Polar Reflection Faraday Effect in Silver and Gold*

JAMES C. MCGRODDY, ARCHIE J. MCALISTER, AND EDWARD A. STERN
University of Maryland, Department of Physics and Astronomy, College Park, Maryland
(Received 30 April 1965)

Measurements of the polar reflection Faraday effect (PRFE) in pure Ag and Au have been made in the visible and near-infrared regions of the spectrum. The measurements are remarkably insensitive to the details of the sample preparation and aging, and can therefore be confidently assumed to represent the bulk behavior. The PRFE measurements are in good quantitative agreement with the values calculated using the known properties of pure Ag and Au, and are strongly influenced by the contact of the Fermi surface at the zone boundaries. Conversely, such measurements can give a quantitative measure of the amount of contact of such Fermi surfaces with the zone boundaries, even for alloys.

I. INTRODUCTION

WHEN plane polarized light is reflected from a metal with a magnetic field normal to the surface, the reflected light is found to be slightly elliptically polarized with the major axis of the ellipse rotated slightly from the initial polarization direction. This effect is known for nonferromagnetic metals as the polar reflection Faraday effect (PRFE), and the study of this effect can yield valuable information on the band structure of metals and alloys. For ferromagnetic metals the effect is much larger and is called the magneto-optic Kerr effect. In a recent paper[1] the theory of this effect as well as a detailed study of the effect in aluminum were presented. The results of this study indicated that, at least for aluminum, the effect is independent of the details of sample preparation or aging, in marked contrast to the case of optical constants. Thus the experimental results can be confidently taken to represent the bulk properties of the metal being studied. In the present work we report a comprehensive study of the PRFE in Ag and Au in the visible and near-infrared regions of the spectrum. These results can be interpreted in terms of the models of the Fermi surfaces of the noble metals proposed by Roaf and Shoenberg.[2] In the case at hand, no new information on the band structure is obtained, but we confirm experimentally the sensitivity of the PRFE to contact of the Fermi surface with the zone faces. In fact, the existence of contact could easily be deduced from a measurement of only the PRFE and the optical constants. It is planned to use this sensitivity to contact of the Fermi surface with the zone faces as a probe of the band structure of the Ag-Au alloy system. Inasmuch as the interesting information on this alloy system comes from a measurement of the PRFE in the visible and near-infrared portions of the spectrum, the condition $\omega\tau\gg1$ can be realized even for a disordered alloy. Here ω is the angular frequency of the light, and τ is the electron relaxation time. When this condition is satisfied the effects of collisions of the electrons can be neglected.

In the next section, we briefly discuss the theory of the PRFE, especially as applied to the noble metals. Section III gives the experimental results which are interpreted in Sec. IV. Conclusions are summarized in Sec. V.

II. THEORY

There is a wealth of information, both theoretical and experimental, available on the band structure of Ag and Au. Extensive de Haas–van Alphen effect measurements and impedance measurements in the anomalous skin region have been fitted to analytical surfaces by Roaf.[2] The magneto-acoustic effect in these metals has been studied by Bohm and Easterling,[3] and recently cyclotron resonance has been studied in Au by Langenberg and Marcus[4] and in Ag by Kip and Howard.[5] More recent low-field de Haas–van Alphen measurements in Ag and Au have been performed by Joseph and Thorsen.[6] An analysis of the optical properties of silver has been performed by Ehrenreich and Philipp.[7] The results of all these studies are consistent with a model for the Fermi surface which consists of a distorted sphere with necks making contact with the zone faces in the $\langle111\rangle$ directions. The spherical "belly" portion is more distorted and there is a larger neck contact in the case of Au than for Ag. The main "belly" distortions consist of a bulging of the sphere in the $\langle100\rangle$ directions and a flattening in $\langle110\rangle$ directions.

In the case of cubic metals and weak magnetic fields, the only new information obtained from a measurement of the PRFE is the off-diagonal element of the conductivity tensor σ_1. In a region where there are no interband transitions, and where $\omega\tau\gg1$, σ_1 can be expressed as

$$\sigma_1=(e^3H/6\pi h^2c\omega^2)I_{\mathrm{FS}}, \quad (1)$$

* Supported in part by the Advanced Research Projects Agency under Contract SD-101.

