Conduction-electron Zeeman splitting on the X-pocket holes in platinum

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The conduction-electron Zeeman splitting on the X-pocket holes in platinum has been investigated, with the use of the de Haas–van Alphen technique. The amplitude for the fundamental and second harmonic has been studied in the (100) and (110) planes. Two spin-splitting-zero contours on the second harmonic have been mapped out. A complete picture of the variation of the g_c factor over the X pocket has been achieved, and its absolute value was found to range from 0.5 to 3.5.

I. INTRODUCTION

Measurements of the conduction-electron cyclotron-orbit g factor (g_c) have recently been performed on the fcc platinum-group metals, rhodium,[1] palladium,[2] iridium,[3] and platinum.[4] These studies have shown that the g_c factor exhibits strong anisotropic behavior on the Γ-centered Fermi-surface sheet ($\Gamma 6$), which these transition elements have in common. In contrast to this anisotropy stands the very isotropic behavior of the g_c factor on the α orbit centered around the W point on the open hole sheet in palladium and platinum. Another part of the Fermi surface which these four metals have in common is the X-centered hole pockets ($X4$). A complete mapping out of the variation of the g_c factor has so far only been performed for the X pockets in palladium [5] where a large anisotropy is reported. A natural continuation of investigations of spin splitting in platinum-group metals is therefore to study g_c factors on the X pockets and to make comparisons between the different metals.

An excellent probe for these studies is the de Haas–van Alphen (dHvA) effect. In the theoretical expression for the amplitude of the dHvA signal, derived by Lifshitz and Kosevich (LK),[6] there is a cosine function, also called the spin-splitting factor, with the argument

$$k\pi R = k\pi E_Z / E_L = k\pi g_c m_c / 2 \ , \tag{1}$$

where k is the harmonic number of the signal, E_Z is the cyclotron-averaged Zeeman splitting, E_L is the Landau-level spacing energy, and m_c is the cyclotron effective mass expressed in free-electron mass units. Whenever $kR = n + \frac{1}{2}$ (where n is an integer) the spin-splitting factor is equal to zero and the dHvA signal for this harmonic will vanish. This absence of amplitude is called a spin-splitting zero (SSZ) and can be found as contours of constant kR over a Fermi-surface sheet. A first attempt to use SSZ contours to determine g_c factors can be found in Ref. 7. For different neighboring SSZ contours on a Fermi-surface sheet the values of n can be identical or differ by ± 1. This uncertainty can be resolved by studying the change of the locations of the SSZ's in a sample diluted with a small amount of magnetic impurities. This will also give the slope of R at the SSZ's. The mapping out of SSZ contours is a straightforward and accurate technique but needs to be used together with complementary methods, such as the absolute amplitude or the harmonic ratio method, to achieve a full picture of the g_c factor. For an early account of these techniques see, for example, Randles[8] and Crabtree *et al.*[9] In this work the SSZ contours and the amplitude have been studied. The g_c factor appears in the cosine function and therefore there is an ambiguity in its absolute value, but the dHvA effect offers a unique possibility to measure its anisotropy. For further details, see Ref. 5.

For the transition elements, with a considerable exchange enhancement, a deviation of the g factor from 2 is expected. An average bulk g factor value for platinum deduced from susceptibility measurements is 4.2.[4] The g_c factor is also enlarged by a cyclotron-orbit Stoner enhancement which is unique for each orbit on a specific sheet of the Fermi surface.[10] As the density of states for the X pockets in platinum contributes less than 1% to the total density of states,[11] its g_c factors may deviate largely from the bulk value.

Although the topology of the Fermi surface of the X pockets in platinum (ellipsoids of revolution) and their effective masses are known from the work by Ketterson and Windmiller[12] (KW) and Dye *et al.*,[13] very little has been reported of their g_c factors. Cavalloni *et al.* have studied magnetostriction and dHvA oscillations in palladium and platinum,[14] and their measurements on the X pockets reveal SSZ's on the second harmonic in platinum at approximately 53° and 78° and a maximum on the fundamental at approximately 65° from [001] in the (010) plane (for the X pocket centered at $00\frac{1}{2}$). In this work we present measurements of the dHvA amplitude over the X pockets in platinum and together with measurements of the amplitude in a platinum sample diluted with a small amount of iron (100 ppm) this leads to a complete picture of the variation of the g_c factor. A determination of its absolute value is also presented.

