Determination of the Fermi Surface of MgB$_2$ by the de Haas-van Alphen effect

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We report measurements of the de Haas-van Alphen effect for single crystals of MgB$_2$, in magnetic fields up to 32 Tesla. In contrast to our earlier work, dHvA orbits from all four sheets of the Fermi surface were detected. Our results are in good overall agreement with calculations of the electronic structure and the electron-phonon mass enhancements of the various orbits, but there are some small quantitative discrepancies. In particular, systematic differences in the relative volumes of the Fermi surface sheets and the magnitudes of the electron-phonon coupling constants could be large enough to affect detailed calculations of $T_c$ and other superconducting properties.

There has been rapid progress in understanding the physical properties of magnesium diboride since the discovery of superconductivity at 39 K just over two years ago. The accepted consensus is that it is an s-wave, phonon-mediated superconductor but with some highly unusual properties. The most important of these are the anomalously high $T_c$ and the existence of two, almost distinct, superconducting gaps. This understanding is based on many experiments and on theoretical calculations of the unusual electronic structure of MgB$_2$. [1, 2, 3, 4].

There have been two direct experimental probes of the Fermi surface (FS) structure of MgB$_2$; angle-resolved photoemission spectroscopy [5] and the de Haas-van Alphen effect (dHvA). Our previous dHvA study [5] showed good agreement between theory and experiment regarding the areas of the orbits that were observed and their electron-phonon mass enhancements. However, only three out of nine predicted dHvA orbits were observed (labelled 1, 2 and 3 in Fig. 1) and so information was only obtained about two of the four FS sheets predicted by band calculations. It was not entirely clear whether the unobserved orbits were missing simply because of their relatively short mean-free-paths, or whether the topology of the other sheets was substantially different. This issue clearly affects calculations of many physical properties (especially $T_c$). In this Letter, we report new measurements of the dHvA effect in magnetic fields up to 32 Tesla for two single crystals. These new results give evidence for orbits on all four sheets of the Fermi surface.

Experiments were conducted on two different crystals grown by different groups. Sample B is one of the crystals studied previously [5] and was grown in Tokyo by high pressure synthesis using natural boron. Sample K was grown in Zürich by a similar technique but isotopically pure boron-10 was used as a starting ingredient [7]. Other crystals from the same batches were found to have anomalously high $T_c$ (onset) of 38.0 K and 37.7 K for the Tokyo and Zürich crystals respectively. dHvA oscillations were observed by measuring the torque ($\Gamma$) with a sensitive piezo-resistive cantilever technique [8]. Both samples were studied extensively at Bristol in fields up to 20.5 T and temperatures down to 0.3 K, and in fields up to 32 T at the NHMFL.

The first harmonic of the oscillatory part of the torque for a 3D Fermi liquid is given by [9, 10]

$$\Gamma_{osc} \propto \frac{1}{C} \frac{dF}{d\theta} B^* R_D R_T R_S \sin \left( \frac{2\pi F}{B} + \gamma \right)$$  \hspace{1cm} (1)

where $F$ is the dHvA frequency [$F = (h/2\pi c)A$], $A$ is the extremal orbit area in k-space; $C$ is the curvature factor, $\gamma$ is the phase; $R_D$, $R_T$ and $R_S$ are the damping factors from impurity scattering, temperature and spin splitting respectively. The Dingle factor, $R_D = \exp \left( - \frac{\pi m}{eB^2} \right)$, where $m_B$ is the unenhanced band mass [4, 10] and $\tau$ is the scattering time. An equivalent expression for $R_D$ is $R_D = \exp \left( - \frac{\pi mass}{eB^2} \right)$, which shows...
clearly the increased damping with shorter mean-free-path \( \ell \) and increased average Fermi wavevector \( k_F \) of the orbit. \( R_T = X/(\sinh X) \) where \( X = \frac{2\pi g}{k_B \bar{m}_e} \), \( \bar{m}_e \) is the quasi-particle effective mass. Finally, the spin splitting factor is given by \( R_S = \cos(\pi n g m_B (1 + S)/2m_e) \) where \( (1 + S) \) is the orbitally averaged exchange-correlation (Stoner) enhancement factor, \( g \) is the electron \( g \)-factor, \( m_e \) is the free-electron mass and \( n \) is an integer.

