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Interaction of Electrons with Impurities and the $k \approx 0$ Longitudinal Optical Phonon in Metal-Insulator-Semiconductor Tunnel Junctions*†

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Tunneling electrons have been observed to interact with phonons in metal-insulator-semiconductor (MIS) tunnel junctions including mass-defect phonons, $k \approx 0$ longitudinal and transverse optical phonons, and zone-boundary phonons. Well-known diffusion techniques have been used to introduce phosphorus into boron-doped silicon producing MIS tunnel junctions having two types of observable mass-defect phonons. These experiments suggest that the mass-defect phonons seen in tunneling arise from an interaction of tunneling electrons with screened, ionized impurity atoms in the reserve region of the semiconductor.

I. INTRODUCTION

Tunneling electrons have been observed to interact with phonons in metal-insulator-semiconductor (MIS) tunnel junctions. Interactions have been observed with local[1–3] and mass-defect[4] phonons, $k \approx 0$ longitudinal[1–3,5–8] and transverse[9] optical phonons, and zone-boundary phonons.[5] A central problem in the understanding of the MIS tunnel junctions is the identification of the mechanisms by which

the electrons and phonons interact.

The present theoretical understanding of the nature of the electron–phonon interactions in MIS tunnel junctions can be briefly summarized as follows: The zone-boundary phonons, emitted in MIS tunnel junctions fabricated with n-type germanium[5] is a well understood special effect due to the band structure of germanium, the direct conduction band being only 140 meV above the indirect conduction-band minimum. Zone-boundary phonons are emitted to conserve k vector as the electrons transfer from an evanescent intermediate state associated with the Γ conduction band to the indirect conduction bands at the L position of the Brillouin zone. Although the phonon emission can occur anywhere in the semiconductor, the strength of the interaction can be adequately accounted for by assuming that the interaction occurs near the tunnel junction.

The theoretical understanding of other phonon effects is not as complete. A theory has been advanced to explain the $k \approx 0$ longitudinal optical phonon interaction, which is observed in many other semiconductors, i.e., n-[4] and p-type[1-3] silicon, p-type germanium,[5] n- and p-type gallium arsenide,[6] n-type cadmium sulfide,[7] n-type gallium antimonide,[8] and n-type tin oxide.[8] It involves the many-body modification of the electron dispersion relation in the electron gas of the bulk semiconductor and treats the spatial dependence of the self-energy at the surface of the semiconductor, also a many-body bulk effect.[10] This theory seems unable, however, to account for the doping dependence of the line shape observed by Cullen[2,3] in MIS tunnel junctions fabricated with p-type silicon. These data show that the line shape at negative bias corresponds to a decrease in conductance (a peak in the derivative of conductance) at the highest doping of 2.3×10^{20} cm^{-3} boron while at 2.0×10^{19} cm^{-3} boron concentration the line shape corresponds to a change in slope in conductance (a step-up in derivative of conductance). Tsui has suggested that the observation of the $k \approx 0$ transverse optical phonon in p-type gallium arsenide[9] tunnel junctions is due to deformation potential coupling in the bulk, again, a many-body bulk effect. Phonons associated with mass defects have been observed in n-[4] and p-type[1-3] silicon and n-type 6H silicon carbide.[4] Davis and Duke[10] have suggested that the same theory which explains the electron – $k \approx 0$ longitudinal optical phonon interaction, can be used to explain the electron–local-mode phonon interaction, simply with a change in coupling constants. However, the observations that the conductance changes are stronger for that bias for which the majority carriers are tunneling from the semiconductor into the metal are not accounted for by this theory. It was also observed[4] that the half-widths of the peaks are not strongly temperature dependent. In n-type silicon, the half-width of the principal mass-defect phonon peaks of phosphorus, carbon, and oxygen are 6% of their energy. It is clear, then, that there remain unanswered questions regarding the nature of the electron–phonon interactions in MIS tunnel junctions.

Boron local-mode phonons have been observed in MIS tunnel junctions fabricated with p-type silicon,[1-3] and phonons arising from the mass-defect phosphorus have been observed in MIS tunnel junctions fabricated with n-type silicon.[4] It was conjectured that the selected addition of impurities into a tunnel junction and their detection would shed light on the mechanism by which the electrons and mass-defect phonons interact. For example, it was supposed that the introduction of phosphorus by standard diffusion techniques into boron-doped silicon would produce the possibility of observing local and mass-defect modes due to both boron and phosphorus in a single tunnel junction. Further, if the concentration of the added compensating impurity could be varied, this would allow an examination of the effect of majority carrier concentration upon the electron–phonon interactions. Such experiments are reported in this article. They have yielded valuable information, not only concerning the electron–mass-defect phonon interaction, but also concerning the interaction of the electrons with the $k \approx 0$ longitudinal optical phonon.

