APPROACH TO MAGNETIC SATURATION AND THE MAGNETIC MOMENTS OF NUCLEI

Amikam Aharoni
Department of Electronics, The Weizmann Institute of Science, Rehovoth, Israel
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Large angles between the local direction of magnetization and that of the applied magnetic field, which have been found in experiments involving the embedding of excited nuclei in Fe or Ni foils, are explained in terms of the crystalline imperfections caused by the foreign nucleus when it penetrates the ferromagnetic lattice. A rather crude estimation yields the same results for line or point defects, and it is concluded that the sine of these angles should decrease, with increasing magnetic field H, roughly as $1/H$.

Many experiments have shown[1] that in a magnetic field of the order of 100 Oe, soft magnetic materials like Ni or Fe are almost completely saturated in the direction of the applied field. Introduction of lattice imperfections by plastic deformation[1] or by neutron irradiation[1,2] increases considerably the portion of the magnetic material magnetized at some angle to the applied field, but this is still a very small percentage in a field of several hundred oersteds. On the other hand, Ben-Zvi et al.[3] have found that the magnetic field at the site of nuclei recoiling into Fe or Ni foils is at a rather large angle to an applied magnetic field in the plane of the foil of[4] about 1000 Oe. The measured angles (in degrees) were 36 ± 2 for ^{186}W in Ni, 30 ± 6 for ^{186}W in Fe, and 24 ± 6 for ^{148}Nd in Fe.[3] Assuming that the field at the site of the embedded nucleus is proportional to the magnetization of the Fe or Ni lattice (which is the usual conclusion[5] from Mössbauer-effect experiments), this observation seems at variance with the magnetic measurements. Moreover, the experiment of Ben-Zvi et al.[3] indicates a concentration of the magnetization in the direction of the generating vector of a cone making the angle of 30° or so with the direction of the applied magnetic field without any magnetization lying parallel to the applied field, which seems very strange even for fields much smaller than 1000 Oe.

These apparent discrepancies are removed if one bears in mind the fact that the recoil nuclei in the Ben-Zvi experiment[3] not only introduce imperfections into the Fe or Ni lattice, as is the case in the usual study[1,2] of radiation damage, but actually come to rest in such an imperfection. These nuclei therefore measure the magnetization of the ferromagnetic material at the relatively few imperfections only, whereas macroscopic magnetic measurements[1,2] yield the average of the magnetization in these lattice sites and in most of the material where the magnetization is more inclined towards the direction of the applied magnetic field. This experiment thus serves as a kind of magnification of what is usually a small effect, or rather as a direct measurement of the amplitude of a deviation, for which other measurements yield only the spatial average, in the effect of imperfections on the approach to magnetic saturation. This interpretation of the experiment will be made somewhat more quantitative in the following.

The effect of imperfections on the magnetization in its approach to saturation was first studied by Brown[6] in general terms of concentration of forces, using very simple geometrical configurations. Later extensions[7-9] to more complicated force fields at line or point defects retained the basic assumptions that the magnetization is almost parallel to the applied field and therefore neglected all terms which were of second order in the transverse component of the magnetization. This could be justified when one was interested only in an average over a large crystal, for which case an error in a small region near the defect itself could not contribute much. The problem is quite different when one is interested in the direction of the magnetization vector in the close vicinity of the imperfection, where it seems very doubtful if the linearized theory is dependable. The reason is that in that vicinity the linearized theory yields angles between the magnetization and the applied field which are not small and which vary very rapidly,[10] so that one cannot justify neglecting second-order terms in this vicinity. Besides, even if the results of the linear theory were dependable in principle, they are not easy to obtain from most of the published calculations[8,9] because a Fourier transformation is introduced at a very early stage, so that only an average over an infinite material is calculated. In fact, for many applications[11] it turns out[12] that one can ignore the dislocation structure and replace it (as far as the average over the whole

sample is concerned) by an effective anisotropy term. Obviously, such an approach cannot help solve the problem that concerns us here.

However, for a rough estimation one need not go into any fine details. Just consider the high concentration of forces which must exist near the imperfection. These might be due to dislocations created by the recoil nucleus, or just result from the mechanical strains caused by the very existence of an impurity atom which has a different "size." In any case, one can approximate the actual spatial dependence of the forces f_1 and f_2 by assuming that they have some constant values inside a small region R of unspecified shape surrounding the point or line imperfection. The forces then decrease outside this region, more or less abruptly. Let

$$\lambda^2 = HM_S/C, \tag{1}$$

where H is the applied magnetic field, M_S is the saturation magnetization of the material, and C is the exchange constant. In the context of the linearized theory,[6] the direction cosines α and β of the transverse magnetization and the magnetostatic potential u should be the solutions of

$$(\nabla^2-\lambda^2)\alpha - \partial u/\partial x = f_1, \tag{2a}$$

$$(\nabla^2-\lambda^2)\beta - \partial u/\partial y = f_2, \tag{2b}$$

$$\nabla^2 u = \frac{4\pi M_S^2}{C}\left(\frac{\partial\alpha}{\partial x}+\frac{\partial\beta}{\partial y}\right). \tag{2c}$$

It is readily seen that in the region R, where f_1 and f_2 are assumed to be nonvanishing constants, these Eqs. (2) have the particular solution

$$\alpha = -f_1\lambda^{-2}, \quad \beta = -f_2\lambda^{-2}, \quad u = 0, \tag{3}$$

and the solution outside this region, or the boundary conditions there, do not make much difference in the present context.

