Method to determine absolute amplitudes in the de Haas—van Alphen effect

L. Nordborg, P. Gustafsson, H. Ohlsén, and S. P. Hörnfeldt*
Department of Solid State Physics, Uppsala University, Box 534, S-751 21 Uppsala, Sweden
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A new method to determine absolute amplitudes in the de Haas—van Alphen effect is presented. The method is based on magnetic interaction effects and the field-modulation technique. It is applied to the α orbit on the open hole sheet and the closed Γ_6 sheet for $\mathbf{B}\|[100]$ in platinum. The results are found to agree well with calculated values.

I. INTRODUCTION

Several physical quantities can be determined using the de Haas—van Alphen (dHvA) effect without knowledge of the absolute values of the detected oscillatory magnetization amplitudes.[1] For some applications it is, however, advantageous to also determine the absolute amplitudes. Shoenberg and Vanderkooy[2] performed absolute-amplitude measurements of the neck oscillations in the noble metals to confirm the theoretical results of Engelsberg and Simpson[3] that many-body effects do not influence the dHvA amplitude predicted by the Lifshitz and Kosevich (LK) independent-quasiparticle model.[4] Absolute-amplitude measurements can also be useful to determine the spin-splitting factor (see Sec. II) and from that the orbital g factor. This can be done by comparing the measured amplitude with the calculated amplitude and thereby obtaining the spin-splitting factor. Often the detection system must be calibrated in order to measure the absolute amplitude. This is easily done when using a torque magnetometer.[2] However, the field-modulation technique is the most frequently used method for detecting the dHvA effect. When calibrating the detection system for this technique several methods were developed (see, e.g., Refs. 5—9) demanding, for example, a measurable harmonic content or making use of the modulation coil as a pick-up coil. It is obvious that when absolute amplitudes are to be determined it is advantageous to use a method that does not depend on the gain of the detection system nor on the coupling between the sample and the pick-up coil.

Alles and Lowndes[10] have discussed how one dHvA frequency through magnetic interaction will modulate the amplitude of another higher dHvA frequency that is simultaneously present. They demonstrate how the low-frequency modulates the Bessel-function argument for the high frequency when the large-amplitude field-modulation technique is employed. In view of the works of Crabtree,[11] Phillips and Gold,[5] and Shoenberg[12] this modulation of the Bessel-function argument is discussed in this paper and how it can be used to determine the absolute amplitude of the lower frequency, and from that, also the absolute amplitude of the higher frequency. The method described is used to determine the absolute amplitudes of the oscillations from the α orbit and Γ_6 orbit in platinum for $\mathbf{B}\|$ [100]. For these orbits, all of the quantities necessary for making a direct comparison between theory and experiment possible are known.

II. THEORETICAL CONSIDERATIONS

We consider a case in which two dHvA frequencies are simultaneously present. We assume for simplicity the following.

(i) We have one high frequency F_H and one low frequency F_L with a ratio between the frequencies of at least five.

(ii) The magnetic field is orientated along a direction of symmetry both for the crystal and for the orbits. This means that the magnetization is parallel to the applied field and the problem can be treated in one dimension.

(iii) The shape of the sample is such that a demagnetizing factor (N) is obtainable.

From the LK theory, the oscillating magnetization from the fundamentals of the two frequencies can be written as

$$M_i = A_i \sin\left[\frac{2\pi F_i}{B}\right], \tag{1a}$$

where F is the dHvA frequency, i is either of the two frequencies H and L, B is the applied magnetic field, and

$$A_i = -\left[\frac{\lambda T F \exp(-\alpha m_c T_D/B)\,|\cos(\pi g_c m_c/2)|}{B^{1/2}\,|\partial^2 S/\partial k_z^2|^{1/2}\sinh(\alpha m_c T/B)}\right]_i, \tag{1b}$$

where $\alpha = 14.69$ $(\mathrm{T\,K^{-1}})$ and $\lambda = 0.6523$ $(\mathrm{A\,m^{-1}\,K^{-1}\,T^{-1/2}})$, m_c is the cyclotron effective mass expressed in units of the free-electron mass, T is the temperature, T_D is the Dingle temperature, $|\partial^2 S/\partial k_z^2|$ is the curvature factor, and $|\cos(\pi g_c m_c/2)|$ is the spin-splitting factor including the cyclotron orbit g factor.

