JAN 31 1968

AEC-copy 4

upper lower

(Renewal Proposal for a Research Grant)

(Contract No. AT(11-1)-1574)

MASTER

Properties of Rare-Gas Solids.

Submitted to the U.S. Atomic Energy Commission

Department of Physics

Michigan State University

East Lansing, Michigan

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

ENDORSEMENT

This proposal is respectfully submitted by:

Principal Investigator

Gerald L. Pollack
Associate Professor of Physics

Department Chairman

Sherwood K. Haynes
Chairman, Department of Physics

Authorized Business Officials

Philip J. May
Vice-President for Business and Finance

Milton E. Muelder
Vice-President for Research Development

Michigan State University

East Lansing, Michigan

Title: Properties of Rare-Gas Solids

Desired Starting Date: March 1, 1968 (Expiration date of present grant is February 29, 1968).

Project Summary

The aim of the proposed research is to continue to study some problems of the physics of rare-gas solids. Of primary interest are problems of thermodynamic properties, surface physics, and vacancy and defect structure of solid argon, krypton, xenon, and neon. For these elements the vapor pressure from the triple points down as low as possible are being measured. The experiment emphasizes equilibrium at low temperature between the solid and gas phases. Results are examined for information on vacancy concentrations, the interatomic forces and anharmonicity. Appropriate theories are being studied concurrently with the experiments. Work is continuing on using liquid helium films to study surface structure and interactions of condensed rare-gases. An experiment is described in which the flow rates of liquid helium films over argon substrates are being measured. Preliminary data from this experiment have been obtained and give information on the interaction between argon and helium atoms. The experiments are being extended to the other rare-gas solids. Theoretical work on waves on static liquid helium films is also described. The nature of the waves depends critically on the force between the substrate and helium and the experimental data are used to study these forces. Work on some problems

of Kapitza resistance which has been started in the past year is described. This phenomenon may be used for studying surface physics and lattice dynamics of all solids.

1. Technical Progress Report

The work on properties of rare-gas solids and related subjects, which is being carried through during the present renewal period of the grant, is concentrated on six problems. Progress on each of these is discussed below. The proposed technical program for these problems in the forthcoming year is described under heading 3.

A. Vapor Pressure Measurements

Measurements of the vapor pressure of argon, krypton, and xenon, and associated theory is to be the Ph. D. thesis project of Mr. Charles W. Leming, a half-time research assistant employed under this Grant since September 1, 1966. This research has been proceeding satisfactorily and, although no data are yet available for publication, a few key measurements have been made on argon. The essential idea to this experiment is that the measurements are to be made as accurately as possible. This means making frequent calibrations and internal consistency checks and, consequently, progress is likely to be a little slow at first.

In the region from 90° K to 15° K, temperature is being measured with a Platinum-resistance thermometer. A Pt-resistance thermometer calibrated in this region at the National Bureau of Standards is available in our laboratory. We have built a suitably shielded calorimetric bomb for calibrating our thermometers against this secondary standard and the calibration above 60°K is essentially complete.

In addition some calorimeters for measuring the vapor pressure have been built and tested with solid argon, and many interesting difficulties have been overcome. Our measurements show that even in a carefully designed apparatus, the time to establish suitable equilibrium between the solid and its vapor near the triple point is about 25 minutes. The design of an adiabatically shielded container which maintains constant temperature for such long times has been a challenging one. Progress has been good, however, and we now have some working designs. These are presently being further tested.

Vapor pressure itself is being measured with a manometer, Bourdon gauge, or McLeod gauge depending on the substance and temperature. The first two of these are functioning and suitably connected to the low-temperature system. Our present experiments on this part of the system are aimed at studying the characteristics of the Bourdon gauge and properly calibrating it. Our idea is to carefully calibrate the pressure and temperature systems while working on argon, then measurements on krypton, xenon, neon, mixtures of rare gases, etc. will be more straightforward.