This is not quite rigorous because usually a solution of the homogenous Eqs. (2) has to be added to (3) in order to fulfill the conditions of continuity on the boundary of the region R. However, it seems that neglecting this correction is not less justified than the basic assumptions of this theory, at least when the magnetostatic self-energy is not too large. One can, therefore, use (3) as an approximation for the magnetization in the near vicinity of the foreign atom and deduce from it, using (1), the following expression for the angle θ between the magnetization and the direction of the applied field;

$$\sin\theta = (\alpha^2+\beta^2)^{1/2} \propto H^{-1}, \tag{4}$$

which is a result that should be checked by experiment.

In a way, the treatment presented here is very similar to the older treatment of the approach to saturation against which Brown[6] argued. However, the argument of Brown applies to the use of solutions like (3) for an average over the whole crystal, for which the assumption of constant f_1 and f_2 cannot be justified. Here it was used only for a small region surrounding the defect, and for this region the model must be at least as good as any used in the theory of the approach to saturation so far.[10]

In this calculation the magnetocrystalline anisotropy has been neglected. Moreover, contamination layers originating from the pumping system, and heating by the bombarding particles, are known[13] to cause mechanical stresses in the foil, which must be equivalent on the average[11] to an extra anisotropy. However, the effect of the anisotropy decreases more rapidly, as H^{-2}, and can therefore be neglected for fields where (4) is still important. In fact, it has been shown that for a polycrystalline ferromagnet approaching saturation, the magnetization varies on the average as

$$M = M_S(1-cK^2H^{-2}M_S^{-2}), \tag{5}$$

where K is the anisotropy constant (for iron, for example, $K/M_S = 280$ Oe), and c is a slowly varying function of H, which can be taken as approximately 0.1, for both iron and nickel.[14] Using (5), for H of at least 500 Oe the contribution of the anisotropy to the magnetization is at most a few percent for Ni or Fe, even if a reasonable value is assumed[13] for the contamination layer. One can thus neglect the anisotropy as was done in (2).

It should be particularly emphasized that the result obtained here is not sensitive to the number of the defects in the sample or to their average distance, which usually[15] affects strongly the functional dependence on the field H of macroscopic magnetic measurements which involve averaging over the whole sample. It is also insensitive to the shape of the defect, or to the force field around it. Moreover, for the particular solution (3), even the use of the linear theory can be partly justified. This is seen by comparing (2) with the nonlinear Brown equations[16] according to which (3) remains a solution in the region just around the defect, even when one introduces the rigorous nonlinear expressions for the exchange and magnetostatic energies. The magne-

tocrystalline anisotropy energy has been neglected anyway, and although the justification for dropping this term is based on the linear approximation, the average for defects in different crystallites cannot be much different than in the calculation of Holstein and Primakoff[14] (i.e., besides the local strong variations, which have been approximately taken into account). One is thus left with the magnetostrictive energy term, which essentially gives rise to the right-hand side of (2a) and (2b). But for this energy term, the linear approximation is used only in a small region near the defect, and in a small region almost everything is linear.

It is very difficult to estimate the proportionality constant in (4), and therefore it is not quite clear if the theory presented here can actually account for the 30° or so measured[3] experimentally. However, some deviations from saturation were measured in Ni in fields up to 6000 Oe,[17] so that rather large angles at the peak, in a field of about 1000 Oe, sound plausible. But, of course, it would be nicer to check (4) by repeating the measurements of Ben-Zvi et al.[3] in various fields H larger than 500 Oe. Another check could come from repeating the Low and Collins experiment[18] of scattering neutrons by impurities in Fe or Ni in a larger field. The field applied in this experiment was not measured, but the sample magnetization was[19] $95\pm 5\%$ of the saturation magnetization, which does not sound like a very high field. If our theory is correct, the angle θ of (4) should have been quite large in this experiment near the impurity atoms, i.e., where it mainly counts.

Because of the particular way in which measurements are subtracted,[18] an angle of 30° should change the measured cross sections by a factor of 2, and this factor tends to infinity for θ (or α, in their[18] notation) tending to 45°. One should, therefore, expect quite a large change in repeating the measurements in larger fields. There is also a possible contribution from diffuse magnetic scattering since the absence of these was checked by comparing with the pure element, disregarding the possibility that the pure element was saturated while the alloy was not. All the conclusions[20] from such measurements might, therefore, have to be modified if the experiment is done in a larger magnetic field.

