Zeitschrift: Archives des sciences [1948-1980]

Herausgeber: Société de Physique et d'Histoire Naturelle de Genève

Band: 14 (1961)

Heft: 10: Colloque Ampère

Artikel: The measurement of magnetic fields in the surface layer of a ferromagnet by the method of paramagnetic resonance

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DOI: https://doi.org/10.5169/seals-739609

The Measurement of Magnetic Fields in the Surface Layer of a Ferromagnet by the Method of Paramagnetic Resonance

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Abstract.

A description is given of the method of measuring magnetic fields in small volumes by the method of paramagnetic resonance. The apparatus permits fields to be measured in a volume of 10^{-8} cm^3. It is shown how this method can be used to determine the magnitude of the demagnetizing fields of ferromagnetic ellipsoids. The determination of the mean intensity of the stray fields in the surface layer of a ferromagnet by means of electron paramagnetic resonance of a film of a free radical, applied directly to the ferromagnetic surface, is described. It is shown that the stray fields determined in this way greatly influence the value of the ferromagnetic resonance curve width, particularly in the case of metallic macroscopic single crystals.

Zusammenfassung.

Es wird eine Methode der Magnetfeldmessungen in kleinen Volumina mit Hilfe paramagnetischer Resonanz beschrieben. Mit dieser Anordnung kann man Felde in Volumina 10^{-8} cm^3 messen. Diese Methode dient auch zur Messung des Entmagnetisierungsfeldes von ferromagnetischen Ellipsoiden. Die Bestimmung des Mittelwertes der Streufelder in der Oberflächenschicht von Ferromagnetika geschieht durch die paramagnetische Resonanz der dünnen Schicht des freien Radikals, die fest auf der ferromagnetischen Oberfläche liegt. Es wird gezeigt, dass die Streufelder, die nach dieser Methode bestimmt werden, den Wert der Linienbreite der ferromagnetischen Resonanz wesentlich beeinflussen, besonders im Falle von makroskopischen Einkristallen.

Method of Measuring Magnetic Field in Small Volume Elements

The requirements laid on the measuring method of magnetic fields is that the field is measured in the smallest possible volume with great accuracy. The dem andfor accuracy leads us to choose the resonance method, for which we shall try to use material with a large value of the magnetic moment in a small volume, while the relaxation time of the moment should be as long as possible. These requirements will be satisfied well by the method of measuring the magnetic field by means of the electron paramagnetic resonance of free radicals in the first phase (where $\Delta H \doteq 1.8$ Oe for the case of the compound DPPH).

This radiofrequency method permits the exact measurement of the values of static magnetic fields from tens of Oe to tens of kOe. Its disadvantage, of course, is that more radiofrequency apparatuses are necessary for different frequency bands if measurements are to be made in a wide range. For our problem of measuring magnetic fields around 3 kOe apparatus working in a narrow frequency range (8-12 kMc) is sufficient but considerable demands will be made on its sensitivity. In order to increase the sensitivity of the apparatus we used high-frequency secondary modulation of the magnetic field having a frequency of 120 kHz [1]. By selective amplification and lock-in detection we attained a sensitivity of 10^{13} ΔH spins at room temperature. With such a sensitivity and using DPPH we can measure a magnetic field in a volume of 10^{-8} cm^3. For this purpose we chose microscopic DPPH crystals, obtained by crystallizing DPPH from a solution of pure benzene. Ferromagnetic resonance can be measured in this way even without resonant cavities in a short-circuited waveguide, e.g. on whiskers 2 mm in length and a few microns thick [2]. Rectangular and cylindrical resonant cavities were used for the measurements and high-frequency magnetic modulation was applied to the cavity through a wall made either from silver-plated manganin foil or from vacuum-silver-plated glass. The power reflected from the cavity was separated from the incident power by a ferrite circulator; selected silicon diodes were used for detection. The frequency around 9 kMc was generated by a klystron of the type 2k25 and stabilized by the Pound d-c method with an accuracy of the order 10^{-7}; the accuracy in measuring the frequency by comparison with the harmonics of an exact crystal oscillator is 1×10^{-7}. The magnetic field of the electromagnet is electronically voltage stabilized with a short-period stability to 10^{-4}. The magnetic field of the electromagnet is measured in a wide range by proton resonance using a Pound oscillator with an accuracy of the order 10^{-5}.

It should be noted that the sensitivity of the apparatus can be used very well not only for measuring spatial variations in the magnetic field strength, but also for measuring the parameters of electron, particularly ferromagnetic, resonance at different points of the same samples [3], e. g. in thin films. If, for example, we use a resonance cavity of mode H_{111} with a very small opening in the front wall, we can make a " map " of the different quantities found by the method of ferromagnetic resonance (e.g. the *g*-factor, internal stresses, anisotropy) with the resolving power of the circular area having a diameter of 0.2 mm [4].

Problem of Measuring Magnetic Field in Surface Layer of Ferromagnet.

