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Low-Temperature Compressibilities and Absolute de Haas-van Alphen Frequencies in Potassium, Rubidium, and Cesium

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The absolute de Haas–van Alphen (dH–vA) frequencies and the scaling effect of small hydrostatic pressures on extremal cross sections of the Fermi surface have been measured in freely mounted samples of K, Rb, and Cs. The dH–vA frequencies are in good agreement with those derived from currently accepted values for the low-temperature lattice constants of the three metals at (1.824 ± 0.002) , (1.603 ± 0.003) , and $(1.365 \pm 0.002) \times 10^8$ G, respectively. The low-temperature compressibilities derived from the pressure measurements at $(2.57_7 \pm 0.013)$, $(3.64_4 \pm 0.015)$, and $(4.02_0 \pm 0.02) \times 10^{-2}$ kbar⁻¹, respectively, show discrepancies of -5, -8, and -14% with those measured by more conventional techniques. The characteristic energy B_0V_0 (bulk modulus × atomic volume) is found to be constant at about 1.67×10^5 J/mole in the three metals.

1. INTRODUCTION

Changes in the Fermi surface (F.s.) of a metal as a function of lattice constant may be studied by measuring changes in absolute or relative phase of de Haasvan Alphen (dH–vA) oscillations as a function of hydrostatic pressure in the liquid-helium bath surrounding the sample.^{1,2} Such studies have shown that there are small but measurable *distortions* in the shape of the F.s. in several cubic metals, in addition to the simple scaling effects predictable from the compressibilities of the metals concerned. However, the F.s.'s of the alkali metals potassium (K), rubidium (Rb), and cesium (Cs) differ only slightly from a free-electron sphere,^{3,4} and are thus unlikely to show any appreciable distortion in their shapes under small hydrostatic pressures. On the other hand, since the compressibilities of K, Rb, and Cs are rather large, the *scaling* effect of pressure on their F.s. dimensions will be large and easily measurable. Thus, the low-pressure dH–vA phase shift technique offers a rather precise direct way of measuring what are essentially the *differential* compressibilities of the alkali metals at very low temperatures, using

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pressures that are several orders of magnitude smaller than those used, for example, by Swenson and co-workers,^{5,6} in more conventional investigations of pressure-volume relationships in the alkali metals. Values for low-temperature bulk moduli of the alkali metals may also be derived from elastic constant data obtained by ultrasonic techniques (e.g., by Trivisonno and co-workers^{7–9}), but here again we believe our method is simpler and more precise.

In order to avoid nonhydrostatic effects we have, of necessity, to use freely mounted samples of the alkali metals. This allows us also to determine, with fair precision, the dH-vA frequency corresponding to the extremal cross section of a free-electron sphere in these metals under conditions that have not previously been realized.* Shoenberg and Stiles³ in their study of the F.s.'s of K and Rb, which involved the original use of the dH-vA phase-shift technique in steady high magnetic fields, used single-crystal beads frozen into petroleum jelly in thin-walled glass tubes. Okumura and Templeton,^{4,11,12} in their preliminary measurements on Rb and in their more detailed study of Cs, used single crystals in the form of fine wires crystallized in, and in intimate contact with, thin-walled glass tubes. Both of these techniques, because the thermal contractions of the alkali metals are very much larger than those of either frozen petroleum jelly or glass, could well lead to considerable negative pressures within the single crystals at low temperatures, and thus to erroneously low values of the measured dH-vA frequencies. These effects have been considered by Dugdale and Gugan¹³ and also discussed by Shoenberg and Stiles³ and by Okumura and Templeton.⁴ Their absence in the present experiments offers a closer approach to ideal conditions in making an absolute measurement of dH-vA frequency in an alkali metal.

2. THE PRESENT EXPERIMENTS

2.1. The Phase-Shift Method as Applied to the Alkalis

The sensitivity of the dH-vA phase-shift method in measuring changes of an F.s. cross section with pressure is related directly to the area of the cross section concerned and to the compressibility of the material. The smaller sizes of the alkali-metal F.s.'s as compared with those of the noble metals are more than compensated by the much higher compressibilities of the alkalis: The areas in the alkalis are smaller by about 3 times but the compressibilities are larger by about 50 times. The available pressure range of 25 bar at 1 K could then give a precision approaching 0.1%: it is, in fact, possible to detect the effect of as little as 1/40 bar on an alkali-metal sample. This corresponds to a phase shift of about 1/1000 of a dH-vA cycle at 50 kG.

2.2. Equipment and Techniques

The equipment and experimental techniques used have for the most part been described by Templeton.¹ The original pressure bomb and pickup coil were

^{*}Since these measurements were made, we have become aware of the recent high-precision measurements of absolute dH-vA frequency in potassium by Thomas and Turner¹⁰ that were also made on freely mounted samples. See Table I.