This might also be connected to the depolarization of neutrons, which has been found[21] to be quite large in magnetic fields for which the deviation from saturation is already very small. The common interpretation of this effect is[22] in terms of magnetocrystalline anisotropy in the different crystallites. However, the essence of the interpretation is the different regions of size δ, which is much smaller than the size d of the sample, and this can be readily adopted to our picture of small regions, near impurities or other imperfections, where the magnetization deviates considerably from the field direction.

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CORIOLIS INTERACTION BETWEEN THREE NILSSON BANDS IN Pa

W. Hoekstra
University of Technology, Delft, The Netherlands

and

A. H. Wapstra
University of Technology, Delft, and Institute for Nuclear Physics Research, Amsterdam, The Netherlands

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By comparing new experimental results with calculations, it is shown that the levels above the $\frac{1}{2}^-[5,3,0]$ ground-state band in ^{233}Pa and ^{231}Pa may be interpreted as resulting from the $\frac{1}{2}^+[6,6,0]$, the $\frac{3}{2}^+[6,5,1]$, and the $\frac{5}{2}^+[6,4,2]$ bands involved in a three-band Coriolis interaction.

Interpretation of several states between 80 and 400 keV in ^{233}Pa as due to a three-band Coriolis interaction was first suggested by Browne and Asaro.[1] New information just available from measurements of the ^{237}Np α and ^{233}Th β^- decays[2,3] will be presented here as strongly confirming this hypothesis. Also, we present results of a calculation showing that the very uncommon deformation of one of the observed rotational bands can be caused by a three-band Coriolis interaction, with very acceptable values for the interaction parameters.

In Fig. 1 the alpha feeding of the levels in ^{233}Pa is shown.[4] The α hindrance factors indicate that the 86.4- and the 238.2-keV levels (level energies as following from the present work) are fed by unhindered alpha transitions and therefore contain considerable percentages of the Nilsson level $\frac{5}{2}^+[6,4,2]$, assigned to the ^{237}Np ground state.[5]

In an earlier proposal the 86.4-keV level was interpreted as the $\frac{5}{2}^+[6,4,2]$ state and the 238.2-keV level as the $\frac{5}{2}^+$ member of the $\frac{3}{2}^+[6,5,1]$ rotational band, mixed with the former state by Coriolis interaction. The failure of this model to explain the very low values of the hindrance factors of the alpha transitions to the 212.4-keV level and to the remarkable doublet at 103.7 and 108.5 keV led Browne and Asaro to the new model mentioned above.

In the meantime, our experiments have shown that a level at 94.6 keV is fed by an allowed, hindered or a first-forbidden, unhindered (ah or 1u) beta transition from ^{233}Th. This 94.6-keV level decays to the ground state, to the 6.7-keV level (Fig. 2), and more strongly, by low-energy conversion electrons,[6] to the 86.4-keV level. We found this level to be in coincidence also with the alpha transitions feeding the 103.7- and 108.5-keV doublet; this explains the lack of gamma-ray transitions from this doublet to the $\frac{1}{2}^-[5,3,0]$ band. Taken together these findings make it at-

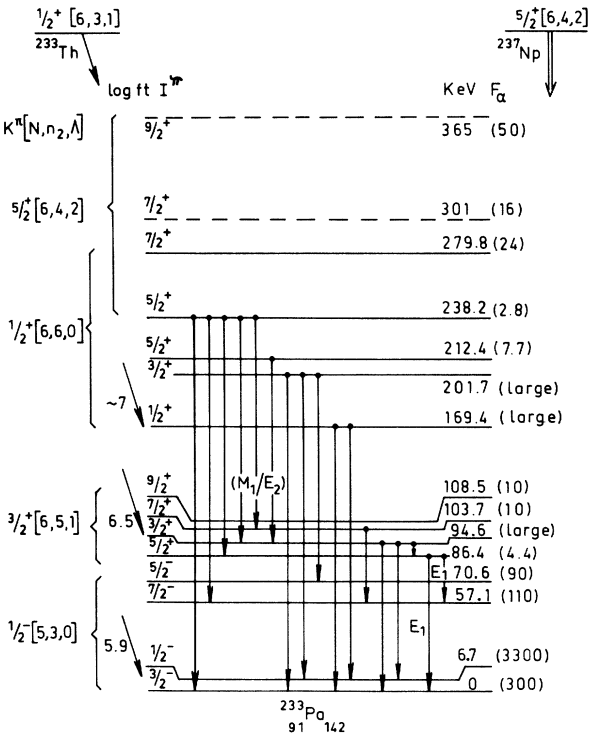

FIG. 1. Level scheme of ^{233}Pa as found from the decays of ^{233}Th and ^{237}Np presented as far as of interest for the accompanying discussion. The hindrance factors F_α refer to the α decay of ^{237}Np.