Stress dependence of the Fermi surface of gold

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Derivatives of the five principal extremal cross sections of the Fermi surface of gold with respect to uniaxial stress along various directions have been determined from simultaneous measurements of the amplitudes of magnetostriction and torque quantum oscillations. The accuracy of the measurements is limited by a spurious magnetostriction signal caused by the crystal bending in response to the torque it experiences when its symmetry axis is misaligned with the magnetic field direction. The results are self-consistent and agree with the results of the direct tension measurements and with stress derivatives calculated from the absolute magnetostriction amplitude measurements. Discrepancies with results from other experimental work are discussed. Angular and tetragonal shear strain derivatives deduced from experiment agree within the combined range of uncertainty with theoretical values. The differences between experimental and theoretical values for gold are similar to those for copper.

INTRODUCTION

The measurement of the derivatives of Fermi surface cross-sectional areas of metallic elements with respect to strain is primarily motivated by the need to evaluate the methods and predictions of theoretical band structure calculations. Noble metals are of particular interest in this regard as their Fermi surfaces are just sufficiently complex to test techniques designed for transition metals. Early experimental work[1] determined the hydrostatic pressure derivatives of the Fermi surfaces of copper, silver, and gold and was subsequently refined to high precision.[2] Uniaxial stress derivatives were measured[3] for noble metal Fermi surface cross sections normal to ⟨111⟩ and ⟨001⟩ by applying direct tension along these directions. A significantly higher value of the stress derivative for the neck cross section in gold was subsequently obtained[4] by applying direct compression to the sample. Absolute magnetostriction amplitude measurements[5,6] have yielded uniaxial stress derivatives of noble metal necks that span the range between the tension and compression results.

In the case of copper, discrepancies among the early measurements have been resolved[7,8] using the indirect technique employed in the present work to achieve a set of self-consistent results of relatively high precision. While the copper results agree quantitatively with theory and confirm the validity of the early direct tension experiments, an extension of these measurements to gold[9] yields values for the stress derivative of the neck that were high compared to theory and in agreement with the direct compression result.

This paper reports the results of a comprehensive experimental study of the derivatives of the five principal cross-sectional areas of the Fermi surface of gold (namely, the ⟨111⟩ neck and belly, the ⟨001⟩ rosette and belly, and the ⟨110⟩ dogsbone) with respect to uniaxial stress along various directions. Measurements of oscillatory amplitudes in magnetostriction and torque yield a self-consistent set of results that are sufficiently precise to provide a meaningful comparison with theoretically calculated shear strain derivatives. Discrepancies in the literature can be understood in terms of systematic errors identified and overcome in the present measurements by the use of a specialized alignment jig.

EXPERIMENT

The technique used here to determine the derivatives of the Fermi surface cross-sectional areas with respect to uniaxial stress is based on a method[10,11] which was recently employed to study the stress dependence of the Fermi surface of copper.[7,8] Of the various techniques,[12,13] simultaneous measurement of the amplitudes of oscillatory magnetostriction and torque is regarded as especially accurate and versatile since it does not require measurement of scattering rates or knowledge of Fermi surface parameters (as do absolute amplitude techniques), while the sign of the stress derivative is uniquely determined (whereas ultrasonic velocity measurements require combinations of measurements to determine the sign).

The expression that relates the amplitudes of oscillatory magnetostriction ε_i and torque τ to the logarithmic derivative of the Fermi surface cross-sectional area A normal to the field with respect to stress σ_i is given by[13,8,12]

$$\frac{\partial \ln A}{\partial \sigma_i} = -\frac{\varepsilon_i}{\tau}\frac{\partial \ln A}{\partial \theta}. \quad (1)$$

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Here θ measures the angle between the magnetic field direction and some reference direction, fixed to the sample and perpendicular to the torque axis. When A is extremal with respect to θ, both the measured component of torque τ and $\frac{\partial \ln A}{\partial \theta}$ vanish. The stress derivative of the extremal area is therefore interpolated from measurements of amplitudes of oscillatory magnetostriction and torque, in the present case, at 12 angles θ that are equally spaced and centered about the angle at which the crystal symmetry direction is aligned with the magnetic field direction.

