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OPTICAL FARADAY EFFECT IN FERROMAGNETIC AND FERRITE FILMS

By R. L. COREN and M. H. FRANCOMBE,
Philco Scientific Laboratory, Blue Bell, Pennsylvania, U. S. A.

Résumé. — On a étudié la dispersion de l’effet Faraday dans les couches de métaux ferromagnétiques et de ferrites oxydées dans le domaine de nombres d’onde s’étendant de $1.4 \times 10^4$ à $3.7 \times 10^4$. Dans les couches métalliques de Ni, Fe, NiFe$_2$O$_4$ et CoFe$_2$O$_4$, la rotation est positive et décroît lorsque le nombre d'onde augmente. Dans les couches de ferrites, la rotation de Faraday est positive pour de grands nombres d’onde et négative pour des nombres d’onde plus petits avec des pics positifs et négatifs. On discute l’absorption optique dans les ferrites.

Abstract. — We have studied the Faraday rotation of the plane of polarization of linearly polarized light by thin films of the ferromagnetic metals, Ni, Fe, NiFe$_2$O$_4$, CoFe$_2$O$_4$, and of the ferromagnetic oxides, NiFe$_2$O$_4$ and CoFe$_2$O$_4$. Hysteresis loops of the ferrite films are displayed using a Faraday effect hysteresigraph. Curves are given for the dispersion of Faraday effect in the metal and oxide films, from $1.4 \times 10^4$ to $3.7 \times 10^4$ wave numbers. In the metal films the rotation is positive and decreases as wave number increases. Superimposed on this general trend are several dispersive anomalies. Faraday rotation in the ferrite films is positive at large wave numbers and negative at lower wave numbers, displaying positive and negative peaks. Optical absorption in the ferrites is discussed.

Introduction. — The Faraday effect in magnetic materials has attracted interest as a means of studying their technical magnetization processes and electronic energy structures. The effect has been employed to examine their hysteretic properties [1, 2] and domain structures [1, 3]. Measurements of the magnitude of the Faraday effect often shows a strong frequency dependence [4-6], sometimes with pronounced resonance type behavior [7]. Since these studies require sample transparency they can only be carried out in certain frequency bands, or with thin single-crystal sections of sufficient purity and perfection, or on materials fabricated in film form. The extreme opacity of ferrites of the first row of transition elements e.g. Ni, Co, Cu has, thus far, made it impossible to study thin bulk samples with visible radiation. It is desirable to do so since crystal field splitting of the 3d electronic energy levels of these divalent metal ions gives rise to energy separations which fall in this range [8]. Transitions between the split states have been correlated [9] with the strong Faraday effect in these materials, making the Faraday rotation a likely means of examining their electronic structure. With the recent development of techniques for producing films of ferrite materials [2, 10] it is now possible to obtain transparent samples and, therefore, to examine their detailed optical properties.

As a result of improved vacuum facilities and techniques, films produced today are more reproducible and reliable than those studied in the past. For this reason, and because previous studies of Faraday effect in magnetic metals have often lacked sufficient precision and detail for a complete determination of its frequency dependence, it is worthwhile to reexamine these materials. This report describes studies we have made of the Faraday effect in thin films of several ferromagnetic metals and alloys and in nickel and cobalt ferrite, over the wavelength range from 2 700 Å to 7 000 Å.

Experimental. — The saturation rotation of the plane of polarization of visible radiation is of the order of $10^4$ degrees/cm for ferromagnetic material and $10^4$ degrees/cm for ferrimagnetic material. Consequently, with a ferromagnetic film 1 000 Å thick or a ferrimagnetic film 10 000 Å thick (the approximate limits of transparency to visible radiation for metals and for Ni and Co ferrites), one expects a rotation of, at most, a few degrees. Because of the smallness of the effect, a configuration with the polarizing and analyzing prisms rotated relatively by 45° produces changes of intensity of the transmitted radiation which are proportional to the Faraday rotation of a sample placed between them. In our experiments the light beam falls on a photomultiplier tube whose output is balanced against a constant d. c. source, amplified, and recorded on the ordinate of an XY Recorder. By turning the analyzer through a high ratio, precision speed reducer this axis can be calibrated directly. The abscissa of the recorder is controlled by the output of a calibrated gaussmeter whose probe is adjacent to the film, so that the system produces a display of the Faraday rotation as a function of applied field.

