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Semi-annual summary research report in physics

Ames Laboratory Staff
Iowa State College

F. H. Spedding
Iowa State College

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Semi-annual summary research report in physics

Abstract
A semi-annual summary research report in physics for the period of July through December of the year 1956.

Disciplines
Physics
UNCLASSIFIED

ISC-833

UNITED STATES ATOMIC ENERGY COMMISSION

SEMI-ANNUAL SUMMARY RESEARCH REPORT IN PHYSICS

For July through December, 1956

by

Ames Laboratory Staff

March 13, 1957

Ames Laboratory
at
Iowa State College
F. H. Spedding, Director
Contract W-7405 eng-82

UNCLASSIFIED
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SEMI-ANNUAL SUMMARY RESEARCH REPORT IN PHYSICS

For the Period July through December, 1956

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

Previous summary research reports in this series are:

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ISC-56
ISC-69
ISC-74
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ISC-79
ISC-113
ISC-130
ISC-133
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ISC-220
ISC-246
ISC-283
ISC-301
ISC-338
ISC-395
ISC-422
ISC-451
ISC-508
ISC-533
ISC-577
ISC-608
ISC-645
ISC-707
ISC-758
Theoretical Physics

1. Orthogonalization Procedures (B. C. Carlson and J. M. Keller)*

A paper entitled "Orthogonalization Procedures and the Localization of Wannier Functions," by B. C. Carlson and Joseph M. Keller, has been submitted for publication in PHYSICAL REVIEW.

Abstract

The method of "symmetric orthonormalization" is shown to have a least-squares property: it constructs those unique orthonormal functions which minimize the sum of squared distances (in Hilbert space) between each initial function and a corresponding function of the orthonormal set. The localization of Wannier functions is a consequence of this property, since they can be obtained from localized atomic orbitals by symmetric orthonormalization. The theorem further implies an optimal resemblance of Wannier functions to atomic orbitals.

2. Fierz Interference in the Beta Decay of P\textsuperscript{32} (B. C. Carlson)

The beta spectrum of P\textsuperscript{32} has been used previously to determine whether or not the beta-decay interaction includes a small amount of axial vector coupling. The theoretical shape of the electron spectrum of P\textsuperscript{32} includes a factor of $1 + rw^{-1}$, where $W$ is the electron total energy in me\textsuperscript{2} units and $r$ is twice the ratio of the axial vector coupling constant to the tensor coupling constant. (If odd interactions are included, $r$ is proportional to $G_T G_A + G_T G_A'$; it is then possible to have a vanishing $r$, and hence no observable Fierz interference, even though the axial-vector coupling constants may be large.)

*Names indicate group leaders in charge of work.
The most precise value for \( r \) was obtained by Pohm, Waddell, and Jensen who measured the spectrum on a thin-lens spectrometer and independently on an intermediate-image spectrometer. They used the upper third of the spectrum to obtain the end-point energy \((1.712 \pm 0.005 \text{ Mev}, \text{ with excellent agreement between the two spectrometers})\) and then used this energy in finding \( r \) from the remaining points of the spectrum.

In order to check the validity of their statistical analysis, their data have been re-analyzed without splitting each spectrum into two energy regions. The statistical problem is to fit the data to a non-linear function of three parameters; it is solved by expanding the normal equations in Taylor series about first estimates of the parameters and carrying out successive approximations. (The convergence is fast.) The results of this re-analysis give essentially the same end-point energy as before, but (as shown in Table I) the values of \( r \) from the two spectrometers are now found to have the same sign. The close agreement for the intermediate-image data is probably fortuitous, since some numerical oversights have been remedied in the re-analysis. An effort is being made to track down the source of the discrepancy in the thin-lens case; it does not seem to be due to numerical errors in either computation.

### Table I

<table>
<thead>
<tr>
<th>Int. Image</th>
<th>Thin Lens</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pohm et al.</td>
<td>0.030</td>
<td>-0.032</td>
</tr>
<tr>
<td></td>
<td>±0.040</td>
<td>±0.045</td>
</tr>
<tr>
<td>Re-analysis</td>
<td>0.033</td>
<td>0.028</td>
</tr>
<tr>
<td></td>
<td>±0.010</td>
<td>±0.018</td>
</tr>
</tbody>
</table>

3. A Modified Deformable Potential for Thermal Scattering of Electrons (J. M. Keller)

A paper entitled "A Modified Deformable Potential for Thermal Scattering of Electrons," by John B. Gibson and Joseph M. Keller was submitted to PHYSICAL REVIEW.

---

Abstract

In order to study thermal scattering of moderately bound electrons in a metal, corrections are considered to a deformable scattering potential. A constant multiple of the dilatation is derived that must be added to account for the correct zero of potential in a homogeneously deformed crystal. In an inhomogeneously deformed crystal (a crystal undergoing thermal vibrations), a function of the strain must be added to take care of charge redistribution. The deformable potential must also be modified to treat umklapp processes consistently. The modification provides that as the phonon wave vector increases in magnitude, some umklapp character must gradually be added to the scattering amplitude.

4. Radiative $\pi^-\epsilon$ Decay and Photodecay of Neutral Pions*

Previous calculations by perturbation theory of the transition probabilities for the processes $\pi^* \rightarrow 2\gamma$ and $\pi^- \rightarrow \pi^- + \nu + \gamma$ (relative to $\pi^- \rightarrow K^- + \nu$) have given results that are larger than the experimental values by factors of 100 and at least 500, respectively. It is found possible to obtain agreement with experiment by modifying the calculations in two respects: (1) including Pauli moments of the virtual nucleon pair, and (2) introducing a cutoff at the vertex of the electromagnetic interaction with the nucleon. The value of the cutoff that gives agreement with experiment is about 0.3 times the nucleon mass; it is interesting that the corresponding Compton wavelength is close to the experimental proton radius as determined from high-energy electron scattering.