The magnetizations from the two frequencies should, however, be added to the applied magnetic field B in the arguments of Eq. (1a) and thus the oscillations will be affected by their own presence. This is often referred to as magnetic interaction (MI). Crabtree[11] has derived an explicit solution to second-order terms of this field problem under the restrictions listed above by applying a total magnetic field $B_{\mathrm{tot}} = B + \mu_0 M + \mu_0 M_D$, where M is the LK sum of the two frequencies and M_D represents the demagnetizing field in the crystal. His result for the magnetization is

$$M = A_H \sin\left[2\pi\frac{F_H}{B}\right] + A_L \sin\left[2\pi\frac{F_L}{B}\right] + A_{H2}\sin\left[2\pi\frac{2F_H}{B}\right] + A_{L2}\sin\left[2\pi\frac{2F_L}{B}\right]$$
$$-K_H\frac{(A_H)^2}{2}\sin\left[2\pi\frac{2F_H}{B}\right] - K_L\frac{(A_L)^2}{2}\sin\left[2\pi\frac{2F_L}{B}\right] - (K_H+K_L)\frac{A_H A_L}{2}\sin\left[2\pi\frac{F_H+F_L}{B}\right]$$
$$+(K_H-K_L)\frac{A_H A_L}{2}\sin\left[2\pi\frac{F_H-F_L}{B}\right], \tag{2}$$

where the labels A_{H2} and A_{L2} refer to second harmonics of the LK signal. The demagnetizing field is contained in K, where

$$K_i = \mu_0\frac{2\pi F_i}{B^2}(1-N) \tag{3}$$

and N is the demagnetizing factor.

When using the field-modulation technique the detected amplitude depends upon the modulation-field amplitude according to a Bessel function $J_n(X)$, where n is the specific harmonic of the modulation frequency on which the detection is based. The Bessel-function argument $X = 2\pi F_i b/B^2$ and b is the modulation-field amplitude. When detecting the dHvA signal with the Bessel-function argument not much larger than that for the first Bessel-function zero, the Bessel-function values for the lower frequencies will be close to zero. The second-harmonic amplitudes A_{H2} and A_{L2} are considerably smaller than the amplitudes for the fundamentals. Thus, the terms 2, 3, 4, and 6 in Eq. (2) can be disregarded in future considerations and we obtain

$$M \propto 2A_H J_n\left[2\pi\frac{F_H b}{B^2}\right]\sin\left[2\pi\frac{F_H}{B}\right] - K_H(A_H)^2 J_n\left[2\pi\frac{2F_H b}{B^2}\right]\sin\left[2\pi\frac{2F_H}{B}\right]$$
$$-(K_H+K_L)A_H A_L J_n\left[2\pi\frac{(F_H+F_L)b}{B^2}\right]\sin\left[2\pi\frac{F_H+F_L}{B}\right] + (K_H-K_L)A_H A_L J_n\left[2\pi\frac{(F_H-F_L)b}{B^2}\right]\sin\left[2\pi\frac{F_H-F_L}{B}\right]. \tag{4}$$

When close to a Bessel-function zero, the Bessel function may be written in a linear approximation:

$$J_n\left[2\pi\frac{F_i b}{B^2}\right] = C\frac{F_i}{B_i^2}\left[1-\left[\frac{B_i}{B}\right]^2\right], \tag{5}$$

where B_i is the magnetic field for which

$$J_n\left[2\pi\frac{F_i b}{B_i^2}\right] = 0\,. \tag{6}$$

Then,

$$\frac{F_H}{B_H^2} = \frac{F_H+F_L}{B_{H+L}^2} = \frac{F_H-F_L}{B_{H-L}^2} \tag{7}$$

and from Eqs. (4), (5), and (7) we obtain

$$M \propto A_H\left[1-\left[\frac{B_H}{B}\right]^2\right]\sin\left[2\pi\frac{F_H}{B}\right] - \left\{A_H A_L K_L\left[1-\left[\frac{B_H}{B}\right]^2\right] - A_H A_L K_H\frac{B_H^2}{F_H}\frac{F_L}{B^2}\right\}\cos\left[2\pi\frac{F_L}{B}\right]\sin\left[2\pi\frac{F_H}{B}\right]$$
$$-\left\{A_H A_L K_H\left[1-\left[\frac{B_H}{B}\right]^2\right] - A_H A_L K_L\frac{B_H^2}{F_H}\frac{F_L}{B^2}\right\}\sin\left[2\pi\frac{F_L}{B}\right]\cos\left[2\pi\frac{F_H}{B}\right]$$
$$-(A_H)^2 K_H J_n\left[2\pi\frac{2F_H b}{B^2}\right]\sin\left[2\pi\frac{2F_H}{B}\right] \Big/ \left[2C\frac{F_H}{B_H^2}\right]. \tag{8}$$

