VI. CONCLUSION

Although the interpretation of the characteristics of the helicon propagation is far more complicated than the interpretation of direct resistance and Hall effect measurements by conventional techniques, the helicon technique is more suitable for low magnetoresistance studies. As the results that we have discussed show, useful information about the topology of the Fermi surface can be obtained from the oscillations arising from magnetic breakdown.

A more detailed study of the quantum oscillations should enable further quantitative checks to be made, not only of the band-structure calculations but also of the theory of magnetic breakdown.

ACKNOWLEDGMENTS

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Splitting of Conduction-Electron Spin Resonance in Potassium

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The 0.5-G splitting of the conduction-electron spin resonance in potassium (at 4200 G) observed by Walsh, Rupp, and Schmidt can be quantitatively explained providing the conduction electrons are in a charge-density-wave (CDW) ground state. The Fermi-surface distortion caused by the CDW energy gap leads to an anisotropic conduction-electron g factor depending on the angle between **H** and the wave vector **Q** of the CDW. The extremal values of g, corresponding to $\mathbf{Q}\perp\mathbf{H}$ and $\mathbf{Q}\parallel\mathbf{H}$, differ by $(3V/8E_F)\Delta g$, where $\Delta g=-0.0025$ is the observed g shift. V is the observed threshold energy of the Mayer–El Naby optical-absorption anomaly, and E_F is the Fermi energy. The predicted maximum splitting is 0.56 G. Interpretation of the data requires the sample to have a macroscopic domain structure, caused by thermal stress and plastic flow when the potassium-Parafilm sandwich is cooled to He temperature. The orientation of **Q** in stress-free regions should be parallel to **H**. In regions of high stress, **Q** is presumed perpendicular to the surface, and therefore approximately perpendicular to **H**.

I. INTRODUCTION

Two years ago, Walsh, Rupp, and Schmidt[1] observed conduction-electron spin resonance (CESR) in extremely pure potassium. Linewidths as narrow as 0.13 G were obtained at a resonance field of 4200 G and a temperature of 1.3°K. The observed g shift, caused by spin-orbit coupling, was $\Delta g=-0.025(4)$, and represents an experimental shift of 5.3 G. A very puzzling feature of their result was that the CESR signal split into two well-resolved components as the magnetic field was tilted away from an initial orientation parallel to the surface of the potassium. The splitting reached a maximum value of about 0.5 G at a tilt angle of 9°. At larger tilt angles the signal intensity diminished, and one component vanished rapidly. The splitting has been observed in a number of specimens[2] having sufficient purity to yield comparably narrow lines.

The possibility that the splitting is caused by an artifact arising from excitation of both faces of a thin sample has been ruled out by Lampe and Platzman,[3] who computed the surface impedance spectra for a variety of configurations. They found line-shape variations but no splitting. The possibility that it is caused by new collective modes (paramagnetic spin waves), resulting from conduction-electron exchange interactions, was ruled out by Platzman and Wolff.[4] They indeed found that such modes exist theoretically and provide a quantitative explanation of the sidebands reported by Schultz and Dunifer.[5] The 0.5-G splitting, however, remained a mystery.[4]

An important characteristic of CESR is that individual electrons undergo several thousand quantum transitions among levels at the Fermi energy during a spin relaxation time T_2. Consequently the resonance field is determined by the average g factor (the average over all levels that contribute to the paramagnetism). Since an averaging process necessarily results in a unique number, a single resonance must always be

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[1] W. M. Walsh, Jr., L. W. Rupp, Jr., and P. H. Schmidt, Phys. Rev. **142**, 414 (1966).

[2] W. M. Walsh, Jr. (private communication).

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[5] Sheldon Schultz and Gerald Dunifer, Phys. Rev. Letters **18**, 283 (1967).