5. Mass Differences of the $\Sigma$ Hyperons*

The $\Sigma^-$ hyperon is known to be heavier than the $\Sigma^+$ by approximately 15 electron masses; the $\Sigma^*$ mass is less accurately known but is probably greater than the $\Sigma^+$ mass by an amount not exceeding 6 electron masses. The possibility of explaining these mass differences as electromagnetic self-energies has been investigated under the assumption that the $\Sigma$'s are Dirac particles having static anomalous magnetic moments. (The same assumption was made by Sudershan and Marshak\footnote{Sudershan and Marshak, Phys. Rev. 104, 267 (1956).} in a paper on the $\Sigma^- - \Sigma^+$ mass difference, \footnote{Of interest to, but not supported by AEC. Work was performed by Hiroshi Katsumori, Institute for Atomic Research, Iowa State College.}
but an error in sign made their numerical results different in the general case.) For a chosen cutoff, experimental values of the two mass differences restrict the unknown magnetic moments $\mu$ of the three hyperons to lie on a hyperbola in the $\mu_-, \mu_+$ plane and a hyperbola in the $\mu_0, \mu_+$ plane. For a cutoff approximately equal to the hyperon mass, the mass differences can be explained with reasonable absolute values of a few nuclear magnetons for all three magnetic moments.

This work is described in more detail in a paper entitled "Mass Differences of $\xi^+, \xi^-, \xi^0$ Hyperons", by Hiroshi Katsumori, which will be submitted to PROGRESS OF THEORETICAL PHYSICS.

Experimental Physics

1. Nuclear Studies

1.1 Time-Jitter Adapter for Single Channel Pulse Height Analyzers (Erling N. Jensen)

The time output of a pulse from a single channel pulse height analyzer is a function of the pulse height. When the pulse height analyzers are used in conjunction with coincidence circuits it is desirable to have this time-jitter reduced to a minimum.

An adapter for single channel pulse height analyzers has been built which reduces the time-jitter in the output of an analyzer by a factor of approximately 6. This reduction enables coincidence studies to be conducted using a correspondingly smaller $2\tau$, where $\tau$ is the resolving time of the coincidence circuit. Without the adapters a $2\tau$ of approximately 1.0 $\mu$sec was used in order to insure against the loss of true coincidences as the window height settings are changed. The use of the time-jitter adapters makes it possible to reduce the $2\tau$ to less than 0.2 $\mu$sec with a resulting effective gain in the ratio of true to accidental coincidences.

The use of the adapters also permits the measurement of shorter half-lives of metastable states than is possible without the use of adapters when only a simple coincidence circuit is utilized. With the time-jitter adapter it is possible to measure half-lives as short as perhaps 0.03 $\mu$sec.

Mihelich, Ward, and Jacob\(^1\) have reported a metastable state in Tm\(^{169}\) with a half-life of $4.5 \times 10^{-8}$ sec. Our preliminary results indicate a half-life closer to $6 \times 10^{-8}$ sec.

\(^1\)Mihelich, Ward, and Jacob, Phys. Rev. 103, 1285 (1956).
1.2 Electrostatic Analyzer (C. L. Hammer)

A 10-inch, 120° electrostatic analyzer for resolving and measuring proton energies in the region below 300 kev has been built and a calibration experiment performed on it. A method for reproducibly aligning the deflection plates was developed, with the result that the theoretical value of the analyzer constant, computed from the geometry, can be determined to about 0.1%. The value found is 13.117 ± 0.014 ev per volt. Maximum theoretical resolution, unless the beam is externally collimated, is about 700, corresponding to an energy spread of 0.14%.

For the calibration experiment the thick target yield from the reaction $^{211}(p, \gamma)^{212}$ was measured as a function of the analyzer deflecting voltage, using analyzed protons in the energy region near the well-known low-energy resonance. The analyzer slits were set to give a theoretical resolving power of about 0.6%. The data were fitted to the functional form predicted by resonance theory, including corrections for the proton wave length and the Coulomb penetrability, by a least-squares method involving 5 parameters. The resonant energy, computed from the resultant curve and the theoretical analyzer constant, is 163.0 ± 0.3 kev, in good agreement with the values reported by other laboratories. The experimental width, uncorrected for the spread in energy of the analyzed beam, is 6.6 kev. This value is also in agreement with the values reported by other laboratories.

1.3 Ionization Yields for Fission Fragments (C. L. Hammer)

The experimental work on the fission fragment ionization yield experiment (see ISC-707) has been completed. The ratio of fission fragment ionization yield to alpha-particle ionization yield was measured in argon, helium, and helium plus three-tenths % argon with a parallel-plate gridded ionization chamber. Energies found for the most probable light and heavy fragments by multiplying these ratios by the alpha energy are shown in Table II. About 0.5

<table>
<thead>
<tr>
<th>Gas</th>
<th>Energy (Mev)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Light</td>
</tr>
<tr>
<td>Argon</td>
<td>93.7 ± .4</td>
</tr>
<tr>
<td>Helium</td>
<td>96 ± .4</td>
</tr>
<tr>
<td>Helium + Argon</td>
<td>97.6 ± .3</td>
</tr>
</tbody>
</table>
Mev should be added for source thickness correction. Natural uranium was fissioned by D-D neutrons from the kevatron. Saturation and rise-time effects were studied in detail. The energy difference between argon and helium agrees with that predicted by Knipp and Ling on the basis of atomic collisions. The addition of sufficient argon to discharge the metastable atoms in helium appears to be a convenient method for studying the relative amount of energy absorbed as excitation in the stopping of the fragments and the alpha particles.

