**Smoluchowski, R.**: The main problem is to explain the similarity between the resistivity anomaly in alpha and delta plutonium which have different crystal structures and 20% different densities. I feel that the antiferromagnetic model (Friedel and Rocher 1960) is untenable because of lack of magnetic or neutron diffraction evidence. On the other hand the interband scattering model (Smoluchowski: 1962) requires only that the $5f$ level forms a narrow band superposed on a broad conduction band. Only the latter would be appreciably altered on changing the lattice structure.

**Mendelssohn, K.**: I quite agree that the Friedel-Rocher model appears inapplicable but I also think that an unusual band structure may be changed considerably when going from the monoclinic $\alpha$-phase to the f.c.c. $\delta$-phase. We have investigated $\delta$-Pu where the accumulation rate is $\frac{1}{4}$ of that in $\alpha$-Pu. I have at this stage no suggestion to make concerning the nature of the co-operative phenomenon involved but the existence of an unfilled lower shell somehow suggests some type of magnetic ordering.

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# The Interaction between Irradiation-Induced Defects and Magnetic Structure

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Hysteresis loop measurements have been conducted on several commercial iron-nickel alloys during neutron irradiation at 90°K and ambient reactor temperature in both zero and saturating magnetic field. Isochronal annealing tests were also performed on the alloys after irradiation. The low temperature irradiation results indicated that magnetic properties were affected not by the point defects themselves but by their motion and eventual disposition. The radiation-induced changes appeared similar to those observed in these alloys upon magnetic annealing, *i.e.*, short-range atomic ordering. However, in all cases the radiation-induced changes were annealed out at temperatures below the usual ordering temperature. This recovery process appeared to be dependent upon the state of magnetization of the sample.

## I. Introduction

It is well known that the magnetic properties of iron-nickel alloys, having a nickel concentration of 50% or more, can be significantly influenced by thermal treatment or magnetic annealing[1),2)]. The effect of such treatment is usually greatest in alloys in which the sum of all the magnetic anisotropies is small. It is considered that magnetic annealing introduces an additional uniaxial anisotropy that tends to increase the remanence and, hence, the squareness of the hysteresis loop.

Many models have been proposed to explain the resultant properties, the most recent being based upon short-range directional ordering proposed by Néel[3] and Taniguchi[4]. This model must be considered along with the discovery of Heidenreich and Nesbitt[5] concerning the necessity for the presence of a minimum amount (about 0.001%) of oxygen which promotes the formation of stacking faults along (111) planes during magnetic annealing. In all previous annealing experiments on soft magnetic materials, as well as in the present investigation, sufficient oxygen or other impurities probably were present.

When iron-nickel alloys are irradiated with neutrons, significant modification of their properties has been found. Paulevé *et al.*[6] noted a large increase in magnetic anisotropy when iron-nickel (50-50) was irradiated in a magnetic field by neutrons. Schindler *et al.*[7] studied changes in the hysteresis loops of iron-nickel alloys upon neutron irradiation. Irradiation in the demagnetized condition generally results in an increase in coercive force, $H_c$, in a decrease in remanence, $B_r$, and in a constricted or kinked hysteresis loop. On the other hand, irradiation in a saturating magnetic field results in a more rectangular loop with increases in both coercive force and remanence. The similarity of these latter results to those occurring for similar alloys thermally annealed in the absence or presence of a magnetic field, taken in conjunction with the Néel-Taniguchi short-range directional order model, suggests[7] that neutron irradiation induces short-range directional ordering. The production of ordinary short-range order by irradiation has already been found and studied in non-magnetic alloys by other investigators[8),9)].

According to the Néel-Taniguchi model, the internal field acting on the diffusing atoms causes anisotropic migration which results in local or short-range directional ordering of like-atom pairs. In the case of thermal annealing, the iron-nickel alloys must be heated to the vicinity of 500°C, presumably to create a sufficient number of thermally activated defects to promote diffusion. Neutron irradiation, on the other hand, can create defects at any temperature. However, these irradiation-induced defects will not promote diffusion unless the temperature of irradiation is high enough for them to be mobile. The role of the prerequisite trace amount of oxygen is not understood, but possibly there may be some relationship between any impurity atoms, such as oxygen, and induced defects which acts to promote diffusion.

