Semi-annual summary research report in physics

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Iowa State College

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Iowa State College

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UNITED STATES ATOMIC ENERGY COMMISSION
RESEARCH AND DEVELOPMENT REPORT

SEMI-ANNUAL SUMMARY RESEARCH REPORT IN PHYSICS

For January through June, 1958

by

Ames Laboratory Staff

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Iowa State College
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SEMI-ANNUAL SUMMARY RESEARCH REPORT IN PHYSICS

For the Period January through June, 1958

This report is prepared from material submitted by the group leaders of the Laboratory

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ISC-758
ISC-833
ISC-901
ISC-975
1. **Shape of $^{32}$P Beta Spectrum (B. C. Carlson)**

Several corrections to the theoretical spectrum of $^{32}$P beta decay have been investigated. There still remains a discrepancy between the theoretical shape ($1 - 0.04 \, W$) for VA interaction and the experimental shape ($1 - 0.01 \, W$) found at this Laboratory and at the Argonne National Laboratory.

The modification of the vector interaction proposed by Feynman and Gell-Mann seems unlikely to have an appreciable effect in $^{32}$P decay. The correction for "weak magnetism" is unimportant in this case unless the transition magnetic moment is large compared to the allowed beta-decay matrix element. Both quantities are $\ell$-forbidden, but their ratio is expected to be close to unity, aside from possible exchange-current effects. The latter cannot be ruled out completely until the lifetime and branching ratio of the analogue state in $^{32}$S (at 7 Mev) are measured.

The relativistic corrections given by Alaga, Sips, and Tadic\textsuperscript{1} have been evaluated for $^{32}$P; the only terms that have a significant

\textsuperscript{1} Nuclear Physics \textbf{6}, 305 (1958).
magnitude are almost exactly independent of energy. These terms have been checked by a method that requires only a differentiation of the radial wave functions of the electron.

Interference terms between the two principal second-forbidden matrix elements from the A interaction have been calculated. They produce a slight effect in the wrong direction to improve agreement with the experimental slope.

The non-relativistic equivalent of \( \int \rho \) is found to be

\[
\int \rho + \frac{1}{2M^2} \int \rho \cdot \mathbf{p} \cdot \mathbf{p},
\]

where the second term comes exclusively from the small components of the nucleon wave function. For a harmonic oscillator potential, \( \frac{\mathbf{p} \cdot \mathbf{p}}{M^2} \) can be replaced by \( \omega \mathbf{r} \mathbf{r} \) (provided that the number of oscillator quanta does not change in the transition), where \( \omega \) is the oscillator frequency and is approximately 20 in natural units. Further calculation shows that the second term has the value 0.013 while the experimental fit value requires the sum to have magnitude 0.008. The signs must be opposite to fit the observed spectrum shape; hence the first term, which would be zero without configuration mixing, is -0.021. If the \( \text{P}^3\text{P}^2 \) nucleus is spherical, the only configuration admixtures that seem likely to contribute to the decay are a \( \text{P}^3\text{P}^2 \) configuration with a d5/2 proton excited to the s1/2 shell, and possibly an \( s^3\text{P}^2 \) configuration with an s1/2 proton and a d5/2 neutron both excited to the d3/2 shell.
A detailed report (ISC-1006) "Shape of the Phosphorus 32 Beta Energy Spectrum" by George Henton and B. C. Carlson was distributed.

2. **Interatomic Exchange** (J. M. Keller)

Exchange interactions between electrons give rise to a spin dependent coupling of the type \( \mathbf{s}_1 \cdot \mathbf{s}_j \), where \( \mathbf{s}_1 \) indicates the spin on electron 1, etc. It is generally believed that there is a corresponding coupling between atoms of the type \( \mathbf{S}_A \cdot \mathbf{S}_B \), which is simply the sum of electron exchanges between the atoms, since each electron of one atom can exchange in turn with each electron of the other atom. (\( \mathbf{S}_A \) is the total spin of atom A, etc.) Our investigation showed that while the dominant exchange coupling is indeed of this type, there are added interactions, due to the simultaneous exchange of two or more electrons between atoms. These give rise to small additional terms of the type \( (\mathbf{S}_A \cdot \mathbf{S}_B)^2 \), etc.

3. **Massless Particles—Anomalous Types** (C. L. Hammer and R. H. Good, Jr.)

In the usual theories of particles with zero rest-mass the wave function is built up from the independent components of a symmetrized spinor which has all indices of the same type. The theories of the two-component neutrino and photon are in this class. Such a theory describes a particle with a definite spin and which can exist in two states: spin parallel or
antiparallel to the momentum. However there also exist wave
equations involving spinors with mixed indices and the properties
of the corresponding particles are being studied. So far the
two-index and three-index cases have been considered. They
correspond to particles of mixed total spin and component
of spin in the momentum direction $0$ and $\pm \frac{1}{2}$ respectively.
The theories can be quantized and the usual connection between
spin and statistics is found.

4. Interpretation of Magnetic Properties of Dysprosium (S. Legvold
and R. H. Good, Jr.)

Dysprosium is ferromagnetic below $85^\circ$K, antiferromagnetic
between $85$ and $178^\circ$K, and paramagnetic above $178^\circ$K. The
magnetic properties in all three regions have been measured by
Behrendt, Legvold, and Spedding. It has been found possible
to interpret all the experimental results on the basis of a
two-sublattice model with the spontaneous magnetic moment,
anisotropy, and exchange interaction between the sublattices
chosen to fit the data at each temperature. Detailed agreement
for the magnetization curves in the ferromagnetic and
antiferromagnetic regions is obtained, giving confidence in the
model.