Two separate optical systems were used: a Faraday effect hysteresigraph, with which to study
the B — H loop properties of the films, and a Faraday effect " spectrometer " for studying the spectral dispersion of the Faraday effect. In the hysteresigraph white light passes through the film at an angle of 45° to its plane while the magnetic field is parallel to the film in the plane of incidence. Since the Faraday effect is proportional to the component of magnetization along the light path [11], magnetic changes in the film plane change that component proportionally, so that as the external field is varied the recorder traces the planar hysteresis loop of the sample. To investigate planar magnetic anisotropy the film can be rotated about its normal.

The Faraday effect " spectrometer " is constructed so that the magnetic field and the monochromatic light beam both pass normally through the film. The field is supplied by an electromagnet with 4 inch cylindrical poles, separated by 0.5 inches, and which have 1/8 inch axial holes for the light path. As the applied field increases the magnetization component perpendicular to the film increases proportionally [12], so that the optical rotation changes linearly with applied field. When the applied field $H = 4\pi M$ magnetic saturation occurs, so that the ferromagnetic Faraday effect reaches its maximum value and remains constant as $H$ is further increased. Superimposed upon this behaviour is the Faraday rotation due to the substrate. Since the substrate is non-ferromagnetic its Faraday effect is proportional to the applied field and can be subtracted.

The metal films studied were prepared by cathodic sputtering from the appropriate element or alloy source, using substrates in the form of microscope cover slips or vitreous silica discs, 12 mm in diameter. NiFe$_2$ and CoFe$_2$ alloy films on silica were subsequently oxidized in air at 1000 °C to produce the ferrites. Details of the sputtering and oxidation processes, as well as of the magnetic and structural properties of these films have been described before [2,13].

Results. — Previously reported [13] measurements on sputtered films of Ni-Fe alloys have shown that, provided suitable vacuum techniques are used, their magnetic properties are highly reproducible and correspond well with those of bulk material at thicknesses down to 100 Å. Magnetic tests on the metal films described here indicate essentially similar behavior. Analysis by electron and X-ray diffraction show the metal and oxide films to be structurally uniform, polycrystalline and single phase.

Magneto-optic hysteresis data for NiFe$_2$O$_4$ and CoFe$_2$O$_4$ films are shown in figure 1. In obtaining these curves the amplitude of the measured ordinate signal depends upon the absorption and specific rotation of the material over the wavelengths range used, the intensity of the light source and the sample thickness. Consequently, the curves in figure 1 have been normalized to a scale of bulk saturation magnetization. Discussion of the coercive force values obtained from such loops has been included in a previous report [2]. Both the loop shapes and coercive forces are very reproducible and show little variation with preparation temperature, providing it is high enough (e.g. above 700 °C for NiFe$_2$O$_4$), but do depend upon film thickness, approaching a constant value above 5 000 Å.

The results of Faraday rotation measurements on several ferromagnetic metals and alloys are shown in figure 2. Of the four metallic ferro-
magnetic films reported here only nickel has a value of $4\pi M$ low enough to permit its being saturated in the 14 000 gauss maximum field of the magnet. With the other films the saturation rotation could not be determined directly, as only the low field Faraday line can be measured. Consequently, the ordinate on figure 2 is the slope of the Faraday curve, after subtracting the rotation due to the substrate. This slope has been labeled $\nu t$ by analogy with the relation for Faraday effect in nonmagnetic materials [1]: $\varphi = \nu t H$; $\nu$ is the apparent Verdet constant, $t$ the sample thickness and $\varphi$ is the Faraday rotation in field $H$. The region with the bar at the bottom of the figure was measured on films of 300 Å to 350 Å thickness. The portions outside of this bar were measured on thinner samples and normalized to the thick film curves.