B. General Activity on Rare-Gas Solids

During the term of the present grant a general solicited article on rare-gas solids has been accepted for publication by the McGraw-Hill Science Yearbook for 1968. A copy of the manuscript is included with this renewal request.

The principal investigator maintains close contact with other workers in the field through correspondence and through joint work on problems of mutual interest on rare-gas solids and related subjects. One of the groups with which we have maintained close contact is at the U.S. Army Frankford Arsenal in Philadelphia. During the present grant period a paper has been published in the Journal Applied Physics describing results of previous joint experiments on optical, dielectric, and crystallization properties in rare-gas solids. A copy of the paper is included in this renewal request.

The laboratory maintains an up-to-date collection of the literature, preprints, and other reported work on rare-gas solids and other simple molecular solids. We propose to continue this activity.

C. Rare-Gas Solid Surfaces and Liquid Helium Films

During the current renewal period, experiments have been carried out which use flowing liquid helium films to study forces at rare-gas solid surfaces . These experiments have been carried out by the principal investigator and Mr. Carl J. Duthler, who has been employed as a half-time research assistant under the grant since July 1, 1967. Progress on these experiments has been good and it is now expected that they will be the subject of Mr. Duthler's Ph.D. thesis when he has taken his comprehensive exams.

Research on this problem studies: (a) The nature of the force between rare-gas atoms in the condensed state and helium atoms in the liquid state, (b) The nature of liquid helium films and superfluid flow, and (c) Condensation phenomena for thin films of solid argon, krypton, xenon, and neon.

The idea is that the thickness of liquid helium films depends on the force between liquid helium and the substrate. Our experiments in the past year have aimed at studying flow rates of liquid helium films across glass surfaces which are covered with varying thicknesses of argon films. According to the usual Brunauer-Emmett-Teller theory of adsorbed films, the chemical potential per gram (μ) of a film of thickness d may be expressed as $\mu = \alpha/d^3$, where both α and the d^{-3} dependence are determined by the nature of the force between the substrate and the film. For a helium film at height h above the surface of a helium bath the chemical potential may also be written as $\mu = gh$. Therefore the connection between d and h is: $d \simeq (\text{constant})h^{-1/3}$. Both the magnitude of the constant and deviations from the 1/3 power height dependence can be used to study the forces. When the helium film flows out of or into a beaker of this height, then the flow rate is given as: $\dot{V}$ (cm^3/sec) $= V_c d \times$ (circumference) or: $\dot{V} = v_c(\text{constant})\, h^{-1/3}$ (circumference). In this expression v_c is a critical velocity of superfluid flow. In our experiments the circumference and all related geometry is known, and we are measuring $\dot{V}$, with a cathetometer, as a function of h and the absolute temperature T. The experiments on clean glass beakers and on beakers with solid argon layers up to 10,000 A thick have already been carried through. The experiments have been done in the range from 1.6° K up to 2.2° K.

Some preliminary results are already available but they are still very qualitative. We have tested for the $h^{-1/3}$ dependence in $\dot{V}$ and found that the exponent depends on the coverage and varies from about 1/7 up to 1/3. The results are to be made more certain in the forthcoming contract period by measurements on more pure systems of argon on glass and then extended to studies of krypton, xenon and neon surfaces and films. The results which have already been obtained are consistent with results of other investigations on related problems. Our curves for $\dot{V}$/(circumference) as a function of T, for pure glass beakers show the maximum of 7.0×10^{-5} cm^3/sec.-cm. at about 1.85° K characteristic of clean systems. When layers of pure Ar 1000A or more thick are condensed on the glass beaker, the flow rates show the same temperature dependence but are about two times faster.

D. Waves on Liquid Helium Films

A study of the theory of waves on liquid helium films which was begun two years ago has been completed during the present contract period. The results have been published in a series of articles, in the Physical Review, of which the most recent appeared during the present grant period. A reprint of the article is included in this report.