Experimental Physics

1. Nuclear Studies

1.1 180° Permanent-Magnet Spectrometer (E. N. Jensen)

A 180° permanent magnet spectrometer of the Brookhaven Type with a magnetic field of about 100 gauss has been constructed and put into operation. Some of the transition energies in the decay of Yb$^{169}$ have been determined with this instrument. The Yb$^{169}$ was evaporated from a hot filament onto a 4 mil tungsten wire. A source of Th(B+C+C") was then collected on the wire. The A, F, and I internal conversion lines were used for calibration purposes. Calibration marks on the camera and their images on the film were used to correct for the shrinkage of the film. The energies of some of the transitions in Tm$^{170}$ as determined with the 180° spectrometer are listed below and compared with those determined by Hatch, Boehm, Marmier, and DuMond$^1$ with a curved-crystal gamma-ray spectrometer. The agreement seems to be quite good.

Tm$^{169}$ Transition Energies in kev

<table>
<thead>
<tr>
<th>Line</th>
<th>180° Spectrometer</th>
<th>Hatch et. al</th>
</tr>
</thead>
<tbody>
<tr>
<td>63L$_1$</td>
<td>63.05</td>
<td></td>
</tr>
<tr>
<td>63L$_2$</td>
<td>63.07</td>
<td>63.12</td>
</tr>
<tr>
<td>63L$_3$</td>
<td>63.03</td>
<td></td>
</tr>
<tr>
<td>93K</td>
<td>93.56</td>
<td></td>
</tr>
</tbody>
</table>

$^1$ Hatch, Boehm, Marmier, and DuMond, Phys. Rev. 104, 745 (1956).
1.2 **Electron Beam Control** (C. L. Hammer and L. J. Laslett)

A paper entitled "Electron Beam Control in a Conventional Synchrotron" by C. L. Hammer and L. J. Laslett was accepted for publication in the proceedings for the Second Geneva Conference for the Peaceful Uses of Atomic Energy. (A/Conf. 15/P/726)

**Abstract**

Precise control of the rate at which the electron beam of a conventional weak focusing synchrotron strikes
a target internal to the synchrotron vacuum chamber is sometimes essential to the performance of a physical measurement. For example, the excitation and measurement of short half lives or neutron time of flight measurements demand a beam burst that is as short as possible, sometimes of the order of milli-microseconds. At the other extreme, coincidence measurements between nuclear fragments from the photo-dissociation of nuclei demand an extended beam burst of the order of milliseconds. In addition to these considerations, it is also desirable to be able to extract the electron beam from the synchrotron with as high a yield as possible. Toward these ends perturbation techniques have been developed and tested using the Iowa State College 75-Mev Synchrotron as a model.

To obtain short beam bursts two methods have been employed, one using a half integral resonance and the other using an integral resonance. The first method has the advantage of causing the electron beam to expand onto a target in approximately ten revolutions. Furthermore, this expansion is azimuthally dependent making the method also useful for beam extraction. The second method, while causing beam destruction more slowly than the first (about 5 microseconds) is more applicable to

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activation experiments where precise control of the direction of the resulting Bremsstrahlung is desirable. Both methods utilize a magnetic deflection scheme.

The usual method for obtaining protracted beam pulses by amplitude modulation of the synchrotron rf system is not applicable to the lower energy machines since radiation losses are low. It is found, however, that frequency modulation of the rf system at the peak of the magnetic guide field gives beam pulses of the order of two milliseconds duration. This is to be compared to a total acceleration time of six milliseconds.

1.3 The Decay of Tb$^{156}$ (L. J. Laslett)

In the activity observed after irradiation of terbium by 64-Mev bremsstrahlung, 18 transitions have been detected, with energies between 89 kev and 2060 kev. The lowest eight of these agree exactly with transitions found by Mihelich et al.\textsuperscript{1} to occur in Gd$^{156}$ with energies of 89.10, 111.9, 155.2, 199.4, 262.7, 296.7, 356.6, and 422.2 kev. Of the remaining ten, which have energies of 534, 767, 934, 1061, 1143, 1216, 1423, 1650, 1826, and 2060 kev, six agree well with lines seen by Handley and Lyon.\textsuperscript{2} This agreement in energy, the similarity

\textsuperscript{1} Mihelich, Harmatz, and Handley, Phys. Rev. 108, 989 (1957).
of the spectrum when observed in coincidence with the K x-radiation from gadolinium, and the absence of any evidence for beta decay or positron emission all lend support to the belief that the transitions occur in Gd$^{156}$ following orbital electron capture by Tb$^{156}$ with a half-life of 5.6 days.

The absence of annihilation radiation suggests little, if any, positron emission, and no evidence of beta decay was found. The spectrum observed with the anthracene crystal is attributed to the production of secondary electrons by electromagnetic radiation from the source material.

The relatively small fraction of 5.5-hr. activity, although surprising at first, is believed to be due in large measure to the thickness of the sources and the softness of the radiations associated directly with the Tb$^{156m}$ state. Mihelich et al.\(^1\) having shown that the isomeric transition involves strong L-conversion.

It seems possible that some of the gamma-ray lines reported by Dillman, et al.\(^2\)\(^,\)\(^3\) and not observed in this investigation may have originated in nuclides other than Tb$^{156}$ due to the high energy of the bremsstrahlung employed in their irradiations.

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1.4 Lifetime of Positrons in Metals (M. G. Stewart)

A fast coincidence circuit of the Bell, Graham, Petch type\textsuperscript{1} has been constructed and is to be used for measuring the lifetime of positrons in various metals. As a check on the apparatus the mean life of positrons in teflon at room temperature was measured. A value of $\tau_2 = 3.2 \times 10^{-9}$ seconds was obtained for the long lived component in good agreement with the value reported by Bell and Graham\textsuperscript{2}.

The mean life in pure yttrium metal was measured to be $\tau \sim 2.0 \times 10^{-10}$ seconds and in $\text{Na}_x\text{WO}_3$ ($x = 0.789$), $\tau \sim 2.6 \times 10^{-10}$ seconds. These values were obtained by a comparison to the lifetime in aluminum which was assumed to be $1.5 \times 10^{-10}$ seconds.