[1] E. A. Stern, J. C. McGroddy, and W. E. Harte, Phys. Rev. **135**, A1306 (1964).

[2] D. Shoenberg, Phil. Trans. Roy. Soc. London **255**, 85 (1962); and D. J. Roaf, *ibid.* **255**, 135 (1962).

[3] H. V. Bohm and V. J. Easterling, Phys. Rev. **128**, 1021 (1962); **125**, 812 (1962).

[4] D. N. Langenberg and S. M. Marcus, Phys. Rev. **136**, A1383 (1964).

[5] Kip and Howard (unpublished) by private communication from J. F. Koch.

[6] A. S. Joseph and A. C. Thorsen, Phys. Rev. Letters **13**, 9 (1964).

[7] H. Ehrenreich and H. R. Philipp, Phys. Rev. **128**, 1622 (1962).

where I_{FS} is an integral over the Fermi surface, which for our purposes in the ensuing discussion can be most usefully put in the following forms:

$$I_{FS}=3\int dk_z \oint d\phi v_\perp^2 \tag{2a}$$

$$=\int_{FS} v^2\left(\frac{1}{\rho_1}+\frac{1}{\rho_2}\right)ds, \tag{2b}$$

where the line integral in (2a) is done around a cross section of the Fermi surface perpendicular to the magnetic field, which is taken to be in the z direction, $v_\perp$ is the component of the Fermi velocity at a point on this curve, and ϕ is the angle between some fixed direction in this plane and the normal to the Fermi curve. The integral over k_z means to sum over all such cross sections. In the second form, the integral is over the whole Fermi surface, v is the velocity at a point, and ρ_1 and ρ_2 are the principal radii of curvature of the surface at that point, i.e., the radii of curvature in two mutually perpendicular planes whose intersection is normal to the surface. The combination $(1/\rho_1+1/\rho_2)$ is invariant with respect to rotation of these two mutually perpendicular planes about the line of their intersection. From the form (2b) it is clear that this integral is independent of the location of the crystal axes relative to the z direction.

In order to make a reasonable estimate of the effects of zone face contact, a simplified model of the Fermi surface of the noble-metal type has been constructed. Insofar as only one new number is obtained from the PRFE data, while the model used has several free parameters, the usefulness of the model might be questioned. However, the model is most useful in that it shows that the general effect of zone face contact is independent of the details of velocity distribution for any reasonable distribution, and that the PRFE is indeed sensitive to this contact. Using parameters for the model which we derived from the Roaf-Shoenberg Fermi surfaces and the measurements of Joseph and Thorsen[6] gives a satisfactory agreement with the experimental results.

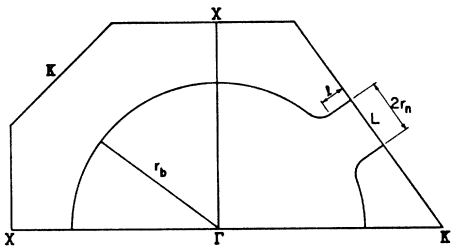

FIG. 1. The model of the Fermi surface used in the calculations to compare with the PRFE and the optical measurements for pure silver and pure gold. The necks consist of a cylindrical section joined smoothly onto a spherical belly. Sections of cross sections in the (100) and (110) planes are shown.

The details of the model follow, and a picture of the model is given in Fig. 1. The central section is spherical, and cylindrical necks are in contact with the ⟨111⟩ zone faces. The necks and the central section are joined by a surface which is a portion of a toroid, and is tangent to both the cylinders and the sphere at their points of contact. For comparison, the Roaf surfaces of Ag and Au are shown in Fig. 2.

