PHONON INTERFERENCE IN THIN FILMS OF LIQUID HELIUM

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Acoustic wave interference has been observed at frequencies between 18 and 58 GHz in thin films of liquid helium absorbed on surfaces of CaF_2, SrF_2, and BaF_2. The thickness of the films, ranging from 10 to 200 Å, could be measured to within a few percent in most cases. The dependence of the van der Waals potential on the film thickness is also discussed.

Following a suggestion by Rudnick,[1] we have used our spin-phonon spectrometer[2] to observe acoustic wave interference in thin films of liquid helium adsorbed on surfaces of CaF_2, SrF_2, and BaF_2. Measurements were made at frequencies between 18 and 58 GHz, where the wavelengths of acoustic waves in liquid helium are 130 and 40 Å, respectively. The results provide new measurements of the film thickness for saturated and unsaturated films, which have proven to be about 40% thinner than is currently assumed.[3]

We have shown that partial saturation of an EPR resonance line of divalent thulium, present in the crystals used for these experiments at a few hundredths of a mole percent, produces monochromatic phonons at the resonant frequency.[4] In the present experiments, these phonons leave the crystal by passing through a thin layer of liquid helium, held on to the surface by the van der Waals force, into a surrounding bath of helium gas kept at 1.35°K. When the thickness of the film is equal to an odd multiple of a quarter wavelength of the acoustic waves, a standing wave is set up which enhances the transmission of the phonons into the gas[5] or liquid. This lowers the steady state density of the phonons in the crystal and cools the partially saturated spins. These changes can be detected by optically monitoring the temperature of the same spins using circular dichroism.

Phonon interference is observed as standing waves even though the phonons approach the crystal-film interface at all angles. This is because the velocity of sound in the film is an order of magnitude smaller than that in the crystal so that even those phonons traveling nearly tangential to the interface are refracted upon going into the liquid to within a few degrees of the normal direction.

Most of the crystals used in the experiments were cleaved on all sides in air just before placing them at the shorted end of a K-band waveguide. A piece of polystyrofoam was pushed on top of the crystal to hold it in place. A glass can was sealed over the end of the waveguide and the interior of the guide and can was pumped to a pressure below 2×10^{-4} Torr before being immersed in liquid helium. Helium gas could be introduced or removed from the interior of the system by way of the waveguide and a separate tube from the glass can went to a Pirani pressure gauge located just outside the Dewar system. The Pirani gauge was calibrated against a McLeod gauge. The monitor light passed through windows in the Dewars, the glass can, two small holes in the waveguide, and the crystal. A 12-in. electromagnet provided the uniform magnetic field necessary to tune the spins. Since in these experiments the crystals were located completely inside the waveguide the spin polarization could also be measured in a standard fashion using the reflected microwave power, but we found the optical system more convenient since a wide range of frequencies was used.

At the start of each run the interior of the system was filled with helium gas until the pressure was saturated and a very small amount of liquid condensed on the bottom of the can. At this point, all the surfaces inside the can and, in particular, the crystal surfaces were coated with a (saturated) film of liquid helium. A fixed level of microwave power was continuously applied to heat the spins ~30%, and the spin temperature was then recorded as the helium gas was slowly pumped away. No changes were observed until excess liquid in the bottom of the can was removed, at which point the film would begin to thin. Each time the film thickness went through one of the resonant lengths, the spin temperature would cool a small amount, as shown in Fig. 1. The reverse pattern was observed when the gas was reintroduced; the point where the film stopped growing was generally abrupt and obvious. The shape of each curve in Fig. 1, and especially the changes with frequency, are what one expects for the phenomena described above.