1.5 Photoprotons from Be\textsuperscript{9} (M. G. Stewart)

The photoproton yield from Be\textsuperscript{9} was measured by observing the beta activity of the daughter nucleus, Li\textsuperscript{8}. The X-ray energy from the Iowa State College Synchrotron was varied in small steps from the threshold for this reaction (16.9 Mev) up to 60 Mev. Several breaks in the yield curve near the low energy end were observed, indicating resonance absorption.

\textsuperscript{2} Bell and Graham, Phys. Rev. \textbf{90}, 644 (1953).
into levels of Be$^9$. More data are being taken to enable the energies of these levels to be more accurately determined.

1.6 Nuclear Spectroscopic Studies of Ba$^{133}$ and Xe$^{133}$ Decay (D. Lu)

The study on the decay scheme of Ba$^{133}$ - Cs$^{133}$ was continued. Essential results were reported$^1$ at the Am. Phy. Soc. Washington meeting of 1958. Some aspects of the experiment were repeated afterwards to check on possible discrepancies. To study the problem further, the Xe$^{133}$ - Cs$^{133}$ decay was studied with a strong Xe$^{133}$ source. Coincidence studies on the $\gamma$-rays with scintillation counters reveal the existence of two 81 kev $\gamma$-rays in cascade, instead of only one as has been previously accepted.$^2$ The 162 kev cross-over transition was too weak to be positively identified. These results are in agreement with the results of the Ba$^{133}$ study.

2. Solid State Studies

2.1 Thermal Diffusivity and Electrical Resistivity of Copper (G. C. Danielson)

The thermal diffusivity and electrical resistivity of Johnson-Matthey copper (spectrographically pure) have been measured in the temperature range 50°C to 1000°C. The data

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are shown in Fig. 1 and Fig. 2. As shown in Fig. 1, the maximum deviation from the curve of any single measurement in thermal diffusivity is less than two percent. The electrical resistivity values (Fig. 2) are accurate to better than one percent.

The electrical resistivity does not vary exactly linearly with temperature. The increase in resistance at high temperatures is undoubtedly caused by expansion of the lattice. The increase in distance between atoms reduces the force constant between atoms and thus reduces the Debye temperature. According to free electron theory, the resistivity varies inversely as the square of the Debye temperature, and hence the resistivity will be increased.

The non-linear decrease in thermal diffusivity at high temperatures is likewise probably caused by expansion of the lattice, since the thermal diffusivity varies directly as the thermal conductivity, which varies inversely as the resistivity according to the Wiedemann-Franz law. The non-linear decrease in the thermal diffusivity from 50°C to 300°C may be caused by the relatively high Debye temperature (340 K) of copper. The specific heat at constant volume would still be increasing slightly at room temperature. This increase would decrease

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1 Mott and Jones, Theory of Metals and Alloys, page 268.
Fig. 1. Thermal Diffusivity of Copper
Fig. 2. Electrical Resistivity of Copper
the thermal diffusivity, which varies inversely as the specific heat. The approximate linear decrease in thermal diffusivity from 50°C to 1000°C is probably caused by the approximate linear increase in specific heat at constant pressure (even though the specific heat at constant volume were constant).

2.2 Infra-red Transmission of Mg$_2$Si (G. C. Danielson)

The infra-red transmission spectra of magnesium silicide has been studied at temperatures from 85°K to 370°K. The absorption edge, corresponding to the energy gap between the valence band and the conduction band, occurred at a wavelength between 1.7 and 2.0 microns. The shift in absorption edge with temperature coefficient has been measured for two samples, and the temperature coefficient of the energy gap calculated in each case.

We assume that the energy gap (E$_g$) decreases linearly with increasing temperature (T). Thus, $E_g = E_0 - AT$, where $E_0$ is the energy gap at 0°K, and A is the temperature coefficient of the energy gap. For Mg$_2$Si sample 28B04, we found $E_0 = (0.793 \pm 0.004)$ ev, $A = (4.00 \pm 0.15) \times 10^{-4}$ ev per degree K. For Mg$_2$Si sample 23B01, we found $E_0 = (0.789 \pm 0.006)$ ev, $A = (4.10 \pm 0.24) \times 10^{-4}$ ev per degree K. From Hall data, Morris, Redin and Danielson$^1$ obtained $E_0 = (0.78 \pm 0.20)$ ev.

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An absorption peak at 0.40 ev was observed in all samples. The height of this peak increased with increasing temperature; this may be due to an electron trapping level located near the middle of the energy gap.

In the long wavelength region (7.5 to 12.5 microns) the absorption was directly proportional to the square of the wavelength as predicted by the Drude theory for absorption by free charge carriers.

2.3 Thermoelectric Power of Mg₂Si (G. C. Danielson)

Additional measurements have been made of the thermoelectric power of Mg₂Si from 300°K to 1000°K. The results indicate that the electrical properties of Mg₂Si may be altered by prolonged heating while measurements are being made. For this reason the previously reported\(^1\) value of 4.3 for the ratio of electron-to-hole mobility in the intrinsic range is not valid. Measurements made using a method which does not subject the crystals to prolonged heating yield a reproducible value of 3.0 for the mobility ratio. From these results it may be concluded that heat treatment of Mg₂Si crystals may introduce acceptor defects, which contribute holes and reduce the number of conduction electrons provided by the donor impurities. This same

\(^1\) ISC-975, page 31 (1958).
effect takes place in germanium; but, in silicon or boron\textsuperscript{1}, a heat treatment increases the number of electrons rather than the number of holes. In Mg\textsubscript{2}Si the increase in the number of holes resulting from heating of the crystal could be caused by a change in stoichiometry due to loss of magnesium. Another possible explanation is the introduction of acceptor defects by diffusion of copper from the electrodes into the crystals. Such a process has been reported by Fuller and Whelan\textsuperscript{2} to take place in semiconducting crystals.