Tsui and Dunkleberger[11] have recently stated that tunneling peaks which we attributed to the phosphorus resonant-mode phonons reflect the silicon phonon density of states. Since the phonon density of states dominates the determination of both band and resonant-mode phonons, it is true that the experiment essentially measures the silicon phonon density of states. It is also true, of course, that phonons are observed in the doped material because the impurity atom couples the tunneling electrons to the phonons. Since the distinction between band and resonance modes is essentially a matter of the phonon lifetime, and since we do not measure the lifetime directly, we have referred to these as "mass-defect" phonons in this paper.

These experiments represent a new method of observing mass-defect phonons which supplements the usual infrared absorption measurements. Accurately compensated material is not required to detect mass-defect phonons by this technique. These experiments also represent an extension into the MIS system of tunnel junctions of the work of Lambe and Jaklevic[12] and Thompson[13] who succeeded in detecting impurities introduced between the surfaces of metal-insulator-metal (MIM) tunnel junctions, the work of Gaiver and Zeller[14] who detected phonons of II-VI compounds that were used as the insulator in MIM tunnel junctions, and the work of Salaneck and co-workers[15] who observed local modes arising from impurities in lead tellu-

ride p-n tunnel junctions.

II. EXPERIMENTAL PROCEDURE

Phosphorus was diffused into samples of freshly cleaved p-type silicon doped with 2.3×10^{20} cm^{-3} boron atoms by the open-tube diffusion method[16]: A carrier gas (argon) flowed through an open tube past a P_2O_5 source heated to 250°C carrying P_2O_5 gas to the silicon samples held at a temperature of 1200°C. The one hour treatment was expected to diffuse phosphorus into a depth of approximately one micron with a phosphorus concentration of 10^{21} cm^{-3} at the surface.[16,17]

The silicon was then etched for 10 min in 10% HF to remove the thick oxide, washed in alcohol, and nickel plated by standard techniques.[18] Solder contacts were made to the nickel and the contacts were coated with a mixture of Styrofoam dissolved in toluene[19] and allowed to dry for 30 min (a stop-etch for HNO_3). The samples were then put into concentrated HNO_3 for 30 to 60 min, removing the nickel plating from the cleaved face. The silicon was finally put into 10% HF to remove the oxide formed by the HNO_3. This exposed a clean surface which was oxidized by exposure to air. Finally, metal dots were evaporated onto the surface producing a MIS tunnel junction.

If the HNO_3 etch failed to produce good tunnel junctions, the surfaces were lightly etched. This was accomplished in two steps. First, the metal dots were removed: The back contact was coated with the stop-etch for HNO_3, allowed to dry for 30 min, placed in HNO_3 for 30 min, and finally, washed in alcohol. Second, for the etching, the sample was placed in 10% HF for 5 min to remove the oxide, washed in alcohol with no exposure of the sample to air, etched 5–10 sec with constant stirring, washed in alcohol, placed in 10% HF for 5 min, and finally, washed in alcohol. The etches used were 250:1 HNO_3:HF (6×10^{-3} mil/min) and 500:1 HNO_3:HF (3×10^{-3} mil/min). The calibration of the etch rates was accomplished by etching a similar piece of silicon for several hours and measuring the change in a dimension with a micrometer readable to 10^{-1} mil. Since the phosphorus diffusion will probably alter the etch characteristics,[20,21] these calibration rates must be considered as approximate only.

The phosphorus-diffused p-type silicon samples that ended up n-type on the surface had a p-n junction approximately 1 μ deep. At 4.2 K this junction is essentially nonconducting. All current passing through the tunnel junction on the surface flowed through the n region along the sides of the sample and finally to the nickel plating. Since this n region is thin (about 1 μ) but has low resistivity (10^{-3} to 10^{-4} Ω cm, it presented 1–10-Ω series resistance to the junction. A slight shift of peaks to higher energy was observed in such junctions.

