A SUMMARY OF THE INTERNATIONAL DISCUSSION MEETING ON RADIATION EFFECTS ON SUPERCONDUCTIVITY

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INTRODUCTION

The effects of radiation on the properties of superconductors was the subject of an International Discussion Meeting held at Argonne National Laboratory, June 12-16, 1977, and was attended by 80 scientists from 7 countries. The program focussed on radiation effects on reversible and irreversible properties, fundamental fluxoid-defect interactions, radiation considerations for fusion magnets, and ion implantation in superconductors.

It is well known that the properties of a superconductor depend on the arrangement of the atoms in a crystalline solid. It is possible to change the positions of the atoms, i.e., introduce defects, by irradiating the superconductor with various particles. The changes in the superconducting properties depend on the type of defect structure introduced, which in turn depends on the type and energy of the irradiating particle, the irradiation temperature, and the characteristics of the target material. Radiation is a convenient method to study the role of defects on the superconducting properties since size and number density can be introduced in a controlled fashion, and the role of defects can be investigated without changing the chemical composition. Although research has been conducted in this field for fifteen years, this was the first meeting on this topic and was held at this time due to the increased interest in A-15 compound superconductors and the superconducting composites that will be used in fusion magnets.

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The sessions began with an invited talk that summarized the knowledge in an area, and was followed by contributed papers on current research. All of the papers will appear in the proceedings of the meeting in the February, 1978 volume of the Journal of Nuclear Materials and all unreferenced statements are implicitly made to those proceedings. In this brief article, I will mention some of the recent advances in the field as they were discussed at the Argonne Meeting. In the summary I will mention the major topics of discussion and speculate on useful areas of investigation for the future. As a useful introduction to this field, the reader is referred to recent reviews and conferences on superconductivity [1-3] and radiation damage [4,5].

FUNDAMENTAL INTERACTIONS

After an introductory talk on radiation damage in metals, the subject of fundamental fluxoid-defect interactions was addressed. This problem consists of three steps:

(1) calculate the fundamental fluxoid-defect interaction for the defect structure of interest,

(2) sum appropriately these interactions to get a volume pinning force density F_p

(3) compare this F_p to the experimental value that can be directly obtained from a measurement of the critical current density J_c.

As was vigorously discussed at an earlier flux-pinning conference [3], step (2) was and continues to be the source of considerable controversy. In many different types of experiments discussed, the experimentally determined F_p is an order of magnitude or more above the theoretical F_p calculated using the Labusch summation. This summation utilizes the fact that the lines of magnetic flux exist in the form of a flux line lattice. There is a stiffness to the lattice, which is represented by an effective elastic constant, which does not allow the full pinning force of each defect to be utilized. The experimental value is below the value found by a direct summation of the individual point pins that clearly overestimates F_p. The direct summation essentially says that a flux line can be pinned fully at every defect center. Thus, this summation completely ignores the well-known lattice nature of the flux lines. Figure 1 is a plot of the experimentally determined specific pinning force (F_p divided by the number density of defect structures) versus the calculated maximum fluxoid-defect interaction f_p in Nb for various types of defects. The predictions of a direct summation and the Labusch statistical summation are indicated. That the experimental values lie below the direct summation, yet above the Labusch summation, shows that the lattice nature of the flux lines is important but over estimated by Labusch. A promising idea being pursued that would resolve the problem invokes the use of defects in the flux line lattice. This would retain the lattice nature of the flux lines, but would result in increased pinning due to the enhanced flexibility of the flux lines lattice because the defects in the lattice would lower the elastic constants.

The effect of voids on flux pinning in Nb and V was the subject of recent investigations. This system is especially appealing since the size and number distribution of the voids can be controlled by the irradiation parameters and characterized by electron microscopy. Therefore, the parameters required for calculating the fundamental interaction are available directly.

Of particular interest in these studies is the observation of a regular void lattice that results in enhanced pinning which is attributed to a matching of the void and fluxoid spacing.