The curves in figure 3 show the experimental normal Faraday rotation of a film of NiFe$_2$O$_4$ with two different frequencies of light. (Note that the vertical scale is different in the two cases.) It is seen that as the field is increased, the signal from the phototube changes in opposite directions when radiations of 0.29 $\mu$ and 0.44 $\mu$ wavelength are used, indicating that the directions of rotation are of opposite sense in these cases. Taking the usual definition of positive rotation as that of a right hand screw advancing along the field direction, we have established that the figure at 0.29 $\mu$ corresponds to positive rotation and that at 0.44 $\mu$ to negative rotation.

The complete frequency dependence of the saturation rotations of nickel ferrite and cobalt ferrite are shown by the solid curves in figure 4. Both curves are taken from films which are about 1 600 Å thick. The curve for nickel ferrite displays a negative maximum at about 23 000 cm$^{-1}$ and a positive maximum at about 30 000 cm$^{-1}$. Cobalt ferrite has a positive maximum at approximately 28 000 cm$^{-1}$ and drops off in a manner similar to the nickel ferrite. The measurements have not been carried far enough to locate a negative maximum.

The absorption coefficient $\alpha$, from the Beer-Lambert law $I = I_0 \exp(-\alpha t)$, of these ferrites is shown in figure 5 as a function of wave number. These values were determined from the transmittance through films of different thicknesses.

**Discussion.** — The use of the Faraday effect to display magnetic hysteresis loops is interesting in that it illustrates the versatility of the optical rotation technique. In addition, it permits us to make comparisons of the magnetic properties of our films and bulk material. We refer to these structural and magnetic studies and to similar
investigations of the metallic films in order to indicate that the films reported on here are reproducible and exhibit properties corresponding to more massive samples.

The curves shown in figure 2 for the dispersion of Faraday effect in ferromagnetic metals exhibit a general trend of decreasing rotation with increasing wave number. This is in agreement with earlier work on nickel and iron films; the points shown on the figure are taken from Siertsema [5] for nickel and iron, and the dashed curve for iron is taken from Skinner and Tool [4]. These data have been adjusted to fit our results at 17,000 cm⁻¹. The necessity of this correction is not surprising since many factors can change the scale of the measured Faraday rotation. The most important of these is probably uncertain knowledge of the thickness values of the films. Others might include the high degree of strain which exists in metal films, and which has recently been shown to alter the Faraday rotation in cobalt films [14], and anisotropy arising from structural texture (e.g. fibre structure) in the films, which would affect their magnetic and optical properties. While these and other factors might shift the absolute level of the observed Faraday rotation, it is not expected that they will alter the validity of the relative spectral changes shown on figures 2 and 4.

Superimposed on the overall trend in figure 2 are several regions of anomalous dispersion. This type of structure has been observed in garnets [6], where it is attributed to particular electron transitions [9], but has not previously been reported in metals. While correlations can be seen between these anomalous regions in the same and different metals the data are presently too incomplete for a proper analysis to be made.

The Faraday rotations in films of NiFe₂O₄ and CoFe₂O₄ are shown on figure 4. This is the first time such data are available for these materials in the visible range. The most striking feature of the curves is the fact that the rotation is negative over a considerable range. While negative Faraday rotations have been observed in garnets [6], these are connected with ferrimagnetic compensation behavior, which is not the case here. Rotational behavior similar to ours has been observed by Skinner and Tool [4]; some of their results are shown by the dashed curve on figure 4. These data are taken from iron films purposely prepared under poor vacuum conditions and which are reported as being "dark" rather than metallic and quite transparent in the visible. They undoubtedly contain large amounts of oxide which dominate their optical behavior.