The experimental measuring procedures have been reported elsewhere.[2–5] Measurements are reported on the dependence of conductance di/dV and the differential conductance d^2i/dV^2 upon applied bias V. A dc bias and an ac modulation of frequency f were applied to the sample. The current measured across a small series resistor at frequency f is proportional to di/dV and at $2f$ to d^2i/dV^2. Standard lock-in detection techniques were used. Errors in the absolute magnitude of d^2i/dV^2 were negligible for the circuit parameters chosen. Data presented here were duplicated on junctions of several samples.

It was observed that tunneling was the current-carrying mechanism, through observation of the superconducting energy gap, when the chemical techniques described here were followed. Extra peaks in the d^2i/dV^2 data, repeatable on a given sample but not repeatable from sample to sample, often appeared. Such peaks were substantially eliminated in some of the experiments after slightly etching the surface. However, the interesting peaks associated with the local- and resonant-mode phonons and the $k\approx0$ longitudinal optical phonon always appeared the same for all junctions on a given material, regardless of the presence of the extra peaks. The data shown here are the best data obtained and represent direct tracings of the recorded data without correction or modification.

III. RESULTS

Shown in Figs. 1 and 2 are data taken on phosphorus-diffused p-type silicon that was initially doped with 2.3×10^{20} cm^{-3} boron. Such a phosphorus diffusion of p-type silicon will produce a heavily compensated sample. One of these samples had a section of its cleaved surface p type and the rest of the surface n type. This was determined by observing the minimum in the conductance di/dV vs voltage data. For the voltage sign convention chosen, the minimum occurs in negative bias for n-type material and in positive bias for p-type material. Various parts of this material had minima varying from -120 to $+60$ mV.

Data on the n-type material are shown in Fig. 1. The minimum in the conductance curve, which is a measure of the Fermi degeneracy of the bulk semiconductor, indicates that there are approximately 6×10^{19} cm^{-3} electrons in the conduction band. The concentration of diffused phosphorus is therefore 2.9×10^{20} cm^{-3}, 2.3×10^{20} for the compensation of the boron, and 6×10^{19} cm^{-3} for the electron concentration. The data look in many respects like data taken by Schein and Compton[4] on MIS tunnel junctions fabricated with n-type silicon, except that these data are somewhat noisier (attributable to the chemical treatment), and the peaks are shifted slight-

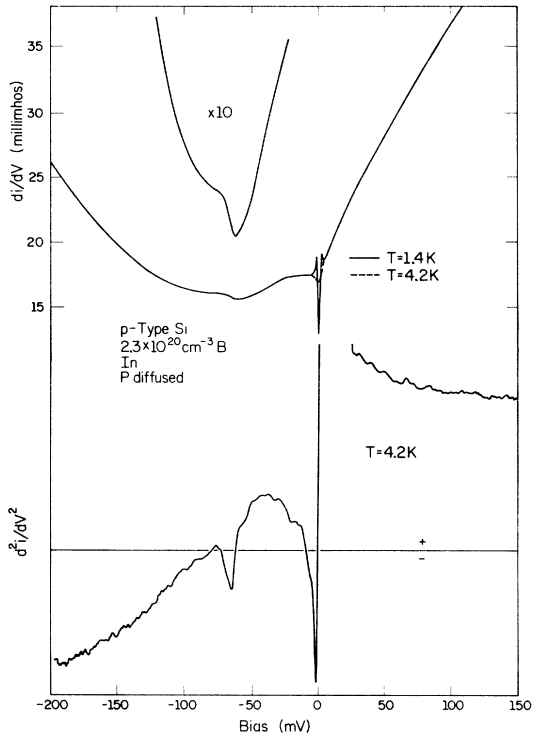

FIG. 1. Tunneling characteristics of a phosphorus diffused p-type silicon MIS tunnel junction. The semiconductor was made n type.

ly to higher energy due to the series resistance of the n-type channel leading to the back contact. One of the properties of the electron–mass-defect phonon interaction, a large peak only in the bias direction for which the majority carriers tunnel from the semiconductor into the metal, is clearly evident. The peak at -60 mV is due to electron interactions with the phosphorus mass-defect phonon. Although near the noise level, there are indications of the other phosphorus resonant-mode peaks at -19 and -52 mV, also previously observed. These three peaks were observed reproducibly in all the junctions fabricated with the n-type material. The carbon local-mode peak at -80 mV is within the noise in this sample. The oxygen and carbon-oxygen local-mode peaks, previously observed at -148 and -138 mV were not observed. We believe that this may be accounted for by the observation of Hrostowski and Kaiser[22] that heating a pulled silicon sample to 1250 °C for 1 h (the temperature treatment used for the phosphorus diffusion) reduced the interstitial oxygen concentration by a factor of 2, the oxygen precipitating into SiO_2 clusters. There is a zero-bias anomaly present at 4.2 K. What is most interesting, however, is that there is no indication of a peak due to the boron local mode, even though there are 2.3×10^{20} cm^{-3} boron atoms present.