II. EXPERIMENTAL AND THEORETICAL CONSIDERATIONS

The measurements were performed in a cryogenic system at a temperature of 0.55 K and a maximum magnetic field of 7 T. The preparation of the samples is described in Ref. 4. The sample can be rotated 180° with a resolution of 0.1° around an axis perpendicular to the magnetic field. As an additional feature the magnet can be tilted ±3.5°, which makes it possible to adjust the direction of the magnetic field relative to the symmetry planes with an accuracy of 0.2°. The magnetic field was modulated with a frequency of 210 Hz and the second harmonic was used for detection on the X pocket and the eighth harmonic for the $\Gamma 6$ sheet. The dHvA signals from the $\Gamma 6$ sheet were used to align the magnetic field with the symmetry planes. A data-acquisition system and a Fourier transform were used to obtain the amplitudes of the X-pocket signals. At each direction of the magnetic field 250 data points were taken during 10–15 oscillations.

The symmetry of the X-centered hole pockets is tetragonal and the irreducible part is $\frac{1}{16}$ of the total surface (see Fig. 1). In each Brillouin zone there are three X pockets in the different $\langle 100 \rangle$ directions, which means that when measuring in a (110) plane one will only detect two different frequencies, while for a (100) plane there will be three different frequencies. With the magnetic field in a [100] or a [110] direction there exist two frequencies, and in a [111] direction only one. With the direction of the magnetic field in the vicinity of these directions the analysis routine cannot resolve the frequencies lying close together, thus giving regions where the spin-splitting factor could not be determined.

The experimental amplitudes at each field direction were corrected with respect to the direction of the magnetization relative to the pickup coils and the Bessel-function values for the present frequencies. In order to optimize the signal the first Bessel-function maximum and the largest accessible modulation field were used, giving an available field range of 2.5–3.0 T.

Comparison of the experimental amplitudes with the LK amplitudes, calculated with the cosine factor omitted, gives values proportional to the cosine factor. At a SSZ for the second harmonic the cosine factor for the fundamental has the value $1/\sqrt{2}$, giving the possibility of calibrating the amplitude for the fundamental. The values of the dHvA frequency and effective cyclotron electron mass used in the calculations were taken from KW. The experimental results of KW reveal that the ϕ dependence of the frequency can be neglected and it is assumed that this is the case also for the effective mass. Recent linear muffin-tin orbital (LMTO) calculations[15] show that the variation of the frequency and the cyclotron mass as a function of ϕ is less than 1%. Dingle temperatures were measured both for the pure platinum sample and for the diluted sample. In the platinum sample the value 0.2 K was found, and in platinum-iron 0.4 K. The Dingle temperature was considered isotropic over the X-pocket surface, as this was found to be the case for palladium.[5] The curvature factor was calculated from the geometry of an ellipsoid of revolution.

In the determination of the cosine factor, errors are introduced both from the measurements of the amplitudes and in the calculation of the LK expression. The error in the cosine factor is less than 0.1. When R is calculated an inverse cosine function is used, thus giving different errors for different parts of the curve, the upper limit varying from ±0.04 to ±0.15. The maximum error in a single g_c value ranges from ±0.2 to ±0.8.

III. RESULTS

The amplitude measurements in the (010) and $(1\bar{1}0)$ planes resulted in a variation similar to that presented by Cavalloni *et al.*[14] The fundamental exhibits minima at [001] and around the waist of the ellipsoid and maxima at $\theta = 64°$. The second harmonic has maxima at [001], around the waist, and at $\theta = 64°$, and SSZ's at $\theta = 50°$ and 80° in the (010) plane and at $\theta = 48°$ and 77° in the $(1\bar{1}0)$ plane (see Fig. 1). The two spin-splitting-zero contours were also observed in the nonsymmetry (110) planes, but with less accuracy (see Ref. 5). This restricts R to $n - \frac{1}{2} < R < n + \frac{1}{2}$ and gives four different possibilities for its variation. To resolve this ambiguity the amplitudes were measured for a platinum sample diluted with a small amount of iron. The magnetic impurities increase the Zeeman splitting, leading to a shift of R towards higher values. By comparing locations of SSZ's and relative amplitudes it may then be possible to resolve the ambiguity (see, e.g., Ref. 2). Since the amplitudes were