The integral in form (2b) can easily be performed for this surface, using velocities which are constant over each portion of the surface. One can also calculate the intraband dielectric constant for this model. A useful quantity to consider is the ratio of the integral given in (2a) and (2b) to the square of the integral involved in the intraband dielectric constant[8] $\int_{FS} vds$. Such a quantity is unchanged if the velocity at each point is scaled by a constant factor. We define the combination

$$R=6\pi^3 n I_{FS}\Big/\left(\int_{FS} vds\right)^2, \tag{3}$$

where n is the density of conduction electrons. This quantity has the property that for a spherical Fermi surface it is unity. The quantity R as a function of neck radius calculated from our model for several sets of parameters is shown in Fig. 3. The sets of parameters used in the calculation include the extremes of the topological parameters and velocity distributions vary-

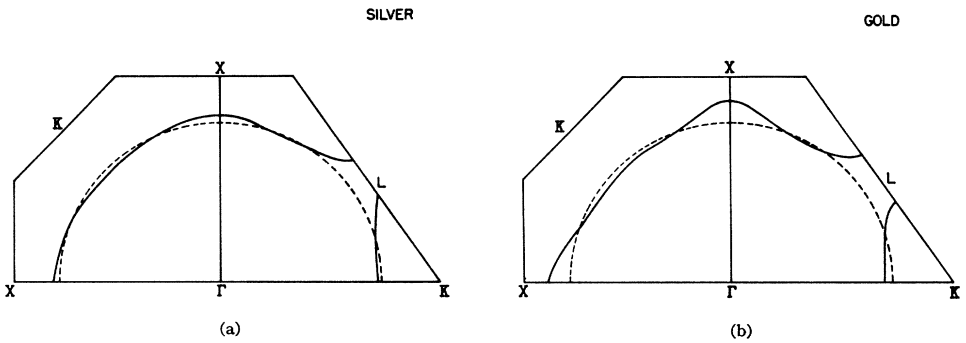

FIG. 2. The actual Fermi surfaces of pure Ag and pure Au after Roaf. The same cross sections are shown as in Fig. 1.

[8] M. H. Cohen, Phil. Mag. 3, 762 (1958).

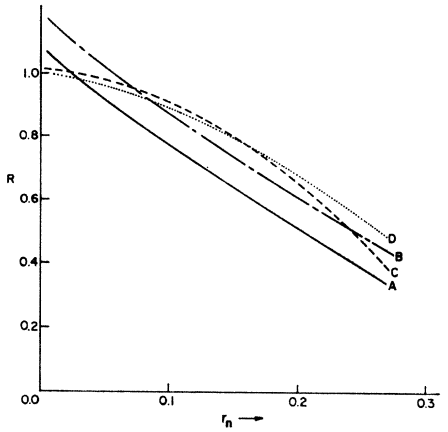

FIG. 3. The parameter R versus the neck radii in units of the free-electron sphere radius r_n for different values of the other parameters. The A curve is for the case where the velocity is a constant over the whole surface and the length of the cylindrical portion of the neck is zero; i.e., the neck consists entirely of the joint section. The B curve is again for the case where the velocity is a constant over the whole surface but the neck consists entirely of the cylindrical portion and the joint radius is equal to zero. The C and D curves are the more physical situations where the velocity is permitted to vary. The velocity is assumed to have a constant value over the belly, another constant value over the joint, and a third constant value over the cylindrical portion of the neck. The ratio of the velocity on the cylindrical portion of the neck to the belly velocity is assumed to be directly proportional to r_n/r_b and is chosen to equal one at $r_n/r_b=0.3$. The joint velocity is always taken to be the average of the cylindrical portion and belly velocities. The C curve is for the extreme case where the neck consists entirely of joint and the D curve is for the other extreme where the neck consists entirely of the cylindrical portion. Note the insensitivity of the result to this particular assumption.

ing from being uniform over the entire surface to the more physical situation permitting a variation of the neck velocity as the neck radius changes. One expects roughly that the neck velocity will be directly proportional to the neck radius, and in Fig. 3 we have plotted R for this case assuming that $v_n/v_b=1$ for a neck radius of 0.3 the belly radius. The joint velocity v_j is taken to be the mean of v_n and v_b. Here the subscripts n, j, and b refer to the neck, joint, and belly regions of the Fermi surface as explained in the caption to Fig. 1. Figure 3 shows that the quantity R is always a monotonically decreasing function of the radius of contact with the zone face. The effects of additional distortions of the Fermi surface such as the bulges in the ⟨100⟩ directions in Au would be to increase the value of R by a small amount.

