de Haas–van Alphen Effect in Gadolinium

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We have measured the de Haas–van Alphen effect in a single crystal of Gd, refined by solid-state electrolysis, observing four frequencies (in the range 10^5–3×10^7 G) near the c axis and three frequencies in the basal plane. The faster oscillations identify with orbits on the third-zone sub-band Fermi surfaces of the relativistic augmented plane-wave calculations, if the sub-band energies are displaced by ± 0.57 eV by the ferromagnetic internal field.

Although the Fermi surfaces of most metals and a number of intermetallic compounds have now been experimentally determined, there is almost a complete absence of experimental information on the Fermi surfaces of the trivalent rare-earth metals. These Fermi surfaces are, in fact, of particular interest because the variety of magnetic structures found in the rare-earth metals is usually attributed to quite small differences in their Fermi surfaces.[1,2] This paper presents results on the de Haas–van Alphen effect in gadolinium, and represents the first direct experimental measurement of this Fermi surface. Indeed, it is the first direct experimental determination, apart from the single de Haas–van Alphen frequency reported for lutetium,[3] in any trivalent rare-earth metal. The results will be interpreted in terms of the band structures calculated for heavy rare-earth metals by the relativistic augmented plane-wave (RAPW) method,[1,4] allowance being made for the energy difference between spin-up and spin-down sub-bands in a ferromagnetic material.

The purification of gadolinium by solid-state electrolysis and the growth of a single crystal have already been described.[5] The specimen, of resistance ratio 240,[6] was spark machined into a sphere of radius 3 mm. This sphere was afterwards smoothed by chemical polishing, but there was no attempt to make it accurately spherical by mechanical lapping. The measurements were performed by the usual modulation method, using an 80-kG superconducting solenoid (Oxford Instruments) and a Brookdeal phase-sensitive detector, at a modulation frequency of 230 Hz and a temperature of approximately 0.9 K. The normal field-measurement technique of *in situ* NMR could not be used with a ferromagnetic sample, so the field measurement was made using a calibrated copper magnetoresistor.

In a ferromagnetic material the periodicity of the de Haas–van Alphen effect is not in the reciprocal of the applied field B_a, but in the reciprocal of the total magnetic induction in the specimen. This is the sum of B_a and a shape-dependent term B_0 which is proportional to the magnetization and is thus constant once saturation has been established. The values of B_0 at the symmetry axes, obtained by fitting the best straight line to graphs of oscillation number versus $1/(B_a+B_0)$, were all in the range 6–8 kG. For field directions away from symmetry axes a value of 7 kG was assumed for B_0, and used in evaluating the de Haas–van Alphen frequencies. We assume throughout that $\vec{B}$ is parallel to the applied $\vec{H}$.[7]

The experimental frequencies are shown in Fig. 1 and Table I. The specimen was initially oriented by x rays, but the actual [0001] and $[2\bar{1}\bar{1}0]$ axes were located to ± 1° from the symmetry of patterns in the actual results. Figure 1(a) shows frequencies of four oscillations, all of which were well resolved at [0001]. Of the two faster oscillations, the larger amplitude at [0001] was exhibited by B. However, B had completely disappeared when the field was 25° from [0001] towards $[10\bar{1}0]$, whereas A displayed its largest amplitude at 55° and then fell rapidly to disappear sharply (± 1°) at 60° from [0001]. C disappeared quite sharply between 13° and 17°, but the disappearance of the very low-frequency oscillation D, which was best resolved at low fields, was not very clear cut—being masked by an increasing background of steady variation in the magnetization.

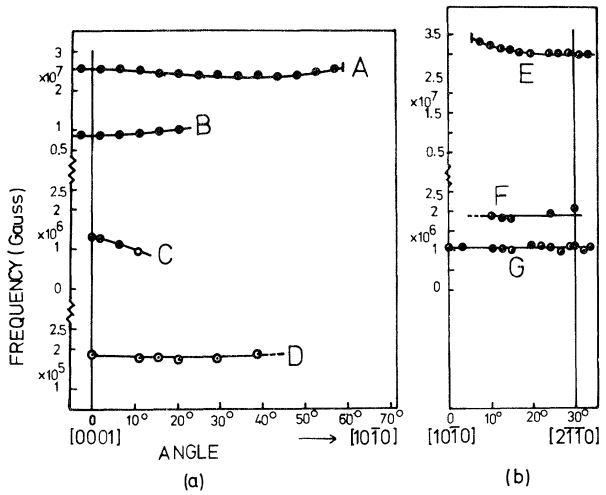

FIG. 1. Observed de Haas–van Alphen effect frequencies plotted against the orientation of the applied field (a) in the (2$\bar{1}\bar{1}$0) plane and (b) in the (0001) plane. It should be noticed that the vertical scale is not constant, the figure being split into three regions of different scale.