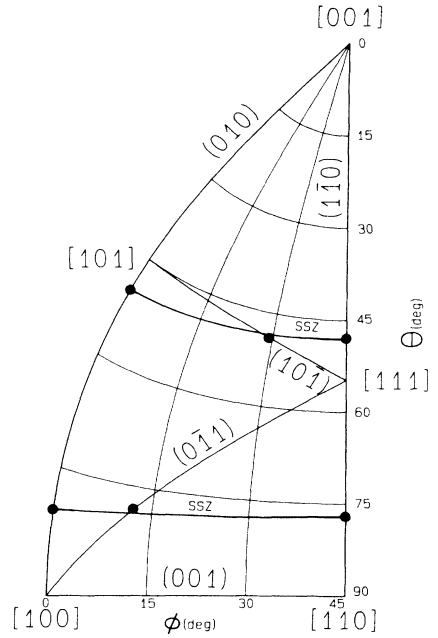

FIG. 1. The basic irreducible $\frac{1}{16}$ wedge for the X pocket centered at $00\frac{1}{2}$, with the two spin-splitting-zero contours for the second harmonic (solid lines) displayed. The solid circles indicate the detected SSZ's. The SSZ's in the nonsymmetry planes are measured with less accuracy.

rather low any SSZ's for the second harmonic were difficult to observe. However, the amplitude for the fundamental at [001] in the diluted platinum sample was found to be relatively larger and a new developed SSZ was found 33° out from [001] as can be seen in Fig. 2. This new SSZ and the shift in R for the diluted sample support only one of the four possibilities mentioned above and we are left with the interpretation of R displayed in Fig. 3. The measured amplitudes have been adjusted according to the procedure presented in Sec. II and the values of the cosine factor have been transformed into values of R both for the fundamental and the second harmonic in the symmetry planes.

Two separate measurements in the [001] and [101] directions were performed to establish the absolute phase at infinite magnetic field for the X-pocket dHvA oscillations. Owing to the presence of other signals special precautions were taken to make it possible to follow a single frequency over a considerable field interval. The results for the phases in the [001] and [101] directions were 0.23 and 0.17 in units of 2π, respectively, when the oscillations are represented by cosine functions. In this case the LK theory predicts the values $\frac{3}{8}$ and $\frac{7}{8}$ depending on whether the cosine factor is positive or negative and our results show that the cosine factor is positive (see Ref. 5). This implies that the integer number in the presented curves of R may take only even values. In the analysis of these measurements the following values of the two frequencies were found: in the [001] direction, 112.0 T, and in the [101] direction, 133.5 T, to be compared with 111.5 and 133.7 T from KW.

It is interesting to note that the results from the diluted sample show that the change in R caused by the impurities, $\Delta R(\theta,\phi)$, is also anisotropic. ΔR is large around [001] and small close to the waist of the pocket. This is in line with the results found for palladium doped with nickel[16] and may be an indication of where exchange interaction effects are large.

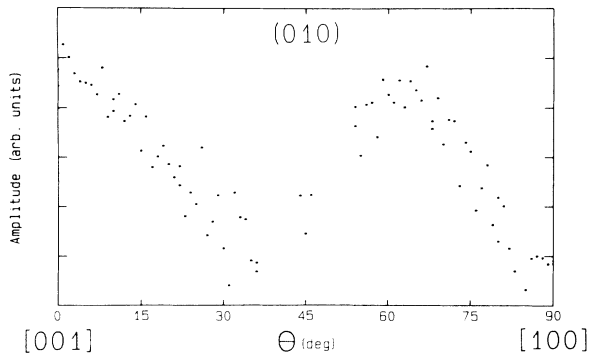

FIG. 2. The de Haas–van Alphen amplitude for the fundamental harmonic in platinum diluted with iron as a function of magnetic field direction, with the magnetic field in the (010) plane. A spin-splitting zero is located at $\theta=33°$, to be compared with pure Pt which has no spin-splitting zeros for the fundamental.

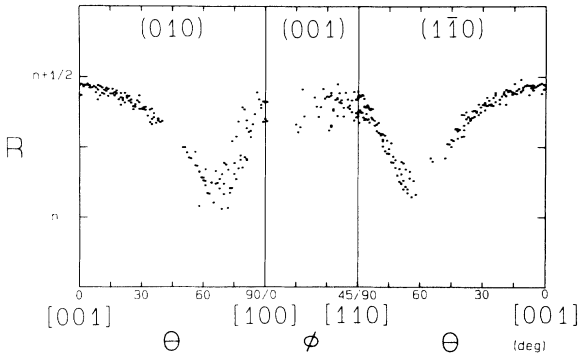

FIG. 3. $R(\theta,\phi)$ in the symmetry planes on the X pocket, calculated by comparing the experimental amplitudes with the calculated LK amplitudes.

