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Oscillatory metallic field effect and surface magnetoelasticity in thin ferromagnetic films

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The metallic field effect is the change of surface conductance linear with electrostatic surface charge q. In thin Permalloy films this signal is sensitive to the direction θ and magnitude H of an applied magnetic field, showing an overall 4θ dependence for an in-plane field, and varying as $1/H$. Comparison with an analogous bulk signal proportional to q^2 indicates that the linear signal originates in the magnetoconductivity modulation of a thin surface layer caused by interfacial stresses linear in q. Detailed interpretation of the data in terms of this model allows determination of the magnetic properties of the surface layer. The interfacial stresses deduced from the data agree with theoretical estimates.

I. INTRODUCTION

The metallic field effect (MFE) is the change of surface conductance of a metal with electrostatic charging. Recent studies have established that it is caused by the change in surface scattering of current carriers when the metal surface becomes charged.[1,2] but without specifying a detailed mechanism for this interaction. Several proposed models[3-5] have failed to account for one or another of the major observed features of the effect such as its sign, its linearity in applied surface charge density, or its temperature dependence. The study presented here was undertaken to extend MFE data to ferromagnetic metals, which had not been investigated before. They also offer the magnetic state of the sample and of its surface as an additional parameter that might influence the MFE.[6] In particular, observed changes in spin-dependent surface scattering would contribute supplementary information of the type needed to elucidate the origin of the MFE.

The basic MFE experiment consists of making a thin ferromagnetic film one plate of a parallel-plate capacitor and of measuring its change of conductance as a function of the charge on the capacitor; the magnetic state of the sample is controlled by an in-plane magnetic field $\vec{H}$ at an angle θ to the current. With a surface charge density varying like $q\sin\omega t$, the observed change of film conductance at ω and 2ω is given by

$$\delta\Sigma = -\delta\Sigma(\omega)q\sin\omega t + \delta\Sigma(2\omega)q^2\cos 2\omega\tau\,, \qquad (1)$$

with both $\delta\Sigma(\omega)$ and $\delta\Sigma(2\omega)$ sensitive to $\vec{H}$ and θ.

As shown in the preceding paper[7] (which from now on will in this report be referred to as I), the second-harmonic amplitude $\delta\Sigma(2\omega)$ describes a bulk effect arising from the modulation of the conductivity and of the magnetoconductivity of the sample by electrostrictive strains transmitted from the dielectric substrate to the film. In the same measurements, the first harmonic $\delta\Sigma(\omega)$ was confirmed to be a surface effect and independent of sample thickness, just as in nonmagnetic metals. It represents a true MFE.

Except for this one important difference in their thickness dependence, the magnetization-dependent parts of $\delta\Sigma(\omega)$ and $\delta\Sigma(2\omega)$ show strikingly similar behavior. To emphasize this similarity, we have presented the data on both amplitudes of Eq. (1) together in the figures of I, even though in that paper the emphasis was on interpreting the second harmonic. In this paper we will analyze the first-harmonic data given in I, as well as present and discuss additional information on $\delta\Sigma(\omega)$.

We will show that the form of the magnetic field dependence of $\delta\Sigma(\omega)$ follows from a magnetoconductivity modulation in the surface region as a result of strains in a thin surface layer caused by interfacial stresses linear in q. Therefore, it results from the full surface analogue of the corresponding bulk effect that causes $\delta\Sigma(2\omega)$, and it must reflect the magnetic and elastic properties of the surface region. Thus an analysis of $\delta\Sigma(\omega)$ enables us to deduce the anisotropy energy, its easy axis, and its strain dependence in this region. In addition, the field-independent part of $\delta\Sigma(\omega)$ can be used to obtain good estimates of the magnitude of the charge-induced interfacial stresses.

We find, for example, that in going from bulk to the surface region, of all magnetic properties only the strain dependence of the anisotropy energy changes markedly. Furthermore, the charge-induced interfacial stresses have magnitudes agreeing with theoretical estimates, but varying somewhat with Permalloy composition. The dependence of all these parameters on static strains in the sample is also reported.

Section II gives a brief review of the experimental results already presented in I. In Sec. III we interpret the data within the model outlined above, and Sec. IV extends the analysis to statically strained Permalloy samples.