The quantity R can be deduced from experiment using the formula for the PRFE given in Ref. 1;

$$\chi+iQ=\frac{4\pi i\sigma_1/\omega}{(\epsilon_0-1)\{\sin^2\theta/\cos\theta+(\epsilon_0-\sin^2\theta)^{1/2}\}}, \quad (4)$$

where ϵ_0 is the dielectric constant in the absence of a magnetic field, σ_1 is the conductivity defined in Eq. (1), ω is the angular frequency, and the square root is defined to have positive real and imaginary parts. χ is the PRFE angular rotation and Q the PRFE ellipticity for initially s-polarized light. In a region where interband absorptions are not important and well below the plasma frequency and $\omega\tau\gg1$, we have χ a constant, independent of ω and $Q=0$. Thus experimental determination of the frequency dependence of χ and Q together with the wave-length dependence of ϵ_0 enables one to determine the region where σ_1 has the simple form given by (1) and (2). From this the quantity R defined in Eq. (3) can be calculated which then measures the amount of contact of the Fermi surface with the zone boundary.

III. EXPERIMENTAL

The experimental setup used to measure the PRFE is practically identical to that described in Ref. 1, except for the substitution of a piece of birefringent materal for the phase shifter described in that paper. The samples were prepared by evaporation in vacuum onto polished pyrex substrates. The material used for the evaporation had a stated purity of 99.995%. Pressures immediately before evaporation were about 5×10^{-6} Torr.

The optical constants of the samples were measured by the Drude method immediately after evaporation. The samples were exposed to the atmosphere for a total of approximately one or two hours before being placed in the measuring apparatus. While in the measuring apparatus, the samples are stored at a pressure of approximately 0.05 Torr.

Measurements of the PRFE were made on two different gold samples and on two different silver samples. No difference in the measurements outside of the experimental uncertainties of about 1% was found on the same Ag sample when repeated after an interval of about eight months. During this interval, the specimen was stored in a desiccator under a nitrogen atmosphere. In addition there was no appreciable difference in the PRFE values for the two different silver specimens. For the gold samples, there was about 4% difference in the PRFE values for the two different specimens. The results are presented in graphical form in Figs. 4(a) and 4(b). The error bars represent one standard deviation and include the differences from one sample to another, but do not include errors in calibration, which are estimated to be less than 2%. These results emphasize the rather remarkable fact that the PRFE is very insensitive to the details of sample preparation and aging, while the ordinary optical constants are quite strongly influenced by these same factors.

IV. INTERPRETATION

As was mentioned above, it is possible to obtain an estimate of the extent of contact of the Fermi surface with the zone boundaries from a measurement of the PRFE and the ordinary optical constants. If measure-