In the next step we will especially study what will happen when $\cos(2\pi F_L/B)\sim\pm1$, which means that $\sin(2\pi F_L/B)\sim0$. Keeping in mind that K_L is much smaller than K_H, we can further simplify the expression. The detected amplitude of the fundamental for the high-frequency signal, incorporating $K_H=2\pi\mu_0(1-N)F_H/B^2$ and multiplying by $1/A_H B_H^2$, may then be written as

$$M_H \propto \left[\frac{1}{B_H^2}-\frac{1}{B^2}+2\pi\mu_0(1-N)\frac{F_L A_L}{B^4}\cos\left[2\pi\frac{F_L}{B}\right]\right] \times\sin\left[2\pi\frac{F_H}{B}\right] . \tag{9}$$

By observing the locations of the magnetic field for which the amplitude vanishes it is possible to identify the fields B_+ and B_- for which the following relation holds:

$$\frac{1}{B_-^2}\left[1+\mu_0\frac{(1-N)2\pi F_L A_L}{B_-^2}\right] =\frac{1}{B_H^2}=\frac{1}{B_+^2}\left[1-\mu_0\frac{(1-N)2\pi F_L A_L}{B_+^2}\right] . \tag{10}$$

If $\mu_0(1-N)2\pi F_L A_L/B^2 \ll 1$, B_H will be close to the mean value of B_+ and B_- and Eq. (10) can be reduced to

$$(1-N)A_L=\frac{B_-^2-B_+^2}{4\pi\mu_0 F_L} . \tag{11}$$

The effect of the lower frequency on the higher frequency can in a simplified manner be interpreted according to Alles and Lowndes.[10] The presence of the lower frequency magnetization H_L will give an effective Bessel-function $J_n(X)$ argument for the higher frequency:

$$X=\frac{2\pi F_H}{B^2}b\left[1+\mu_0(1-N)\frac{\partial M_L}{\partial B}\right] . \tag{12}$$

Compared with M_H, $\partial M_L/\partial B$ is slowly varying with B and can easily be found in the experiment through an effective modulation of the high-frequency oscillation. By selecting b such that the Bessel function is close to a zero, the effect from $\partial M_L/\partial B$ will dominate the detected signal. In the experiment, b can be kept constant while B is slowly increased. In the detected signal, the positions in B, where $\partial M_L/\partial B=0$ or has its extreme values, are easily recognized (see Fig. 1). By plotting the amplitude of the high-frequency oscillations at these field positions versus the magnetic field positions, three Bessel-function zeros will result. The amplitudes for which $\partial M_L/\partial B=0$ will give B_H, the position in B of the Bessel-function zero caused by b in the absence of $\partial M_L/\partial B$. Plotting the extremes of $\partial M_L/\partial B$ will give two satellites to B_H which we then label B_- and B_+. These satellite magnetic field values correspond to the same Bessel-function argument as for B_H but where the maximum and minimum values of $\partial M_L/\partial B$ must be included in Eq. (12). This gives a relationship identical to Eq. (10) for determining $\partial M_L/\partial B$. Since this amplitude measurement determines the sign of A_L (see Fig. 1), a measurement of the low-frequency signal in the same field interval will reveal if the detected amplitude signal is phase shifted 180° or not, thus giving a sign calibration for the detection system.