Although previous experiments have been interpreted on the basis of directional ordering, changes might be expected to result from the interaction between irradiation-induced defects and magnetic domain walls. In fact, an interaction between aligned divacancies and domain walls has recently been proposed by Klein and Kronmüller[10].

In order to study the effects of irradiation-induced defects on magnetic properties, 60-cycle hysteresis loop measurements were made on a number of magnetic materials before, during, and after neutron irradiation.

## II. Experimental Procedures

Five different commercial iron-nickel alloys which had been prepared to have optimum magnetic properties, *i.e.*, a minimum hysteresis loop area, were selected for the irradiation experiments. The alloys were in the form of 2 mil tape, 1/8 inch wide, wound into toroids with an outer diameter of 3/4 inch. The toroids had the following nominal compositions:

No. 1 79% Ni—16% Fe—5% Mo
No. 2 48% Ni—52% Fe
No. 3 78% Ni—15% Fe—5%Cu—2% Cr
No. 4 47% Ni—50% Fe—3% Mo
No. 5 43% Ni—54% Fe—3% Si

The irradiation facility used in the Oak Ridge Graphite Reactor had a fast flux of about $4.5\times10^{14}$n/cm$^2$/hr and could be maintained at low temperatures (~90°K). Arrangements were made so that hysteresis loops could be obtained on each of the samples, in succession, while they were being irradiated. After irradiation, the toroids could be withdrawn and annealed in a lead-shielded furnace.

*Experiment 1. Low Temperature Irradiation*

Toroids of alloys 1—5 were irradiated in the demagnetized state for seven weeks at ~ 90°K (integrated fast flux of about $5\times10^{17}$ n/cm$^2$). After irradiation, the samples were removed from the neutron flux, and the temperature was gradually increased to 10°C during a period of five hours. The samples were then kept at room temperature for three weeks, after which isochronal annealing studies were made. Hysteresis loops were made during the irradiation, during the warm-up, and during the subsequent "isothermal" room temperature anneal. For the isochronal studies the samples were heated for one hour periods at temperature intervals of 25C° from room temperature up to a maximum temperature of 450°C. Hysteresis loop measurements were made at the elevated temperature and again at room temperature following each step anneal. Similar unirradiated control samples were also subjected to the isochronal anneals, and subsequent measurements on the control samples indicated essentially no change in their room temperature magnetic properties as a result of the thermal treatment.

*Experiment 2. Ambient Temperature Irradiation*

Irradiations were performed for 11 weeks at 70°C±15°C in the low temperature facility which could also be operated at ambient reactor temperature. The integrated fast flux was about $7.5 \times 10^{17} n/cm^2$. Pairs of toroids (1′, 2′) having the same composition as 1 and 2 of the previous experiment, as well as an additional pair of permalloy toroids (79% Ni—17% Fe—4% Mo) were irradiated. One sample of each pair was irradiated in the demagnetized state while the other member of the pair was irradiated in the presence of a D.C. magnetic field which resulted in technical magnetic saturation. Hysteresis loops were made during the irradiation. After the irradiation isochronal annealing studies, similar to those conducted in the first experiment, were made up to a maximum temperature of 550°C. During these anneals the magnetic state of each sample was maintained as it had been during the irradiation, *i.e.*, the demagnetized samples were kept demagnetized, and the magnetized samples were kept saturated. The samples were always returned to 70°C for hysteresis loop measurement.

## III. Results

Typical results of the low temperature experiment are illustrated in Figs. 1 and 2. The room temperature measurements of $B_r$, following the isochronal annealing steps for both the low temperature irradiated samples and unirradiated control samples, are shown in Fig. 1. This treatment left the unirradiated control specimens relatively unaltered. (Consequently, only the initial and final values of $B_r$ for the control samples are shown, and are depicted with horizontal lines drawn through the symbols.) The two data points for each sample at 25°C are the values of $B_r$ before and after the "isothermal" room temperature anneal. It can be seen that the remanence decreased during the isochronal anneals until annealing was performed in the vicinity of 350°C. Further anneals above this temperature caused $B_r$ to approach the pre-irradiation value. Similar variations but of opposite direction, *i.e.*, increases instead