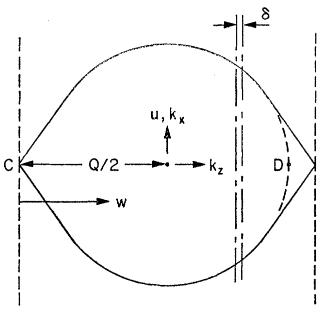

FIG. 1. The lemon-shaped Fermi surface of a CDW ground state. The vertical dashed lines through the conical points, e.g., point *C*, are the planes in *k* space where the energy gaps caused by the CDW potential occur. The conical-point distortion is exaggerated by about a factor of 3.

anticipated. This is strikingly confirmed by resonance experiments on dilute paramagnetic alloys.[6] Paramagnetic Mn dissolved in Cu or Ag produces a single line with *g* factor intermediate between that of the Mn ion and the host metal. Spin exchange between conduction-band levels and localized paramagnetic levels is sufficiently rapid to merge the contributions into a single, sharp resonance line.

We conclude therefore that a split CESR indicates that the metal sample is heterogeneous. That is, we suggest that a K sheet (~0.02 cm thick) sandwiched between layers of Parafilm divides into contiguous regions having either of two possible *g* factors. This hypothesis requires that there are two distinguishable variants of K, and that both occur in samples where CESR splitting was observed. Had this occurred in Na, an immediate explanation would have been possible. Below 35°K, Na partially transforms martensitically[7] from bcc to hcp structure. Conceivably the two structures would have slightly different *g* factors. However, there is no similar transformation in K. It must also be appreciated that the splitting, though only 0.5 G, is relatively large, ~10% of the spin-orbit shift, which is the true measure of such an effect.

The purpose of this paper is to show that the foregoing hypothesis is a natural one provided the electronic ground state of the metal is a spin-density-wave (SDW) or charge-density-wave (CDW) state. For alkali metals a CDW state is preferred for theoretical[8] and experimental reasons. In this case, as with other alkali-metal anomalies that have been explained quantitatively[9] by assuming the ground state has an exchange instability wave, the interpretation is the same for either a CDW or SDW. The theoretical characteristic of either state is that each electron experiences a self-consistent periodic potential,

$$U(\mathbf{r})=V\cos\mathbf{Q}\cdot\mathbf{r}. \tag{1}$$

The wave vector **Q** is unrelated to the lattice periodicity, and for a simple metal, has a length just right for critical contact between the Fermi surface and the two energy gaps of magnitude *V*. This distorts the Fermi surface into a lemon shape as shown in Fig. 1.

An immediate consequence of the distorted Fermi surface and modified electron wave functions, arising from (1), is that the average *g* factor depends on the angle between **Q** and the magnetic field **H**. The principal axes of the *g*-factor tensor will of course correspond to $\mathbf{Q}\|\mathbf{H}$ and $\mathbf{Q}\perp\mathbf{H}$, and will be associated with the extremal values of *g*. In Sec. II we show that

$$g(\mathbf{Q}\perp\mathbf{H})-g(\mathbf{Q}\|\mathbf{H})\cong(3V/8E_F)\Delta g, \tag{2}$$

where E_F is the Fermi energy. Derivation of this result does not require an accurate theory of the conduction-electron *g* factor. All one needs is the simple result, valid for alkali metals,[10] that the *g* shift of a nearly-free-electron Bloch state **k** is

$$\Delta g(\mathbf{k})\cong-\mu k^2\sin^2\theta, \tag{3}$$

where θ is the angle between **k** and **H**. For a nearly spherical Fermi surface,

$$\Delta g=\langle\Delta g(\mathbf{k})\rangle_{\mathrm{av}}=-\tfrac{2}{3}\mu k_F{}^2. \tag{4}$$

We need not compute the coefficient μ since the experimental Δg can be used in Eq. (2).

The CDW energy gap *V* in K is 0.6 eV, the threshold energy of the Mayer–El Naby optical-absorption anomaly.[11] $E_F=2.12$ eV. Consequently Eq. (2) predicts that the maximum splitting is 10.6% of the observed Δg, which implies a maximum observed splitting of 0.56 G.

In Sec. III we indicate why a thin K sheet subjected to severe thermal stress on cooling would likely divide into contiguous patches having **Q** either parallel to **H** or perpendicular to the surface (and therefore perpendicular to **H**). The remarkable quantitative agreement between the predicted maximum splitting and that observed provides significant additional evidence for a CDW state in K.