1.4 Extraction of Electrons From a Conventional Synchrotron
(C. L. Hammer)

The method for extracting electrons from a conventional synchrotron which has been proposed by Hammer and Bureau has been successfully used on the Iowa State College Synchrotron. Preliminary data indicate that 30% of the circulating beam is extracted. The principal advantage of the method is that no internal deflectors, either electrostatic or magnetic, are necessary to the extraction process, thereby permitting full use of the guide field aperture at injection.

1.5 Isomeric State of W^{181}
(C. L. Hammer)

A paper entitled "An Isomeric State of W^{181}," by A. J. Bureau and C. L. Hammer was submitted for publication in PHYSICAL REVIEW.

Abstract

An isomeric state of W^{181} is excited by a (γ,n) reaction on W^{182} using the X-ray beam of the Iowa State College 75-Mev Synchrotron. The energy and half-life of the isomer are found to be 366 ± 4 kilovolts and 14.4 ± 0.3 microseconds, respectively. A measurement of the K shell conversion coefficient yields a value of 0.3 ± 0.03. These experimental data suggest that the transition is of mixed multipole order, 28% E1 + 72% M2. This assignment is in agreement with the transition probabilities computed from Nilsson's wave functions for the first excited state and the ground state of the W^{181} nucleus.

1.6 Proportional Counter Spectrometer (D. J. Zaffarano)

The characteristics of a large proportional counter suitable for the study of the decay of radioactive sources\(^1\) are being reinvestigated in an attempt to extend the measurable energy range and resolution of the instrument. The counter is operated in an axial magnetic field which constrains the beta-ray orbits to helical paths. To date only argon-methane mixtures have been used as the filling gas. A resolution (full width at half-maximum) of less than 4% has been achieved with the internal conversion line from Cs-137. This represents an improvement, by a factor of 2, over results previously reported in the literature. The "summing" feature of the spectrometer enables one to observe coincidence relations between internal conversion lines. This has proven useful in the study of the decay of Ba-133.

1.7 Isomeric Transitions in the Rare Earth Elements (M. G. Stewart)

A paper entitled "Isomeric Transitions in the Rare-Earth Elements," by C. L. Hammer and M. G. Stewart was submitted to PHYSICAL REVIEW for publication.

Abstract

A search was made for isomeric transitions in the rare-earth elements with half-lives greater than 5 microsec. and less than \(\sim 30\) minutes which arise in the decay of photoexcited nuclei. Four isomeric transitions were found and identified as follows: \(\text{Tb}_{158}, 50 \text{ kev} < E < 200 \text{ kev}, 11.0 \pm 0.1 \text{ sec.}, \text{M}_3 \text{ or E}_3; \text{Ho}_{163}, 305 \text{ kev}, 0.8 \pm 0.1 \text{ sec.}, \text{E}_3; \text{Er}_{165}, 167, \text{or E}_{169}, 213 \text{ kev}, 2.5 \pm 0.1 \text{ sec.}, \text{E}_3; \text{Lu}_{174}, 133 \text{ kev}, 75 \pm 2 \text{ microsec.}, \text{E}_2.\) These results are compared with the predictions of the nuclear models which have been proposed by Nilsson and Gottfried.

2. Solid State Studies

2.1 Semiconducting Intermetallic Compounds \(\text{Mg}_2\text{Si}\) and \(\text{Mg}_2\text{Ge}\)

(G. C. Danielson)

2.1.1 Crystal Growing

Single crystals of \(\text{Mg}_2\text{Si}\) and \(\text{Mg}_2\text{Ge}\) have been grown successfully by the Bridgman method. Owing to the high vapor

pressure of magnesium, attempts to pull crystals from the melt have not been successful. The only crucible material found to be suitable for Mg2Si was graphite. A fairly large temperature gradient (about 25°C per cm) seemed to be necessary for best crystal growth. Owing to porosity of the graphite, the freezing temperature was moved rather rapidly from the bottom to the top of the melt; the total solidification time was less than 1 hour. It was necessary to cool the crystals slowly from the melting temperature to room temperature in order to reduce fractures in the crystals; about 50°C per hour was found to be satisfactory. When the crystals were grown in an argon atmosphere and the heating time was reduced to a minimum, little magnesium was lost, in spite of its high vapor pressure. In most runs, pure compound was formed with stoichiometric proportions of the two constituents, and one atomic per cent excess magnesium (to compensate for losses) would produce a magnesium-rich eutectic mixture.

Ordinarily the crystals grown were n-type semiconductors. By doping with silver, however, we have been able to produce single crystals of p-type Mg2Si. The purity of the crystals is indicated to some extent by the carrier concentrations. In the case of Mg2Si carrier concentrations were about $10^{17}$ electrons/cm$^3$; in the case of Mg2Ge, carrier concentrations as low as $3(10^{15})$ electrons/cm$^3$ have been obtained. This latter sample was therefore 100 times purer than any sample previously reported.

### 2.1.2 Resistivities and Hall Coefficients

The resistivities and Hall coefficients at 300°K of our samples of Mg2Si and Mg2Ge are compared with those of other investigators in Tables III and IV. The samples reported by Winkler were polycrystalline but that of Ellickson and Nelson was a single crystal. In general, the single crystals have higher purities as shown by the higher Hall coefficients. The p-type single crystal of Mg2Si was quite impure, however.

