NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

Technical Memorandum 33-653

# Energy Storage in Solid Helium

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(NASA-CR-135716) ENERGY STORAGE IN SOLID HELIUM (Jet Propulsion Lab.) ~~14~~ 15 p HC $3.00 CSCL 20L

N73-32541

Unclas

G3/23 18714

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Prepared Under Contract No. NAS 7-100
National Aeronautics and Space Administration

# PREFACE

The work described in this report was performed by the Space Sciences Division of the Jet Propulsion Laboratory.

ACKNOWLEDGMENT

The author wishes to express his appreciation to Drs. Laurent Caron, Nathan Jacobi, Melvin Saffren, and Sandor Trajmar for their helpful discussions.

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ABSTRACT

The problem of storing large amounts of energy in electronically excited solid helium, briefly He IV, is discussed. Important physical processes in He IV are identified, and a study program is proposed with the aim of making energy storage in He IV a practical reality.

## INTRODUCTION

It is known that the $2^3S_1$ excited state of helium is metastable with a lifetime theoretically estimated at about $10^4$ seconds or 2.3 hours.[1,2] The low atomic mass of the abundant helium isotope (A = 4) and the high excitation energy (~20 eV) of the $He(2^3S_1)$ state make helium an extremely attractive species for energy-storage applications. To achieve large concentrations of stored energy, it is desirable to use liquid or solid phases of helium. The collisional quenching of excited states is expected to be less severe in the solid in view of the greater localization of helium atoms in this phase. For all these reasons, a study of solid helium for energy-storage applications appears to be quite promising and is being initiated at JPL. A preliminary study on solid helium has been made and is presented in this report. The aims of the study were twofold: to identify those physical processes in solid helium which are important in determining its energy-storing properties and, on the basis of this identification, to suggest problems for further detailed study.

In general, one is interested not only in the problem of storing energy in a system but also in the problems of efficiently transferring energy into and from the system. The latter two problems, although very important, are beyond the scope of this report. Indeed, we feel that it is not worthwhile to be concerned about these two problems unless one has reasonable assurance that the system is able to store energy efficiently in the first place.

## ORDINARY AND ELECTRONICALLY EXCITED SOLID HELIUM

Experimentally, it is known that both normal and superfluid helium (He I and He II) can be solidified at sufficiently low temperatures by applying pressures in excess of 25 atm.* The resulting solid, referred to here as *ordinary solid helium* or He III, can have a bcc, hcp, or fcc lattice structure, depending

*25 atm = $25.33 \times 10^5$ N/m$^2$.

on pressure and temperature. All the atoms in He III are, of course, in their ground states ($1^1S_0$). Physical properties of He III have been extensively investigated, both experimentally and theoretically, and excellent reviews and monographs on the subject exist in the literature.[3,4] Our interest here centers on electronically excited solid helium which, as far as we know, has previously not been studied.

The long lifetime of the excited $2^3S_1$ state of atomic helium leads one to expect that samples of solid helium with an appreciable fraction of atoms in this excited state can be produced and maintained "alive" for a considerable length of time, so that these samples may readily be studied experimentally. This expectation is somewhat moderated by the presently unknown effect of the host lattice on the lifetime of an He($2^3S_1$) atom, a point to be investigated.

From a practical point of view, one would like to be able to produce samples of solid helium with most of the atoms excited. This leads one to the notion of a completely electronically excited solid helium, henceforth referred to as He IV, regarded as an idealized version of a highly excited solid helium. One expects that He IV will be a useful theoretical construct, or model, for studying the energy-storage problem in solid helium, much like the electron-gas model is useful for understanding physical properties of real metals. In the remainder of this report we discuss the energy-storage problem in solid helium in the context of the He IV model. The possibility that something closely approximating He IV can be realized experimentally in bulk quantities should not be discounted. Experiments to create He IV will no doubt be attempted if theoretical studies indicate promise.