Single crystals of gold were obtained from the National Research Council of Canada.[14] The crystal was first aligned to within $\pm 0.5°$ using Laue x-ray back reflection. Secondly, a parallelepiped sample ($4.6\times3.8\times6.1$ mm) was spark cut from the bulk with a wire saw so that the resulting faces were perpendicular to the [111], $[1\bar{1}0]$, and $[11\bar{2}]$ crystallographic directions. The saw marks were removed from the faces normal to [111] and $[1\bar{1}0]$ by polishing these faces against an optically flat diamond stone. The sample was then annealed by cycling the temperature between 980 °C and 1040 °C once every hour for 24 hours (in a manner similar to that used for copper[15]) in order to remove dislocations introduced by handling or polishing.

Oscillations in sample length were detected using a three-terminal capacitance technique.[16] The capacitance cell that measured magnetostriction and the sample mounting procedure were as described previously.[8] It was found that mechanical polishing of the sample face after mounting in the cell was required to ensure that this face be coplanar with the tops of the cell walls.

Torque was measured by mounting the capacitance cell into the torquemeter as shown in Fig. 1. The torque acting on the sample causes the crossed springs to flex slightly, thereby changing the gap between the static and dynamic capacitor plates. The signal was calibrated by comparing it to the signal resulting from a known torque applied through one of the calibration coils. The design of the crossed spring system was adapted from an older version.[8] While the torquemeter compliance (10^{-7} rad dyn^{-1} cm^{-1}) was about 20% higher than that of its predecessor, nonlinear interaction effects due to torquemeter compliance[17,18] were determined to be negligible on the basis of the interaction parameter (≤ 0.06)

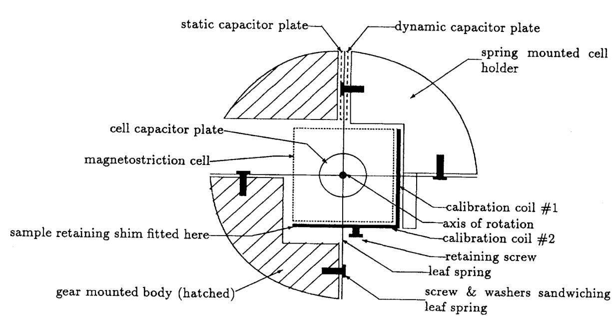

FIG. 1. Schematic representation of the new torquemeter showing essential features (not to scale).

defined in Eq. (5a) of Ref. 18 in analogy with the magnetic interaction parameter a of Eq. (2) below.

Reliable application of Eq. (1) requires that the complicated factors that determine the torque and magnetostriction amplitudes cancel out in the amplitude ratio. Dingle temperatures inferred from the field dependencies of both torque and simultaneously measured magnetostriction, however, have been found to be discrepant by 0.2 K for a sample used for preliminary measurements. The discrepancy is thought to be due to the way in which different regions of an inhomogeneous crystal contribute to the measured components of vector torque and tensor magnetostriction. The measurements reported here are therefore based on a sample characterized by a Dingle temperature of 0.93 K determined consistently from both torque and magnetostriction field dependencies.

All experiments were performed with the magnetic field near 12 T and the temperature at about 4.2 K in order to limit the magnetic interaction parameter a, given by

$$a = 4\pi \left| \frac{dM}{dH} \right| = 8\pi^2 F \tau \left(\frac{\partial \ln F}{\partial \theta} H^3 \right)^{-1} ; \qquad (2)$$

a was 0.16 for the [111] belly orbit and at least an order of magnitude lower for other orbits. Reducing the magnetic interaction parameter in this way ensured that frequency mixing did not interfere with the resolution of individual peaks in the Fourier transforms of the data.

A NMR magnetometer[19] was implemented to characterize the field inhomogeneity along the solenoid axis. By locating the sample in a field region that was determined to be homogeneous to better than 4 parts in 10^5, a potential source of phase smearing was eliminated.