The temperature dependences of the resistivities and Hall coefficients of Mg2Si are shown in Fig. 1 and Fig. 2. The resistivities in the intrinsic range are in good agreement for all samples except that of Whitsett's, which was cut from a portion of the Mg2Si crystal quite near the carbon crucible and may have been badly contaminated by the carbon. The Hall coefficients of sample 21B-1 were positive up to 650°K. Above this temperature the Hall coefficients were negative. The mobilities of the electrons in Mg2Si must therefore be greater than the mobilities of the holes.
### Table III
Resistivities and Hall Coefficients of Mg$_2$Si at 300°K

<table>
<thead>
<tr>
<th>Sample</th>
<th>Resistivity (ohm-cm)</th>
<th>Hall coefficient (cm$^3$/coulomb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>19B-1</td>
<td>0.085</td>
<td>-21</td>
</tr>
<tr>
<td>22B-1</td>
<td>0.25</td>
<td>-93</td>
</tr>
<tr>
<td>22B-3</td>
<td>0.18</td>
<td>-73</td>
</tr>
<tr>
<td>23B</td>
<td>0.08</td>
<td>-24</td>
</tr>
<tr>
<td>21B-1</td>
<td>0.02</td>
<td>+0.7</td>
</tr>
<tr>
<td>Ellickson$^1$</td>
<td>0.1</td>
<td>-30</td>
</tr>
<tr>
<td>Winkler$^2$</td>
<td>0.07</td>
<td>-7</td>
</tr>
<tr>
<td>Whitsett$^3$</td>
<td>0.03</td>
<td>-5.5</td>
</tr>
</tbody>
</table>

### Table IV
Resistivities and Hall Coefficients of Mg$_2$Ge at 300°K

<table>
<thead>
<tr>
<th>Sample</th>
<th>Resistivity (ohm-cm)</th>
<th>Hall coefficient (cm$^3$/coulomb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>7B-1</td>
<td>7.3</td>
<td>-2000</td>
</tr>
<tr>
<td>7B-3</td>
<td>9.4</td>
<td>-2200</td>
</tr>
<tr>
<td>Winkler$^2$</td>
<td>0.3</td>
<td>-20</td>
</tr>
<tr>
<td>Whitsett$^3$</td>
<td>0.07</td>
<td>-13</td>
</tr>
</tbody>
</table>

---


Fig. 1 - Resistivity of Mg$_2$Si as a Function of Temperature
Fig. 2 - Hall Coefficient of Mg$_2$Si as a Function of Temperature
The temperature dependences of the resistivities and Hall coefficients of Mg$_2$Ge are shown in Fig. 3 and Fig. 4. The far greater purity of our samples 7B-1 and 7B-3 is at once evident. This greater purity is highly desirable since it greatly extends the intrinsic range and allows much greater confidence in the validity of energy gap determinations.

2.1.3 Mobilities of the Charge Carriers

The Hall mobilities of Mg$_2$Si and Mg$_2$Ge as functions of temperature are shown in Fig. 5 and Fig. 6. The mobilities of the electrons at room temperature (300 K) are between 200 and 400 cm$^2$/volt-sec. The low mobilities by Winkler$^1$ and the bending of his curves even at high temperatures indicate very large impurity scattering in his polycrystalline samples. At high temperatures the slopes of all curves are approximately equal, except for the very impure p-type sample 21B-1. If thermal scattering by acoustical modes were the only scattering mechanism at high temperatures, the slope of all the curves should be -1.5. The observed values, however, are between -2.2 and -2.6 for Mg$_2$Si and about -2.0 for Mg$_2$Ge. Hence some other type of scattering is also important at high temperatures. This scattering is probably thermal scattering by optical modes but the quantitative temperature dependence of such scattering from theoretical considerations is, unfortunately, not yet known.

There seems to be an anomaly in each of the mobility curves for the very pure Mg$_2$Ge samples 7B-1 and 7B-3. The sudden change in slope just below room temperature is a strange effect—if it is real. Conceivably, it might be associated with inhomogeneity in the distribution of the sample's impurities or it might be associated with the optical modes.

2.1.4 Energy Gap

The determination of the energy gap between the valence band and conduction band is not a perfectly unambiguous calculation since it depends upon assumptions regarding the mobilities of the charge carriers.

A summary of the results obtained for the energy gap $E_0$ of Mg$_2$Si and Mg$_2$Ge is given in Table V. Three methods were used to

Fig. 3 - Resistivity of Mg₂Ge as a Function of Temperature
Fig. 4 - Hall Coefficient of Mg₂Ge as a Function of Temperature
Fig. 5 - Hall Mobility of Mg$_2$Si as a Function of Temperature
Fig. 6 - Hall Mobility of Mg$_2$Ge as a Function of Temperature
Table V

Summary of Values Obtained for the Energy Gap of Mg$_2$Si and Mg$_2$Ge

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Method (1) $E_g$ (in electron volts)</th>
<th>Method (2) $E_g$ (in electron volts)</th>
<th>Method (3) $E_g$ (in electron volts)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mg$_2$Si</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>19B-1</td>
<td>0.697</td>
<td>0.854</td>
<td>0.813</td>
</tr>
<tr>
<td>22B-1</td>
<td>0.693</td>
<td>0.890</td>
<td>0.791</td>
</tr>
<tr>
<td>22B-3</td>
<td>0.701</td>
<td>0.821</td>
<td>0.737</td>
</tr>
<tr>
<td>23B-3</td>
<td>0.681</td>
<td>0.839</td>
<td>0.859</td>
</tr>
<tr>
<td>(p-type)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>21B-1</td>
<td>0.685</td>
<td>0.811</td>
<td>-</td>
</tr>
<tr>
<td>Average</td>
<td>0.691</td>
<td>0.843</td>
<td>0.800</td>
</tr>
<tr>
<td>Winkler$^a$</td>
<td>0.70</td>
<td>0.76</td>
<td>0.77</td>
</tr>
<tr>
<td>Whitsett$^b$</td>
<td>0.48</td>
<td>0.61</td>
<td>0.61$^c$</td>
</tr>
<tr>
<td>Mg$_2$Ge</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7B-1</td>
<td>0.641</td>
<td>0.704$^d$</td>
<td>0.709</td>
</tr>
<tr>
<td>7B-3</td>
<td>0.641</td>
<td>0.690$^d$</td>
<td>0.693</td>
</tr>
<tr>
<td>Average</td>
<td>0.641</td>
<td>0.697$^d$</td>
<td>0.701</td>
</tr>
<tr>
<td>Winkler$^a$</td>
<td>0.62</td>
<td>0.74</td>
<td>0.74</td>
</tr>
<tr>
<td>Whitsett$^b$</td>
<td>0.64</td>
<td>0.71$^d$</td>
<td>0.62</td>
</tr>
</tbody>
</table>