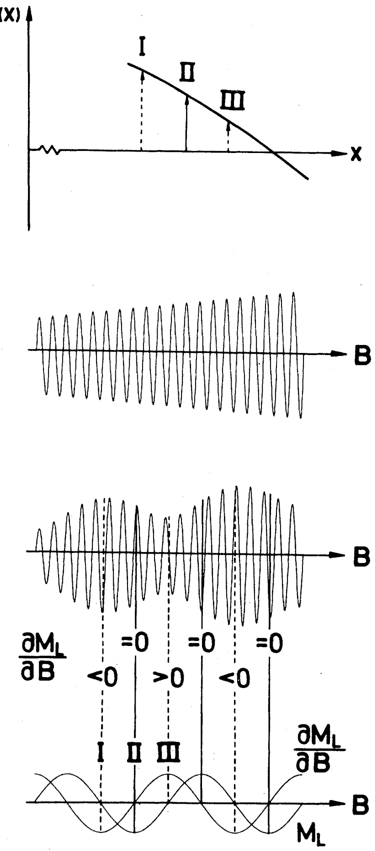

FIG. 1. With increasing field, the argument X of the Bessel function is decreasing. To the left of the zero crossing in (a) the amplitude is therefore increasing. In absence of a low frequency, the detected signal will appear as in (b). In the presence of an M_L oscillation, X will oscillate between two extreme values depending on $\partial M_L/\partial B$ [see Eq. (12)] and will give an amplitude-modulated signal (c). From this [and the knowledge of the sign of $J_n(X)$] the sign of both $\partial M_L/\partial B$ and M_L can be obtained (c), and the detection system sign calibrated.

Once the low-frequency amplitude is known, absolute amplitudes of other frequencies may also be determined. The detected amplitude of the low-frequency signal can be written as $DJ_n(X_L)(1-N)A_L$ where D is the detection constant of the system and $J_n(X_L)$ is the Bessel-function value for the low-frequency signal. Since $(1-N)A_L$ is measured and $J_n(X_L)$ can be calculated, it is possible to determine the constant D. The absolute amplitude may be determined for any other frequency by detecting the signal and calculating its Bessel-function value $J_n(X)$. However, the experimental "Bessel function" might differ from the mathematical[10] and therefore it is preferable to determine the absolute amplitude of the higher frequency in the following way. Choose a sufficiently high modulation field so that the amplitude of the low-frequency signal is easily detected. This will then give $DJ_n(X_L)$. The low-frequency signal has a Bessel argument F_L/F_H times lower than the higher frequency. If the modulation field is reduced by this ratio, the Bessel-function value will remain the same when detecting the high-frequency signal. $(1-N)A_H$ is then easily obtained from the detected signal.

III. EXPERIMENTAL TECHNIQUE

Single crystals of pure platinum were prepared by conventional electron-beam zone refining. The starting material of 99.999% purity (supplied from Johnson and Matthey Co.) in the form of a 1-mm-thick wire was zone refined for 20 zone passages. A final zone passage downwards was made to obtain a single crystal sample about 6 mm long. The single crystal samples were spark cut into 1-mm pieces and etched in hot aqua regia to remove surface damages caused by the spark cutting.

Ordinary field-modulation technique was employed to observe the dHvA signals. The experiment was performed at a temperature of 0.55 K. The platinum sample was mounted in a rotator, so that the magnetic field could be rotated in a symmetry plane. The magnet is installed in such a way that it can be tilted $\pm 3.5°$ when energized. By performing field rotation using the rotator and subsequently tilting the magnet, the field could be aligned parallel with the [100] axis to within two-tenths of a degree. The dHvA signals were recorded by making use of the fourth and eighth harmonic of the modulation frequency (210 Hz).

IV. RESULTS

The method was used to determine the absolute amplitudes of two orbits in platinum. The low-frequency signal is the α orbit existing at the W point on the open hole sheet and the high-frequency signal is the orbit on the closed Γ_6 sheet. The modulation amplitude was selected such that the Bessel argument fell outside the first zero of J_8. Then, a magnetic field sweep was performed towards larger fields and the dHvA signals were recorded. The Bessel-function argument will decrease with increasing magnetic field and the dHvA signal will exhibit the Bessel-function zero and the two satellites caused by the magnetization modulation from the α orbit as described in Sec. II and Fig. 1. The result is presented in Fig. 2. The values of B_+ and B_- were determined by graphic interpolation as shown in Fig. 2(b) and are presented in Table I. From Eq. (11), $(1-N)A_\alpha=-48$ A/m was obtained. The demagnetizing factor for the cylindrically shaped sample was estimated to be $N=0.33$, after Crabtree,[11] and thus $A_\alpha=-72$ A/m.

TABLE I. Amplitudes for the Γ_6 and α orbits. Quantities used from Refs. 13 and 14 are as follows. α: $F_\alpha=2770$ T, $|\cos(\pi g_c m_c/2)|=0.80$, $m_c=1.53$; Γ: $F_\Gamma=28\,800$ T, $|\cos(\pi g_c m_c/2)|=0.94$, $m_c=2.44$.