Although the signals for the field in the basal plane, Fig. 1(b), were less strong than for fields near [0001], the faster oscillation, E, was very well resolved at [2$\bar{1}\bar{1}$0]. The lowest-frequency oscillation, G, showed a greater amplitude than F over the whole range, although F could be detected everywhere except very close to [10$\bar{1}$0]. However, when the field was tipped from [2$\bar{1}\bar{1}$0] towards [0001], G was found to die out after only 12° whereas E and F persisted over the available range of tip of 15°.

Before discussing any theoretical models of the Fermi surface, it is of value to notice that oscillations A and B suggest a form for part of the surface. Thus, A is of almost constant frequency over 60° from [0001] and then disappears sharply, suggesting a spherical surface of radius 0.28 Å^{-1} whose central section is cut off at 60° by necks which extend in the [0001] direction. Assuming that the neck is represented by oscillation B, it will have a minimum radius of 0.16 Å^{-1} which will occur at approximately a distance of 0.16 $\times\cot(30°)$, i.e., 0.28 Å^{-1} from the center of the sphere. This surface is marked I in Fig. 2(b).

Both the relativistic[1] and the nonrelativistic[8] band-structure calculations predict essentially similar Fermi surfaces for nonmagnetic gadolinium, consisting of a [0001] "column" of holes in the third zone and a multiply connected fourth-zone electron surface. Keeton and Loucks (KL) also show their bands for hcp dysprosium, as typical of a heavy rare-earth metal, and these are confirmed by Jackson's[4] RAPW bands for hcp

TABLE I. The de Haas–van Alphen effect frequencies for the applied magnetic field along the principal symmetry axes. The surfaces are identified by 3 or 4 for the zone and r or l for the carriers being on the surfaces of raised or lowered energy in the internal field, respectively.

Field direction	Frequency (Gauss)	Index	Range (degrees)	Identification
[0001]	$2.55 \pm 0.09 \times 10^7$	A	60[a]	3r-α_1
	$0.89 \pm 0.03 \times 10^7$	B	22[a]	3r-α_2
	$1.24 \pm 0.08 \times 10^6$	C	15[a]	31 or 41 ?
	$1.90 \pm 0.13 \times 10^5$	D	40[a]	31 or 41 ?
[2$\bar{1}\bar{1}$0]	$2.96 \pm 0.09 \times 10^7$	E	24[b], >15[c]	31-β
	$2.00 \pm 0.20 \times 10^6$	F	27[a], >15[c]	41 ?
	$1.10 \pm 0.10 \times 10^6$	G	30[a], 12[c]	41 ?
[10$\bar{1}$0]	$1.06 \pm 0.03 \times 10^6$	G	30[b]	41 ?

[a]Towards [10$\bar{1}$0].
[b]Towards [2$\bar{1}\bar{1}$0].
[c]Towards [0001].

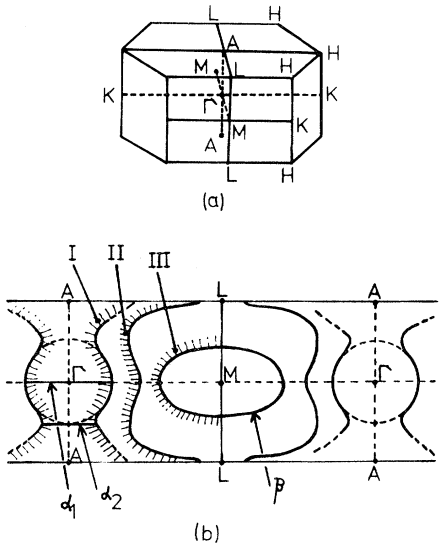

FIG. 2. (a) Symmetry points in the Brillouin zone of an hcp metal. (b) $(2\bar{1}\bar{1}0)$ sections of the third-zone "column" hole surface of Gd. II is the nonmagnetic section predicted by Keeton and Loucks, while I is our experimental section (which we identify with the raised holes), and III is the section which we predict for the lowered holes.