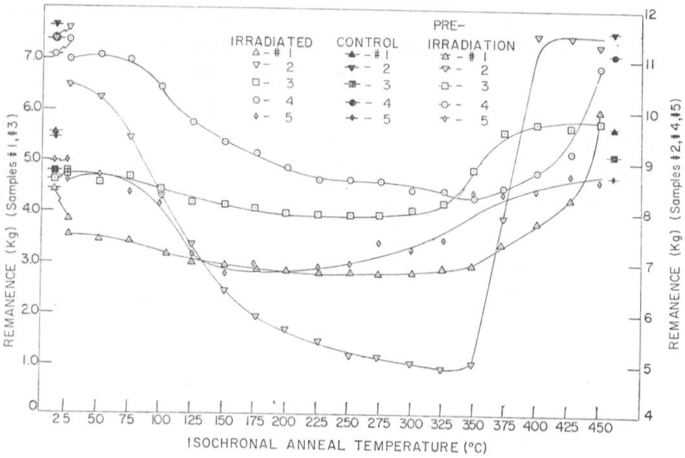


Fig. 1. The effect of isochronal step anneals on the magnetic remanence of samples irradiated at 90°K. All measurements were made at 25°C. The sample compositions are given in the text.

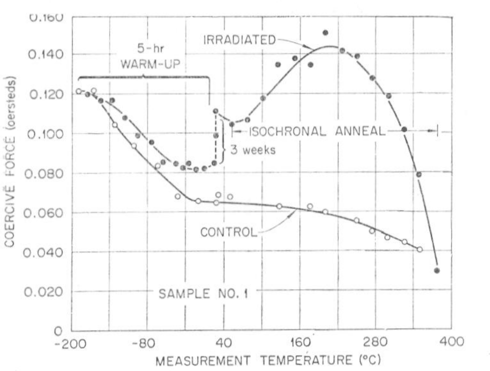


Fig. 2. The variation of the coercive force during warm-up and isochronal annealing of sample No. 1 following 90°K irradiation compared with that of a similar unirradiated sample.

of decreases, were found in values of the coercive force upon isochronal anneal.

Fig. 2 shows the variation of the coercive force with temperature for Sample No. 1 in the irradiated and unirradiated state. The values to the left of the dashed line were taken during the warm-up to room temperature; the dashed line represents the variation during the room temperature anneal; the values to the right of the dashed line represent the values measured at the anneal temperature near the end of the one hour anneal. It should be noted that the largest difference between the irradiated and unirradiated state occurred in the vicinity of 200°C.

The results of the low temperature irradiation experiments can be summarized as follows:

1. Irradiation at 90°K does not affect the shapes of the hysteresis curves measured at 90°K.

2. In all cases, the coercive force increased and the remanence decreased during room temperature anneals and during the initial isochronal anneals.

3. Both $H_c$ and $B_r$ reached extreme values after isochronal anneals in the vicinity of 350°C; further isochronal anneals at higher temperatures caused these parameters to tend to return to their pre-irradiation values.

Typical results of the ambient temperature irradiation (~70°C) and isochronal annealing studies are shown in Figs. 3 and 4. Significant changes occurred during the irradiation, and the variation in $B_r$ and $H_c$ is shown as a function of time of irradiation on the left-hand portion of the curves. The right-hand portion of the curves depict the values of $B_r$ and $H_c$ taken at 70°C following each isochronal annealing cycle at the temperature indicated. The solid symbols represent the variation of the parameters for the samples treated in the magnetized state, while the open symbols are those of the samples treated in the demagnetized state.

The results of the ambient temperature (70°C) irradiation experiment can be summarized as follows:

1. Both $B_r$ and $H_c$ increased as a function

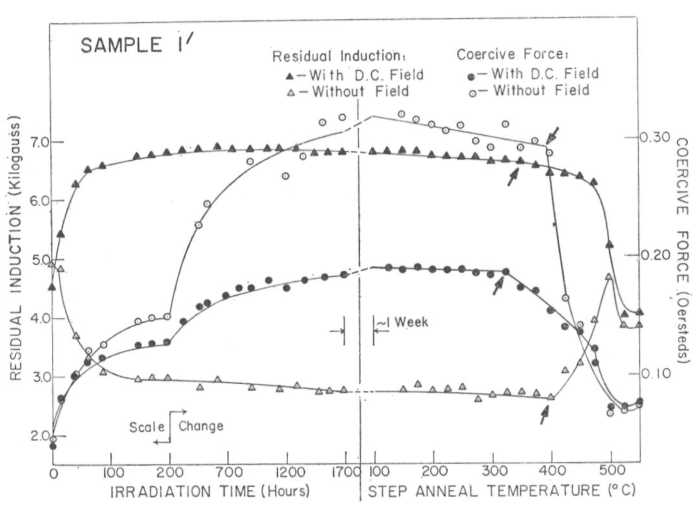


Fig. 3. Remanence and coercive force values for Sample No. 1′ as a function of irradiation time (left) and isochronal anneal (right). All measurements taken at ~70°C.