$^c$Computed by Morris and Redin from Whitsett's data.

$^d$From a plot of $\log \rho T^{-1/2}$ vs $1/T$ since observed Hall mobility varied as $T^{-2}$. 
obtain \( E_0 \):

1. From the slope of \( \log \rho \) vs \( 1/T \) plot.
2. From the slope of \( \log \rho /T \) vs \( 1/T \) plot.
3. From the slope of \( \log RT^{3/2} \) vs \( 1/T \) plot.

Method (1) assumes that the hole and electron mobilities vary as \( T^{-3/2} \). This has not been observed. Method (2) assumes that the hole and electron mobilities vary as \( T^{-5/2} \). This is closer to the observed temperature dependence. Method (3) assumes that the hole and electron mobilities have the same temperature dependence. All three methods assume that the energy gap is a linear function of temperature and that the samples are in the intrinsic conduction range. For method (3) the slopes were determined analytically by a least-squares method. For the other two methods the slopes were determined graphically.

Although method (3) has the largest experimental uncertainty, it involves the least restrictive assumptions and, at present, is regarded as giving the best value for \( E_0 \). The values obtained are in better agreement with those of Winkler than with those of Whitsett. The energy gap in this group of magnesium compounds is seen to decrease with increasing atomic number of the other constituent from 0.80 ev for Mg\(_2\)Si to 0.70 ev for Mg\(_2\)Ge to 0.33 ev for Mg\(_2\)Sn and to less than zero for Mg\(_2\)Pb (which is a metal).

### 2.2 Single Crystals of Tungsten Oxide (G. C. Danielson)

A new method of preparing single crystals of tungsten oxide (WO\(_3\)) has been developed by S. Sawada, visiting professor from Tokyo University. A platinum crucible, filled with tungsten oxide powder, was placed upside down upon a platinum plate and a ceramic weight was set on the base of the crucible. The assembly was inserted into a Globar muffle furnace and was maintained at a temperature of 1370°C for several days. A slight temperature gradient (perhaps 10°C per cm) also seemed to be necessary. Since the platinum crucible and platinum plate adhered to each other very well at this high temperature, a high pressure of tungsten oxide vapor was produced inside the crucible. This vapor resulted in the growth of excellent crystals.

Crystals as large as 10 mm by 8 mm by 1 mm have been obtained by keeping the crucible at 1370°C for 1 week. Single untwinned regions as large as 5 mm by 5 mm in area were easily found. Some preliminary measurements have been made of the index of refraction as a function of temperature and measurements of the electrical properties are in progress.

2.3 Thermal Diffusivity and Electrical Resistivity of High Purity Iron (G. C. Danielson)

The thermal diffusivity of Armco iron was reported in ISC-707. The thermal diffusivity and electrical resistivity of higher purity iron have now been measured over the temperature range 25°C to 1000°C. Both measurements were made on the same sample of spectrographically standardized metal. Impurities present in the iron rod were: carbon 0.03%; oxygen and nitrogen each 0.01%; sodium 0.001%; nickel 0.0005%; and copper, silver, magnesium and silicon each 0.0001%. The sample was annealed at 1000°C after inserting the necessary thermocouples and before any measurements were taken.

The electrical resistivity of iron as a function of temperature is shown in Fig. 7. The electrical resistivity increases from about 10 microhm-cm at 25°C to 115 microhm-cm at 1000°C. The points indicated by circles are for increasing temperature, the points indicated by triangles are for decreasing temperature. Temperature hysteresis effects are observed near the magnetic transformation (Curie temperature is at 765°C) and near the alpha-gamma phase transformation (900°C). The resistivity curve exhibits a definite discontinuity in slope at 765°C and a very small discontinuity in value at 900°C. The sample was cycled several times between 25°C and 1000°C in order to observe the effect of such thermal cycling on electrical resistivity. Changes in resistivity due to such cycling were less than 2 microhm-cm.

The thermal diffusivity of iron as a function of temperature is shown in Fig. 8. The thermal diffusivity decreases from about 0.23 cm²/sec at 25°C to 0.04 at 765°C and increases to 0.08 cm²/sec at 1000°C. The thermal diffusivity curve exhibits a pronounced discontinuity in slope at 765°C and a small discontinuity in value at 900°C. These discontinuities are approximately equal and opposite to the discontinuities observed in the specific heat. Hence the changes in thermal conductivity (which is the product of the thermal diffusivity and the specific heat per unit volume) at the magnetic transformation (765°C) and at the alpha-gamma phase transformation must be very small.
Fig. 7 - Electrical Resistivity of Johnson, Matthey Spectrographically Standardized Iron
Fig. 8 - Thermal Diffusivity of Johnson, Matthey Spectrographically Standardized Iron
2.4 **Specific Heat Measurements by a Pulse Method** (G. C. Danielson)

Modifications have been made in the pulse method for measuring the specific heats of metals, reported in ISC-707, and the new pulse method appears to be very satisfactory. The specific heats of platinum and iron have been measured over the entire temperature range 25°C to 1000°C and the estimated error is about 2%.