terbium. The $[2\bar{1}\bar{1}0]$ section of the third-zone column, as given by KL, is marked as II in Fig. 2(b). Clearly our experimental surface I is a much shrunken, but still recognizable, version of II. In a ferromagnetic metal the kinetic energies of spin-up and spin-down carriers at the Fermi surface are shifted by $\pm\Delta$ from the nonmagnetic Fermi energy, where Δ depends on the internal field. Thus these sub-bands have different Fermi surfaces, corresponding to changes $\pm\Delta$ in the nonmagnetic Fermi energy. We propose that the differences between KL's surface II and our experimental surface I is due to this change Δ in the apparent Fermi energy. To produce this shrinkage of the third-zone hole surface on the ΓMK plane of Jackson's bands requires $\Delta = 0.042$ Ry (0.57 eV).

Besides this surface of "raised holes" in the third zone, we can expect three other Fermi surfaces: the "lowered holes" in the third zone and the raised and lowered electrons in the fourth zone, the raising and lowering referring to the shifts of $\pm\Delta$ in the kinetic energies at the Fermi surface. The effect of lowering the Fermi energy of the third-zone hole surface of Jackson's bands by 0.042 Ry is to expand that surface so that it becomes multiply connected across the $[10\bar{1}0]$ zone face, as is shown in the $[2\bar{1}\bar{1}0]$ section, marked III, in Fig. 2(b). This gives rise to a $[2\bar{1}\bar{1}0]$ neck orbit β with a predicted frequency, assuming it to be an ellipse, of approximately 3.2×10^7 G. Our oscillation E has both the character and the frequency (3×10^7 G) to identify it with this orbit β.

The effect on the fourth-zone electrons of raising the Fermi energy by 0.042 Ry is to produce a multiply connected surface of [0001] tubes. This will give rise to frequencies of order 10^8 G, which is well above the fastest frequency we observe. A better candidate for our slow oscillations C, D, F, and G is the "lowered electron" surface, shrunken to a small volume along the HK line, produced when the electron Fermi energy in the fourth zone is lowered by 0.042 Ry. This is unfortunately a particularly difficult region for predicting the Fermi surface because it comes very close to the point H of the RAPW bands, where four bands are degenerate, and the line HK where the bands are practically flat as well as being partially degenerate. A more detailed calculation in this region will be necessary before attempts to identify our remaining oscillations are of any value. Another possible designation for our oscillations C or D is a neck at H on the surface of "lowered holes" in the third zone, but clearly no positive identification can be made at this stage.

We thus conclude that our three principal oscillations [in the range $(1-3)\times10^7$ G] can be interpreted as orbits on the third-zone hole surface of the typical RAPW band structure of a hcp rare-earth metal. The energy shifts of the magnetic sub-bands, due to the ferromagnetic internal field, required to fit our frequencies are ±0.57 eV. The available band structures are not sufficiently developed to account in detail for the low frequencies (10^5-10^6 G) which we observe.

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[6]In attempting to assess the electronic relaxation time of Gd from measurements of resistance ratio, it must be borne in mind that the room-temperature resistivity of Gd is inflated by about a factor of 2, compared to similar nonmagnetic metals (such as La or Lu), by magnetic disorder scattering.

[7]The small anisotropy energy of Gd [C. D. Graham, J. Appl. Phys. 38, 1375 (1967)] is readily overcome by our applied field.

[8]J. A. Dimmock and A. J. Freeman, Phys. Rev. Lett. 13, 750 (1964).

ERRATUM

OBSERVATION OF A PHASE-MODULATED QUASIPARTICLE CURRENT IN SUPERCONDUCTING WEAK LINKS. C. M. Falco, W. H. Parker, and S. E. Trullinger [Phys. Rev. Lett. 31, 933 (1973)].

On page 933, following Eq. (2), the correlation function should read $\langle \tilde{L}(t+\tau)\tilde{L}(t)\rangle = 2kT(\sigma_0+\sigma_1\cos\varphi) \times \delta(\tau)$ since thermal noise originates in both conductivities σ_0 and σ_1. This correlation function was implicitly assumed, thus leaving all results unchanged.