The electronic structure and dHvA orbits of MgB\(_2\) have been calculated by three different groups \([11, 12, 13]\). The calculated Fermi surface is shown in Fig. 1, together with the expected dHvA extremal orbits \([14]\). The calculations \([1]\) show that the electronic states near the Fermi level arise primarily from the boron atomic orbitals. The calculated Fermi surface is composed of four distinct sheets. Two of these arise from boron \( \sigma \) orbitals and are quasi-two dimensional warped cylinders running along the \( c \)-direction, whereas the other two are tubular networks with larger \( c \)-axis dispersion, that are mainly formed from the boron \( \pi \) orbitals. In total 9 primary extremal orbits have been predicted, and 7 of these are labelled in the Figure (two orbits with frequencies >30kT have been omitted). Calculations of the dHvA frequencies and masses of the various orbits by the three groups are all in good agreement (the differences are typically 100-200 T, and \( \lesssim 5\% \) respectively).

The fast Fourier transforms (FFTs) of the raw torque data between 20 and 32 T at 1.4 K for crystal K are shown in Fig. 2 as a function of angle \( \theta \) as the sample was rotated from \( H \parallel c \) to \( H \parallel a \) (\( H \parallel c \equiv 0^\circ \)). In addition to the frequencies observed in our previous study \([8]\) (\( F_1, F_2 \) and \( F_3 \)) several additional peaks are visible. Some care is needed in interpreting these new frequencies. Because the cantilever is deflected slightly by the torque, the torque measurements are not made at constant angle. This generates spurious harmonics and combinations of the main dHvA oscillations. For crystal K only weak harmonics of \( F_2 \) are observed (just above \( F_5 \)– see Fig. 2), whilst for the larger crystal B, several harmonics of \( F_3 \) and a frequency corresponding to \( F_1 + F_2 \) were observed.

The observed frequencies (omitting those assigned as harmonics or combinations) are shown in Fig. 3 as a function of \( \theta \) for both crystals. The solid lines in Fig. 3 are fits of the observed \( F(\theta) \) values to \( F(\theta) = \sum_{i=1}^{3} \alpha_i \cos^i(\theta - \theta_0) \) (\( \theta_0 = 0 \) or \( 90^\circ \)), which we use to extrapolate the observed frequencies to the symmetry points (we denote these frequencies as \( F_{0n} \)). The assignment of the FFT peaks to the orbits shown in Fig. 1 was achieved by comparing the values of the frequencies obtained by extrapolation, and their angular dependencies, with the calculations.

For crystal K, signals from 6 orbits associated with all
4 sheets of Fermi surface are observed. We are therefore able to verify experimentally the Fermi surface topology predicted by Kortus et al. and shown in Fig. For crystal B, in addition to the 3 orbits observed previously, 3 further frequencies are seen. One of these can be assigned to F₄. The frequency of F₃ (F=4600 T) is close to that predicted for orbit 7 (F=4294 T) and has a similar effective mass, however, the frequency is also close to that expected for an orbit equivalent to F₃ but from the tube oriented along a (i.e., at 60° to a). A subsequent in-plane rotation study showed the latter to be the most likely origin. The origin of F₄ is less clear, but it could arise from a slight warping of the same in-plane tube responsible for F₃.

Table I shows that the values of F⁰ found for the two crystals are in good agreement. In total, we have studied crystals from six batches (5 from Tokyo and one from Zürich) and so far, the dHvA frequencies agree to within 30 T or 0.06 % of the basal area of the first Brillouin zone (50.2 K). This suggests that although Tc is slightly reduced compared with the best polycrystalline samples, any possible Mg deficiency is very reproducible, and probably small.