The most important result of this work is to show that if normal-fluid motion is allowed in liquid helium films, then the films will support two wave modes. One of these is an unattenuated wave mode, called third sound, and the other is a new attenuated wave mode. These two kinds of waves have been described, as functions of temperature and of viscous forces, in the present research. In addition, the connection between the initial conditions of real experiments and the amount of each of the wave modes excited in the experiments, has been established under general assumptions.

For the time being no further direct work on this subject is being undertaken. However, the hydrodynamic properties of liquid helium II are still of interest. In the proposed technical program below we suggest an interesting experiment which is aimed at further investigation of simple nonlinear hydrodynamics and surface physics in liquid helium. This is a measurement of the connection between first and second sound.

E. Specific Heat of Rare-Gas Solids

During the present renewal period work was started on this interesting but difficult problem by Mr. Paul B. Williams, a half-time research assistant employed under the grant for four months, and the principal investigator. During this time a calorimeter was built that is quasi-adiabatic, and preliminary tests were carried out on it. The research assistant was taken off grant support at his request in September 1967, although his progress had been satisfactory. Work on this problem has been suspended, at least temporarily, until a new student who would like to work on it can be found. Meanwhile, the apparatus built during the summer will be used on other projects.

Our experience this summer shows that this experiment is likely to be a difficult one for a graduate student. The specific heat must be measured to considerably better than 2% in order that it be a significant improvement over the previous measurements which were done at the National Research Council in Ottawa. This may be difficult now. However, the vapor pressure measurements are providing much valuable experience along these same directions. We are therefore considering waiting until the vapor pressure measurements are more advanced before trying again to measure the specific heat.

F. Kapitza Resistance

A thorough study of the literature in the theoretical and experimental problems of Kapitza resistance has been undertaken. This phenomenon occurs at all solid-solid and solid-liquid interfaces across which thermal energy flows. The resistance itself is a measure of the efficiency with which thermal energy is transported across the interface and it depends on: (a) The energy distribution on both sides of the interface, (b) The acoustic impedance on both sides of the interface (this is essentially the product of the density and the sound velocity), (c) The quality of the interface (i.e. state of purity, annealing, or electropolishing), and on many other factors which are not yet well understood.

Our study has convinced us that Kapitza resistance is potentially a very useful way of studying surface physics and all the critical unsolved problems associated with surface physics. We have started outlining some interesting experimental and theoretical investigations that are important and deserve further study. Work on a review article on Kapitza resistance, to be published in the Reviews of Modern Physics, has started and the experimental part of the article is now complete. This comprises about the first half of the article. The rest of the article will be finished shortly and publication in the proposed grant renewal period is expected.

Numbered Bibliography for period from March 1, 1967 to present.

4. "Radiation of Sound in He-II Films", by Gerald L. Pollack, in Physical Review 161, 172-178 (5 September 1967). Reprint enclosed.

5. "Inert Gases", by Gerald L. Pollack, accepted for publication in the Yearbook of Science and Technology (McGraw-Hill, New York, 1968). Copy of manuscript enclosed.

6. "Dielectric and Optical Properties of Crystalline Argon", by I. Lefkowitz, K. Kramer, M.A. Shields, and Gerald L. Pollack, in Journal of Applied Physics 38, 4867-4873 (November 1967). Reprints not yet available, a copy of the article is enclosed.

Bibliography for period from March 1, 1966 through February 28, 1967.

1. "Effect of Normal-Fluid Motion on Third Sound in Liquid-Helium Films", by Gerald L. Pollack, in Physical Review 143, No. 1, 103-109 (4 March 1966).

2. "New, Attenuated Wave Mode in HeII Films", by Gerald L. Pollack, in Physical Review 149, No. 1, 72-76 (9 September 1966).

3. "Solid Noble Gases", by Gerald L. Pollack, in Scientific American 215, 64-74 (October 1966).