## PROPERTIES OF He IV

The most fundamental question one can ask about He IV is whether it exists for an appreciable length of time, i.e., whether it is metastable. The answer would be in the affirmative if the lifetime of an He($2^3S_1$) atom, when situated in a lattice of similar atoms, were not drastically different from the lifetime of this atom in isolation. To get some insight into the problem, let us discuss what happens when two isolated excited atoms are brought together. For small interatomic separations, the individual lifetimes of these two atoms will in general be shorter because there is now an extra perturbation (the atom-atom interaction) acting on each atom which can facilitate the $2^3S_1 \longrightarrow 1^1S_o$ transition. This effect is vividly illustrated in the case when the spin magnetic quantum numbers of the two atoms are equal and opposite ($M_S = 1$ and $M_S = -1$). Then, by the electron exchange process $\uparrow\uparrow + \downarrow\downarrow \longrightarrow \uparrow\downarrow + \uparrow\downarrow$, each triplet-state atom can go over into the singlet state, making a transition which is spin-forbidden for atoms in isolation (except for small higher-order effects leading to the observed slow magnetic-dipole photon emission). Clearly, the probability amplitude for the electron-exchange induced $2^3S_1 \longrightarrow 1^1S_o$ transition depends on the degree of overlap of atomic wave functions. It is important to note that this process is inoperative if the spins of the two singlet atoms are aligned. Thus, in the interest of maximal stability, one would prefer to have all spins in a sample of He IV aligned in one direction by applying an external magnetic field.

On the basis of the above discussion, we conclude that two important problems early in the study program of He IV are: first, to compute the potential energy of two He ($2^3S_1$) atoms with parallel spins and, second, to determine the effect of wave function overlap on the lifetime of individual atoms as a function of interatomic separation. The knowledge of the interatomic potential would also provide a basis for estimating the P-V-T properties of He IV.

The possibility that He IV might be ferromagnetic deserves to be investigated, since this question might have an important bearing on the question of meta-stability of He IV. Again, the knowledge of the pair potential is a prerequisite for these investigations.

The low atomic weight of helium and the relatively weak forces between ground-state atoms (beyond the strong repulsive hard-core part of the potential) conspire to make He III a quantum solid, i.e., a solid in which quantum effects are of overwhelming importance. The more open or diffuse electronic cloud structure of the He($2^3S_1$) atom, as compared to He ($1^1S_0$), suggests stronger interatomic He($2^3S_1$) forces. Hence He IV is expected to be more classical than He III. Knowing whether He IV is quantum or classical is important in deciding what mathematical techniques one should use in detailed studies of He IV.

## PHYSICAL PROCESSES IN He IV

In order to estimate the efficiency of He IV as an energy-storing medium, one must understand, first, how energy is stored, and, second, what are the dominant energy loss mechanisms operating in the medium. The first problem is not trivial, contrary to appearances, since excited helium atoms do decay after some time and, if photon losses can be controlled, the emitted photons must come into some sort of equilibrium with the helium atoms and share the burden of energy storage. The physical picture is actually by far more complicated, as we now discuss.

Consider an infinitely large sample of He IV produced at time t = 0. We wish to study the time development of He IV and to inquire what kind of equilibrium states are possible for this system*.

*According to our original definition, He IV is a pure He($2^3S_1$) solid. Nevertheless, it is convenient to be a bit loose and to use the same appellation for the He($2^3S_1$) solid with He($1^1S_0$) and other impurities (excited He atoms in other states, electrons, photons, etc.) present.

A typical excited atom in the He IV lattice can decay to the ground state by emitting a photon with energy ~20 eV. As the photon propagates away from the emitting atom, several things can happen to it. First, it may be absorbed by an encountered ground-state helium atom, one that has itself radiated away a photon earlier. Second, the photon may hit an excited helium atom and photoionize it. Since the ionization potential of an $He(2^3S_1)$ atom is about 5 eV, the ejected electron would have an energy of about 15 eV. Third, the photon may be scattered by helium atoms or ions as well as by photo-emitted electrons.