The single largest potential source of systematic error resulted from the bending of the magnetized sample in response to the torque it experienced in the magnetic field. This bending caused the sample face to tilt relative to the magnetostriction cell capacitor plate. If the equilibrium alignment of the sample face was not exactly parallel to the facing capacitor plate, then this gave rise to a spurious change in the magnetostriction cell capacitance that was proportional to the torque and could not be distinguished from the capacitance change due to the true magnetostriction.[8] In practice, it was found that for orbits perpendicular to $\langle 001 \rangle$, the torque-induced magnetostriction could be larger than the true magnetostriction. In order to accurately determine the stress derivative it was essential that the interpolated value correspond to the point at which the symmetry direction was precisely aligned with the magnetic field direction. To this end, a specialized alignment jig was constructed which is similar in design to the sample holder described in Ref. 20 except that the rotation gear circumscribed the torquemeter system shown in Fig. 1 and was driven by a double worm gear reduction similar to the tilt mechanism. The jig allowed independent angular alignments of the sample to within a precision of 0.01° about two orthogonal axes that passed through the sample center. Alignment of the crystal symmetry direction with the magnetic field direction in the plane of rotation was

TABLE I. Experimental stress derivatives (in units of $10^{-12}\ \text{cm}^2/\text{dyn}$).

Cross section	$\frac{\partial \ln A}{\partial \sigma_{[1\bar{1}0]}}$	$\frac{\partial \ln A}{\partial \sigma_{[111]}}$	$\frac{\partial \ln A}{\partial \sigma_{[11\bar{1}]}}$	$\frac{\partial \ln A}{\partial \sigma_{[101]}}$	$\frac{\partial \ln A}{\partial \sigma_{[110]}}$
[111] Neck	-5.38 ± 0.03	8.57 ± 0.04	-3.82 ± 0.03		
[111] Belly	0.069 ± 0.006	-0.459 ± 0.010	-0.032 ± 0.007		
[110] Dogsbone	-1.74 ± 0.06	1.74 ± 0.02			3.70 ± 0.10
[001] Rosette	0.082 ± 0.014	-0.124 ± 0.002		-0.246 ± 0.019	
[001] Belly	0.38 ± 0.02	-0.125 ± 0.014		-0.460 ± 0.012	

achieved by recording the phase of the magnetostriction and torque signals after each of a succession of angular increments. The symmetry direction was inferred from this phase plot to within a typical angular increment size of 0.05°. If the phase of an individual orbit was readily observed, alignment in the tilt plane proceeded in an analogous manner. Alignment of the rosette and belly cross sections, however, required measurement of oscillatory torque amplitudes as the field was varied for successive settings of tilt angle. A vanishing torque amplitude indicated precise alignment of the sample symmetry direction with the magnetic field direction.

The magnetostriction amplitudes of the gold samples studied were found to decrease as the symmetry direction was rotated away from the field direction much more rapidly than the amplitudes due to the copper samples studied.[7] The measurements were therefore taken over angular intervals, ranging from ±2° to ±10°, on either side of the various symmetry directions. The frequency dependence of the Fermi surface cross-sectional area required in Eq. (1) was determined by fitting a polynomial to the results of a KKR phase-shift pseudopotential model calculation. Measurement of the oscillation phase as the crystal was rotated yielded frequency data that was less precise, but consistent with the theoretical determination. The extremum of the interpolating frequency polynomial was made to correspond exactly to the point at which the interpolated value of torque amplitude is zero.

RESULTS AND ANALYSIS

For each of the five orbits studied, several stress derivatives have been determined independently. The particular stress derivative is determined by the direction along which magnetostriction is measured (in this case along either the ⟨111⟩ or ⟨110⟩ axes), while the rotation axis is either ⟨110⟩ or ⟨001⟩. The experimentally measured stress derivatives for each configuration investigated are set out in Table I. The experimental uncertainties associated with the values for the neck and dogsbone, for which orientation errors are negligible, correspond to estimated random errors. Uncertainties in measurements of the rosette and belly orbits are determined by repeating the stress derivative measurements at the two tilt angle settings that bracket the range of uncertainty in the location of the symmetry direction.

Self-consistency of the stress derivatives of a particular cross section with respect to stress along various directions is demonstrated in Table II. Here, the stress derivatives of Table I are combined with experimentally measured hydrostatic pressure derivatives[2] using Eqs. (22)–(26) of Ref. 8.

In order to facilitate comparison to theory, the experimentally measured derivatives may be expressed in terms of volume conserving angular and tetragonal shear strain derivatives using Eqs. (30)–(38) of Ref. 8 along with pressure data[2] and low temperature elastic constants.[21] The results summarized in Table III are weighted averages of all possible ways of calculating these derivatives from independent measurements of uniaxial stress derivatives in various configurations. The theory entries are those calculated[9,22] using a relativistic KKR phase-shift pseudopotential model.[12]

DISCUSSION

Comparison of directly measured stress derivatives to values deduced from measurements in other geometries,

TABLE II. Stress derivatives in units of $10^{-12}\ \text{cm}^2\text{dyn}^{-1}$ as calculated from Eqs. (22)–(26) of Ref. 8, compared with directly measured values.