II. EXPERIMENTAL RESULTS

The method of measurement of $\delta\Sigma(\omega)$ has already been discussed in I. Most data to be reported here have been obtained on the samples described there, and the experimental curves displayed in Figs. I-2, I-3, I-4, and I-7 are typical of the results obtained.

As formulated in Eq. (1) $\delta\Sigma(\omega)$ measures the response of $\delta\Sigma$ linear in q. It does not vary significantly with film thickness, at a given nominal Permalloy composition, and therefore we must relate it to an interaction of the current carriers with the interface region.

Figures I-2, I-3, and I-4 show that $\delta\Sigma(\omega)$ obeys characteristics closely following those of $\delta\Sigma(2\omega)$. In each case there is a magnetic-field-independent component, on which a field-sensitive signal is super imposed, with a roughly 4θ variation, and with an amplitude increasing as H decreases. The maxima and minima of both signals usually come near the same field angle, although occasionally sharp differences in phase are observed, as for example in Fig. 1.

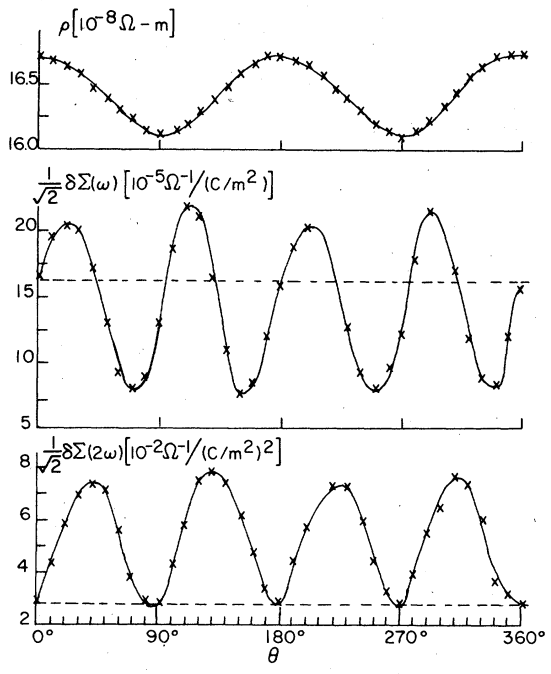

FIG. 1. ρ, $\delta\Sigma(\omega)$, and $\delta\Sigma(2\omega)$ vs θ for a 82-18 Permalloy film of 500 Å, at $H = 20$ Oe. The high-field values, given by the dashed lines, indicate that there is a large phase shift between $\delta\Sigma(\omega)$ and $\delta\Sigma(2\omega)$.

Apart from their thickness dependence, $\delta\Sigma(\omega)$ and $\delta\Sigma(2\omega)$ differ in two important respects. First, the amplitude of $\delta\Sigma(\omega)$ is always of the same sign, even when the oscillations of $\delta\Sigma(2\omega)$ reverse, as, for example, in Fig. I-4. Second, when the sample is mechanically clamped so that there can be no macroscopic distortion with charging, $\delta\Sigma(2\omega)$ is suppressed, but $\delta\Sigma(\omega)$ is completely unaffected (Fig. I-7).

Any good understanding of these data must account for both the similarities and the differences in the two signals.

III. INTERPRETATION

When we focus on the similarities between $\delta\Sigma(\omega)$ and $\delta\Sigma(2\omega)$, it becomes evident that the H and θ dependence of both signals appear to follow the same formal description. This is true to such an extent that we have tried to fit the data for $\delta\Sigma(\omega)$ to an equation of the type shown in Eq. (I-14). Let us therefore write

$$\delta\Sigma(\omega) = B_s(\omega) + A_s(\omega) F(h_s, \theta; \theta_{es}), \tag{2}$$

where the subscript s has been introduced to denote surface quantities. At this stage the amplitudes A_s and B_s cannot be defined further because, in contrast to the structure of $\delta\Sigma(2\omega)$, their composition is unknown. F is the function already defined by Eq. (I-12) [or (I-16)] and now has as variables the reduced surface field h_s and the surface easy axis direction θ_{es}. θ is the in-plane direction of $\vec{H}$ relative to the current. The direction of the surface magnetization $\vec{M}_s$ is determined by an equation like Eq. (I-4), perhaps generalized to include an additional exchange energy contribution proportional to $\vec{M} \cdot \vec{M}_s$ arising from the difference in direction of the two magnetizations.