Data on the p-type material are shown in Fig. 2. The position of the Fermi level, as determined from the minimum in the conductance curve, indicates that there are 3×10^{19} cm^{-3} holes in the valence band. Thus, the 2.3×10^{20} cm^{-3} boron atoms are compensated by 2.0×10^{20} cm^{-3} phosphorus atoms. The boron local-mode phonon at 80 mV and the $k\approx0$ longitudinal optical phonon peaks at ±65 mV are present, as has been previously observed in data taken by Cullen *et al.*[1–3] on MIS tunnel junctions fabricated with p-type silicon. The isotope splitting of the boron local mode is not observable at 4.2 °K, the temperature at which these data were taken. The structure near ±20 mV is due to the lead phonons; the peak at -35 mV was not reproducible. However, even though there are at least 2×10^{20} cm^{-3} phosphorus atoms present, there is no indication of the phosphorus mass-defect phonon. No effects due to a boron local mode perturbed by a phosphorus nearest neighbor were observed in the p-type material, but this is probably due to the lower concentration of such complexes.

IV. DISCUSSION

Consider first the electron–mass-defect phonon

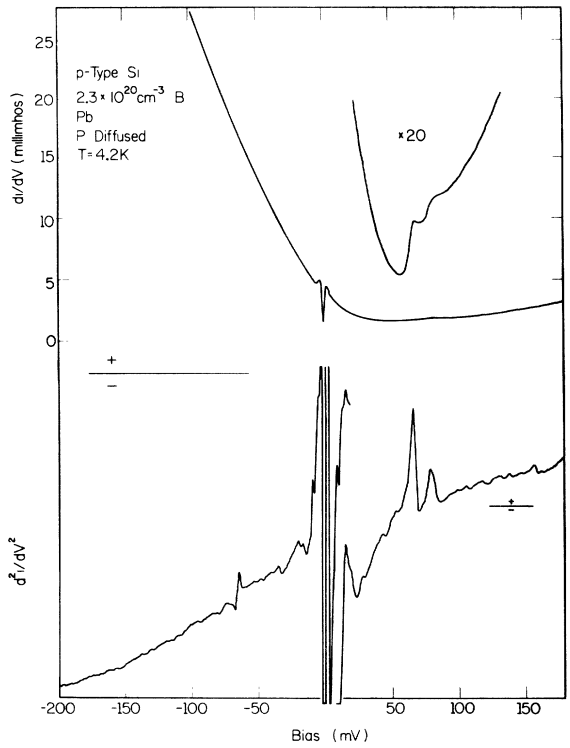

FIG. 2. Tunneling characteristics of a phosphorus diffused p-type silicon MIS tunnel junction. The semiconductor remained p type. Note that the zero on the lower right is shifted vertically with respect to the zero on the lower left.

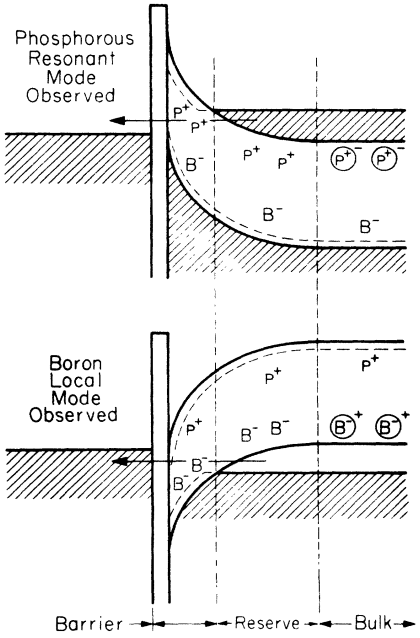

FIG. 3. Schematic energy-vs-distance diagrams of MIS tunnel junctions with the semiconductor p and n type. The junctions are shown with the bias applied in the direction for which the larger electron–mass-defect phonon interaction occurs, that is, in the direction for which the majority carriers tunnel from the semiconductor into the metal. All impurities are ionized in the barrier region. In the bulk the minority impurities are ionized and the majority impurities are screened by the majority carriers, indicated by circles.

interaction. In the n-type material, there is no evidence of a peak due to boron in the data, even though there were 2.3×10^{20} cm^{-3} boron atoms present. Similarly, in the p-type material there is no evidence of a peak due to phosphorus, even though there were approximately 2.0×10^{20} cm^{-3} phosphorus atoms present.