2.4 Measurement of Hall Mobility at Microwave Frequencies

(G. C. Danielson)

The use of microwave power for Hall mobility measurements in semiconductors was first suggested by W. J. Spry of National Carbon Research Laboratories and preliminary measurements have been reported\textsuperscript{3}. This work is being continued by Nishina in this Laboratory. Complete microwave equipment has been assembled, and the theory has been further developed.

A microwave measurement has the following possible advantages over a direct current measurement. (a) No wires or probes need to be attached to the specimen. Such attachments can be a major

\textsuperscript{1} Shaw, Hudson, and Danielson, Phys. Rev. 107, 419-427 (1957).
problem for small single crystals when heated (or cooled) over a large temperature range. (b) No appreciable temperature rise in the sample will occur with the low microwave power level employed. (c) With a high Q cavity ($Q \sim 6000$), the sample can be as small as a few millimeters in diameter. The thickness needs to be only a few times the skin depth. (d) In some semiconductors, the relaxation time of the charge carriers at low temperatures is comparable to the period of the microwave field. In such cases, it may be possible to determine the relaxation time from the difference between the microwave Hall mobility and the dc Hall mobility.

The principle of the measurement, as suggested by Spry, is shown in Fig. 3. The TE$_{101}$ rectangular dual mode cavity is the central part of the experimental equipment. In the cavity space two distinct modes of oscillation may be excited separately at a single resonant frequency. The first mode has the electric field ($E_y$) in the $Y$-direction, and can be excited only by the power ($P_{in}$) from the klystron. The second mode has the electric field ($E_z$) in the $Z$-direction, and will be excited by the Hall effect in the sample. A part of this power can be coupled out to waveguide 2, but not out to waveguide 1. The semiconductor sample is placed on the cavity wall ($Y-Z$ plane) with a good conducting paint.
Fig. 3. Dual mode cavity for microwave Hall mobility measurements
When the microwave power $P_{in}$ is fed into the cavity from waveguide 1, the first mode only is excited and no microwave power comes out waveguide 2. When a magnetic field $B_x$ is applied in the X-direction, however, the Hall effect gives rise to an electric field $E_z$ and the second mode is excited. Some of the power in the second mode is coupled out into waveguide 2. The power ratio $P_{out}/P_{in} = (1/4)(B\mu^2)$, where $\mu$ is the mobility of the charge carriers and $\gamma$ is a constant depending upon the cavity geometry and upon the particular sample being studied.

Nishina and Spry carried out some measurements on n-type germanium at room temperature and at 9121 megacycles per second. The dc mobility was 2670 cm$^2$/volt-sec, the dc resistivity was 16 ohm-cm. The microwave mobility was 2900 cm$^2$/volt-sec with an estimated accuracy of 15 per cent.

Recent work at this Laboratory has involved a more accurate calculation of the constant $\gamma$. The theoretical calculations have now been completed under the following assumptions. (a) The sample does not appreciably perturb the field distribution in the cavity. (b) The sample is isotropic and only one type of carrier contributes to conduction. (c) The field in the sample can be expressed in terms of plane waves. (d) The magnetic field $B$ is sufficiently small so that $B\mu << 1$. (e) The dual
mode cavity has perfect symmetry ($P_{in}$ could come in by waveguide 2, and $P_{out}$ leave by waveguide 1).

2.5 **Electrical Resistivity of Na$_x$WO$_3$ (G. C. Danielson)**

When the electrical resistivity of sodium tungsten bronze (Na$_x$WO$_3$) was plotted as a function of sodium concentration, both Brown and Banks$^1$ and Gardner and Danielson$^2$ found a minimum in the resistivity at a sodium concentration corresponding to $x = 0.75$. The minimum was conspicuous at all temperatures, even at 0°K. The appearance of this minimum has never been successfully explained. Brown and Banks suggested the possibility of an anomaly in the number of charge carriers at $x > 0.75$; but the Hall data of Gardner and Danielson showed conclusively that no anomaly existed at $x = 0.75$, the number of charge carriers was exactly equal to the number of sodium atoms for all values of $x$ from $x = 0.58$ to $x = 0.90$. Juretschke$^3$ suggested a relationship between the resistivity minimum in the bronzes and the resistivity minimum of sodium when the resistivity was plotted as a function of pressure. Gardner and Danielson showed this hypothesis to be untenable, since the minimum

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persisted down to 0°K. Vest, Griffel and Smith\textsuperscript{1} measured the electronic specific heat of the bronzes and found an increase in the density of states at $x > 0.75$. The effect was too small however, to explain the observed minimum in the resistivity at $x = 0.75$.

Gardner and Danielson postulated the existence of long range order for the sodium atoms. In particular, they looked for a superlattice by X-ray diffraction; but the scattering by the tungsten was too great relative to the scattering by the sodium to observe the superlattice, if one existed. Recently, Atoji and Rundle\textsuperscript{2} have discovered a superlattice by neutron diffraction, a cube with twice the lattice parameter has sodium atoms consistently absent at the corner and body center positions. The ordering was increased with a decrease in temperature. (The small displacements of the oxygen atoms observed by Atoji and Rundle strongly suggest that the Na$_x$WO$_3$ crystal was not strictly cubic, in confirmation of the report by Ingold and DeVries\textsuperscript{3}.) The ordering has not been studied as a function of sodium concentration, but it is doubtful if the ordering alone can account for the large decrease in resistivity at $x = 0.75$.

\textsuperscript{2} M. Atoji and R. E. Rundle, Private communication.
\textsuperscript{3} J. H. Ingold and R. C. DeVries, General Electric Report 57-RL-1821.
We have now found an additional explanation, which is very likely to be the dominant cause of the observed minimum in resistivity. The crystals have been found to be ordinarily non-homogeneous, especially at values of $x$ far from $x = 0.75$. The non-homogeneity results in a high resistivity, frequently many times the resistivity of a homogeneous crystal. The minimum in resistivity then results from the crystals with $x$ below $x = 0.75$ and with $x$ above $x = 0.75$ being more non-homogeneous than those with $x = 0.75$. Preliminary measurements on homogeneous crystals indicate that the resistivity minimum is not conspicuous and may be absent altogether.