The saturated-film thickness, measured in acoustic wavelengths, obtained by simply count-

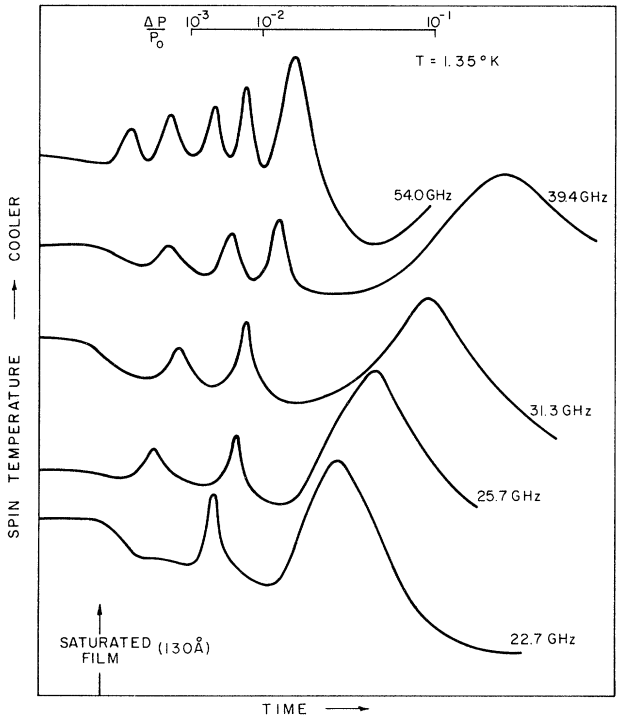

FIG. 1. Acoustic wave interference in thin films of liquid helium adsorbed on cleaved surfaces of SrF_2 detected through its effect on the spin temperature of divalent thulium in the crystal at 0.02 mole%. The spin temperature on the vertical scale is in arbitrary units but the maximum excursion of each curve corresponds to a change of at most 30 m°K. The horizontal scale is uniform in time during which the helium was slowly pumped away and so once past the saturation point the scale represents the film thickness in some nonuniform way. Each of the five runs shown was taken at the different frequencies noted but under otherwise identical experimental conditions, each run taking 20 min. They are aligned along the horizontal axis at the point where the measured pressure changed by 10%, as noted on the top of the figure, and are offset from one another along the vertical axis for convenience of display. The final peak at the right is the simple quarter-wave which is not shown for 54 GHz because it occurs off the horizontal scale.

ing the number of peaks observed and estimating where saturation occurs, is plotted as a function of frequency in Fig. 2. The SrF_2 and CaF_2 data points were obtained in this way. The BaF_2 data were taken using the following more accurate method. The saturated film could be set into oscillation at about 0.2 Hz for a few minutes by the impulsive addition of a small amount of gas. When the microwave frequency was tuned so that the saturated film was an exact resonant length, this low-frequency oscillation would generate a 0.4-Hz modulation of the spin temperature. The

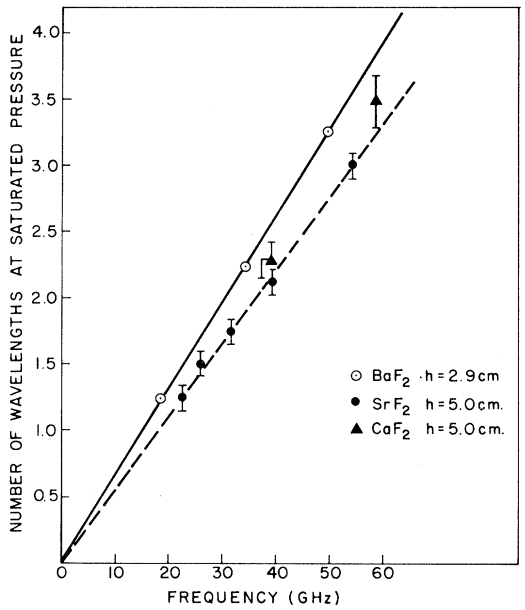

FIG. 2. The saturated-film thickness measured in acoustic wavelengths plotted against the frequency used for the measurement. The BaF_2 results differ only because the saturated film is thicker at the lower height.

saturated-film thickness could thus be measured, in acoustic wavelengths, with a precision of ±1%. These BaF_2 data points are consistent with the dispersion in the velocity being less than 2% over the frequency range. Since the straight line drawn through the data points in Fig. 2 extrapolates through the origin, the order of the resonances are correctly identified.