By combining the measurements of thermal diffusivity and specific heat, we should now be able to determine the thermal conductivity of any metal over the entire temperature range 25°C to 1000°C. The overall error should be less than 5%.

2.5 **Electrical Properties of Carbon Films** (G. C. Danielson)

A paper entitled "Electrical Properties of Arc-Evaporated Carbon Films" by M. D. Blue and G. C. Danielson was submitted for publication in JOURNAL OF APPLIED PHYSICS.

**Abstract**

Uniform thin films of carbon in the thickness range 100 to 2300 Å have been prepared by arc-evaporation in vacuum. Evaporation rates were about 50 Å/sec. The electrical properties of these films suggest that unannealed arc-evaporated carbon had less long-range order and was, therefore, more truly amorphous in structure than any other form of carbon yet obtained. After the films had been heated to 1200°K, however, their properties were similar to ordinary fine-particle carbon blacks, apparently as a result of increased ordering. The electrical resistivity of unannealed films followed ohms law, was not photosensitive, and could be well represented in the temperature range 77°K to 300°K by the equation $R = AT^{-b}$, where $b = 5$. When the carbon films were heated above room temperature, the resistivity and the exponent $b$ were both decreased substantially. After the films had been annealed at 1200°K, the resistivity was reduced by a factor exceeding 800, and could be best represented, in the temperature range 77°K to 1200°K, by the equation $R = \lambda - \beta T$. The values of the resistivity, temperature coefficient of resistivity, and thermoelectric power of the annealed films were then more nearly in agreement with the corresponding values for ordinary fine-particle carbon blacks. Other properties and applications of the films are discussed.
2.6 Electrical Properties of Boron (G. C. Danielson and D. E. Hudson)

A paper entitled "Electrical Properties of Boron Single Crystals" by W. C. Shaw, D. E. Hudson and G. C. Danielson was submitted for publication in PHYSICAL REVIEW.

Abstract

The electrical resistivity, Hall coefficient, and thermoelectric power of microscopic boron single crystals have each been measured as a function of temperature. Rectification and photoconductivity effects were observed at room temperature. The large majority of these single crystals had a high resistivity (about $1.7 \times 10^6$ ohm-cm at 300°K) which decreased by a factor of nearly $10^{10}$ as the temperature was increased from 200°K to 1000°K. A few crystals, however, had a much lower resistivity at 300°K (less than 100 ohm-cm) and at this temperature their resistivity decreased much more slowly with increasing temperature. Such behavior is attributed to donor impurity levels. From data in the high temperature range (800°K to 1000°K) the energy gap between the filled band and conduction band was calculated to be $1.55 \pm 0.05$ ev. Hall, thermoelectric and rectification studies indicated that hole conduction predominated for all crystals at high temperatures but that electron conduction predominated for the low resistivity crystals near 300°K. The mobilities of the charge carriers at 300°K were found to be the order of one cm$^2$ volt$^{-1}$ sec$^{-1}$ with the hole mobility the larger. Non-ohmic conduction was observed and shown to arise from joule heating.

2.7 Mass Spectrometric Studies of Metallic Phase Transitions (D. E. Hudson and F. H. Spedding)

In previous reports we have described the development of a mass spectrometric method of studying the thermodynamic properties of metals at high temperatures. The method is rapid in operation, uses small samples, and has proved to be considerably more accurate than conventional procedures. New studies have been carried out by this method on dysprosium, erbium, holmium, terbium, lutetium, and gadolinium. The new results are as follows ($\Delta H = \text{latent heat of sublimation}$):

**Dysprosium:** Three new runs on high-purity vacuum distilled material gave $\Delta H = 68.5 \pm 0.7$ kcal/mole at 1232°K. This result is in satisfactory agreement with our previous value of $69.3 \pm 0.6$ kcal/mole at 1215°K.
Erbium: The metal is easy to ionize but has a lower vapor pressure than expected. Three runs gave \( \Delta H = 71.6 \pm 2.5 \text{ kcal/mole} \) at 1278°K. This is the first experimental determination of the heat of sublimation of this material, and, therefore, no other values are available for comparison.

Holmium: This metal should have an ionization efficiency about the same as erbium. However, no ion beam was obtained, even at cell temperatures as high as 1475°K. This suggests that the vapor pressure of holmium is abnormally low compared to erbium, and is surprising since the two metals are so similar chemically and structurally.

Terbium: The comments on holmium also apply to this metal.

Lutetium: Seven runs have been made on the purest sample available; however, even this sample appears to have some impurities as manifested by inclusions visible under a microscope. Preliminary results indicate a solid-solid, first-order phase transition near 1287°K. The latent heats of sublimation of the phases were about 77 and 100 kcal/mole. However, this rather unusual behavior may not be characteristic of the pure metal.

Gadolinium: This metal appears to have a rather low vapor pressure. Four runs gave \( \Delta H = 77.5 \pm 3.1 \text{ kcal/mole} \) at 1547°K. These are the first data on this material.

2.8 Study of Charge Trappings in a Diamond Conduction Counter
(D. E. Hudson)

Preliminary results indicate that the space charge traps in the crystal are discharged much more effectively by light of 0.3 microns wavelength than by light of 0.5 microns wavelength. The work verifies some previous results obtained in this laboratory. The existence of traps shallower than 2.5 ev below the conduction band is certain but their distribution is still unknown.