The cause of the non-homogeneity in $\text{Na}_x\text{WO}_3$ crystals has not yet been determined. It may be partly due to non-homogeneity in ordering of the sodium atoms. At present, we believe, however, that macroscopic defects, inclusions, and impurities may be chiefly responsible. Better crystals are being grown by several different methods and highly uniform crystals are being selected for further resistivity measurements.

2.6 Mass Spectrometric Studies of Rare Earth Cohesive Energies (D. E. Hudson and F. H. Spedding)

Latent heats of sublimation were determined for several of the rare earths. In the following the numbers given are the latent heats of sublimation reduced to 298°K.
Eight additional runs on europium gave a value of $42.60 \pm 0.4$ kcal/mole in good agreement with our best previous value of $42.70 \pm 0.40$ kcal/mole and close to the value of 41.9 kcal/mole obtained by Spedding, Hanak, and Daane of this Laboratory.

Erbium and holmium have been measured for the first time anywhere as far as is known. Our results are: erbium (14 runs), 75.8 $\pm$ 0.4 kcal/mole and holmium (13 runs), 74.5 $\pm$ 0.7 kcal/mole. Previous values of their sublimation enthalpies (about 70 $\pm$ 7 kcal/mole) are only estimates and are not suitable for exact experimental comparisons.

Gadolinium has been studied and nine runs give a preliminary value of 81.3 $\pm$ 0.4 kcal/mole. No previous experimental results are available for comparison.

2.7 Surface Conductivity of Diamond (G. C. Danielson)

A diamond (number D-3) was heated for two hours at a temperature of 1000°C and a pressure of $2 \times 10^{-6}$ mm of Hg. Before the heat treatment, the resistance of the diamond in air and at room temperature was greater than $10^{12}$ ohms; after the heat treatment the resistance in air and at room temperature was $10^5$ ohms. When the heat-treated diamond was heated for five minutes in air at 400°C, the resistance of the diamond returned to its high original value. The effect was reproducible with the same diamond.
We have shown that this effect is caused by a thin film of carbon, which is produced at 1000°C by graphitization of the diamond, and which is removed at 400°C by oxidation. After heat treatment at 1000°C, the electron diffraction pattern showed broad diffuse bands that corresponded to the 100 and 110 planes of graphite. The pattern was similar to the electron diffraction patterns of evaporated carbon obtained by Blue and Danielson¹. After the heat-treated diamond had been heated in air at 400°C, the diffraction pattern characteristic of carbon was absent.

After the heat treatment, the diffraction pattern showed very faintly several lines due to SiO₂. The SiO₂ undoubtedly came from the fused silica container in which the diamond was heated. After the carbon was removed (by five minutes of heating at 400°C), the SiO₂ diffraction lines were much more numerous and much more intense. We conclude, therefore, that the amorphous carbon, formed by graphitization of the diamond at 1000°C, must diffuse over the entire diamond surface and cover almost completely any surface contamination such as SiO₂. This conclusion (that carbon atoms have substantial mobility at 1000°C) is consistent with the observation by

Blue and Danielson that the annealing of arc-evaporated carbon films became essentially complete at 1000°C.

2.8 Carrier Trapping in Diamond (D. E. Hudson)

The conduction properties of diamond are being exploited to study directly the trapping levels responsible for carrier capture in diamond crystals. We previously reported a rapid rise in the photoelectric detrapping cross section for photons of 3.0 e.v. energy. We have extended the measurements to 3.4 e.v. and find a continuation of this rapid rise. The most direct interpretation of these phenomena is in terms of the existence of rather deep trapping levels in the band structure. Some recent reports in the literature indicate that gold may be the impurity responsible for the deep traps.

2.9 Superconducting Transition Temperatures

2.9.1 Rare Earth Alloys (S. Legvold and F. H. Spedding)

A sizeable shift in the superconducting transition temperature of lanthanum is observed when a small amount of gadolinium is added to the melt. A half percent of gadolinium shifts the transition temperature from 5.85°K to 3.80°K. It would appear that magnetic impurities of this type could well account for the different transition temperatures for lanthanum reported by various investigators.

2.9.2 Effects of Pressure (L. D. Jennings and C. A. Swenson)

A paper entitled "The Effects of Pressure on the Superconducting Transition Temperatures of Sn, In, Ta, Tl and Hg" by L. D. Jennings and C. A. Swenson was submitted for publication in PHYS. REV.

ABSTRACT

Solid para-hydrogen has been used as a transmitter of approximately hydrostatic pressure to study the effects of pressures up to 10,000 atmos on the superconducting transition temperatures of polycrystalline tin, indium, tantalum, thallium and mercury. The technique which was used allowed an approximate evaluation of the effects of sample deformation and pressure gradients, and the results are considerably more accurate than the high pressure data previously available. The transition temperature data for tin and indium showed considerable curvature when plotted vs pressure, but gave a roughly linear relationship when plotted against volume. No curvature was found for tantalum. The thallium data agree qualitatively with previous work, and show a maximum in the transition temperature vs pressure curve at about 2000 atmos. The
mercury results were anomalous in that two distinct transition temperature vs pressure curves (with different zero pressure transition temperatures) were found; one when the pressure was kept below 4000 atmos, and the other when the sample was cycled from zero to 10,000 atmos. Other experiments have shown that these results are due to two different modifications of solid mercury, each of which can exist at liquid helium temperatures.

