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Report
~~Renewal Proposal~~

~~for a Research Grant~~

Contract No. AT(11-1) - 1574

Properties of Rare-Gas Solids

Submitted to the U.S. Atomic Energy Commission

Department of Physics
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~~Title: Properties of Rare-Gas Solids~~

Proposed Renewed

~~Desired Starting Date: March 1, 1973 (Present Grant Expires on February 28, 1973)~~

Item 1: Technical Progress Report*

During the current Grant Period we have made good progress in our goal of better understanding the physical properties of rare-gas solids. We have succeeded in one of our long-time goals of measuring superfluid He film flow into and out of solid Ar beakers. We have extended the self-consistent phonon theory in an application to our current vapor pressure and sublimation pressure measurements on Ar³⁶. We have made measurements of the thermal conductivity of solid argon in the region between 65°K - 83°K. Finally, we have made calculations of the Kapitza Resistance between He⁴ and Cu, and other solids, using the properties of waves in layered media. Several related problems have also been investigated.

The principal investigator has devoted approximately 50% of his time to this research during the Grant Period since the beginning of the current term. He will continue to devote 50% of his time to this research.

The work for the current year will be briefly described below under these five headings: A. Vapor Pressure of Rare-Gas Solids, B. Thermal Conductivity of Single Crystals of Ar, C. Liquid He Film Flow into and out of Solid Ar Beakers, D. Kapitza Resistance, and E. General Activity on Rare-Gas Solids.

A. Vapor Pressure of Rare-Gas Solids

During the current Grant period we have completed our experimental and theoretical study of the vapor and sublimation pressure of Ar³⁶. The measurements were part of our long-term study of vapor and sublimation pressures of rare-gas solids and liquids. Previously we have studied and reported on Ar, Kr, and Xe. The idea behind the current study is that, by using one isotope of Ar, the effects on the thermodynamic properties, of the different masses of the atoms in normal isotopic mixtures, could be better understood and accounted for.

We made measurements from about 24°K - 87.4°K and analyzed our results for $P(\text{Ar}^{36})$ and $P(\text{Ar}^{36})/P(\text{Ar}^N)$, i.e. vapor pressure and vapor pressure ratio using the improved self-consistent theory of anharmonic crystals.

The results of our experiment, their comparison with those of other workers and with theory, as well as a description of the experiment proper, are described in the enclosed preprint entitled: "Sublimation and Vapor Pressure of Ar³⁶", by Jon L. Opsal, Charles W. Leming, and Gerald L. Pollack. This paper has been submitted for publication to Physical Review B.

B. Thermal Conductivity of Single Crystals of Ar

During the current Grant period we have: completed our thermal conductivity apparatus, grown about 10 polycrystal or single crystal samples of solid Ar, each about 1 cm in diameter and 3.2 cm long, and measured the thermal conductivity as a function of temperature in the range 83 - 65°K. This experiment is the thesis research problem of Mr. David K. Christen, who has

been making good progress with the challenging and difficult, but important, experiment.

The crystals we have grown so far have been solidified from the melt at a growth rate of 0.6 mm/hr. This growth rate is one of the experimental variables, it is controlled by a moving heater collar driven by a synchronous motor. We plan to increase the grain size of our polycrystals by growing at rates close to 0.1 mm/hr and subsequently annealing. The data so far are in the range where the thermal conductivity is proportioned to T^{-1} , and our seven data points are consistent with previous work of others in this range.

Some of the experimental problems which have been overcome are unique to properties of rare-gas solids. Solid Ar has a large linear expansivity and the expansivity is important even down to low temperatures (it is about $10^{-4}/^{\circ}\text{K}$ at 15°K). Since the crystal is grown from the melt just below the triple point (83.8°K), this means that special techniques must be used to insure that contact between the crystal and the thermometers is maintained as the temperature is lowered. We have done this by using a spring bellows whose length is controlled by the vapor pressure of the solid Ar. The bellows is long near the triple point of Ar, where the vapor pressure is large and the crystal is long. The bellows is short at low temperatures, where the vapor pressure is low and the crystal is shorter. The dimensions are carefully chosen to minimize strains in the crystal.