The saturated-film thickness was found to be independent of temperature between 1.35 and 1.80°K. The signals decrease rather rapidly with increasing temperature, making it difficult to obtain meaningful measurements above this temperature. Several SrF_2 crystals as well as some which were exposed to air for extended periods of time gave identical results. Surprisingly, BaF_2 and CaF_2 give the same film thickness within the present experimental uncertainties of a few percent.

The saturated-film thickness could not be measured on crystals with highly polished, rather than cleaved surfaces, as only the quarter-wave resonance and sometimes a very small three-quarter-wave resonance could be observed. Thus, while the cleaved surfaces have large areas which are flat on an atomic scale,[6] this is not true for the polished surfaces.

If the film and gas are in equilibrium, the chemical potentials of the liquid and vapor are equal,

which leads to the equation[7]

$$\mu(d) = mgh + kT\ln(P_0/P),$$

where $-\mu(d)$ is the van der Waals potential for a helium atom on the surface of the film at a distance d from the substrate and is usually given in the form αd^{-n}, mgh is the gravitational potential for an atom at a height h above the lowest point in the can (or the bulk liquid surface in the case of saturation), kT is the Boltzmann constant times the temperature, and P_0/P is the ratio of the vapor pressure of bulk liquid at the system temperature to the pressure at the lowest point in the can. This pressure ratio is equal to the ratio of the saturated pressure to the pressure of the system as both are measured at the Pirani gauge outside the Dewars, and it is this measured ratio we use.

For the saturated-film measurements, $P_0/P = 1$, the van der Waals potential is equal to the negative of the gravitational potential. A measurement of the saturated-film thickness as a function of height above the layer of bulk liquid in the bottom of the can would give the complete functional form of $\mu(d)$. This we have done from 0.9 to 10.0 cm, where the film thickness ranged from 200 to 100 Å. However, we could not go above 10 cm with our present Dewars, and so we also measured the thichness of unsaturated films, $P_0/P > 1$, which is equivalent. In this way, the van der Waals potential could be measured for film thicknesses of about 60 Å, where the Pirani gauge could just measure the change in pressure, to about 10 Å, which is the quarter-wave length of a sound wave in liquid helium at 58 GHz, our present upper frequency limit. The signal-to-noise ratio on the quarter-wave peak is generally very good and so there is no intrinsic reason why measurements could not be extended to higher frequencies. That we see this quarter-wave resonance at 58 GHz is remarkable since 10 Å corresponds to less than three statistical layers of helium atoms.

The van der Waals potential is given in Fig. 3 plotted against film thickness measured as the transit time for an acoustic wave to cross the film. (This is equal to the thickness measured in acoustic wavelengths divided by the frequency.) The transit time can be converted to thickness by multiplying by the bulk sound velocity (2.36×10^4 cm/sec at 1.35°K).[7] The major uncertainty in the thickness arises from the first two layers of helium being denser than the bulk liquid, and