Here we shall first solve the problem of measuring the demagnetizing field of an ellipsoid by the direct method. Obviously, at those points on the surface of an ellipsoid where the tangential surface is parallel to the direction of the vector of magnetization, the value of the magnetic field on the surface of the sample is equal to the value of the demagnetizing field due to the continuity of the tangential components of the magnetic field. In an ellipsoid this point is given by the point of intersection of the surface and the plane normal to the direction of magnetization of the ellipsoid and led through the centre of the ellipsoid. By measuring the magnetic field in points on this line on the surface of the ellipsoid, we can then determine the value of the demagnetizing field by direct measurement. It is obvious that in reality we cannot measure the field directly in a point but only in a finite volume, and then such measurements will only be approximate. Very good accuracy can be achieved by measurements on ellipsoids, the curvature of which in the place measured is small in the direction of the vector of magnetization (i.e. either an oblate prolate spheroid, magnetized normal to the axis, or a prolate spheroid magnetized along the axis).

The actual measurements are made as follows: in the case of metallic ferromagnets the ellipsoids form part of the wall of the resonance cavity and in the case of insulators in the form of prolate ellipsoids they can be placed completely or partly right into the measuring cavity. For such measurements insulators may of course be covered with a thin metal film and treated as conductors. We should like to point out that the EPR signal is not covered by a strong signal of ferromagnetic resonance since the influence of high-frequency demagnetization in most cases shifts the value of the magnetic field by ferromagnetic resonance outside the EPR region. We then measure the intensity of the EPR field without the ferromagnet (or calculate it from the known value of the g-factor) and then at the same frequency measure the intensity of the EPR magnetic field of the material located on the surface of the ferromagnet. The value of the demagnetizing field on flat electrolytically polished rotational elipsoids of permalloy alloys measured by this method agreed with the values calculated magnetostatically from the value of the saturation magnetization with an accuracy of 3%. It should be noted that this method can be used to measure the value of the saturation magnetization of ellipsoids if their demagnetizing factors are known.

Stray Fields on Ferromagnet Surface and Ferromagnetic Resonance Curve Width.

When determining the demagnetizing fields by the above method we observed not only a displacement of the EPR but also a clear widening of its resonance curve when the DPPH element was brought nearer to the surface of the ferromagnet [5]. It is thus obvious that strays fields exist on the surface of a ferromagnet, which widen the EPR curve. Since it is difficult to bring the DPPH crystal ideally near to the ferromagnet surface, we measured the widening of the curves on thin films formed by depositing a solution of DPPH in benzene directly on the ferromagnetic surface. Layers of DPPH, 4 mm in diameter and 1 to 10 μ thick, were placed in the middle of thin (circa 0.1-0.3 mm) electrolytically polished ferromagnetic discs 25 to 30 mm in diameter, which formed part of the wall of micro-wave resonators. For alloys of iron and nickel in a range of 30 to 70% the DPPH curve widening (originally $\Delta H_{\text{infl}} = 1.8$ Oe) was up to 70 Oe. When the DPPH crystal was moved over the surface of the ferromagnet the EPR field only changed in the limits of $\pm$ 6 Oe, since the crystal detects the field only at a certain distance from the surface. It is obvious that the mean value of the intensity of local fields on the surface of a ferromagnet measured in this way is given as the mean of the values over the whole volume of the DPPH layer and that the mean value thus obtained is merely a rough picture of the dispersion of the values of the magnetic field in the surface layer of a ferromagnet. However, the value is remarkable and proves that the distribution of magnetization, at least in the surface layer of a ferromagnetic ellipsoid, is not homogeneous even in a strong external field (here approximately 3200 Oe). Thin films of DPPH located on non-ferromagnetic surface give the original curve width. On the other hand, the EPR width of a DPPH element increases very much (over 100 Oe) when the element is brought closer to a longitudinally magnetized ferrite polycrystalline cylinder (Mn-Mg ferrite), and when applied to the surface it can no longer be detected.

Let us now give the cause which we think are important for the creation of stray fields in the surface of a ferromagnet. Firstly, these may be defects in the surface, e.g. scratches and microscopic flaws. Secondly, there may exist impurities and inclusions, clusters of dislocations, particularly on the edge of the grains of the polycrystalline material. In a polycrystal small deviations occur in the direction of magnetization owing to the different values of the effective field given by the magnetic anisotropy and magnetostriction. Finally, any kind of inhomogeneity of stress in the sample causes the vector of magnetization to deviate from the homogeneous direction (e.g. the mean value of the stray fields for electrolytically polished material annealed before measurement is half that of non-annealed material for a 50% permalloy).

Interesting results are obtained by comparing the value of the stray fields in the surface layer of a ferromagnet, obtained by this semi-quantitative method, with the width of curve of ferromagnetic resonance absorption. It is obvious [6] that inhomogeneity of the magnetic field inside the ferromagnet contributes to a widening of the resonance curve. Of course, such a rough comparison can only be made for the case of ferromagnetic resonance in metals, where the depth of penetration and the thickness of the DPPH layer on the surface of the ferromagnet is approximately the same (circa 1 μ), and when it can be assumed that the distribution of the stray fields does not change very much at the depth of penetration. Another reason for speaking only of a semi-quantitative comparison is the possibility of interaction between the regions of the ferromagnet of different intensities of the internal magnetic field (e.g. the resultant curve may narrow due to dipilar interaction [7]). Secondly, the ferromagnetic resonance width is influenced not only by static stray fields but scattering also occurs with high-frequency fields, which will be influenced by the above-mentioned inhomogeneities of the sample.