Experiment		LK theory	
B_+ (T)	6.604	$T_D(\alpha)$ (K)	0.31
B_- (T)	6.442	$T_D(\Gamma)$ (K)	0.25
A_α (A/m)	-72		-66
A_Γ (A/m)	-45		-42

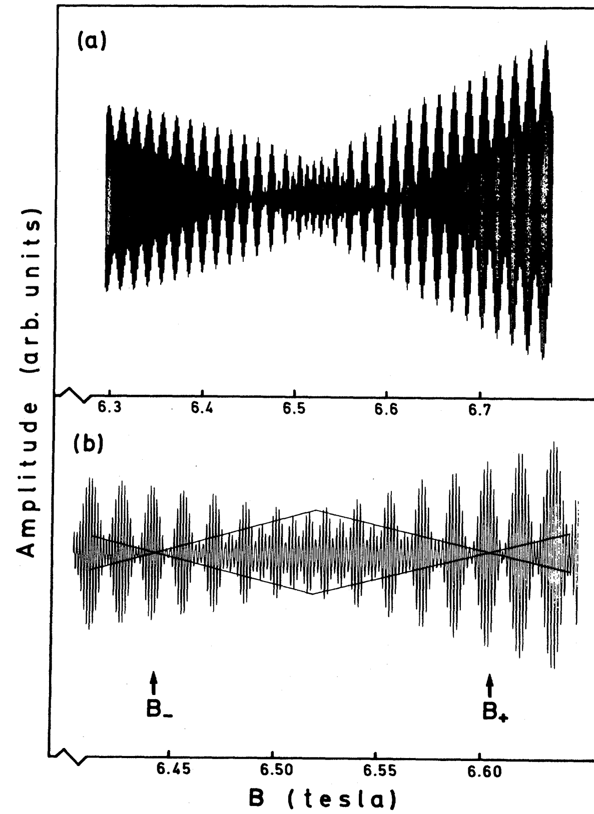

FIG. 2. (a) Amplitude of the detected signal as a function of B, showing how the three Bessel-function zeros appear in a field sweep. (b) Close up of (a) showing the graphic interpolation procedure (weak solid lines) to determine B_- and B_+.

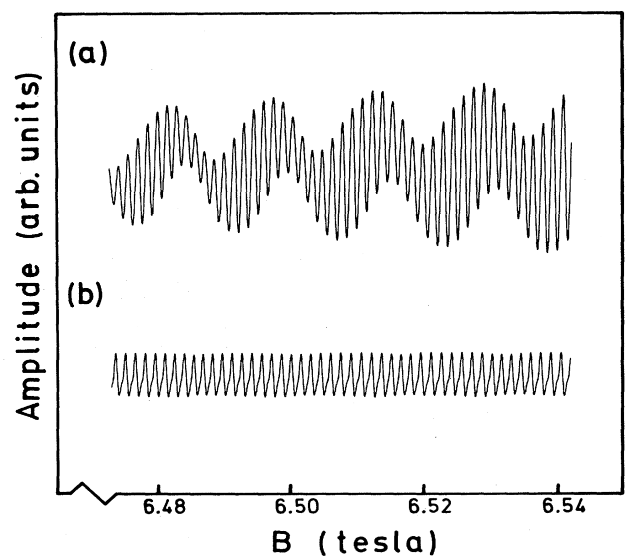

FIG. 3. (a) Amplitude of the detected signal as a function of B, showing the two signals $DJ_4(X_\alpha)M_\alpha$ and the amplitude-modulated $DJ_4(X_\Gamma)M_\Gamma$. (b) Detected signal when the modulation field is reduced by a factor F_α/F_Γ showing the remaining signal $DJ_4(X_\alpha)M_\Gamma$ with a considerable second-harmonic content.

To determine the absolute amplitude of the M_Γ signal according to the procedure described in Sec. II, the field sweep shown in Fig. 3(a) was performed. Here, the fourth harmonic was used to optimize the ratios of the detected signals. Two different signals can be seen; the low-frequency signal $DJ_4(X_\alpha)M_\alpha$ and the amplitude-modulated high-frequency signal $DJ_4(X_\Gamma)M_\Gamma$. This modulation is caused by MI and has the same frequency as M_α. The modulation field was then reduced by a factor F_α/F_Γ giving the signal in Fig. 3(b). Here, $DJ_4(X_\alpha F_\alpha/F_\Gamma)A_\alpha \approx 0$ but since a low Bessel-function argument is used, the signal $DJ_4(X_\alpha)M_\Gamma$ is strongly affected by the second harmonic and therefore an ordinary Fourier transform was used to determine the amplitude of the fundamental. The fraction A_Γ/A_α was found to be 0.63 which gives the absolute amplitude of the M_Γ signal, $A_\Gamma = -45$ A/m.