Stress direction	Cross section	Stress derivative Direct	Stress derivative Calculated	Calculated from eqs. of Ref. 8
[111]	[111] Neck	8.57 ± 0.04	8.70 ± 0.08	(22)
[111]	[111] Belly	-0.459 ± 0.010	-0.444 ± 0.013	(22)
$[11\bar{1}]$	[111] Neck	-3.82 ± 0.03	-3.82 ± 0.05	(22)&(23)
$[11\bar{1}]$	[111] Belly	-0.032 ± 0.007	0.012 ± 0.004	(22)&(23)
$[11\bar{1}]$	[111] Neck	-3.82 ± 0.03	-3.77 ± 0.03	(22)&(23)
$[11\bar{1}]$	[111] Belly	-0.032 ± 0.007	0.017 ± 0.004	(22)&(23)
[110]	[110] Dogsbone	3.70 ± 0.10	3.75 ± 0.08	(24)
[111]	[001] Rosette	-0.124 ± 0.002	-0.120 ± 0.001	(25)
[111]	[001] Belly	-0.125 ± 0.014	-0.119 ± 0.001	(25)
[101]	[001] Rosette	-0.246 ± 0.019	-0.221 ± 0.007	(26)
[101]	[001] Belly	-0.460 ± 0.012	-0.37 ± 0.01	(26)

TABLE III. Angular and tetragonal shear derivatives, calculated by Lee (Refs. 22, 9, and 23) compared with the values derived from the present measurements.

Cross section	Theory (Lee)		Experiment	
	γ_z	γ_{xy}	γ_z	γ_{xy}
[111] Neck	0	4.8 ± 0.2		4.24 ± 0.04
[111] Belly	0	-0.21 ± 0.05		-0.159 ± 0.006
[001] Rosette	-0.40 ± 0.12	0	-0.21 ± 0.04	
[001] Belly	-0.67 ± 0.09	0	-0.55 ± 0.10	
[110] Dogsbone	-1.01 ± 0.17	2.24 ± 0.09	-1.02 ± 0.14	2.48 ± 0.04

as presented in Table II, reveals a high degree of internal consistency with two notable exceptions. First, the directly measured $[11\bar{1}]$ stress derivative of the [111] belly orbit is discrepant with the values calculated from the $[1\bar{1}0]$ and [111] stress derivatives of that orbit, which are self-consistent. This discrepancy is due to inaccurate alignment of the sample in the tilt plane as a result of an ill-refined alignment technique employed for the direct $[11\bar{1}]$ measurement early in the course of the work. It is therefore excluded from further calculations. Second, the stress derivatives of the [001] belly with respect to stress along [101] and $[1\bar{1}0]$ are discrepant slightly beyond the prescribed error estimates. If one assumes that the measured stress derivative is proportional to the magnetostriction amplitude as the tilt angle is varied, then the discrepancy corresponds to a total misalignment of about 0.17°.

The uniaxial stress dependence of the Fermi surface of noble metals has typically been measured in previous experimental work only for stress parallel to the magnetic field direction (with two notable exceptions[8,9]). Comparison of the present data to these results is made in Table IV by taking weighted averages of the appropriate entries from Table II. Although categorically lower in magnitude, the tension results[3] agree to within the combined experimental error with the present results. The compression result[4] is discrepant with all of the other measurements. The claim[4] that the tension results suffer from systematic error due to friction in the vacuum seal is questionable given the agreement that exists between the tension results and both the copper results[8] and the gold results of the present work (both of which display a high degree of internal consistency). While the details of the compression measurement are unpublished, it may be noted that this measurement is susceptible to buckling or rotation of the sample in response to the applied compression (whereas applied tension tends to maintain alignment).

The magnetostriction amplitude results[5,6] are also in agreement with the present work provided they are recalculated using the presently accepted values for effective mass and curvature.[20] With respect to Ref. 6, it has been noted[8] that extrapolation of stress derivative measurements to zero Dingle temperature yields good agreement with more recent measurements[8] in the case of copper. Likewise, this treatment produces good agreement in the case of gold with the present result.