2.10 Magnetic Phenomena (S. Legvold and F. H. Spedding)

2.10.1 Gadolinium Carbide and Terbium Carbide

The magnetic properties of Gd$_3$C and Tb$_3$C have been investigated. Results to date have indicated that Gd$_3$C is ferromagnetic with a Curie point higher than the Curie point of pure Gd. Measurements on Tb$_3$C indicate that this material is antiferromagnetic.

2.10.2 Terbium

A paper entitled "The Magnetic Properties of Terbium Metal" by W. C. Thoburn, S. Legvold and F. H. Spedding was submitted for publication in PHYS. REV.

ABSTRACT

Results of magnetic measurements on metallic terbium over the temperature range from 4° to 375°K and in fields
The metal is found to have a paramagnetic Curie point of 237°K. Tests with fields of from 50 to 800 Oe indicate an order-to-disorder transition at approximately 230°K which is evidently a Néel point. It appears that terbium has a weakly bound antiferromagnetic ordering for temperatures between 218° and 230°K, this ordering being gradually changed to ferromagnetic in fields exceeding 200 Oe. Below 218°K the metal is ferromagnetic. The magneton numbers obtained from the paramagnetic susceptibility measurements and from the extrapolated ferromagnetic saturation moment are in good agreement with values calculated for the free ion.

2.10.3 Hall Effect in Rare Earths

A paper entitled "Hall Effect in Sm, Tm, Yb and Lu" by G. S. Anderson, S. Legvold and F. H. Spedding was submitted for publication in PHYS. REV.

**ABSTRACT**

The Hall effect in Lu, Yb, Tm, and Sm has been studied as a function of temperature for the temperature range 40°K to 320°K in a magnetic field of 5,500 Oe. The Hall coefficient for Yb is positive, while the Hall coefficients for Lu and Tm are negative. Sm has a negative Hall effect at room temperature, but at lower temperatures the sign apparently changes.
2.11 **Heat Transfer Studies (S. Legvold)**

2.11.1 **Heat Transfer in Gases**

A paper entitled "Electroconvectional Heat Transfer in Gases" by Sigurds Arajs and Sam Legvold was submitted for publication in J. CHEM. PHYS.

**ABSTRACT**

Electroconvectional heat transfers have been measured in gaseous \( \text{N}_2, \text{CO}_2, \text{NH}_3, \text{SO}_2 \) and \( \text{CCl}_3\text{F} \). The results are interpreted and correlated according to the theories of Kronig and of Senftleben and Bultmann. The theoretical predictions of Senftleben and Bultmann concerning the dependence of the electroconvectional heat transfer on the pressure of the gas and the magnitude of the electric field are found to fit the experimental results.

2.11.2 **Heat Transfer from a Wire**

A paper entitled "Free Convectional Heat Transfer from a Single Horizontal Wire" by Sigurds Arajs and Sam Legvold was submitted for publication in J. CHEM. PHYS.

**ABSTRACT**

Free convectional heat transfer from single, horizontal, heated, 2 and 3 mil platinum wires was measured in gaseous \( \text{N}_2, \text{O}_2, \text{CO}_2 \) and in liquid \( \text{(CH}_3\text{)}_2\text{CO}, \text{CH}_3\text{OH}, \text{C}_6\text{H}_5\text{CH}_3 \). The measurements, extending approximately from \( \text{GrPr} = 1 \) to \( \text{GrPr} = 10^{-5} \), satisfied the Senftleben correlation.
2.11.3 Senftleben Method

"Comments on the Senftleben Method for the Determination of the Heat Conductivity, the Specific Heat and the Viscosity of Gases" by Sigurds Arajs and Sam Legvold was sent to J. APPL. PHYS. for publication as a Letter to the Editor, and is reproduced here in its entirety.

In 1953 Senftleben\(^1\) suggested a new method for the simultaneous determination of the molecular heat conductivity, the specific heat at constant pressure, and the viscosity of gases as a function of temperature. The method is based on the heat transfer measurements from a horizontal electrically heated wire in a standard gas with known thermal properties and in a gas whose thermal properties are to be determined. Recently a need for viscosity information on some gases led us to examine the Senftleben method. It was found that the proposed technique has a serious shortcoming with respect to the viscosity determination.

The equation from which the coefficient of viscosity can be calculated contains a factor \(a = \frac{Pr^X}{Pr^S}\), where \(Pr = \frac{c_p \eta}{\lambda}\) is the Prandtl number and \(c_p, \eta\) and \(\lambda\) represent the specific heat at constant pressure, the coefficient of viscosity and the molecular thermal

---

conductivity, respectively. The symbol X refers to the unknown gas and the symbol S refers to the standard gas. On the basis of available experimental information and using CO₂ as a standard gas, Senftleben proposed a curve \( a = f(\frac{\lambda_X}{\lambda_{CO₂}}) \) shown in Fig. 4 which should fit data for all gases. By computing the values of \( \frac{Pr_X}{Pr_{CO₂}} \) and \( \frac{\lambda_X}{\lambda_{CO₂}} \) for some gases at various temperatures as indicated in Fig. 4, we have found that data for many gases do not fall on the curve. The deviation for H₂ was so large it was impossible to represent \( a = f(\frac{\lambda_{H₂}}{\lambda_{CO₂}}) \) on Fig. 4; e. g., at 220°K \( \frac{Pr_{H₂}}{Pr_{CO₂}} = 0.876 \) and \( \frac{\lambda_{H₂}}{\lambda_{CO₂}} = 12.9 \). The values of \( \lambda, \rho \) and \( \eta \) for A, CO, H₂, N₂ and O₂ were obtained from the National Bureau of Standards Circular 564.¹ For the other gases the corresponding information was obtained from a Du Pont Technical Bulletin² and Lange's Handbook of Chemistry.³

Although the Senftleben method cannot provide reliable values of the viscosity of gases, our examination using O₂, N₂ and A showed that the coefficient of the molecular heat

Fig. 4. Senftleben's correlation curve compared with experimental data for a number of gases.
conduction and the specific heat at constant pressure could be determined with deviations of about \( \pm 5\% \) from the corresponding values obtained by the standard techniques.