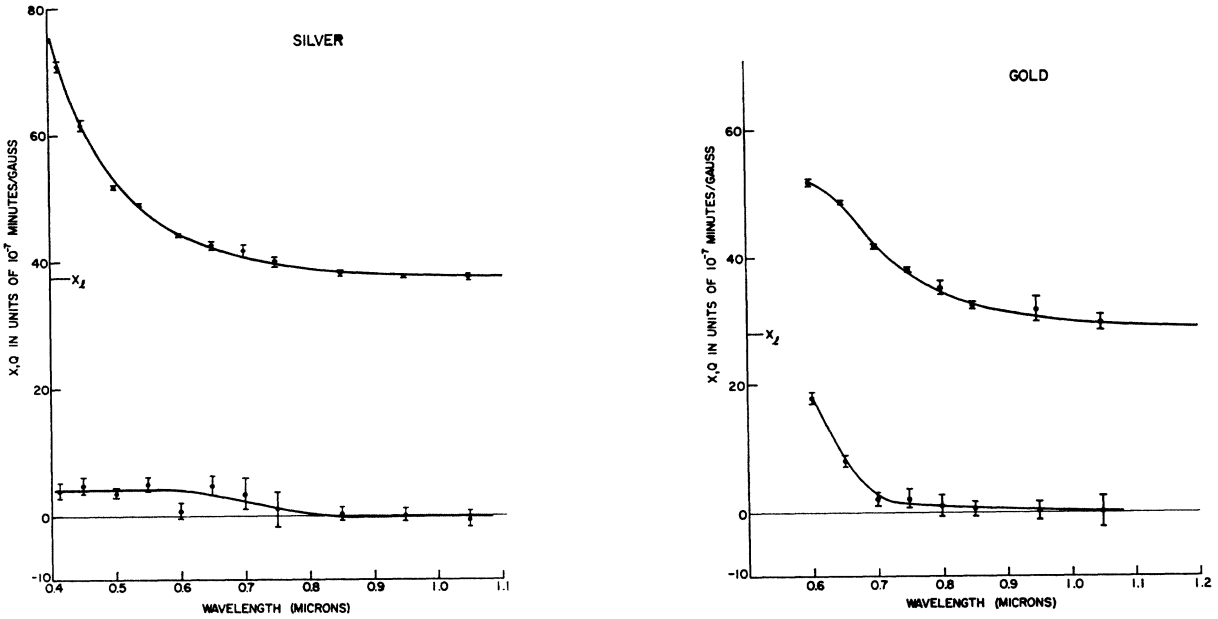

FIG. 4. (a) The values of the measured PRFE angular rotation χ and the ellipticity Q for pure silver as a function of wavelength. The long-wavelength constant value is χ_l given in Table I. (b) The values of the measured PRFE angular rotation χ and the ellipticity Q for pure gold as a function of wavelength. The long-wavelength constant value is χ_l given in Table I.

ments are performed where all of the following conditions are satisfied: The PRFE is independent of wavelength; the ellipticity is zero; the optical constants exhibit no appreciable interband effects or absorptions; and $\sin^2\theta$ in (4) can be set equal to zero, the quantity R defined in Eq. (3) can be obtained from the experimental data as

$$R=\frac{4\pi nec}{\omega}\frac{(-\epsilon_0)^{1/2}}{(1-\epsilon_0)}\frac{\chi}{H}, \tag{5}$$

where all the symbols have been defined previously. The ϵ_0 are now pure real numbers and from the above-mentioned assumptions, have the form

$$\epsilon_0=1-4\pi ne^2/m_0\omega^2, \tag{6}$$

where m_0 is the optical mass. Since it is also assumed that ω has a small enough value so that χ is independent of ω, this implies from (1), (4), and (6) that the first term on the right-hand side of (6) can be neglected compared to the other term. Under these conditions, R can be written,

$$R=(\chi_l c/H)(4\pi nm_0)^{1/2}, \tag{7}$$

where χ_l is the long-wavelength extrapolated value of χ which is independent of frequency. This form has the desirable feature that the optical measurements (which are all summarized by m_0) are not weighted as heavily as the PRFE measurements. Since the PRFE measurements are more reliable than the optical measurements, the value for R calculated from (7) can be expected to have some meaning.

For silver as indicated in Table I, the uncertainty in m_0 is 6% while the uncertainty in χ_l which has to be extrapolated from the measured values to its wavelength-independent value, is about 1%. The total uncertainty in R for Ag is about 3%. There has not been as detailed analysis of the optical data of Au as there has been for Ag so that we estimate the optical mass of Au as follows. Cyclotron resonance data gives belly masses m_b as listed in Table I. We assume that the optical masses for Ag and Au are in the same ratio as the belly masses for these materials. This is reasonable

TABLE I. Summary of data.