The 15 eV electrons can interact with helium atoms in a variety of ways. First, they can recombine with helium ions. Second, they can cause further ionization of excited helium atoms. Third, they can scatter, elastically or inelastically, off helium atoms and helium ions. It is clear that an important part of early studies of He IV is to establish which of these various processes are dominant and which can be neglected. The presence of electrons in He IV also raises the interesting question whether they are trapped in "bubbles" as they are in He III up to pressures of 6660 atm.[5†] Should this be the case, electronic processes in He IV would strongly be influenced by the bubble effect.

As a result of the above discussion, one sees that for $t > 0$, He IV is a very complex physical system consisting of He, He* (various excited states), He+, He +*, and electrons and photons of various energies. Clearly, many-body effects are of paramount importance in determining the properties of this system. Once the dominant physical mechanisms have been identified, an important problem will be to determine equilibrium states of the system by solving the rate equations for the various species. Of course, this approach neglects coherent processes, which may be studied later by more comprehensive techniques.

†6660 atm = $6748.25 \times 10^5$ N/m$^2$.

The assumption of an infinite sample of He IV made in the preceding discussion is of course an idealization, and in practice samples of finite size must be considered. This immediately raises the question of the unwanted outflow of energy across the walls enclosing a finite sample of He IV. If one had perfectly opaque walls, to heat transfer and to radiation of every wavelength, then the problem of energy storage would be solved. One would need only to put the medium into a box with perfectly opaque walls, excite it, and leave it alone until energy extraction is called for. In practice, walls are lossy, and hence the major problem of energy storage is how to choose the characteristics of the system (and the enclosure) so as to minimize the losses. To this end, an understanding of the inherent energy loss mechanisms in He IV is required.

There are basically two types of losses one must contend with: radiation and phonon. As discussed earlier, photons emitted by excited helium atoms have energies ~20 eV and thus wavelengths in the vacuum ultraviolet. Since there do not exist efficient reflectors at these wavelengths, other means must be sought to minimize energy losses due to the 20 eV photon efflux from the system. One possibility is dynamic radiation trapping of the type discussed by Bykov[6] caused by externally induced (e.g., by acoustic perturbations) periodicites in He IV. It is interesting to note that the problem of establishing a periodic structure in liquid helium is of great current interest in connection with NASA's distributed-feedback superfluid helium UV laser program. The technology generated by the UV laser research would undoubtedly be useful for He IV energy-storage applications.

Finally, an understanding of phonon processes in He IV is important for gaining insight into the heat generation and conduction properties of He IV and thus for estimating the rate of the degradation of stored energy into heat. For this purpose, one needs estimates of the phonon-electron, phonon-photon, and phonon-phonon coupling strengths. A rather detailed model of He IV is necessary before such estimates can be made reliably.

## SUMMARY

In the preceding discussion we have examined some of the important physical processes in He IV and have identified a number of problems which should be investigated in order to establish the feasibility of using He IV as an energy-storing medium. The problems, listed very roughly according to their presently estimated priorities, are the following:

1. $He(2^3S_1) - He(2^3S_1)$ potential for parallel spins.
2. Effect of wave function overlap on the $He(2^3S_1)$ lifetime.
3. Lifetime of an $He(2^3S_1)$ atom in a lattice of similar atoms.
4. Quantum or classical nature of He IV.
5. P-V-T properties of He IV.
6. Dominant electron and photon processes in He IV.
7. Existence of electronic bubbles.
8. Equilibrium state of He IV with electron and photon processes taken into account.
9. Radiation trapping.
10. Phonon losses.
11. Ferromagnetism in He IV.

Although in this report we have dealt exclusively with theoretical aspects of solid-helium research, it is clear that experimental work is desirable and should be undertaken in those areas which are indicated as promising by theoretical studies.

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