II. ANISOTROPY OF *g*

In order to compute the conduction-electron *g* factor, starting from Eq. (3), we must know the eigenfunctions and energy spectrum $E(\mathbf{k})$ of the Schrödinger equation having the potential-energy term (1). Compact solutions of this—the Mathieu equation—cannot be written down, so we make use of the following artifice which has sufficient accuracy. The perturbation (1) can be divided into two parts, one which leads to the energy gap for $k_z=-\frac{1}{2}Q$, the gap on the left in Fig. 1, and the other

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which leads to the gap for $k_z=\frac{1}{2}Q$. Each part must be treated with great accuracy because wave functions and k-space occupation are significantly altered near the gaps. The simplifying feature of the problem is that the physical consequences of the two parts are additive. Accordingly, we shall compute the anisotropy of g arising from that part of the perturbation associated with the left-hand gap, and multiply the result by 2.

The part of (1) which we shall treat causes the unperturbed state $\mathbf{k}$ to be mixed with $\mathbf{k}+\mathbf{Q}$ only. That is, we solve the secular equation,

$$\begin{pmatrix} (p^2/2m)-E & \frac{1}{2}V\exp(-i\mathbf{Q}\cdot\mathbf{r}) \\ \frac{1}{2}V\exp(i\mathbf{Q}\cdot\mathbf{r}) & (p^2/2m)-E \end{pmatrix}=0, \qquad (5)$$

within the space defined by the basis states $\mathbf{k}$ and $\mathbf{k}+\mathbf{Q}$. The energy for the solution below the gap is

$$E_{\mathbf{k}}=\tfrac{1}{2}(\epsilon_{\mathbf{k}}+\epsilon_{\mathbf{k}+\mathbf{Q}})-\tfrac{1}{2}[(\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}+\mathbf{Q}})^2+V^2]^{1/2}, \qquad (6)$$

where $\epsilon_{\mathbf{k}}=\hbar^2k^2/2m$, the unperturbed energy. The corresponding eigenfunction is

$$\psi_{\mathbf{k}}=\cos\varphi\exp(i\mathbf{k}\cdot\mathbf{r})-\sin\varphi\exp[i(\mathbf{k}+\mathbf{Q})\cdot\mathbf{r}], \qquad (7)$$

where

$$\cos\varphi(\mathbf{k})\equiv V/[V^2+4(\epsilon_{\mathbf{k}}-E_{\mathbf{k}})^2]^{1/2}. \qquad (8)$$

It is of interest that Eq. (6) is the exact solution of the spiral SDW problem.[12] [For that case, the two terms of Eq. (7) would have opposite spin states.]

The equation describing the Fermi surface is obtained by setting

$$E_{\mathbf{k}}=E_F=[\hbar^2(\tfrac{1}{2}Q)^2/2m]-\tfrac{1}{2}V. \qquad (9)$$

The last equality is imposed because E_F must be the energy of the point of critical contact,[8] point C of Fig. 1. We choose $\mathbf{Q}$ in the z direction and introduce dimensionless variables:

$$u\equiv k_x/Q,\quad v\equiv k_y/Q,\quad w\equiv(k_z+\tfrac{1}{2}Q)/Q, \qquad (10)$$

so point C is (0,0,0). The equation of the Fermi surface in the (u,w) plane is

$$u=[(w^2+\alpha^2)^{1/2}-\alpha-w^2]^{1/2}, \qquad (11)$$

where $\alpha\equiv mV/\hbar^2Q^2$. Note that $u=0$ at $w=0$, which verifies our choice for E_F in (9). This surface has a peculiar shape. Near point C it is a circular cone. But near point D of Fig. 1, the other intersection with the w axis, it is nearly spherical, as indicated by the dashed curve. The coordinate of point D is obtained by setting (11) to zero.

$$w(D)\equiv\gamma=(1-2\alpha)^{1/2}. \qquad (12)$$

The radius r_0Q of the Fermi surface at $w=\frac{1}{2}$, the origin in k space, can be written as a power series in α. To first order, Eq. (11) is

$$r_0\cong\tfrac{1}{2}(1-2\alpha). \qquad (13)$$

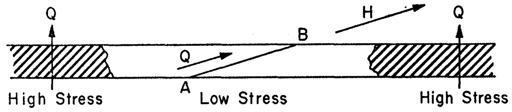

FIG. 2. Schematic conception of the stress-induced Q domains in a thin sheet of K, sandwiched between layers of Parafilm. For large $\omega_c\tau$ spatial diffusion of electrons is very directional, and is parallel to H. For example, an electron originally at point A. will, upon diffusion to the other side, arrive near point B. Subsequent diffusion back will again be along the line from B to A.