The curve fitting of the data to Eq. (2) is very successful. As a matter of fact, for many samples the parameters h and θ_e are exactly the same for fitting the field and angle dependence of both $\delta\Sigma(2\omega)$ and $\delta\Sigma(\omega)$. Two such fits of $\delta\Sigma(\omega)$ are shown in Figs. 2 and 3 for the sample data of Figs. I-2 and I-3, using the identical parameters required for the fit of $\delta\Sigma(2\omega)$ of Figs. I-5 and I-6. For other samples, especially those showing a phase shift between $\delta\Sigma(\omega)$ and $\delta\Sigma(2\omega)$, the fit appears to require a new easy axis direction θ_{es}. However, in view of the strong exchange energy accompanying large changes of direction of the magnetization, this is an unlikely interpretation. Alternatively, the phase shift can be incorporated by generalizing the strain dependence of the anisotropy energy in the surface, K_s, to allow not only changes in magnitude but also in easy axis direc-

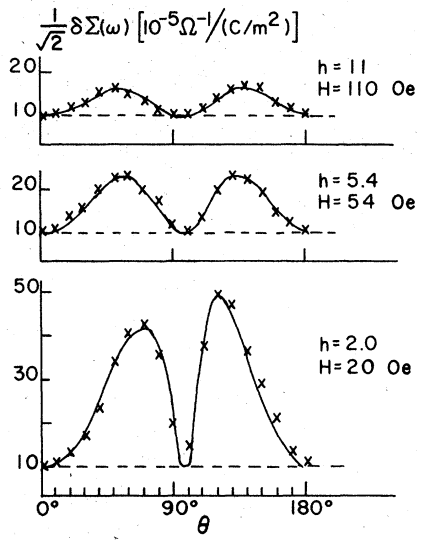

FIG. 2. Match of the experimental points of $\delta\Sigma(\omega)$ of Fig. I-2 to Eq. (I-16) at three fields, using the parameters $H_K = 10$ Oe, $\theta_t = 5°$, $\theta_m = 90°$, $\kappa = 0.1$, with the common amplitude $A_s(\omega) = 2.0 \times 10^{-3}\Omega^{-1}(C/m^2)^{-1}$.

tion. If we write

$$\delta K_s/K_s = \eta_s e_s, \quad \delta\theta_e = \epsilon_s e_s \tag{3}$$

then the form of F of Eq. (I-12) still holds, but with the substitution

$$\theta_{es} = \theta_e + \tan^{-1}(\epsilon_s/\eta_s) \tag{4}$$

and with the addition of a constant h_{sb} (where s is surface and b is bulk) in the denominator to

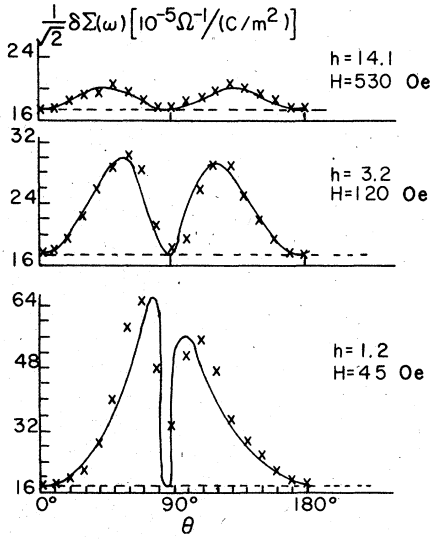

FIG. 3. Match of the experimental points of $\delta\Sigma(\omega)$ of Fig. I-3 to Eq. (I-16) at three fields, using the parameters $H_K = 38$ Oe, $\theta_t = -5°$, $\theta_m = 90°$, $\kappa = 0.03$, and the common amplitude $A_s(\omega) = 1.2 \times 10^{-3}\ \Omega^{-1}(C/m^2)^{-1}$.