IV. DISCUSSION AND CONCLUSIONS

The possible alternatives for the g_c factor for the X pockets in platinum from our measurements are presented in Fig. 4. They have been achieved by making use of the cyclotron effective masses reported by KW. They measured the effective mass in the [100] direction and anticipated that the mass should vary as the frequency over the pocket. This model for the masses has been used although a comparison with calculated masses gives a slight deviation from this model. From calculated masses it is found that the KW value of m_c at [001] is about 6% lower than the calculated. Such a deviation is not large enough to cause any considerable change in the presented curves for the g_c factor. It is important also to note that it is the measured effective mass that has been used, which then includes the electron-phonon enhancement effect. The presented g_c-factor values thus differ from what would have been achieved by using calculated bare band masses. This has been discussed in more detail by Gustafsson *et al.*[4]

Normally the absolute value of the g_c factor is not directly obtainable from dHvA measurements. Howev-

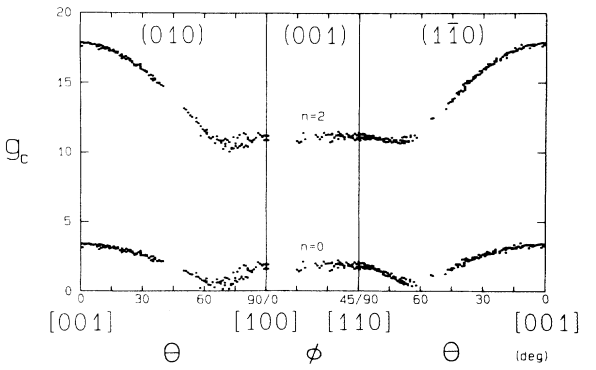

FIG. 4. The values of $g_c(\theta,\phi)$ in the symmetry planes on the X pocket calculated from $R(\theta,\phi)$ with $n=0$ and $n=2$, using the effective electron masses from KW.

er, the measured value of the phase at infinite magnetic field strength which excluded the odd values of the integer n, together with the small effective mass, make this possible in this case. A comparison of the two plausible remaining alternatives for g_c with the bulk value 4.2 favors the choice $n=0$, though one should be aware of the uncertainty due to the small contribution to the density of states as discussed in the introduction. The g_c factor then ranges from a value of less than 0.5 to about 3.5.

Compared to palladium the X pocket in platinum has much smaller effective masses, which means that the difference between the possible alternatives (different integer n) is smaller in palladium than in platinum. The variations found for g_c in the two metals are very similar in their major features though they differ in absolute magnitude. Such a similarity has earlier been shown also for the Γ sheet and the α orbit. For the X pockets, the maximum value is at [001], a minimum at 65° from [001], and an intermediate value at [100] and [110]. They both show deviations from rotational symmetry around the waist (approximately 10% for platinum).

Band-structure calculations of the g_c factors in platinum-group metals have recently been performed by using the LMTO method with a perturbation to the Hamiltonian from spin-orbit coupling and an applied magnetic field.[15] For the X pockets in platinum, values of the g_c factors at [001], [100], and [110] were 0.07, 0.54, and 0.48, respectively. Around the waist the calculated g values show a slight deviation from rotational symmetry by about 15% while the calculated mass and area are constant. The calculated and experimental g_c values show the same degree of anisotropy. The experimental g_c factors averaged over the X pocket are less than the total bulk value. This is also the case when comparing the calculated g values for the X pockets with an averaged calculated g value for platinum (taken from Ref. 17). The detailed variation found for the experimental g factor is not explained by the calculated values. This may indicate that the model used in the band calculations cannot quantitatively account for the anisotropy caused by spin-orbit interaction. It is, however, also possible that the exchange interaction, which is not included in the model, plays an important role for the anisotropy of g_c and not only for its absolute value. The latter has so far not been investigated in detail. Jarlborg and Freeman presented calculations for palladium where exchange interaction is included and their results for the X pockets are enhanced but do not reveal any anisotropy.[18]

The measurements reported here and in Ref. 5 clearly demonstrate the large anisotropy associated with the X pockets in palladium and platinum and that they are similar regarding the variation of the g_c factor. It would be most interesting to investigate the double X pockets in iridium and rhodium, both for the comparison between the two pockets in each metal and for a comparison between all four metals which have very similar band structures. It is also of great interest to improve the band-structure calculation models to see whether it is possible to achieve a better understanding of the X-pocket g-factor behavior.

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