Let us now give a few cases of comparison of the EPR curve width of a thin DPPH film on the surface of a ferromagnetic disc with the ferromagnetic resonance curve width measured on the same region of the sample. On the left of Fig. 1 we see the ferromagnetic resonance curve, i.e. a quantity proportional to the square root of the imaginary part of the effective permeability ($\mu_R^{\frac{1}{2}}$) as a function of the magnetic field strength; on the right we have the EPR curve of a DPPH film about 4 μ thick for 50% permalloy. The mean value of the DPPH film width for this material for several measurements is 30 Oe, the ferromagnetic resonance curve width is 65 Oe. If we add the theoretical values of the ferromagnetic

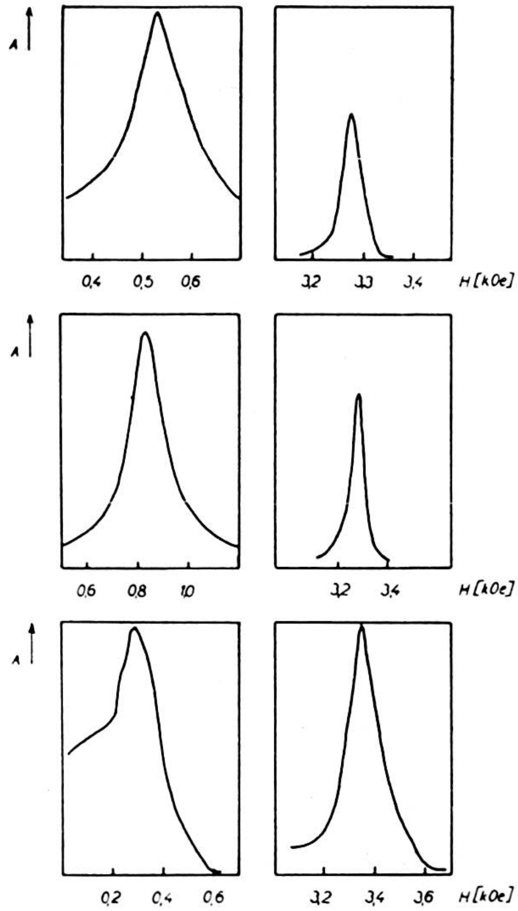

FIG 1
FIG 2
FIG 3

resonance curve width due to skin-effect [8] (20 Oe) and the polycrystalline property of the material (max. 6 Oe) and also the measured mean value of the stray fields, we obtain good agreement with the ferromagnetic resonance curve width. It must be emphasized that this agreement was obtained on the uncertain assumption of the simple addition of widths, caused by different mechanisms. For the analogous case (Fig. 2) of a 78% Ni permalloy the half-width due to skin-effect is 25 Oe and due to the polyscrystalline property is 9 Oe, the EPR width on the surface of the sample is 44 Oe; the agreement with the mean value of the ferromagnetic resonance curve width (95 Oe) is again good. For a metallic single crystal (Fe with 2.5% Si) in the shape of a disc in the (100) plane and in the direction of the static field at an angle of about 10% from the [010] direction (Fig. 3), the EPR curve width is roughly 230 Oe, the ferromagnetic resonance curve width is about 300 Oe. The influence of the skin-effect is about 40 Oe. It is seen that for a macroscopic metallic single crystal, where the ferromagnetic resonance curve width is usually large, the EPR curve width measured on its surface forms a large part of the value of the ferroresonance curve. We have not yet been able to explain the cause of this high value of the stray field. In the case of stray fields measured on thin ferromagnetic films the EPR curve width for a film of 78% Ni permalloy, 3000 Å thick and with a ferroresonance curve width of 30 Oe, is 8 Oe.

It should also be noted that in the above comparisons we were inconsistent in that we measured the dispersion of the surface fields at a value of the static magnetic field around 3200 Oe, while the actual ferromagnetic resonance occurs in lower fields for the frequency used. It follows from other measurements [9], however, that the curve width of metallic ferromagnets having a small value of the magnetic anisotropy changes very little for a change in the measured frequency. The case is different for materials having a large value of the magnetocrystalline anisotropy, where we get double resonance curves [10] in lower fields in a single crystal. Even for this case, however, the value determined by us is an estimate for the value of the dispersion of local internal fields.

From the experiments made up to now it is inferred that the stray fields in the surface layer of a ferromagnet greatly influence the curve width of metallic ferromagnets, particularly with macroscopic single crystals. This conclusion is confirmed by the fact that the curve width of metallic ferromagnets with a small number of defects (e.g. for whiskers [2, 11]) is given by the theoretical value, given by the exchange interaction at the depth of penetration, and that for thin films, where there is no such mechanism, the curve width will fluctuate in units of Oe [12].

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