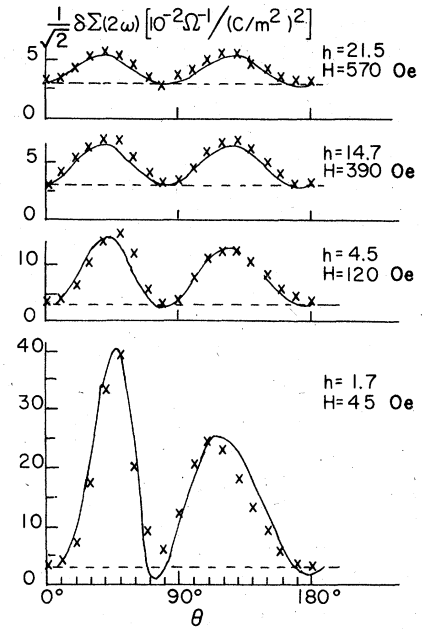

FIG. 4. Experimental curves of $\delta\Sigma(2\omega)$ of a 540-Å 90-10 Permalloy film, and their match, as in Fig. 2, with the parameters $H_K = 26.5$ Oe, $\theta_t = -15°$, $\theta_m = 90°$, $\kappa = 0.6$, and the common amplitude $A(2\omega) = 1.4\ \Omega^{-1}(C/m^2)^{-2}$.

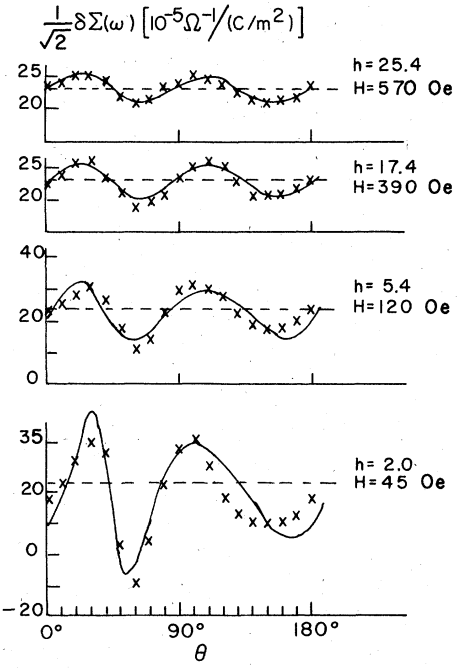

FIG. 5. Experimental curve and fit of $\delta\Sigma(\omega)$ for the film of Fig. 4. The parameters are $H_K = 22.5$ Oe, $\theta_{ts} = -50°$, $\theta_{m=90}°$, $\kappa = 0.6$, $h_{sb} = 0.5$, and the common amplitude $A_s(\omega) = 2.4 \times 10^{-3}\ \Omega^{-1}(C/m^2)^{-1}$.

TABLE I. Experimental amplitudes of the two contributions to the MFE of Eq. (2), for Permalloy films at two compositions. The values of $A(2\omega)$ and $B(2\omega)$ are taken from the data of I.

	t (Å)	$A_s(\omega)$ $[10^{-3}\ \Omega^{-1}(C/m^2)^{-1}]$	$B_s(\omega)$	$A(2\omega)$ $[\Omega^{-1}(C/m^2)^{-2}]$	$B(2\omega)$
90-10	260	1.1	0.52	0.35	0.017
	280	1.2	0.25	0.25	0.027
	540	2.4	0.33	1.4	0.045
		(2.0,1.4)			
80-20	280	3.2	0.18	0.002	0.023
	300	2.0	0.14	0.37	0.018
	300	2.3	0.29	−0.41	0.017
	400	3.0	0.18	−0.05	0.028

characterize the exchange coupling between $\vec{M}$ and $\vec{M}_s$. The constant A_s must then be composed of two contributions

$$A_s = (A_{\eta s}^2 + A_{\epsilon s}^2)^{1/2} \propto \eta_s (1 + \epsilon_s^2/\eta_s^2)^{1/2} . \tag{5}$$

With this slight generalization we have obtained good fits to any films showing phase shifts between $\delta\Sigma(\omega)$ and $\delta\Sigma(2\omega)$. An example of such fit is given in Figs. 4 and 5 for a 90-10 film, with the parameters listed in the captions. The anisotropy field H_K is slightly lower in the surface region, but, generally, surface and bulk parameters are very close.