V. DISCUSSION

The experimental results were compared with the LK theory using Eq. (1). It should be pointed out that even though this formula is theoretically deduced, some of the quantities used are experimentally obtained which introduces an uncertainty in the values. Extremal areas and effective masses were taken from Ref. 13. The Dingle temperatures were determined during the experiment through standard Dingle plots and the values of the spin-splitting factor were taken from Ref. 14. To estimate the geometrical factor we have used the identity

$$\frac{\partial^2 S}{\partial k_z^2} = 2\pi \left[\frac{1}{S_0} \frac{\partial^2 S}{\partial \theta^2} - 1 \right] \tag{13}$$

for the α orbit. From a spherical harmonic expansion of the area,[15] an explicit expression for $\partial^2 S/\partial\theta^2$ was deduced. This gave $|\partial^2 S/\partial k_z^2|$ equal to 1.0. Since $(1/S_0)\partial^2 S/\partial\theta^2$ is close to 1, a small change in $\partial^2 S/\partial\theta^2$ results in a larger relative change of $\partial^2 S/\partial k_z^2$, which indicates an uncertainty in this value. For the Γ_6 sheet, a numerical interpolation scheme with Fermi radii[15] as input was used giving $|\partial^2 S/\partial k_z^2| = 5.2$. The calculated amplitudes for the Γ_6 and α orbit are presented in Table I. The uncertainty in these values is estimated to be 15% for the Γ_6 orbit and 20% for the α orbit. This higher value for the α orbit is mainly due to the uncertainty in the curvature factor. It should be observed that there exist 24 W points in each Brillouin zone and that for a [100] direction the contribution to one zone by the activated α orbits summed together are two orbits. Thus, the calculated value for the absolute amplitude of the α orbit, using Eq. (1b), has to be doubled.

The error limits for the measured amplitudes only depend on a few parameters. The last term in Eq. (8) will dominate the total signal at fields close to B_+ and B_- since the rest of the terms in Eq. (8) summed together are close to zero in the vicinity of these fields and will thus introduce an error in the graphic interpolation used to determine B_+ and B_-. The magnitude of this "second harmonic" will depend on the order of the Bessel function in the experiment but this error does not exceed 1%. Crabtree[11] gives the demagnetization factor N as a function of the ratio between the length and diameter of the sample and the angle between the cylindrical axes of the sample and the direction of the magnetic field. An error of, for example, 10% in the estimate of the ratio for the sample used in this experiment will give a change in N of only 3%. The uncertainty in the angle between the field and the cylindrical axis of the sample comes from how well the sample and the sample holder are aligned. A reasonable estimate is that this is done within a few degrees which gives an error of less than 2%. From this we estimate the error in N to less than 4%. The error in the dHvA frequency used is not greater than 0.2% and hence the error in the measured absolute amplitude of the α orbit is less than 6%. For the Γ_6 sheet there is an additional error arising from the uncertainty in the Bessel argument adjustment. This is especially important at low arguments where $J_4(X) \propto X^4$. Therefore the uncertainty for the A_Γ value can be 10%.

We find that the experimental and the calculated values for each orbit tabulated in Table I agree well within the error limits. The advantages of the method are its simplicity and its need of only the few parameters as described by Eqs. (10) and (11). One application for this method of measuring absolute amplitudes will be to find values for the spin-splitting factors. The ratio between the measured absolute amplitude and the LK amplitude with $|\cos(\pi g_c m_c/2)| = 1$ will give the value of the spin-splitting factor. An extension of this type of measurement would be studies of diluted materials. A small amount of magnetic impurity will change g_c and hence also the value of the spin-splitting factor while the extremal areas and the effective masses remain unaffected. With knowledge of the Dingle temperature the change in the absolute amplitude will give the change in g_c due to a specific concentration of impurity.

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*Present address: ASEA Research, S-721 83 Västerås, Sweden.

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