a crystal at volume 24.1 cm^3/mole. The absolute value of the velocity was taken to be 500 m/sec for the purposes of this plot. Its actual value was known only to within about ± 10%. The temperature was not varied monotonically with time for these data.

A total of 27 pure crystals with molar volumes between 23.9 and 24.7 cm^3 were grown and useful measurements were obtained on eight of these. The values of ϕ ranged between 0.2 and 0.3 K. The values of α ranged between 0.4×10^{-5} and 0.7×10^{-5} K. Neither quantity showed a systematic dependence on volume within these limits. The variation of both was apparently due to variations in the nature and density of crystal defects, crystal axis orientation, or some undetermined parameters.

Figure 3 shows the most commonly observed behavior of the velocity and attenuation during cooldown of the crystals containing 158 ppm ^{4}He. Velocity and amplitude decrease proportionately as a function of temperature below a critical temperature which ranged between 50 and 30 mK for the samples studied. The variation takes place within a temperature range of between 10 and 20 mK. At lower temperatures both variables become independent of temperature. The ratio of the fractional change in velocity to the change of the amplitude can vary by more than a factor of 2 between thermal cycles of a given crystal. Curves of the type shown were obtained for a range of cooling rates between 0.05 and 0.25 mK/min and are therefore assumed to be equilibrium curves. As in the pure sample, if the crystal is disturbed mechanically or cooled too rapidly, irregular behavior is observed and

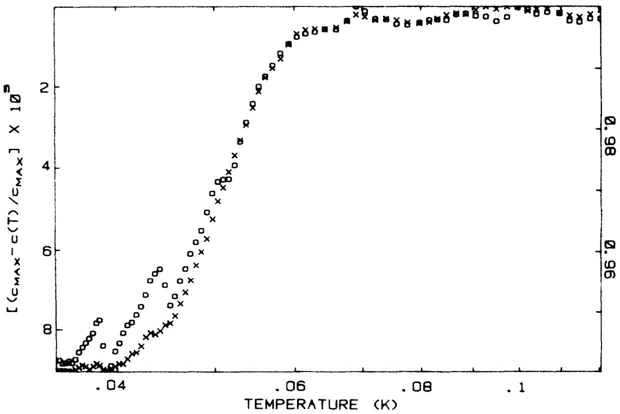

FIG. 3. Fractional change in velocity and amplitude during cooldown of a crystal with 158 ppm of ^{4}He. The volume is 24.2 cm^3/mole.

velocity and amplitude do not vary proportionally. No systematic dependence on density or sound pulse amplitude is observed over the measured range of molar volumes (23.2 to 24.2 cm^3). Upon warming, the samples displayed a complicated temperature hysteresis involving nonequilibrium states with relaxation times of many hours.

The behavior of the impure samples can be explained in terms of a relatively complicated model involving thermal relaxation of screw dislocations and the redissolution of ^{4}He impurities. Previous experiments have observed a variety of phenomena in the range of concentrations around our value but usually at somewhat higher temperatures.[7,8] The workers have all interpreted their results to be a consequence of the combined effects of phase separation and dislocations using the theory of Granato and Lücke.[9] In our case the phase separation takes place over a finite temperature range (as in the original observations of Edwards, McWilliams, and Daunt[5]) and begins at a temperature lower than the 86 mK predicted by regular solution theory. The finite temperature range seems to indicate that the separation does not proceed by nucleation. Previous workers have suggested that it might be an example of a spinodal transformation.[10] Regular solution theory would indicate that the spinodal would occur at a much lower temperature, 0.25 mK. However, the strain energy in the neighborhood of dislocations where the impurities are clustered could raise this temperature.

The behavior of the pure samples is easier to interpret qualitatively but the quantitative details raise new questions. The temperature dependence of the velocity is due to the contribution of the exchange energy to the bulk modulus. Therefore the sound velocity can be calculated from the parametrized partition function[11] (in zero field): $\ln Z = \ln 2 + e_2/8T^2 - e_3/24T^3$, where $e_2 = 12J^2$ in the simple case when all exchange parameters equal J. If we define $\gamma = d\ln e_2/d\ln V$ and retain only first-order terms in $1/T$, the adiabatic bulk modulus is given by $B = B_T - (Nk/16VT)\gamma e_2$, where $B_T = (Nke_2/8T)[\gamma/V + (d\gamma/dV)_T]$. We write the velocity as $c = [(B_0 + B)/\rho]^{1/2}$ where B_0 is the bulk modulus independent of the exchange contribution and $B_0 >> B$. We assume that $(d\gamma/dV)_T << \gamma/V$ and set c_0 equal to the maximum observed velocity. Then we find

$$\frac{c_0 - c(T)}{c_0} = \frac{B}{2B_0} = \left(\frac{Nk}{16VB_0}\right)\frac{e_2\gamma^2}{T}.$$

Previous measurements of the exchange constants yield[8] $e_2 = 6.8\ \mathrm{mK}^2$ (at $V = 24.22\ \mathrm{cm}^3/\mathrm{mole}$) and[12] $\gamma = 36$. Changes in volocity are measured accurately by our experiment but the absolute velocity is not. Therefore we compare our results with the above calculation by finding the ratio of experimental values to calculated values of $\alpha = [c_0 - c(T)]T/c_0$. If γ were truly independent of density this ratio would range between 0.4 and 0.8. The recent work of Chapellier *et al.*[4] finds γ decreasing with increasing volume, passing through zero at about 24.6 $\mathrm{cm}^3/\mathrm{mole}$ and becoming negative at still larger volumes. Thus values of γ determined from their data would predict smaller values of α than we have measured. On the other hand, their values of the exchange, J, are determined from measurements of the nuclear spin-lattice relaxation times assuming a specific form for the spectral density of the exchange. The J computed in this manner may be incorrect for determining the velocity of acoustic waves.

The temperature dependence of the attenuation is due to the interaction of the acoustic waves with some type of thermally activated excitations. However, the nature of these excitations is uncertain in view of the small measured value of the activation energy. Although the influence of thermally activated vacancies has been observed on a number of other properties, that activation energy has been found to be strongly volume dependent and at least an order of magnitude larger than the one found here. The small value which we find could arise from a number of mechanisms such as weak binding of vacancies to dislocations, the binding of ^{4}He impurities to dislocations, or dissipation due to formation of kinks on dislocations or to some previously unidentified family of low-energy excitations. Additional experimental and theoretical work will be required to resolve the question.

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