Using these prescriptions for fitting the data, we have obtained the amplitudes A_s and B_s of Eq. (2) for the entire group of samples whose second-harmonic response was analyzed in I. These values are listed in Table I, which also contains the corresponding constants at 2ω derived from the results already discussed in I. We observe some fluctuation in properties from sample to sample, but there is a clear increase of A_s with composition as the percentage of iron increases, while the opposite trend is seen in B_s. The one additional set of values for A_s indicated in parentheses refers to the two constants A_{η_s} and A_{ϵ_s} discussed in Eqs. (3)–(5).

The important conclusion derived from Figs. 2–5 and from Table I is that the same theory that has successfully explained the values of $A(2\omega)$ in I gives a systematic fit to the magnetization-dependent contribution to $\delta\Sigma(\omega)$. Effectively, this comparison forces that we interpret $\delta\Sigma(\omega)$ in terms of a very similar model.

Such a model must have the following features: (i) Surface charging of the metal induces a strain in the immediate surface region: (ii) The strain is linearly proportional to the surface charge density q; (iii) the strain pattern decays away from the surface, and cannot be suppressed by macroscopic clamping of the metal sample; (iv) in the region in which it exists, the strain can alter the local magnetic anisotropy energy just as in bulk, and thus affect the direction of the local magnetization; (v) the magnetoresistance of the affected layer responds to the changing direction of the local magnetization, giving rise to a characteristic change of surface conductance.

Such a model would give a self-consistent interpretation of the magnetic field dependence of $\delta\Sigma(\omega)$. But, unless we know the detailed form of the strains resulting from the surface stress, and their spatial variation, as well as the electrical and magnetic properties of the surface region, it is impossible to formulate the MFE of this model quantitatively. At this stage, it is best to apply the model to evaluate some of its most important parameters from the experimental data, in order to see that they are consistent and of reasonable magnitude. For this purpose, for instance, we can assume that the strains active in the surface exactly parallel those induced by electrostriction of the mica, and that the average strain e_s proportional to q acts throughout an effective thickness t_s. Under these simplifications we can use exactly the same formulation as in I and apply it to the surface layer of thickness t_s. In analogy with Eq. I-14, we therefore write

$$\delta\Sigma(\omega) = \Sigma_s s_s \left(\frac{2\alpha}{1-\alpha} + \gamma + \frac{2\Delta\sigma}{\sigma_0} \eta_s F(h_s, \theta; \theta_{es}) \right) \tag{6}$$

where Σ_s is the conductance of the surface layer, $\Sigma_s = \sigma_0 t_s$, and $s_s = e_s/q$ couples the strain e_s to q. α is Poisson's ratio of the metal, γ is the metal's strain dependence of conductivity, and $\Delta\sigma/\sigma_0$ is its magnetoconductivity coefficient. All of these quantities should be characteristic of the surface layer, but we may assume them not to be too different from their bulk values. On the other hand, η_s, the strain dependence of the anisotropy energy K_s, given in Eq. (3), is very different in the surface region. This two-layer model can be applied to electrical conduction since the carrier mean free path in Permalloy is probably smaller or at most equal to t_s, so that size effects play a small role, and since the actual transition between layers is gradual so that there should be very little interlayer scattering.

By comparing Eq. (6) with Eq. (2), we can express the constants A_s and B_s in terms of the parameters of the model. In particular, using the corresponding constants $B_s(\omega)$ and $B(2\omega)$, we find

$$\Sigma_s s_s = \tfrac{1}{2}(\Sigma s)(B_s/B) , \tag{7}$$

and by substituting Eq. (7) in the expression for A_s, we can determine η_s from the experimental

TABLE II. Strain coefficient of surface magnetic anisotropy energy, η_s, and surface stress per unit surface charge density X_s/q of the Permalloy films of Table I, according to Eqs. (8) and (10). The value $s_{11}(1-\alpha) = 5.4\times10^{-12}$ $(N/m^2)^{-1}$ was taken to be that of nickel (Ref. 9).