2.12 Magnetic Field and Gradient Meter

A paper by W. C. Thoburn entitled "A Portable Magnetic Field and Gradient Meter" was submitted for publication in REV. SCI. INSTR.

ABSTRACT

A portable instrument for measuring magnetic fields and field gradients is described. The measurements depend on comparing the alternating emf's induced in two search coils oscillated synchronously at about 3 cps through angles of less than 3°, one in the field to be measured and the other in the reference field of a permanent magnet in the instrument. A precision helipot is used as a potentiometer, in which a measured fraction of the signal from the reference coil is balanced against the signal from the probe coil. The null is obtained through an electronic amplifier and galvanometer. A field gradient can be measured with the same instrument by use of a search coil which is given translatory oscillations in the direction of the gradient.
2.13 Phase Transitions (C. A. Swenson)

2.13.1 Cerium

A paper entitled "The Temperature Dependence of the Phase Transition in Cerium" by R. Herman and C. A. Swenson was submitted for publication in J. CHEM. PHYS.

ABSTRACT

The cubic-condensed cubic transition in cerium has been observed as a function of temperature and pressure from 360°K and 11,000 atmos to 78°K and zero pressure. The data which were obtained definitely link the high pressure, room temperature and the low temperature, zero pressure transitions which have been observed previously. While the data show some unexplained features, the calculated value for the latent heat of transition at 288°K (1150 ± 100 cal/mol) agrees as to order of magnitude with a directly measured value (880 ± 40 cal/mol) by other workers.

2.13.2 Mercury

A paper entitled "The Phase Transition in Solid Mercury" by C. A. Swenson was submitted for publication in PHYS. REV.

ABSTRACT

A polymorphic transition in solid mercury which was initially discovered by Bridgman has been studied at lower...
temperatures and pressures than those previously used. The transition would occur at zero pressure and 79°K if these data are extrapolated. However, the transition begins to show time effects and large pressure hysteresis at much higher temperatures, and below 93°K it can only be made to run irreversibly and in the α-β direction, and then only upon the application of several thousand atmos pressure. The changes in the molar volume with both temperature and pressure from 4.2°K to 200°K and from zero to 12,000 atmos have been determined in addition to the changes in the thermodynamic parameters at the transition. In particular, these results can be used to explain an anomaly which was found in work on the effects of pressure on the superconducting transition in mercury.

2.14 Nuclear Quadrupole Resonance Spectrum of TiBr₄

(R. G. Barnes)

A preliminary study of the dependence upon external pressure of the Br⁷⁹ and Br⁸¹ nuclear quadrupole resonances in TiBr₄ found these to possess a negative pressure coefficient in the temperature range -20° to -30°C. This result is consistent with general theoretical arguments concerning the dependence of quadrupole resonance frequencies on temperature and pressure.
In order to arrive at an understanding of the unusual maximum in the temperature dependence of these resonances, the equation of state (in the form of PV isotherms) has been determined experimentally with the help of Dr. C. A. Swenson of this Laboratory. Data obtained cover the pressure range to 10,000 atm and the temperature range 180° to 310°K. A beryllium-copper pressure bomb has been constructed and auxiliary equipment assembled so that the pressure dependence of the resonances may be studied at pressures up to approximately 3000 atm.

2.15 **Electron Coupled Nuclear Spin-Spin Interactions**

(R. G. Barnes)

A theoretical investigation of the effect of electron coupled nuclear spin-spin interactions on the nuclear quadrupole resonance spectrum of Cl\textsuperscript{35} in TlCl\textsubscript{3} has shown that the magnitude of this effect must be at least an order of magnitude smaller than that which has been observed. (See ISC-975). A careful re-examination of the Cl\textsuperscript{35} quadrupole resonance spectrum in HgCl\textsubscript{2}, using a high resolution spectrometer, failed to show any sign of fine structure attributable to this type of interaction although the expected magnitude of the effect would be close to that in TlCl\textsubscript{3}. We conclude that the pair of resonances observed in TlCl\textsubscript{3} are crystallographic in origin and that the large ionic character of the metal-chlorine bonds in TlCl\textsubscript{3} and HgCl\textsubscript{2} causes the electron coupled nuclear spin-spin interaction to be unobservably small.
APPENDIX I: LIST OF REPORTS FROM THE AMES LABORATORY

1. Reports for Cooperating Laboratories

ISC-780 Joseph Zimmerman and C. V. Banks. Some Properties of Sulfoacetic Acid and 3-Sulfopropionic Acid and Their Use as Analytical Masking Agents.


ISC-831 Fred Barson, S. Legvold and F. H. Spedding. Thermal Expansion of Rare Earth Metals.

ISC-851 Henry J. Hettel and V. A. Fassel. Quantitative Separation of Small Amounts of Rare Earths from Thorium, Uranium and Zirconium by Ion-Exchange.


ISC-873 Charles V. Banks and Gerald L. Weiss. Annotated Bibliography of 2-(2-Hydroxy-3,6-Disulfo-1-Naphthylazo)-Benzearsonic Acid.


ISC-927 Philip J. Lynch and D. J. Zaffarano. Tests and Analysis of Magnetic Quadrupole Lenses.

ISC-928 Paul E. Woerner and P. Chiotti. Precipitation of Thorium as Thorium Hydride from Thorium-Magnesium Solutions.

ISC-929 William J. Burkhard and J. D. Corbett. Solubility of Water in Molten Alkali Chlorides.


ISC-998 Lester Knapp, Roman Schoenherr, John Barghusen and Morton Smutz. A Polyethylene Box-Type Mixer-Settler Extractor.