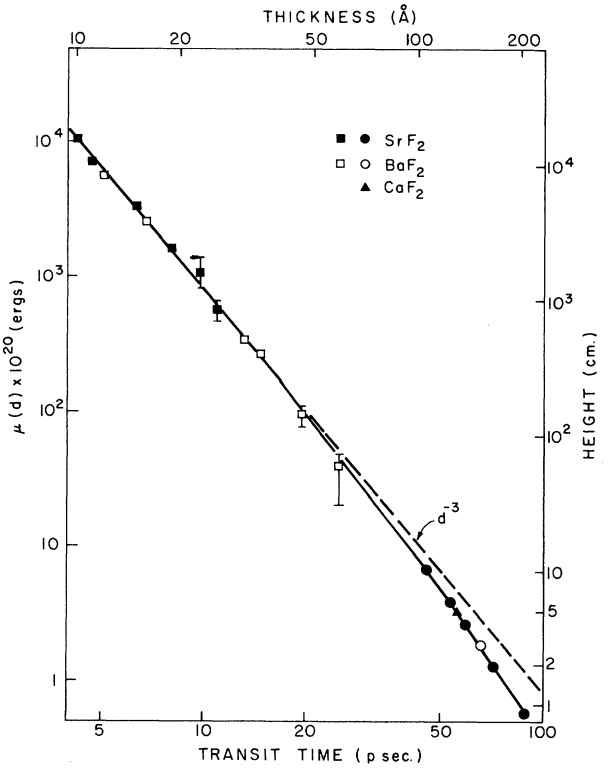

FIG. 3. The potential energy of a helium atom on the surface of the film as a function of the transit time for a phonon to cross the film. The thickness scale in Angstroms on the top of the figure was obtained by multiplying the transit time by the bulk-liquid sound velocity. The data represented by circles and one triangle are from saturated films, the squares are from unsaturated films.

so the thickness scale may be off by several Angstroms.

The value of α, the coefficient of the van der Waals potential in the region where it clearly obeys a simple inverse-cube law, $n = 3$, is a useful quantity for comparing these results with previous measurements. Recently it has been fashionable to give α in degrees Kelvin and measure the thickness in layers of helium, one layer equaling 3.6 Å. Our results give $\alpha = 16$°K (layers^3) which is significantly smaller than the values of 87-98°K presently used,[3] which means the films are 40% thinner. The most likely explanation is that the van der Waals potential is smaller for these fluorides than it is for the substrates most previous measurements have used. Our results do agree quite well with those given in a brief note by Hemming.[8] The deviation of the potential from the simple inverse-cube law for thick films is consistent with retardation effects.[9]

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PAIR TUNNELING AS A PROBE OF FLUCTUATIONS IN SUPERCONDUCTORS*

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We suggest a method of using the average pair field of one superconductor to probe the fluctuation pair field of a second metal at temperatures above the latter's superconducting transition point. The two metals should be fabricated as a strongly coupled tunnel junction and the dc I-V characteristic measured as a function of both the bias voltage V and the amplitude of a magnetic field H parallel to the junction interface. The resultant I-V characteristic is predicted to give a direct measurement of the frequency and wave-vector dependence of the Δ susceptibility characteristic of the superconducting transition.

In most second-order phase transitions the order parameter can be directly coupled to an external field and the response of the order parameter to this coupling determines the characteristic susceptibility associated with the onset of order. The frequency and wave-vector dependence of this characteristic susceptibility provide the most direct probe of the fluctuations associated with the phase transition. Two well-known examples of this are magnetic transitions, where the magnetization is coupled to a magnetic field giving the magnetic susceptibility, and the liquid-gas transition, in which the density is coupled to the pressure giving the compressibility. In a superconducting system, the analogous characteristic susceptibility involves coupling to the pair field Δ which is off-diagonal in electron number space. For this reason one cannot couple to Δ with a classical field, and the question of whether, in principle, the Δ susceptibility of a superconductor can be directly measured has been raised.[1] The electrical conductivity[2] and magnetic susceptibility,[3] which have been observed near the superconducting transition temperature, involve convolutions of Δ susceptibilities. Recently, however, Ferrell[4] has suggested that the Δ susceptibility could be obtained by measuring the frequency-dependent conductivity of a Josephson junction in which one side of the junction is near its transition temperature while the other is well below its transition temperature. In this case, the average pair field of the higher transition temperature superconductor provides the necessary off-diagonal coupling.

Here we explore this idea of using the average pair-field amplitude of one superconductor to probe the fluctuation pair field of a second metal at temperatures above its superconducting transition temperature. We show that a measurement of the dc I-V characteristic can provide a direct determination of frequency and wave-vector dependence of the Δ susceptibility. The frequency is set by the bias voltage and the wave vector is determined by the application of a magnetic field parallel to the junction interface. Whether this proposed experiment will in fact provide a useful probe of the Δ fluctuations depends critically upon the ability to fabricate strongly coupled tunnel junctions. Some estimates of the parameters which enter this type of measurement are discussed in the conclusion.

Although a direct microscopic calculation starting from the tunneling Hamiltonian can be carried out, the following phenomenological approach introduced by Ferrell[4] provides more insight into the underlying physics. In particular