Since the thermal conductivity of Ar is so low near the triple point the thermal relaxation times are very long, so that

it was necessary to use a solid with a large diameter (1 cm). This has made the relaxation times reasonable, but each data point still requires 24 hours to obtain and during this time the apparatus must be almost constantly attended for minor adjustments.

In general, progress on this experiment has been quite satisfactory. The results have been a little slow in coming, due to inherent difficulties of the experiment, but the results look very good in comparison with previous theoretical and experimental work. Our current goals are to make measurements below 65°K and down to 2°K and to improve the quality of the crystals so as to get a large maximum in thermal conductivity at very low temperature. We have already made several experimental investigations of problems we are likely to encounter and we believe that all the important difficulties are under our control.

C. Liquid Helium Film Flow into and out of Solid Argon Beakers.

During the current Grant period progress with this problem has gone very well and the main goals have been reached. Mr. T. O. Milbrodt, the research assistant whose Ph.D. thesis this is, has succeeded in growing 5 leak-free solid Ar beakers. We have measured superfluid He II film flow rates σ (cm³/sec-cm of circumference) into and out of these beakers for both filling and emptying as a function of temperature between 1.4K - 1.8K, level difference (Z), and film thickness, i.e. height from the source level to the beaker rim.

We have also done some theoretical analysis of our results

and this part of the work is still continuing. In general our T dependence of σ is slower than ρ_s/ρ , the expected dependence. Our results for $\sigma(Z)$ follow the form $\sigma = 1/(A-B \ln Z)$, in qualitative agreement with the Langer-Fisher-Iordanskii theory. This means that fluctuations are playing a role in establishing the velocities of flow. There are still questions about what should be called the critical velocity, since in the theory the flow rate for zero chemical potential ($Z = 0$) is $\sigma = 0$, i.e. there is no critical velocity. Very recent experiments which measure flow rates under very low chemical ^{potential} gradients have tended to agree with this. Our experiment measures the product of $v_s d$, (superfluid velocity) $\times$ (film thickness). We have been tentatively assuming the Leiden rule, $v_c d^{1/2} = 1$ (c.g.s units), to get estimates of d from our data, but this is only approximate and we are seeking better values than this can give.

One Goal of our experiment was to see whether we could measure the substrate dependence of the He film flow rate. We have observed and measured this effect by observing lower rates for film flow out of Ar beakers than have been observed for other substrates; we believe that our values of σ are probably the lowest which have been reported on any substrate. We can use two theories to calculate the He film thickness over Ar: We have used the Van der Waals forces theory of Schiff and Frenkel. This theory derives the force between substrate and He by considering the intermolecular Van der Waals potential. There is now a more complete theory due to Lifshitz which has newly been applied by Anderson and Sabisky to alkali halide and Ar inter-

actions with He. This theory is based on the electronic polarizability as a function of frequency, as determined by spectroscopic absorption measurements. We have been working on comparing our data with both kinds of theories and so far the results are quite satisfactory.

Much of this work was presented at the 13th International Conference on Low Temperature Physics in Boulder, Colorado in August 1972. A preprint of that work is included with this report. It is entitled: "He II Film Transfer Rates Into and Out of Solid Ar Beakers", by T. O. Milbrodt and Gerald L. Pollack (Publication number C00-1574-30). ^{removed} A more detailed description of our work and results is given there. A more complete description of our research is being currently prepared for submission to The Physical Review. This will be ready in two or three months.

As we mentioned above, the main goals of this research have been largely met. It is proposed to continue the research into the forthcoming Grant Renewal Period. During this time we plan to complete our theoretical analysis, publish the complete results, and attempt to extend the experiments to include He film flow out of solid Ne beakers. During the renewal period Mr. Milbrodt will get his Ph.D. degree and we shall obtain a suitable position for him.

D. Kapitza Resistance

During the current Grant period, we have started serious calculations on Kapitza resistance, the thermal boundary resistance associated with heat flow between a solid and liquid He II.

This problem is the thesis problem of Mr. Jon L. Opsal, a research assistant on this Grant.