This radius must equal the free-electron-sphere radius to order α,[13] since the effect of the perturbation on states near the belly of the lemon is of order α^2. Consequently,

$$Q/k_F\cong 1/r_0\cong 2(1+2\alpha). \qquad (14)$$

Together with the definition of α given above, this implies

$$Q\cong 2k_F[1+(V/4E_F)]. \qquad (15)$$

This result then provides an expression for α depending only on V/E_F:

$$\alpha\cong(V/8E_F)/[1+(V/4E_F)]^2. \qquad (16)$$

The geometry of the semideformed Fermi surface has now been completely specified.

The g shift for the case $\mathbf{Q}\|\mathbf{H}$ can now be computed from Eq. (3):

$$\Delta g(\mathbf{Q}\|\mathbf{H})=-\mu\langle k_x^2+k_y^2\rangle_{\text{av}}. \qquad (17)$$

The density-of-states average over the Fermi surface is the one required. For the case $\mathbf{Q}\perp\mathbf{H}$, with $\mathbf{H}$ in the y direction,

$$\Delta g(\mathbf{Q}\perp\mathbf{H})=-\mu\langle k_x^2+k_z^2\cos^2\varphi+(k_z+Q)^2\sin^2\varphi\rangle_{\text{av}}. \qquad (18)$$

The reason why (18) is more complex than (17) is that the wave vector $\mathbf{k}$ in Eq. (3) is not the wave-vector label of the state, but the actual eigenfunction component wave vector. Since (7) has two components, $\mathbf{k}$ and $\mathbf{k}+\mathbf{Q}$, they both must be included in proportion to their probabilities. There is no contribution to (18) from the interference term of the two components. The reason for this depends on the fact that $\mathbf{Q}$ is incommensurate with the reciprocal lattice. For the alkali metals the conduction-electron g shift can be considered to originate from the corelike p functions admixed into the (otherwise) pure plane waves by the process of core orthogonalization. If one were to examine the g-shift contribution attributable to a particular atomic cell of the metal, there would be an interference term dependent on the relative phase of the $\mathbf{k}$ and $\mathbf{k}+\mathbf{Q}$ components at that cell. However, the crystal sum of the interference term vanishes because $\mathbf{Q}$ is not a simple rational fraction of a reciprocal-lattice vector.

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[13] The reader should not concede this point, however, without verifying that the volume enclosed by the surface, Eq. (11), differs from that of a sphere of radius r_0 only by terms of order $\geq\alpha^2$.

The axial symmetry of the Fermi surface about the z axis makes the density-of-states average a very simple operation. Consider a slice of k space perpendicular to the z axis and having thickness δ, as shown in Fig. 1. The number of quantum states of energy $\leq E$ within the slice is

$$Z=\pi t^2\delta/8\pi^3=(m\delta/4\pi^2\hbar^2)[E-E_0(K)]. \qquad (19)$$

Here t is the radius of the energy surface E at $k_z=K$, and $E_0(K)$ is the z component of energy, Eq. (6) evaluated at $(0,0,K)$. The density of states N in the slice is

$$N=(\partial Z/\partial E)_K=m\delta/4\pi^2\hbar^2. \qquad (20)$$

Since this is constant, the density-of-states average of an axially symmetric function F is merely

$$\langle F\rangle_{\text{av}}=\gamma^{-1}\int_0^\gamma F(w)dw, \qquad (21)$$

where $F(w)$ is the value of F at $k_z=(w-\frac{1}{2})Q$ on the Fermi surface.