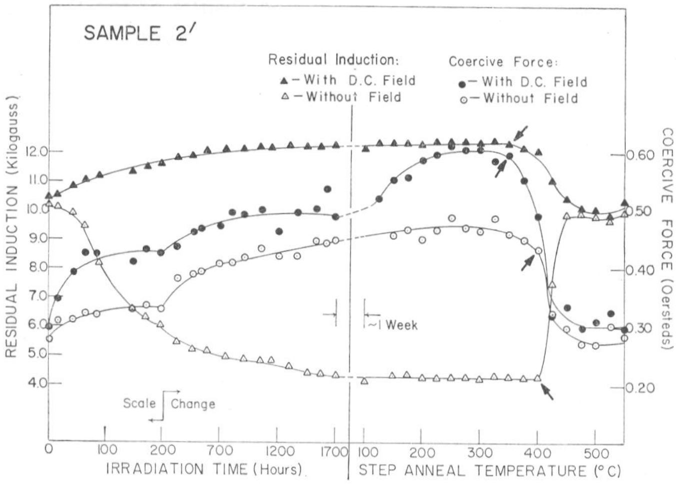


Fig. 4. Remanence and coercive force values for Sample No. 2′ as a function of irradiation time (left) and isochronal anneal (right). All measurements taken at ~70°C.

of time of irradiation for all the samples irradiated in the magnetized state, *i.e.*, rectangular hysteresis loop characteristics were exhibited.

2. For samples irradiated in the demagnetized state, $B_r$ decreased and $H_c$ increased as a function of irradiation time.

3. In all cases the initial variation of $B_r$ and $H_c$ during irradiation was rapid but later appeared to be asymptotically reaching a limiting value.

4. Beyond some critical annealing temperature, the 70°C value of both $B_r$ and $H_c$ approached their pre-irradiation values. There was an indication that this critical temperature is approximately 50°C lower for the samples annealed in the magnetized state.

## IV. Discussion

Although these measurements have been made upon commercially obtained samples in which the knowledge of certain parameters, such as composition and initial state of disorder, are not accurately known, it seems clear that certain conclusion can be made. The defects produced by the neutron irradiation are quite immobile at liquid nitrogen temperatures. This can be ascertained from the fact that both $B_r$ and $H_c$ are essentially unchanged by the low temperature irradiation. However, during the warm-up to room temperature, the room temperature isothermal anneal, and the beginning of the isochronal annealing treatment, the mobility of the defects increases and defect migration proceeds. It is therefore clear that changes in $B_r$ and $H_c$ are not caused by isolated irradiation-induced defects, *per se*, but by their movement and concomitant enhancement of a diffusion process. Ostensibly, the initial variation of $B_r$ and $H_c$ during the post-irradiation treatments is similar to the variation of these same parameters as a function of increasing short-range ordering. In an experiment which will be reported on more fully at a later date, an activation energy of 0.30 eV was obtained for this radiation-induced ordering process. In the experiment performed at ambient temperature, the defects produced are mobile and similar changes of $B_r$ and $H_c$ occur during the irradiation.

The results of the isochronal annealing studies are more difficult to interpret. In particular, it is noted that following the low temperature and ambient temperature irradiations, $B_r$ and $H_c$ for all the samples begin to revert to their pre-irradiation values after an isochronal anneal at temperatures which are considerably below the usual order—disorder temperature (490°C). Paulevé *et al.*[6] have suggested the possibility of an irradiation-induced ordered state for NiFe with a critical temperature of 330°C. This might explain the observed reversion found here for samples of composition 2, but cannot be used as an explanation for the behavior of the other samples. In addition, a further small decrease in the critical anneal temperature seemed to be indicated for samples irradiated in the magnetized state. The observed annealing behavior suggests some tvpe of interaction between defect clusters and magnetic structures.