The ideas behind our calculations are based on the experimental observation that the measured Kapitza resistance (R_k) is much less, often an order of magnitude less, than the R_k calculated by the Khalatnikov theory. In addition the observed T dependence appears to be more complicated than that predicted by theory, R_k proportional to T^{-3} .

Our current calculations consider the effect of adsorbed He layers on the surface of a typical solid, say Cu. It is known from specific heat measurements and measurements of adsorption isotherms what the approximate thickness, structure, and properties of these layers are. We are attempting to put this into the theory. Since the sound velocity and the density in the intermediate adsorbed layers are intermediate between those in the solid and those in liquid He II, the adsorbed layer serves as a matching acoustic impedance. This has the effect of decreasing the thermal boundary resistance since the acoustic mismatch at the boundary is less with the adsorbed layer than it is when the solid terminates abruptly into the liquid.

We are examining this idea quantitatively by putting in a suitable density profile for the adsorbed He layer. Roughly speaking, we take the first adsorbed layer, i.e. first $5\text{\AA}$, to have a constant density equal to twice the bulk liquid density. We then assume that the density falls off smoothly, reaching the liquid value at about $15\text{\AA}$ from the wall. In our calculations we split up the adsorbed He into thinner layers of constant density, a maximum of 32 layers, and we apply the theory of wave

propagation in layered media. This is a theory which has been developed and applied to geophysical problems, for example, and much of those results and formalism can be applied to our problem. Our calculations are extensions of the earlier work of Challis, Dransfeld, and Wilks.

Briefly what we have done is to consider monochromatic phonons incident at various angles from the liquid onto the surface of the solid. These waves are refracted and reflected by the adsorbed layers; this is what we calculate by applying the theory of wave propagation in layered media. This calculation gives us the transmission coefficient as a function of frequency and angle of incidence, $TR(\omega, \theta)$. The calculation assumes conservation of the tangential component of the wave vector, i.e. Snell's law for acoustic waves. We predict a critical cone, for transmission, with half-angle θ_c approximately 0.103 radians for a Cu-He⁴ interface.

The calculations are still in an early stage, but we have been able to compare our results with other theories and experiments. Our calculations give approximately and qualitatively the same T dependence as observed. We calculate R_k proportional to T^{-3} for T below about 0.4K and R_k proportional to T^{-3} for T greater than 1.3°K. Between 1.3K and 0.4K the Kapitza resistance increases more rapidly with decreasing T than does T^{-3} . There are many aspects of this calculation and of Kapitza resistance which we still want to investigate. Our overall goal in this problem is to understand the phenomenon and the related surface physics problems as well as we can. In par-

ticular we should like to get an understanding of the key problem in Kapitza resistance studies, as described above, why do experimental values and theoretical predictions of R_k differ so much?

E. General Activity on Rare-Gas Solids

During the current Grant period the principal investigator and his students have continued to maintain close contact with several other laboratories working on the properties of rare-gas solids, liquid helium, and related fields. We have also maintained an up-to-date comprehensive literature survey and bibliography of research work in these fields.

In August of the current Grant period, the principal investigator and the three research assistants supported by this Grant attended the Thirteenth International Conference on Low Temperature Physics in Boulder, Colorado. At this Conference we presented our work on He II flow into and out of solid Ar beakers and we discussed our research and exchanged research ideas with many, many researchers in these fields. This exchange of ideas was very fruitful for us and we all profited considerably from this Conference.

The principal investigator is Colloquium Chairman for the Michigan State University Physics Department for the 1972-73 academic year. This means that several visiting scientists come to the laboratory and we have a valuable opportunity to exchange ideas with them, too.

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Numbered Bibliography for Current Grant Period

29. "Sublimation and Vapor Pressure of Ar³⁶", by Jon L. Opsal, Charles W. Leming, and Gerald L. Pollack. Submitted to Physical Review B.
30. "He II Film Transfer Rates Into and Out of Solid Ar Beakers", by T. O. Milbrodt and Gerald L. Pollack. To appear in the Proceedings of the 13th International Conference on Low Temperature Physics.
31. Technical Progress Report for Grant AT(11-1)-1574.