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Precursor effects to the transition in ReO₃

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The pressure dependence of the β and γ_1 de Haas-van Alphen (dHvA) frequencies of ReO₃ for the [011] direction show a nonlinear change with pressure in the range 0-3 kbar. This behavior has an electronic origin and is precursory to the transition between 3 and 5 kbar observed in all the dHvA frequencies

A pressure-induced change in the Fermi surface of ReO₃ has been of considerable interest recently. It was detected by Razavi *et al.*¹ in de Haas—van Alphen (dHvA) effect experiments as a 0.5% change in dHvA frequencies between 3 and 5 kbar for the [001] and [111] directions. No additional dHvA freqencies at high pressure that would indicate a crystallographic phase transition were found. Possible explanations of this effect that were considered in this first work were a change in electronic band width and a displacement of the oxygen atoms from their octrahedral sites.

These results were confirmed by later measurements of the α dHvA frequency for the [011] direction.² Evidence was also presented for a discontinuous increase in the compressibility by a factor of about 7 at 5 kbar. The change was interpreted in terms of an ususual "compressibility collapse." Further measurements of the first- and second-order quadrupolar effects of the NMR of Re indicated that the high-pressure phase has lower symmetry but remains cubic.³ It was suggested that the increased compression is related to buckling of the Re-O chain. This proposed structure was consistent with the results of a dHvA experiment⁴ at 5 kbar with a [001] magnetic field direction in which a new, low dHvA frequency was observed.

The compressibility of ReO_3 has also been determined from the change in length as a function of pressure up to 30 kbar.⁵ A striking divergence of the compressibility near 5 kbar was found. However, the lattice was quite soft at pressures above the divergence. These results suggested an electronic change in the band structure as well as a possible buckling of the Re-O chain. Thus, there is still uncertainty about the origin of this effect and whether it is electronic or structural in origin.

We have carried out additional dHvA experiments with the apparatus used previously¹ and have measured frequencies for the [011] direction up to pressures of 5 kbar. Data were taken in the field range 3-5.5 T with a sample temperature of 1.2 K. The frequency branches were labeled according to the scheme of Graebner and Greiner.⁶ The α , β , and γ_1 frequencies that are reported were derived by Fourier analysis from the same recording of dHvA oscillations. The crystal orientations and conditions at any pressure are therefore the same for all frequencies. The pressure dependence was checked by realigning the crystal and obtaining similar results a second time.

The α frequency shown in Fig. 1 increases linearly with pressure up to 3 kbar according to a Fermi surface area compressibility of 3.8×10^{-4} kbar⁻¹. The change of frequency with pressure is much greater above 3 kbar. The area compressibility obtained by considering the change above 3 kbar to be linear is 3.4×10^{-3} kbar⁻¹. These re-



FIG. 1. α -dHvA frequency of ReO₃ as a function of pressure with the magnetic field along the [011] direction.

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FIG. 2. γ_1 -dHvA frequency of ReO₃ as a function of pressure with the magnetic field along the [011] direction.

sults for the α frequency are in reasonable agreement with those found by Schirber and Morosin for the same frequency and are similar to the results obtained by us¹ for the [001] and [111] directions.

Figure 2 shows the pressure dependence of the γ_1 frequency. The pressure dependence is not linear below 3 kbar but rather shows a maximum at approximately 1.6 kbar. The frequency at 3 kbar is about the same as that at zero pressure. The increase of the γ_1 frequency with pressure above 3 kbar is similar to that of the α frequency.

The largest and most unusual change with pressure occurs for the β frequency as shown in Fig. 3. There is a local maximum at 1.5 kbar followed by a minimum, which is less than the frequency at zero pressure, at 3.2 kbar. The β frequency increases with pressure from 3.2 to 5 kbar with some scatter in the data points. The β frequency between 1 and 2 kbar was very sensitive to the degree of alignment of the magnetic field to the [011] direction and changed appreciably in different experiments. The error bars in Fig. 3 are the uncertainties of determining the frequencies from the data of one experiment.

The γ_1 and β frequencies are expected to be the most sensitive to electronic changes in ReO₃. This is because the energy bands are so close that there is magnetic breakdown between them to form the



FIG. 3. β -dHvA frequency of ReO₃ as a function of pressure with the magnetic field along the [011] direction.

 γ - β orbit when the magnetic field is aligned exactly along the [011] direction.⁷ The β Fermi-surface piece contains electrons and is roughly spherical centered at Γ . The γ Fermi surface consists of [100] directed electron cylinders intersecting at Γ and forming a multiple connected surface in the extended zone scheme. Small electronic redistribution will make a large relative change in the small energy gap between these surfaces at the [011] direction. There is a large energy gap between other orbits. The small electronic changes with pressure will therefore affect the γ_1 and β frequencies without influencing the other frequencies. It is therefore reasonable to assign the nonlinear changes with pressure below 3 kbar to changes in electronic properties, either from a change in bandwidth or an electronic redistribution in the band structure. This appears precursory to the transition between 3 and 5 kbar which is observed in all the dHvA frequencies and in the compressibility. These results, therefore, indicate an electronic origin of some of the unusual changes with pressure in ReO₃ in addition to a possible change in structure.

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