	Au	Ag
m_b	1.1[a]	0.95[b]
m_n	0.30[c]	0.40[c]
r_n/r_b	0.186[d]	0.14[d]
m_n/m_b	0.27	0.42
v_n/v_b[e]	0.69	0.33
v_j/v_b[f]	0.85	0.66
m_0	1.2±0.15[g]	1.03±0.06[b]
n	5.90×10^{22}	5.86×10^{22}
χ_l	$28\pm1\times10^{-7}$ min/G	$37.5\pm0.3\times10^{-7}$ min/G
R [calculated case (a)]	0.66	0.83
R [calculated case (b)]	0.69	0.86
R (measured)	0.69±0.05	0.86±0.03

[a] Reference 4.
[b] Reference 5.
[c] Reference 6.
[d] Reference 2.
[e] Calculated from $v_n/v_b=r_n m_b/r_b m_n$.
[f] Calculated from $v_j/v_b=(v_n+v_b)/2v_b$.
[g] Calculated by assuming that the m_0 for Ag and Au are proportional to their respective m_b.
[h] Reference 7.

since the shape of the Fermi surfaces of the two metals are so much alike. By this means we obtain the optical mass with about 13% uncertainty. The uncertainty in the wavelength-independent extrapolation of χ for Au is larger than for Ag since the extrapolation is further and is estimated to be 4%. In this manner and using the data listed in Table I we find from (7) the values of R listed in Table I as R_{meas}.

In calculating the expected value of R from the model discussed in Sec. II it is necessary to fix the various parameters in the model. From experiment the ratio of the neck to belly radii r_n/r_b and the ratio of the neck to belly mass m_n/m_b are known as listed in Table I. From this data the ratio of the neck to belly velocities can be calculated by

$$v_n/v_b=r_n m_b/r_b m_n.$$

The velocity on the joint between the neck and belly v_j is not given directly by experiment, but we assume that v_j is the average of the neck and belly velocities.

The remaining quantity in the model to be fixed is the length of the cylindrical region of the neck l. This also determines the joint radius connecting the cylinder to the belly. The result that one obtains is not very sensitive to the assumed length l. The table shows the calculated results in the two extreme cases that (a) the neck is composed entirely of a joint region and $l=0$, and (b) the neck is composed entirely of a cylindrical portion and is attached to the belly by a joint of zero radius. In case (b) the joint still gives a contribution to R. All the R values calculated from the model agree within the experimental uncertainty with the measured values.

As was mentioned in the Introduction, no new information on the band structure of Ag or Au has been obtained from this experiment. However, we have provided experimental confirmation of the sensitivity of the PRFE to contact of the Fermi surface to the zone boundaries. In fact, it is not unreasonable to conclude that from the results of this experiment alone one would have predicted such contact, and could even have obtained a semiquantitative estimate of it. The optical constants of Ag and Au in the infrared are about the same so that the angle of rotation χ in the PRFE is approximately proportional to R. The fact that χ for Ag is larger than that for Au is a direct consequence of the fact that the Ag Fermi surface has a smaller contact with the zone faces than that of Au.

This agreement between theory and experiment for pure Ag and Au gives us confidence that the same analysis applied to the Ag-Au alloy system will permit a determination of the shape of the Fermi surface in this alloy system. For most of the range of this alloy system the more powerful methods for determining the shape of the Fermi surface such as magnetoresistance, de Haas–van Alphen effect, and cyclotron resonance, do not work because of the small relaxation time of the electrons. Because such high frequencies are used in the PRFE, the small relaxation time found in these alloys does not cause any difficulty and this effect can be used on such alloys.

V. CONCLUSIONS

In this paper we have presented a detailed study of the PRFE in Ag and Au in the visible and near-infrared regions of the spectrum. It is found that the PRFE is remarkably insensitive to the details of sample preparation and aging, and can therefore be confidently assumed to represent a bulk property. The results are in good agreement with the accepted band structures of these metals, and are strongly influenced by the contact of the Fermi surface at the zone boundaries. Thus, the PRFE can be used to study such contact in metals and alloys.

ACKNOWLEDGMENT

The authors take pleasure in acknowledging the technical assistance of O. H. McKim.