The differences between the measured F⁰ values and those predicted by theory is a significant fraction of F⁰ in some cases, but we note that 100 T discrepancy only amounts to ~ 0.3 % of the basal area of the Brillouin zone. The volumes of the tubes are proportional to the average of the two extremal areas and these are both ~ 16% smaller than the calculations, implying a corresponding reduction in the number of holes in these two tubes. This may have a significant effect on calculations of physical properties such as Tc, its dependence on alloying or pressure, and the London penetration depth.

It is instructive to calculate the Fermi energy shift ΔE which would bring the theoretical frequencies in line with experiment. As the dHvA band mass is defined as m_B = \frac{\hbar^2}{2\pi^2} \frac{\partial^2 E}{\partial k^2}, the necessary band shifts are given by ΔE = \frac{\hbar e}{m_B} ΔF (where ΔF = F_Th - F_exp). There is remarkable consistency between the ΔE values for the orbits, with the values roughly falling into two groups (Table I). For the σ sheet orbits (1,2,5,6) the average shift is 83 ± 4 meV, whereas for the π sheet orbits (3,4) it is 61 ± 5 meV. Because of the high degree of reproducibility of the frequencies between samples, it is unlikely that this discrepancy is caused by sample impurities or non-stoichiometry. Instead, it seems to imply a shortcoming of the LDA calculations.

Quasiparticle effective masses were determined by performing field sweeps at different temperatures and fitting the amplitudes to Eq. We our experimental values of m* are compared with the calculated band masses in Table I. In MgB₂ the dominant source of mass enhancement is the electron phonon interaction. If we assume this is the only source of enhancement we can calculate an upper bound for the electron-phonon coupling constants λ, from λ = m*/m_B - 1. The results (Table I) show that the values of λ on both the σ sheets are approximately a factor three larger than those on the π sheets.

A detailed comparison with the orbit-resolved theoretical values of λ is also shown in the table. Generally our values are slightly smaller than the theoretical ones. The most significant differences are for the larger orbits on the σ tubes for which λ values which are ~ 20% smaller than theory. The small shifts in Fermi level described above do make small differences to the calculated band masses, increasing the λ values by ~ 6%, but the differences remain significant. These differences could have a relatively large effect on the detailed calculations of Tc. For example, using the isotropic McMillan equation, with parameters appropriate to MgB₂, we estimate a 20% reduction in λ would imply a ~ 8 K reduction in Tc. It is likely that the reduced λ values we observe are caused by phonon anharmonicity, which has been shown to reduce the average value of λ by around 20 %.

Scattering rates (τ^-1) were calculated by fitting the raw torque versus field data to Eq. The results are also shown in Table I. It can be seen that the mean free path (ℓ) for sample K is significantly longer for orbits F₂, F₁ on the c-axis tube but not for F₄ on the in-plane...
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17 T, and for crystal K, \(B_{c2}^{||} = 2.5\) T, \(B_{c2}^{\perp} = 12\) T (all values quoted at \(T = 0.3\) K).

The longer mean free path of the smaller \(\sigma\) tube (\(F_1\) and \(F_2\)) in crystal K, has allowed us to track the amplitude of the signal over a very wide range of angle (up to 67° and 81° respectively). The data shown in Fig. 4 show pronounced minima at \(\theta = 51 \pm 1^\circ\), \(\theta = 53 \pm 1^\circ\), \(\theta = 75.7 \pm 0.5^\circ\) for \(F_1\), \(F_2\), and \(F_3\) respectively, which we attribute to the spin-zero effect (the \(R_e\) damping term in Eq. 1). Using the calculations of \(m_B(\theta)\) by Harima [11] we can deduce the enhancement of the spin susceptibility (1 + \(S\)) for these three orbits (at the spin zero angle). Taking \(g = 2\), we find that \(S = 0.07, 0.12\) and 0.45 for the three orbits respectively. The reason for the significantly larger enhancement on the \(\pi\) band is not clear. Band structure calculations [12] predict \(S = 0.31\) and 0.26 on the \(F_2\) and \(F_3\) orbits respectively. The calculation therefore overestimates \(S\) on the \(\pi\) sheet by a factor 2.5 and underestimates it on the \(\sigma\) sheet by a factor 1.7.