Distinct mass-defect phonons due to both phosphorus and boron in the same crystal have been observed by infrared absorption measurements.[23] One is therefore led to consider why, in a tunneling experiment, only one type of phonon is detected depending on whether the material is n type or p type. Shown in Fig. 3 are schematic energy vs distance diagrams of MIS tunnel junctions with the semiconductor p and n type. The junctions are shown with the bias applied in the direction for which the larger electron–mass-defect phonon interaction occurs, that is, in the direction for which the majority carriers are tunneling from the semiconductor into the metal. The semiconductor is divided into three regions. The reserve region is located between the position at which the majority carrier concentration and the electric field vanish. It is clear that whether the semiconductor is p or n type, the tunneling particle must pass through a region with positive phosphorus and negative boron atoms. The interaction is observed to change when the bulk is changed (from p to n type), while the nature of the charges in the barrier remain unchanged. This strongly suggests that the electron interaction with mass-defect phonons does not occur in the barrier region.

Since it has been argued that the interaction is not due to electrons interacting with ionized atoms in the barrier, one is led to conclude that the interaction involves the screened ionized atoms in the reserve or bulk regions of the semiconductor. We believe that the reserve region is involved, for the electrons in the reserve region must have some combination of the properties of the conduction-band electrons and electrons bound to hydrogenlike states, and would therefore be sufficiently coupled to the ionized impurity atoms that they could contribute to the electron–mass-defect phonon interaction. This would naturally explain why the boron local mode is not observed in n-type material and the phosphorus mass-defect mode is not observed in p-type material, for there are no electrons in hydrogenlike states around a negatively charged ionized boron atom and there are no holes in hydrogenlike orbits about positive charged ionized phosphorus atoms. Such states in the reserve region of a tunnel junction have been used by Wolf and Losee[24] to explain the characteristics of a type of zero-bias anomaly.

The variation in the size of the Bohr radii among the semiconductors on which MIS tunnel junctions have been fabricated may be of importance in determining the strength of this interaction. For example, if the interaction with the majority carriers in the reserve region in hydrogenlike states is a deformation potential interaction, it would be expected that the interaction would be stronger for larger l/a, where l is the decay length of the mass-defect phonon and a is the Bohr radius of the majority carrier about the impurity atom. n- and p-type silicon and n-type 6H silicon carbide, in which electron–mass-defect phonon interactions have been observed, have smaller Bohr radii than either n-type germanium or n-type gallium arsenide in which electron–mass-defect phonon interactions have not been observed. If this suggestion is valid, one would expect that tunnel junctions fabricated with p-type silicon carbide, a material in which holes are very tightly bound to the impurity atoms, would exhibit large electron–mass-defect phonon interactions.

The size of the change in conductance of the boron local mode for both isotopes observed in MIS tunnel junctions fabricated with p-type silicon is approximately 3% at a doping of 2.3×10^{20} cm^{-3}, becoming unobservable at 2×10^{19} cm^{-3} (less than 0.5%).[2,3] In Fig. 2 the change in conductance is

6%. The larger size may arise from the heat treatment and rapid quench to room temperature accompanying the phosphorus diffusion. Since there are 3×10^{19} cm^{-3} holes in the valence band and at least 2.3×10^{20} cm^{-3} boron present, the size of the boron local-mode peak indicates that the mass-defect phonon interaction depends on the concentration of impurities and not the hole concentration.

Next, consider the electron – $k\approx0$ longitudinal optical phonon interaction. The negative-bias line shape has been previously observed[2,3] to be an increase in conductance (or a peak in derivative of conductance) at a boron doping level of 2.3×10^{20} cm^{-3} and a change in slope in conductance (or a step-up in derivative of conductance) at a doping level of 2.0×10^{19} cm^{-3}. The negative-bias line shape shown in Fig. 2, taken on material with 2.3×10^{20} cm^{-3} boron atoms and 3×10^{19} cm^{-3} holes, exhibits a change in slope in conductance (or a step-up in derivative of conductance). Detailed comparison with Fig. 7 of Ref. 3 clearly indicates that the line shape of the $k\approx0$ longitudinal optical phonon interaction depends on the hole concentration and *not* on the concentration of impurity atoms. This is opposite to the electron–mass-defect phonon interaction. Data taken on these samples have produced some of the other line shapes shown on Fig. 7 of Ref. 3, which also correlate with the hole concentration, as determined by the Fermi degeneracy.