2.9 Magnetic Measurements on Neodymium and Dysprosium
(S. Legvold and F. H. Spedding)

Magnetic measurements were made on single crystals of neodymium and dysprosium which were grown by the Bridgman method; i.e. by lowering a molten sample in a tantalum crucible slowly into a region below a furnace coil so that the sample would slowly congeal from the bottom end.
In the case of neodymium the susceptibility obtained at and above 20.4°K with the field perpendicular to the c axis of the hexagonal crystal is greater than that obtained with the field perpendicular to the c axis. No anisotropy was observed in the plane perpendicular to the c axis (the basal plane). However, at 4.2°K magnetic anisotropy is observed in the basal plane and the magnetization data suggest antiferromagnetic ordering. The orientation of the magnetic moments at 4.2°K is not revealed by the data but the direction of easy magnetization appears to be along an a axis in the lattice.

Magnetic measurements above 178.5°K (the Néel point) were made on a single crystal of dysprosium weighing 0.5 gm and below this temperature a spherical single crystal weighing 0.1 gm was used. Use of the two different samples in this manner was dictated by the method of measurement and by the accuracy desired in the paramagnetic range and the antiferromagnetic and ferromagnetic ranges which were known to exist for the metal.

Below the Néel point the individual magnetic moments in dysprosium are aligned perpendicular to the c axis of the hexagonal lattice. The moments are aligned in an antiferromagnetic manner from 178.5°K down to 85°K and in a ferromagnetic manner below 85°K. It is believed that the moments in a plane perpendicular to the c axis are all parallel and the moment vector lies in the plane for all temperatures below 178.5°K. Adjacent planes have their moments antiparallel from 178.5°K down to 85°K and below 85°K adjacent planes have parallel moments. These conclusions are based on the fact that measurements of the magnetic moment with the applied field directed parallel and perpendicular to the c axis reveal magnetic anisotropy at all temperatures. In addition the direction of easy magnetization is along an a axis while in the c direction the crystals are extremely "hard" magnetically, the magnetization curves being linear (as for a paramagnetic) from 4.2°K up to room temperature. Below 110°K dysprosium also shows anisotropy in the basal plane.

From extrapolations of the magnetization curves to infinite field the saturation moments were found as a function of temperature. These saturation moments followed a T^2 law.

2.10 Superconductivity of Lanthanum and Lanthanum-Rich Alloys
(S. Legvold and F. H. Spedding)

Meissner effect measurements on pure lanthanum have revealed two superconducting transition temperatures. It is quite clear that the two transitions are associated with different allotropic forms.
Cubic lanthanum has a transition temperature of 5.9°K while the hexagonal form (with abac stacking) has a transition temperature of 5.0°K. It has not been possible to completely transform a sample to the hexagonal form, but strong evidence (from two knees in the data for a mixed sample) support the latter transition temperature.

With regard to lanthanum alloys, a shift of the transition temperature with concentration of Lu, Y, or Yb has been observed. Results to date appear in Table VI.

Table VI

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<th>Alloy</th>
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<td>100% La</td>
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<td>5.9°K</td>
</tr>
<tr>
<td>100% La</td>
<td>hex (abac)</td>
<td>5.0°K</td>
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<tr>
<td>95% La 5% Y</td>
<td>cubic</td>
<td>5.4°K</td>
</tr>
<tr>
<td>85% La 15% Y</td>
<td>hex (abac)</td>
<td>3.1°K</td>
</tr>
<tr>
<td>75% La 25% Y</td>
<td>hex (abac)</td>
<td>2.6°K</td>
</tr>
<tr>
<td>60% La 40% Y</td>
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<td>1.8°K</td>
</tr>
<tr>
<td>80% La 20% Lu</td>
<td>hex (abac)</td>
<td>3.4°K</td>
</tr>
<tr>
<td>95% La 5% Yb</td>
<td>hex (abac)</td>
<td>4.0°K</td>
</tr>
</tbody>
</table>

In the case of pure cubic lanthanum it was possible to obtain critical magnetic field data. It was found that

\[
\frac{dH_c}{dT} \bigg|_{T = T_S} = 550 \text{ oersted/deg.}
\]

and that for \( T = 0°K \), \( H_0 = 1600 \) oersteds.

From these characteristics it appears that lanthanum may be classed with the hard superconductors.
2.11 Thermal Expansion of Rare Earth Metals (S. Legvold and F. H. Spedding)

A paper by F. Barson, S. Legvold and F. H. Spedding entitled "Thermal Expansion of Rare Earth Metals" was submitted for publication in PHYSICAL REVIEW.

Abstract

Thermal expansion measurements at temperatures ranging up to 900°C for polycrystalline samples of La, Ce, Pr, Nd, Gd, Tb, Dy, Er, and Yb metals are reported. La, Ce, Pr, and Nd exhibit plastic flow properties at temperatures well below their melting points. High temperature phase transformations are reported for Pr, Nd, and Yb. The coefficient of expansion for Yb is found to be three times as large as the coefficients for the other metals reported. Negative coefficients are observed near the Curie points of Gd, Tb, and Dy.

2.12 Nuclear Quadrupole Resonance Spectra of Group IIIB Trihalides (R. G. Barnes)

The nuclear quadrupole resonance spectra of the IIIB trihalides indicate that AlBr₃, AlI₃, GaCl₃, GaBr₃, GaI₃, InBr₃, and InI₃ are dimeric in the solid phase.¹ The gallium and indium resonances in GaBr₃, GaI₃, and InI₃ have now been detected, providing an estimate of the departure of the bonding from tetrahedral symmetry at the metal atoms. In addition, solid phase transitions have been noted in GaI₃ and InI₃ below room temperature. The halogen-pure quadrupole spectra of TlCl₃ and TlBr₃ have been detected and show no lines attributable to the bridging atoms characteristic of the IIIB dimers.