	t (Å)	η_s	X_s/q $[(N/m)(C/m^2)^{-1}]$
90–10	260	0.14×10^3	4.0
	280	0.54	1.3
	540	0.63	2.0
		($\epsilon_s = 0.44$)	
80–20	280	1.9	1.1
	300	1.5	1.2
	300	0.6	2.5
	400	1.5	1.3

numbers:

$$\eta_s = (\sigma_0/\Delta\sigma)(B/\Sigma s)(A_s/B_s). \tag{8}$$

The values of η_s obtained in this fashion for the samples of Table I are given in Table II, using any of the other needed film properties from Table I of I. All η_s at any one composition are very similar, but change significantly with composition. (Incidentally, if oxide clamping of the value of $B(2\omega)$ has to be taken into account, as suggested in I, all η_s may be somewhat larger.) Compared to the bulk values η of Table I of I, the most important difference is that there is no change in sign for η_s at the 80-20 composition. In fact, since H_K itself is practically unchanged in going from the bulk to the surface, the behavior of η_s at the two compositions indicates that at 90-10 the response to strain of the anisotropy energy δK_s becomes smaller, while at 80-20 it often becomes larger, but always remains of the sign characteristic of the bulk properties of the nickel-rich compositions. Evidence for such a nickel-rich metallic surface layer, of a depth of about 25 Å (but also dependent on the method of film preparation) has been obtained from Auger studies.[8]

Thus the numbers for η_s come out reasonably, and, in fact, should be able to shed new light on some of the magnetic surface properties of Permalloy films. Our results indicate that (a) the main parameters controlling magnetic behavior, such as H_K and θ_e, are the same as in bulk, but (b) the strain dependence of the anisotropy energy differs significantly in the surface, and may involve changes in both magnitude and direction. It is important to recall, however, that the values of η_s deduced in Table I rely on specific assumptions concerning the strain distribution and the electrical properties of the surface layer.

In the spirit of the same model we can also use our results to make an estimate of the magnitude of the surface stress produced by the charge density q. If the surface stress X_s is isotropic in the surface, it gives rise to an average strain within the thickness t_s given by

$$e_s = s_{11}(1-\alpha)X_s/t_s, \tag{9}$$

where s_{11} is the appropriate elastic constant of the metal. Using the relation $e_s = s_s q$, and replacing s_s from Eq. (7), we can write

$$\frac{X_s}{q} = \frac{s}{2s_{11}(1-\alpha)}\,\frac{B_s}{B/t}. \tag{10}$$

The values of the unit surface stress for each of the samples of Table I calculated from Eq. (10) are also listed in Table II. Very little is known experimentally about such surface stresses. They must arise from a surface piezoelectric effect of the metal, or the underlying substrate, or both. In either case, such effects can appear if these surface regions lose their bulk symmetry to allow a polar response. They are known to exist on the surface of insulators,[10] and they have also been invoked to explain the interaction of single electrons with metal surfaces.[11] In fact, Herring's order-of-magnitude predictions for the surface

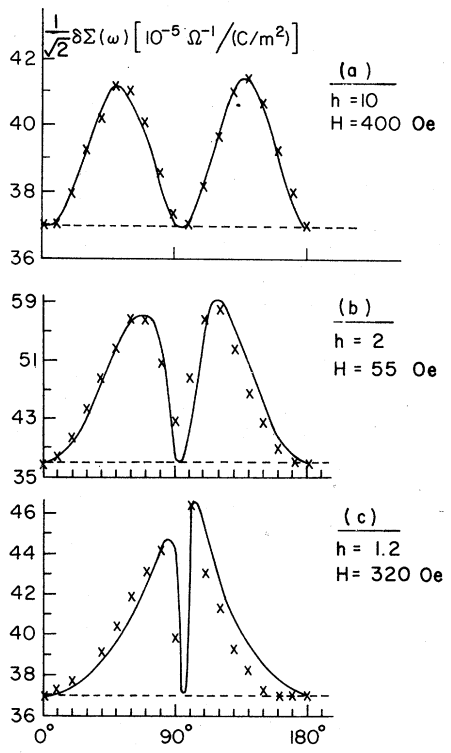

FIG. 6. $\delta\Sigma(\omega)$ of a 260-Å 90–10 film under static strains (a) $e=0$, (b) $e_{\parallel}=2\times10^{-4}$, and (c) $e_{\perp}=10^{-3}$. The parameters for the match at these strains are listed in Table III.

TABLE III. Static strain dependence of the MFE parameters of the 90-10 Permalloy film of Fig. 6.