The solid lines in Fig. 4 show a fit to the data for \(F_1\) and \(F_2\) with Eq. 1 using the \(m_B\) values of of Harima [11]. There are 3 free parameters: the overall amplitude, \(\tau^{-1}\) (assumed constant as a function of angle), and \(S\). The fit is remarkably good and the values of \(\ell\) were found to be 410 Å and 900 Å for \(F_1\) and \(F_2\) respectively which are close to the values obtained from a fit to the field dependence at a single angle given in Table 1. However, it was not possible to fit the data for \(F_3\) because the abrupt fall for \(\theta \lesssim 68^\circ\) cannot be explained simply in terms of the angular dependence of \(m_B\) and a constant scattering rate (Dingle plots show that \(\tau\) is approximately constant with angle).

In conclusion, our data strongly support the overall topology of the predicted electronic structure of \(\text{MgB}_2\) and the calculations of the electron-phonon coupling constants for the different orbits. Our data give direct evidence that the electron-phonon interaction is large on both \(c\)-axis \(\sigma\) sheets and much smaller on both \(\pi\) sheets. We have therefore obtained conclusive evidence in favor of the two key ingredients in the two-gap model of superconductivity in this compound, namely the Fermi surface topology and the disparity in the electron-phonon coupling for the \(\sigma\) and \(\pi\) bands.

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![FFT amplitude versus angle for frequencies F1, F2 and F3 in crystal K. The solid lines are fits to the data as described in the text. The dotted line is a guide to the eye.](image)

References:

[1] J. Kortus, I. I. Mazin, K. D. Belashchenko, V. P. Antropov, and L. L. Boyer, Phys. Rev. Lett. 86, 4656 (2001).
[2] A.Y. Liu, I. Mazin and J. Kortus, Phys. Rev. Lett. 87, 087005 (2001).
[3] H.J. Choi, et al. Nature 418, 758 (2002); Phys. Rev. B 66, 020513(R) (2002).
[4] I. I. Mazin et al. Phys. Rev. Lett. 89, 107002 (2002).
[5] H. Uchiyama et al. Phys. Rev. Lett. 88, 157002 (2002).
[6] E.A. Yelland, J.R. Cooper, A. Carrington, N.E. Hussey, P.J. Meeson, S. Lee, A. Yamamoto and S. Tajima, Phys. Rev. Lett. 88, 217002, (2002).
[7] Raman studies of crystals from the same batch give a ^10^B substitution level of \(\sim 70\%\) (E. Liarokapis, Private communication).
[8] C. Bergemann, Ph.D. Thesis (University of Cambridge) (1999).
[9] D. Shoenberg, *Magnetic Oscillations in Metals*, (Cambridge University Press, 1984) ISBN 0521 224802
[10] A. Wasserman and M. Springfield, Adv. Phys. 45, 471 (1996).
[11] H. Harima, Physica C378-381, 18 (2002); and private communication.
[12] I.I. Mazin and J. Kortus, Phys. Rev. B 65, 180510 (2002).
[13] H. Rosner, J.M. An, W.E. Pickett and S.L. Drechsler, Phys. Rev. B 66, 024521 (2002).
[14] Note that \(F_\tau\) was only predicted by one author [11], and that this work also predicts two additional frequencies close to \(F_3\) and \(F_\tau\) which arise from slight undulations of the Fermi surface.
[15] J.R. Cooper et al., Physica (Amsterdam) 385C, 75 (2003).