APPENDIX I: LIST OF REPORTS FROM THE Ames LABORATORY

1. Reports for Cooperating Laboratories

ISC-673 G. A. Guter and G. S. Hammond. Steric Effects on the Formation Constants of Metal Chelates of \( \beta \)-Diketones.


ISC-703 D. L. Horrocks and A. F. Voigt. The Formation of Thallium Chloride Complexes and Their Extraction into Ether.


ISC-737 Charles V. Banks and Howard B. Nicholas. Annotated Bibliography of 1,2-Cycloheptanedionedioxime.

ISC-738 Charles V. Banks, Howard B. Nicholas and James L. Pflasterer. Annotated Bibliography of 1,2-cyclohexanedionedioxime.


ISC-793 E. L. Koerner and M. Smutz. Separation of Niobium and Tantalum - A Literature Survey.


2. Publications

Banks, C. V. and C. I. Adams
The Preparation of 3,4-Bis(2-Furyl)-1,2,5-Oxadiazole. J. Org. Chem. 21, 815 (1956).

Banks, C. V., P. G. Grimes and R. I. Bystroff

Banks, C. V. and Donn W. Klingman

Banks, C. V. and J. W. O’Laughlin

Banks, C. V., John L. Spooner and Jerome W. O’Laughlin

Banks, C. V. and Joseph Zimmerman

Beyer, G. H. and F. M. Jacobsen

Deal, B. E. and H. J. Svec

Duke, F. R. and Bernard Bornong

Fassel, V. A. and R. W. Tabeling

Fritz, James S. and Sally Ann Sutton
Titration of Mercury with Bis(2-hydroxyethyl)dithiocarbonate. Anal. Chem. 28, 1300-1303 (1956).

Griffel, Maurice, R. E. Skochdopole and F. H. Spedding

Guter, G. A. and G. S. Hammond
Hansen, R. S., R. E. Minturn and D. A. Hickson

Hansen, Robert S. and Doris O. Stage


Jones, J. T., Jr., and J. M. Keller

Kross, R. D. and V. A. Fassel

Kross, R. D., K. Nakamoto and V. A. Fassel

Layton, E. Miller, Jr., R. D. Kross and V. A. Fassel

McMullen, R. K. and J. D. Corbett

Margerum, D. W., R. I. Bystroff and C. V. Banks

Mitchell, R. F. and D. S. Martin

Peterson, D. T., P. F. Diljak and C. L. Vold

Puri, R. P., F. R. Duke and J. Lomnes
Rundle, R. E.

Shull, Harrison and Per-Olov Lowdin

Smith, J. F., O. N. Carlson and R. W. Vest

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

Spedding, F. H., A. H. Daane and K. W. Herrmann

Svec, Harry J.

Voigt, A. F.
# APPENDIX II: LIST OF SHIPMENTS

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<th>Item</th>
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</thead>
<tbody>
<tr>
<td>Laramie, Wyoming</td>
<td>1 gm lanthanum oxide</td>
</tr>
</tbody>
</table>

Purdue University
Lafayette, Indiana

Dr. Frank A. Kanda
University of Syracuse
Syracuse, New York

Dr. W. C. Koehler
Oak Ridge National Laboratory
Oak Ridge, Tennessee

University of Kentucky
Chemistry Department
Lexington, Kentucky

Stephen M. Shelton
Oregon Metallurgical Corp.
Albany, Oregon

Dr. R. S. Milliken
University of Chicago
Chicago, Illinois

Dr. Joseph G. Graca
Iowa State College
Ames, Iowa

University of Wisconsin
Madison, Wisconsin

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
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</tr>
</thead>
<tbody>
<tr>
<td>1 gm cerium oxide</td>
<td>113 gm pure calcium</td>
</tr>
<tr>
<td>1 gm praseodymium oxide</td>
<td>1 bar polycrystalline neodymium</td>
</tr>
<tr>
<td>1 gm neodymium oxide</td>
<td>15 gm cerium metal</td>
</tr>
<tr>
<td>1 gm samarium oxide</td>
<td>1 lb. yttrium fluoride</td>
</tr>
<tr>
<td>1 gm gadolinium oxide</td>
<td>3 gm N&lt;sup&gt;15&lt;/sup&gt;</td>
</tr>
<tr>
<td>1 gm yttrium oxide</td>
<td>273 cc 5% NH₄Nd versinate</td>
</tr>
<tr>
<td>100 mg terbium oxide</td>
<td>273 cc NH₄ EDTA (ethylenediaminetetraacetate)</td>
</tr>
<tr>
<td>100 mg dysprosium oxide</td>
<td>200 cc each of 5.0% aqueous solutions of cerium, lanthanum praseodymium, and neodymium chloride</td>
</tr>
<tr>
<td>100 mg holmium oxide</td>
<td>200 cc each of cerium chloride versinate and lanthanum chloride versinate</td>
</tr>
<tr>
<td>100 mg erbium oxide</td>
<td>66 gm neodymium metal</td>
</tr>
<tr>
<td>100 mg ytterbium oxide</td>
<td>66 gm samarium metal</td>
</tr>
<tr>
<td>100 mg thulium oxide</td>
<td>77 gm erbium metal</td>
</tr>
<tr>
<td>100 mg lutetium oxide</td>
<td></td>
</tr>
</tbody>
</table>
**Destination**

Tufts University  
Chemistry Department  
Medford, Massachusetts

Walter Reed Army Medical Center  
Washington, D. C.

**Item**

3 gm lanthanum metal  
3 gm gadolinium metal  
10 gm cerium oxide