A discrepancy exists between the present work and the measurements of Klimker, Perz, and Lee.[9] This discrepancy may be partly due to the analysis procedure used[9] which ignores the lack of symmetry about the [111] direction in the $(1\bar{1}0)$ plane. Repetition of these measurements as a preliminary introduction to this work has yielded better agreement with the present results when the cur-

TABLE IV. Experimentally measured uniaxial stress derivatives from all known sources. Stress is applied parallel to the magnetic field which defines the normal to the Fermi surface cross section.

Cross section	Derivative (10^{-12}cm^2/dyn)	Source
[111] Neck	9.5 ± 0.4	preliminary—old apparatus
	8.63 ± 0.11	this work
	11.5 ± 0.3	Klimker, Perz, and Lee (Ref. 9)
	7.4 ± 1.2	Shoenberg and Watts (Ref. 3)
	12.3 ± 1.2	Gamble and Watts (Ref. 4)
	8.2 ± 1.0	Aron (Ref. 5) recalculated (see text)
	10.1 ± 1.1	Slavin (Ref. 6) recalculated (see text)
	(9.6 ± 0.9)	Slavin (recalculated $T_D = 0$ value)
[111] Belly	-0.43 ± 0.01	Ostapiak (Ref. 23)
	-0.452 ± 0.012	this work
	-0.32 ± 0.05	Shoenberg and Watts
[001] Rosette	-0.65 ± 0.07	Ostapiak
	-0.54 ± 0.06	this work
	-0.49 ± 0.12	Shoenberg and Watts
[001] Belly	-0.63 ± 0.04	Ostapiak
	-1.3 ± 0.2	this work
	-1.0 ± 0.1	Shoenberg and Watts

rent method of analysis and refined frequency data are employed. The small remaining discrepancy may be understood in terms of a misalignment of the sample by about 1° (as determined through x-ray photographs) in the tilt plane that the early apparatus could not correct.

Agreement of experiment with theory is generally satisfactory but marginal for the rosette and neck orbits. It is worth noting that the theoretically calculated values are larger in magnitude for all orbits but the dogsbone. This pattern is identical to that observed in comparisons of copper measurements to theory (except for the [111] belly angular shear derivative for which the experiment could not resolve the sign with certainty). The errors associated with the theoretical values reflect the sensitivity of the model to the uncertainty in the Fermi energy parameter and the spin-orbit parameter.[9] A reduction of the Fermi energy parameter could improve agreement between the measured and calculated values, but the extent of the differences cannot be spanned by adjustment of this parameter alone.

CONCLUSION

From simultaneous measurements of quantum oscillations in magnetostriction and torque, the values of uniaxial stress derivatives of the five principal cross sections of the Fermi surface of gold have been determined for stress along various directions. A specialized alignment jig and experimental technique have been developed in order to allow alignment of the crystal in the magnetic field to a precision necessary to allow accurate determination of the stress derivative. Although angular adjustments to better than ±0.01° are possible, the measurement accuracy is limited by the uncertainty in locating the symmetry direction relative to the magnetic field direction. The larger experimental uncertainties associated with the stress derivatives of the belly and rosette reflect this uncertainty. The derivatives of individual Fermi surface cross-sectional areas with respect to stresses along various directions are found to be internally consistent when combined with experimental values of hydrostatic pressure derivatives. The early results of direct tension experiments,[3] although systematically lower in magnitude, agree just within the combined error with the present measurements. The early magnetostriction measurements[5,6] on the neck cross section are also consistent with the present work. While the value measured by Klimker, Perz, and Lee[9] may be rejected due to imprecise analysis, repetition of the experiment of Klimker, Perz, and Lee by the author has yielded results that are in better agreement with the current values. Only the very high value of neck stress derivative measured by direct compression[4] cannot be reconciled with the stress derivatives measured by other researchers.

Angular and tetragonal shear strain derivatives for each cross section determined by phase-shift pseudopotential calculations are generally larger in magnitude but agree within the combined range of uncertainty with the values deduced from the experimentally determined stress derivatives.

The uniaxial strain response of the Fermi surface of gold can be interpreted in terms of a dilation as well as angular and tetragonal shears. The tetragonal and angular shear strain derivatives of the Fermi surface of gold are appreciably greater than those of copper.

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