The information on the electron–mass-defect phonon interaction can be summarized as follows: (i) The change in conductance is much stronger for that bias for which the majority carriers are tunneling from the semiconductor into the metal. (ii) The interaction does not occur in the barrier region of the semiconductor. (iii) The interaction depends on the concentration of impurity atoms, not on the concentration of majority carriers. (iv) The half-widths of the peaks in derivative of conductance data are not strongly temperature dependent. In n-type silicon the half-widths of the principal peaks are equal to about 6% of the energy of the peaks.

The information on the electron – $k\approx0$ longitudinal optical phonon interaction can be summarized as follows: The interaction depends on the concentration of majority carriers, not on the concentration of impurity atoms. Thus, the interaction between electrons and $k\approx0$ longitudinal optical phonons is different from the interaction of electrons and mass-defect phonons.

These observations make it unlikely that the electron–mass-defect phonon interaction can be explained by the many-body bulk effect that describes the coupling of the electrons and the $k\approx0$ longitudinal optical phonons. We suggest that the interaction of the tunneling electrons and the mass-defect phonons occurs in the reserve region of the semiconductor and that the pseudobound states of the impurities in this region of the semiconductor are important in the coupling of the electrons and the mass-defect phonons.

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Study of the Transverse Magnetoresistance of Polycrystalline Potassium†

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The impurity, strain, temperature, and size dependence of the transverse magnetoresistance of polycrystalline potassium has been investigated in fields up to 100 kG. The results of both low-field helicon experiments on large rectangular plate samples and four-terminal measurements on long wires show the existence of a nonlinear low-field behavior or "knee" previously reported in the magnetoresistance. The shape of the "knee" is observed to be strongly dependent on sample annealing and purity and, to a lesser extent, on temperature. In all samples under all experimental conditions, there is a linear magnetoresistance at high fields. The well-annealed wires exhibit a regular pattern in the impurity dependence of the high-field Kohler slope which differs from that reported for other simple metals. The Kohler slope is nearly independent of temperature below 4.2 K for samples of a wide range in purity. The experimental results are compared with predictions of macroscopic theories of a linear magnetoresistance due to sample geometry and inhomogeneities and with more recent proposals for an intrinsic linear magnetoresistance of potassium.

I. INTRODUCTION

Although theory predicts a saturating magnetoresistance for potassium, experimental work has produced a host of conflicting results. A linear magnetoresistance has generally been reported at high fields; however, there has been little agreement on the magnitude of the linear term and no satisfactory elucidation of the parameters determining its magnitude. Recently, there has also been disagreement as to whether there is a nonlinear low-field behavior or "knee" similar to that observed in the magnetoresistance of other closed-orbit metals such as indium and aluminum. While much of the uncertainty over early work centered on the effects of voltage probes and sample geometry, probeless methods and measurements in a wide variety of sample geometries have produced similarly conflicting results. Consequently, there has been increasing attention given to other factors, such as lattice defects and inhomogeneities, which could be of importance in a metal as soft and reactive as potassium.

Thus two primary reasons have led us to a paper of considerable length on measurements of the magnetoresistance of potassium. The first is the complicated nature of this phenomenon, which depends on several parameters and which has given rise to the variety of results having been reported. It requires a detailed analysis both to report our results and assess the extent to which they agree with previous work. The second reason is the crucial significance the magnetoresistance of potassium has for the semiclassical transport theory of metals. Potassium is generally regarded as one of the simplest metal systems, having a single band of conduction electrons with a nearly spherical Fermi surface. As such, it should have a saturating magnetoresistance at high fields. Yet there have been several intrinsic theories proposed in the last few years to explain the high-field linear magnetoresistance of potassium. We attempt to give a comprehensive assessment of these theories based on experimental data from a large number of samples. It is our hope that by providing this rather exhaustive "catalog" of experimental results we can give a sounder basis to future theoretical study of this important phenomenon.

In order to establish a basis for comparison with the work described in this paper, we briefly outline some of the more recent experimental results. Penz and Bowers[1] observed a linear magnetoresistance in the high-field regime $\omega_c\tau \gg 1$, where ω_c is the cyclotron frequency and τ the relaxation time determined from the zero-field resistivity.