Clogston [9] has pointed out that optical Faraday effect in the garnets may arise either from charge transfer between the transition metal ions and the surrounding oxygen anions, or from internal transitions between cation levels which are split due to crystalline fields, and that these two mechanisms lead to opposite rotations. He indicates that a strong Faraday effect in NiFe₂O₄ and CoFe₂O₄ could only be ascribed to internal transitions such as 3d → 4p. In order to study the pertinent transitions it is of interest to examine the absorption in these materials. Curves of the absorption coefficients of NiFe₂O₄ and CoFe₂O₄ are presented on figure 5. The dashed curve represents the measurements of Skinner and Tool [5] on their "dark" iron films. It is seen that both ferrites have an absorption band in the region where the Faraday effect is negative or is becoming negative. At smaller wave numbers the absorption rises again. α in these ferrites seems to be 10 to 100 times greater than in yttrium iron garnet [6] at the same frequency. The stronger interaction between ferrite and radiation indicated by this absorption also manifests itself in a larger Faraday rotation; the saturation rotation is about 100 times that in the garnets [6].

It should be noted that the data presented in this report have, so far, only been obtained from rather thin films. Until these investigations are carried out over a larger thickness range it will be difficult to determine whether any of the observed properties are unique to the film geometry (such as e.g. the magnetic resonance modes reported in thin films [15]) or characteristic of bulk. More studies are now being undertaken on films of different thicknesses for this purpose.

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Discussion

M. ZAININGER. — How are your thicknesses determined?

Réponse : The metal film thicknesses are determined from the time of sputtering, since thickness is proportional to sputtering time [13]. The proportionality is determined by weighing thick samples. The ferrite thicknesses are computed from the thickness of the unoxidized metal films and the relative cell dimensions, per metal atom, of the ferrite and metal alloy.

M. HEAVENS. — How do your measured values of 4πM agree with bulk values?

Réponse : The method of measuring 4πM by magnetic saturation normal to the film plane is sensitive to strain, crystalline effects, shape effects which alter the demagnetizing field, e. t. c. [12]. In general, however, the measured 4πM values of our metal and oxide films are in good agreement with tabulated values.
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APPLICATION DES COUCHES MINCES EN OPTIQUE

Par P. GIACOMO,
Faculté des Sciences de Caen.

Résumé.
On essaye de dégager les difficultés générales (calculation, contrôle, uniformité, stabilité, matériau), comment elles guident le choix des solutions adoptées et comment elles limitent la qualité des résultats. Anti-reflets, miroirs semi-transparents et opaques, filtres colorés, polariseurs, filtres d'amplitude sont analysés de ce point de vue.

Abstract.
An attempt is made to disentangle the general difficulties (calculation, control, uniformity, durability, materials), how they guide the choice of the methods adopted, and how they limit the performance. Anti-reflection coatings, semi-reflecting and opaque mirrors, color filters, polarizers, amplitude filters are reviewed in this respect.

Il a été souvent question, dans les communications précédentes de mesures précises des propriétés optiques des couches minces : pouvoir réflecteur R (ou R'), transmission T, déphasages correspondants Ar{ (ou A,,), At, auxquels il nous sera utile d'ajouter l'absorption A = 1 - R - T (ou A' = 1 - R' - T). Ces mesures visaient à obtenir des renseignements sur les propriétés des matériaux constituant les couches et sur leur structure.

Pour le physicien paresseux, il est tentant d'utiliser toute cette documentation dans un autre but : on a souvent besoin de modifier les propriétés optiques d'une surface, par exemple d'augmenter son pouvoir réflecteur, ou, au contraire, de le diminuer, sans modifier ses propriétés géométriques ; en cherchant bien dans les nombreux exemples déjà étudiés, on a quelque chance de trouver une couche mince susceptible de rendre le service attendu. Comme la chose semble facile, on pourra même empiler les unes sur les autres plusieurs couches et obtenir ainsi des propriétés intéressantes ou amusantes.

Cette dernière idée élargit effectivement beaucoup.