ISC-318(Del) F. H. Spedding. The Molten-Metal Fuel Reactor.
2. Publications

Anderson, G. S., S. Legvold and F. H. Spedding
Superconductivity of Lanthanum and Some Lanthanum

Arajs, Sigurds and Sam Legvold
Comments on the Senftleben Method for the Determination
of the Heat Conductivity, the Specific Heat and the

Atoji, Masao, Karl Gschneidner, Jr., A. H. Daane, R. E. Rundle
and F. H. Spedding
The Structures of Lanthanum Dicarbide and Sesquicarbide
80, 1804-1808 (1958).

Banks, Charles V., John L. Spooner and Jerome W. O'Laughlin
Differential Spectrophotometric Determination of Rare

Behrendt, D. R., S. Legvold and F. H. Spedding
Magnetic Properties of Dysprosium Single Crystals.

Bochinski, J., M. Smutz and F. H. Spedding
Separation of Individual Rare Earths by Liquid-Liquid
Extraction from Multicomponent Monazite Rare Earth

Bray, P. J., Saul Moskowitz, H. O. Hooper, R. G. Barnes and
S. L. Segel
Pure Quadrupole Resonance Determinations of Molecular
Charge Distributions. I. Substituted Quinones,
Quinolines, Pyridines, Pyrimidines and Triazines. J.

Chiotti, P. and H. E. Shoemaker
Pyrometallurgical Separation of Uranium from Thorium.
Ind. and Eng. Chem. 50, 137-140 (1958).

Corbett, John D. and Alex Hershaft
The Lower Oxidation States of Gallium. II. The
Ga₂Br₄-GaBr System. J. Am. Chem. Soc. 80, 1530-1532
(1958).
Corbett, John D. and Pyrtle X. Seabaugh

Duke, F. R. and M. L. Iverson

Duke, Frederick R. and Arthur S. Doan

Elleman, Thomas S., John W. Reishus and Don S. Martin, Jr.

Fassel, Velmer A. and William A. Gordon

Fassel, Velmer A., William A. Gordon and Ray W. Tabeling

Fritz, James S. and Evelin Carlston Bradford

Fritz, James S., Richard T. Oliver and Donald J. Pietrzyk

Gibson, E. D., B. A. Loomis and O. N. Carlson

Haefling, James F. and A. H. Daane
Hansen, Robert S., Mary E. Purchase, Terry C. Wallace and Robert W. Woody

Hansen, Wilford N. and Maurice Griffel

Heidel, Robert H. and Velmer A. Fassel

Junk, Gregor and Harry J. Svec

Katsumori, H.

Laity, Richard W. and Frederick R. Duke

Lowry, Ralph A. and Glenn H. Miller

Mickelson, Robert and David Peterson

Morris, R. G., R. D. Redin and G. C. Danielson

Ray, A. E. and J. F. Smith
Redin, R. D. and G. C. Danielson

Redin, R. D., R. G. Morris and G. C. Danielson

Rhodes, B. L., S. Legvold and F. H. Spedding

Richardson, A. E. and A. F. Voigt

Rogers, B. A., D. F. Atkins, E. J. Manthos and M. E. Kirkpatrick

Smith, H. G. and R. E. Rundle

Spedding, F. H., J. J. Hanak and A. H. Daane

Svec, Harry J. and H. Gene Staley

Vest, R. W., M. Griffel and J. F. Smith

Williams, D. E. and W. H. Pechin
## APPENDIX II: LIST OF SHIPMENTS

<table>
<thead>
<tr>
<th>Destination</th>
<th>Item</th>
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<tr>
<td>U. S. Radiation Laboratory Livermore, California</td>
<td>4 gm neodymium 4 gm samarium 4 gm thulium 4 gm lutetium</td>
</tr>
<tr>
<td>General Electric Company Richland, Washington</td>
<td>1 3/4 lbs cerium metal</td>
</tr>
<tr>
<td>Nuclear Metals, Inc. Cambridge, Massachusetts</td>
<td>1 yttrium billet</td>
</tr>
<tr>
<td>U. S. Naval Research Laboratory Washington 25, D. C.</td>
<td>1-3 gm samarium metal 1-3 gm dysprosium metal</td>
</tr>
<tr>
<td>Massachusetts Institute of Technology Cambridge, Massachusetts</td>
<td>1 gm yttrium metal</td>
</tr>
<tr>
<td>M. Neal Paris, France</td>
<td>5 gm gadolinium metal 5 gm dysprosium metal 5 gm erbium metal</td>
</tr>
<tr>
<td>University of Denver Denver, Colorado</td>
<td>1 lb special cerium metal</td>
</tr>
<tr>
<td>National Chemical Laboratory Poona, India</td>
<td>2 gm lanthanum metal 2 gm cerium metal 2 gm neodymium metal 2 gm yttrium metal 1 gm praseodymium metal 1 gm samarium metal 1 gm gadolinium metal 1 gm dysprosium metal</td>
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<td>General Electric ANP Dept. Cincinnati, Ohio</td>
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<tr>
<td>New Brunswick Area Office New Brunswick, New Jersey</td>
<td>10 gm europium oxide 200 gm europium oxide</td>
</tr>
<tr>
<td>Union Carbide Nuclear Company Oak Ridge, Tennessee</td>
<td>12 gm scandium metal</td>
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<tr>
<td>Physikalisches Institut der Universität Heidelberg Heidelberg, Germany</td>
<td>10 gm europium oxide</td>
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<tr>
<td>Pratt &amp; Whitney Aircraft Middletown, Connecticut</td>
<td>10 gm ytterbium metal</td>
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<tr>
<td>University of Illinois Urbana, Illinois</td>
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<td>Destination</td>
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<td>Walter Reed Army Medical Center, Washington 12, D. C.</td>
<td>100 gm yttrium oxide</td>
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<td>Institute for Theoretical Physics, Copenhagen, Denmark</td>
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<tr>
<td></td>
<td>1 gm samarium oxide</td>
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<tr>
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<tr>
<td>Union Carbide Nuclear Company, Oak Ridge, Tennessee</td>
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