Strain	Units	$e=0$	$e_{\parallel}=2\times10^{-4}$	$e_{\perp}=10^{-3}$
H_K	Oe	40	27.5	270
θ_t	deg	5	5	5
κ		0.2	0.1	0.01
$A(2\omega)$	$\Omega^{-1}(C/m^2)^{-2}$	0.35	0.38	0.06
$B(2\omega)$	$\Omega^{-1}(C/m^2)^{-2}$	0.017	0.021	0.014
$A_s(\omega)$	$10^{-3}\Omega^{-1}(C/m^2)^{-1}$	1.1	1.1	0.2
$B_s(\omega)$	$10^{-3}\Omega^{-1}(C/m^2)^{-1}$	0.52	0.52	0.52
X_s	$(N/m)(C/m^2)^{-1}$	4.0	3.2	4.9
η_s		140	140	25
η		1400	1500	220

stress of metal surfaces with charging are in excellent agreement with the values of Table II. This table indicates, furthermore, that X_s varies from one material to another, thus suggesting that the MFE may offer a direct method for investigating surface stresses due to surface charging as a function of the two materials making up the interface. That subject is taken up in another publication.[12]

We conclude that with respect to the two most important new variables of our proposed model, namely, η_s and X_s, our data give consistent and plausible results.

IV. DEPENDENCE OF THE MFE PARAMETERS ON STATIC STRAIN

As a further test of the consistency of our interpretation, we have studied the variation with static strains of all four signal amplitudes appearing in the MFE of ferromagnetic films. The experimental data, exemplified for one film by Figs. 6 and I-8, lead to the parameters listed in Table III for two applied strains $e_{\parallel}$ and $e_{\perp}$ parallel and normal to the easy axis.

As already discussed in I, the variation of the bulk parameters H_K, θ_e, and η is as expected. That the analysis of the data is in each case successful using the same set of parameters to characterize $\delta\Sigma(\omega)$ and $\delta\Sigma(2\omega)$ confirms the earlier conclusion that the magnetic properties in the surface region are close to those of bulk. The last three lines of Table III give the interpretation of the experimental coefficients in terms of Eqs. (8) and (10), and of the η dependence of the coefficient $B(2\omega)$.

The strain dependence of these coefficients has some interesting implications, even given the uncertainties of the model within which they were derived. First, the specific interfacial stress X_s caused by q is roughly independent of macroscopic static strain. This also implies that there is no strain dependence of the coefficients γ, so that for strains up to 10^{-3}, the conductivity σ_0 is linear in the strain. Second, η and η_s are influenced by static strain in the same way, becoming significantly smaller at high strain. On the other hand, the products $H_K\eta$ and $H_K\eta_s$ stay more or less constant under all strain conditions. This suggests that if Eq. (I-7), where the strain dependence of the anistropy energy K is defined, is altered to read

$$\delta K = \sum_i \bar{\eta}_i e_i , \tag{11}$$

the new strain coefficients $\bar{\eta}_i$ are now independent of the value of K_0, and δK is linear in the strains at least up to 10^{-3}.

V. CONCLUSIONS

The magnetic field dependence of the linear MFE in Permalloy films follows the quadratic signal discussed in I to such an overwhelming extent that an exactly parallel analysis of the data is required. The linear signal, independent of sample thickness and not suppressed by macroscopic clamping, must therefore arise from the magnetoconductivity modulation in a surface layer produced by an interfacial strain linear in the applied surface charge density. Simple estimates of the magnetic surface properties in terms of this model indicate that (in the magnetically annealed samples studied here) only the strain dependence of the anisotropy energy differs markedly from its bulk value, while this energy itself, and the easy axis direction, remain unchanged. Thus the magnetic field dependence of the MFE offers a new tool for investigating the surface properties of ferromagnetic metals. As an additional application, we have determined the linearity of the change of anisotropy energy with strain, both in the surface and in bulk, up to strains of 10^{-3}.

The success in interpreting the magnetic field

dependence of the linear MFE as caused by surface strains suggests that the field-independent contribution has a similar origin. This intepretation allows an estimate of the surface stress caused by charging, in good agreement with theoretical predictions. It thus appears that the field-independent MFE is a means of exploring interface stresses caused by electrostatic charging.

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