With Eqs. (10), (17), (18), and (21) in view we define two subsidiary functions:

$$A(\alpha)\equiv\int_0^\gamma u^2dw,$$

$$B(\alpha)\equiv\int_0^\gamma [(w-\tfrac{1}{2})^2\cos^2\varphi+(w+\tfrac{1}{2})^2\sin^2\varphi]dw. \qquad (22)$$

Furthermore, we define the fractional g-factor anisotropy f of the CDW state:

$$f(\alpha)\equiv[g(\mathbf{Q}\perp\mathbf{H})-g(\mathbf{Q}\|\mathbf{H})]/\Delta g, \qquad (23)$$

where Δg is the g shift for $\mathbf{Q}\|\mathbf{H}$. Inspection of (17), (18), and (22) allows us to conclude

$$f(\alpha)=2\{[\tfrac{1}{2}A(\alpha)+B(\alpha)]-A(\alpha)\}/A(\alpha). \qquad (24)$$

The factor 2 accounts for the anisotropy introduced by the other energy gap, at $k_z=\frac{1}{2}Q$, as anticipated in the first paragraph of this section. The factor $\frac{1}{2}$ inside the square brackets follows from the obvious relation $\langle k_x^2\rangle_{\text{av}}=\frac{1}{2}\langle k_x^2+k_y^2\rangle_{\text{av}}$. The integral $A(\alpha)$ is easily performed after using Eq. (11). The integral $B(\alpha)$ is likewise elementary after the functional dependence of $\cos^2\varphi$ on w is found. From (6), (8), and (10),

$$\cos^2\varphi=\tfrac{1}{2}[1+w(w^2+\alpha^2)^{-1/2}]. \qquad (25)$$

The final result, with the help of (12), is to terms linear in α

$$f(\alpha)\cong 3\alpha. \qquad (26)$$

Use of Eq. (16) to order V/E_F completes the derivation of Eq. (2), $f\cong 3V/8E_F$.

The question remains as to what fraction of f is caused by repopulation of k space and what fraction is caused by admixture of $\mathbf{k}\pm\mathbf{Q}$ components into $\psi_{\mathbf{k}}$. This is easily answered by setting $\varphi\equiv 0$ in (22) and repeating the balance of the calculation. One obtains $3V/8E_F$ again. Thus, to lowest order, the entire g-shift anisotropy is caused by repopulation of k space. The $\mathbf{k}\pm\mathbf{Q}$ admixture contribution is of order $(V/E_F)^2$.

The tedious analysis of this section was of course necessary. However, with the understanding that inevitably is acquired one can surmise the final result in a single glance: The fractional increase in $\int k_z^2dk_z$ is just three times the conical-point distortion, $p=V/4E_F$. The factor 3 comes from the binomial coefficient of $(1+p)^3$, associated with the upper limit of integration in k_z. This increase is diluted by 50% due to the invariance of $\int k_x^2dk_z$. Consequently $f=\frac{3}{2}p$.

III. STRESS-INDUCED Q DOMAINS

In Sec. I we concluded that the only possible explanation of CESR splitting in K was a macroscopic division of the sample into two species having different g factors. Our purpose here is to elaborate the required physical characteristics of such a domain structure and to show how its origin is easily understood on the basis of a CDW ground state. This will not require any novel or new subsidiary assumptions.

K has a very large thermal-expansion coefficient. Its lattice constant[7] at 5, 78, and 293°K is 5.225, 5.247, and 5.344 Å, respectively. Consequently, the linear expansion between 5° and 78°K alone is 0.4%. The elastic moduli are known.[14] It follows that a negative pressure of 4.5 kg/mm² would be required to prevent the contraction below 78°K. The polycrystalline yield stress has been measured[15] throughout this temperature range and is only 0.1 kg/mm². Consequently, a sheet of K cooled to He temperatures between layers of another material to which it adheres will undergo severe thermal stress and plastic deformation. It is reasonable to assume that the stress state of such a sample will be macroscopically heterogeneous. Some regions where a great deal of slip has occurred will be relatively stress free. Others will be in planar tension.