Two possible explanations appear plausible for the observed annealing behavior. It should be noted that reversion occurred at annealing temperatures close to the Curie temperatures of the samples. This suggests that a possible interaction between defect clusters and the internal magnetic field results in anisotropic defect configurations. Such configurations could induce a magnetic anisotropy which would modify the hysteresis loops. At annealing temperatures above the ferromagnetic Curie temperature, this interaction ceases and irradiation-induced defects assume isotoropic configurations or disappear entirely.

Secondly, an interaction between defect clusters and domain walls is proposed that would, in addition, explain the small difference in critical anneal temperature which appears between demagnetized and magnetized samples. In the domain walls, magnetostrictive stresses exist which can aid in the nucleation of dislocation loops production by a precipitation of uniformly scattered point defects. A calculation has been made which indicates that these stresses are of the order of $10^8$ dynes/cm$^2$. Near any inclusion within a domain wall, the usual stress concentration effect will raise this stress to $10^9$ dynes/cm$^2$. Since it is difficult to prepare high melting point material without small inclusions of foreign matter, one can reasonably expect large mechanical stresses to exist within domain walls which are comparable to the chemical stress produced by the existence of non-equilibrium concentration of point defects. The combination of the mechanical stress and the chemical stress can lead to preferential nucleation of dislocation loops within domain walls.

The resultant dislocation loops grow in size by absorbing excess point defects from within the domains. In the demagnetized alloys (with many domain walls), a larger number of nucleated dislocation loops compete for the point defects, and both the growth rate and average size of the loops at the elevated temperature will be reduced. The optimum size of the dislocation loop necessary for the pinning of domain walls, of the order of the domain wall thickness, will therefore occur at a higher temperature for the sample treated in the demagnetized state. In both mechanisms suggested, the reversion of magnetic properties upon thermal annealing would not be based upon a disordering model.

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## DISCUSSION

**Chikazumi, S.**: You assumed that the stresses induced by magnetostrictive strain inside domain wall may cause preferential nucleation of vacancy collapse loops. If this is true, we should expect that the effect should depend upon the magnitude of magnetostriction of the specimen which you have used. Do you have any experimental evidence for this?

**Schindler, A.**: The effect should depend upon the magnetostriction constants of the samples. However, the resultant modifications of the magnetic properties would depend upon other physical properties of the material, *i.e.*, intensity of magnetization, magnetic anisotropy. We have not made the examination you suggest. Perhaps the best check of the mechanism we propose would be a direct examination of the defect collapse loops by means of electron microscopy. We hope such an examination will soon be made.

**Okada, K.**: I'm very interested in your research, because there are similar changes of properties after irradiation to those of radiation damaged *ferroelectric* crystals.

According to our experiments on ferroelectric Rochelle salt crystals irradiated by X-rays, the most remarkable changes of properties are as follows: that is, ferroelectric hysteresis loops of a multi-domain crystal or a single-domain crystal change into a double loop or a biased single loop respectively, dielectric constants are reduced, the Curie point is shifted to lower temperatures, stabilization of domain structure, that is, domain wall would not move until strong external fields 2 or 3 times larger than ordinary coercive fields are applied, and so on.

May I ask you whether you observed changes of the magnetic properties corresponding to these changes of ferroelectric substances or not?

**Schindler, A.**: We have in fact found similar modifications in magnetic properties. In particular we have found large increases in the magnetic coercive force which would indicate stabilization of domain walls resulting from the neutron irradiation. We have also found a constricted hysteresis loop after irradiation in the demagnetized state, which superficially resemble your double loop. However because of the dissimilar nature of metal alloys and Rochelle salts, I would hesitate to suggest that the model we propose for the modification of magnetic properties in alloys would also apply to X-irradiated Rochelle salt.

**Brandon, D. G.**: Can you tell us what the metallographic structure of these alloys is before irradiation and whether there are any ordering effect with the heat treatments you use to prepare the samples?

**Schinder, A.**: We have not made metallographic examinations of the samples. Some X-ray measurements we made indicate that the samples all have the face centered cubic structure, both before and after irradiation. They are obtained commercially, and have received the standard treatment for optimum magnetic properties. This includes a quick cool from above the critical ordering temperature. Presumably the samples are in the disordered state prior to irradiation.