We believe that in relatively stress-free regions **Q** will tend to align parallel to **H**. This is not a new assumption, since it is also required[16] for the explanation of other anomalous phenomena. However, we also assume that severe stress prevents alignment parallel to **H**. The striking stress dependence[17] of the high-field magnetoresistance establishes elastic stress as a crucial variable affecting electronic properties. The directional effect of stress on orientation of **Q** can possibly be surmised from the conflicting data on the optical anomaly. Hodgson[18] failed to observe it. An

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important difference between his experiment and that of Mayer and El Naby[11] is that the former was carried out by reflection from an evaporated metal layer on a glass interface, whereas the latter was by reflection from a bulk-metal vacuum interface. The theory[9] of the anomaly based on a CDW or SDW model predicts that it should be observed only if **Q** has a component parallel to the reflecting surface. Hodgson's failure to see the anomaly can therefore be explained if **Q** were perpendicular to the surface of his evaporated layer. Such a layer is probably under severe planar tension because of the large differential expansion between K and glass. We therefore postulate that the similarly stressed patches of the CESR samples also have **Q** nearly perpendicular to the specimen surface. Since **H** is approximately parallel to the surface, these patches will have **Q** nearly perpendicular to **H**.

The dependence of **Q** orientation on **H** and uniaxial stress that is needed to explain the CESR splitting is identical to the behavior that has been postulated previously to explain other anomalies in K. It should also be appreciated that similar behavior has been established experimentally in other systems. For example, the ability of magnetic field[19] or uniaxial stress[20] to orient the SDW **Q** of Cr has been conclusively demonstrated by neutron diffraction.

A schematic illustration of stress-induced **Q** domains in a thin K sheet is shown in Fig. 2. The linear size of the domains in the plane of the sheet must be about 1 or 2 mm, as shown below. To understand the behavior of CESR as **H** is tilted slightly from an orientation parallel to the surface, one must recall the salient properties of conduction-electron diffusion in a magnetic field.[21] The diffusion constant parallel to **H** is $D_{\|}=\frac{1}{3}v_F{}^2\tau$, where v_F is the Fermi velocity and τ the conductivity relaxation time. Diffusion perpendicular to **H** depends on $\omega_c\tau$, where ω_c is the cyclotron frequency. For $\omega_c\tau\gg1$, $D_{\perp}\approx D_{\|}/(\omega_c\tau)^2$. The relevant quantity for CESR is the spin diffusion length $L=(DT_2)^{1/2}$, which is the average distance an electron will diffuse before the phase coherence of its spin polarization is lost. T_2 is the transverse spin relaxation time. For the extremely pure specimens of Walsh, Rupp, and Schmidt $L_{\|}\sim1$ cm. Since $\omega_c\tau\sim40$, $L_{\perp}\sim0.025$ cm.

Suppose now that **H** is perfectly parallel to the surface of the specimen. The electrons travel in tight helical paths about the field lines. Since their spin diffusion length parallel to **H** is about 1 cm, the volume average g factor of the stress-induced **Q** domains will characterize the CESR. There will be a single resonance line, as observed.[1] Suppose that **H** is now tilted as shown in Fig. 2. The extensive diffusion in the plane is suppressed. An electron originally at point A of Fig. 2 will diffuse along the line AB. After reaching point B, it can only diffuse back to A, etc. Such electrons are trapped in the low-stress domain and will contribute a CESR with $g=g(\mathbf{Q}\|\mathbf{H})$. Geometrical considerations govern the orientation at which a significant splitting can occur. No splitting should occur if the domain size is too small, i.e., smaller than $L_{\perp}$. Since the observed maximum splitting occurred for a tilt angle of $\sim\frac{1}{6}$ rad, the average domain size was probably ~10 times the thickness of the sheet.

It is clear that CESR splitting in K is a phenomenon that is sensitively dependent on the metallurgical preparation of the sample. Even when preparation techniques are reproduced, the precise behavior of the resonance with tilt angle will vary from sample to sample.[2] The ideal specimen—one prepared in a stress-free condition—should not exhibit any splitting.

Since $\Delta g<0$, Eq. (2) predicts that $g(\mathbf{Q}\|\mathbf{H})>g(\mathbf{Q}\perp\mathbf{H})$. Therefore the low-field CESR component must be attributed to the stress-free domains, which have **Q** parallel to **H**. One puzzling feature of the data is the rapidity with which this component disappears for tilt angles greater than 9°. One can only speculate about the cause. The stress-free domains are presumably those in which extensive plastic flow has occurred. Consequently slip bands will have caused originally smooth surface to become irregular, providing a greater likelihood for contamination and surface spin relaxation. As tilt angle increases, the low-field component reflects more uniquely the relaxation behavior peculiar to its own domains.

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