RADIATION EFFECTS ON REVERSIBLE PROPERTIES OF SUPERCONDUCTORS

An area that generated considerable discussion and apparent disagreement was the nature of the defect in the A-15 compound superconductors. It has been known that the metastable A-15's are very sensitive to radiation. Figure 2 shows the effect of fast neutron irradiation at 140 C on the critical temperature (T_c) of various A-15's [6]. The values of the unirradiated T_c (T_{co}) are in the range of 18 to 23 K. Previous neutron diffraction experiments have correlated this decrease in T_c with a decrease in the long-range order parameter S. In this A_3B crystal structure a sample is less than fully ordered (i.e. $S < 1$) when A atoms are on B sites or conversely. This disordering idea is appealing since the enhanced superconductivity of the A-15's have been attributed to the orthogonal chains of A atoms in the (100) direction. Disordering would result in B atoms on A sites which would reduce the integrity of these chains and, presumably, reduce T_c. In this connection, it has been shown that stoichiometry plays a vital role in determining T_c in Nb_3Ge. A compound at other than the 3:1 ratio (and, therefore, intrinsically disordered) has a less than maximum T_c [7]. The data for various A-15's could be plotted on a master plot of $T_c/T_{c\ max}$ versus dose with the role of stoichiometry being represented by an equivalent dose for each compound. In contrast to the A_3B compounds where B was a nontransition element, was Mo_3Os where both Mo and Os are transition elements. This showed very little sensitivity to irradiation indicating that it is the presence of a nontransition atom in the A chain that is required for T_c degradation.

Work involving α particle irradiation of A-15's has led to different interpretation of the defect responsible for the T_c degradation. T_c decreased with dose and saturated i.e., showed no further decrease with dose, a fact subsequently observed for neutron irradiation. Measurements of the absolute structure factors in x-ray diffraction experiments in Nb_3Sn indicated large static displacements of both the Nb and Sn atoms from their equilibrium positions. The displacements of ∿0.2 Å result in bond-bending distortions and can be viewed as resulting in a buckling of the chains of atoms. Further work on channeling in α-damaged V_3Si has supported this view. There appeared, however, to be some disagreement on the procedure whereby the large displacements of the atoms were unfolded from the original channeling data. It is also necessary to determine if these measurements are primarily probing the surface as compared to the bulk neutron diffraction measurements.

A number of recent investigations were reported that support the disordering model. The drop in T_c was shown to occur simultaneously with an increase in the normal state resistivity (ρ) during 20 K fast neutron irradiation. An annealing or recovery stage was seen at room temperature for both T_c and ρ, indicating defect motion, and presumably, reordering at this temperature. An extension of this work to large doses showed that both ρ and S varied exponentially with dose, as predicted by earlier theoretical work. Neutron irradiated Nb_3Sn was viewed on superlattice reflections to reveal the presence of highly disordered regions ∿35 Å in size in a much less disordered matrix. The observed T_c decreases in this highly inhomogeneous sample were explained by the proximity effect.

The T_c decreases in the A-15's were fit by a model that invoked a smearing out of the anisotropy of the energy gap. In this and in other discussions, it was observed that annealing experiments should be very helpful in probing the nature of the defects responsible for the T_c decreases in the A-15's. During isochronal annealing, a temperature is reached where the defect is mobile and it either annihilates or agglomerates. The new state of the defect will be reflected to a different extent in the different properties, e.g., ρ, T_c, channeling or x-ray intensity. Therefore, comparison of the change in two or more properties during production and annealing should give information about the defects. Various investigations have studied in detail the annealing behavior of T_c, S and lattice parameter after fast-neutron and fission-fragment irradiation of V_3Ga, Nb_3Sn, Nb_3Al and Nb_3Ge. At this early stage, uncertainties still exist in trying to assign the motion of a specific defect or activation energy to an annealing stage, but considerable detail in the annealing is observed up to 900°C. Disagreement exists in that not all experiments observe steps or stages in the annealing curve. Also, experimental disagreement exists concerning the presence of a threshold at low doses during production, i.e. some experiments saw no T_c decreases until a threshold dose had been reached.

Low temperature oxygen ion irradiations of various elemental superconductors show a minimum in the T_c versus dose curve, and subsequent increase in T_c at higher doses. In Nb this decrease is attributed to the decrease in the energy gap anisotropy with the increase attributed to strain (which changes the electron-phonon coupling) resulting from defect agglomeration.

CRITICAL CURRENT CHANGES IN ALLOY AND COMPOUND SUPERCONDUCTORS

A more difficult area to understand fundamentally is the effect of irradiation on the critical currents of alloy and compound superconductors. Since the critical current (or flux pinning) depends on the geometrical arrangement of the defects and not just the density or electronic mean free path (that T_c apparently depends on), there is a large role played by the various parameters such as sample type and metallurgical condition, irradiating species and energy, and irradiation temperature. The initial talk on radiation damage in metals is particularly relavent here since the defect configuration (point defect, cascade, loop, void) is very important when determining flux pinning (as it is for determining f_p for the elemental superconductors). Many of the results in this area are primarily useful for predicting the results of irradiation. For example, irradiating heavily cold-worked NbTi at 4 K with fast neutrons to a dose of 4 x 10^{18} n/cm^2 will result in a T_c decrease of about 10% [8]. The difficulty lies in explaining the results which in this case is in terms of a decrease of the effectiveness of the pinning by the cell walls due to an increase in defect density within the cell cores.

Information can be derived from experiments that vary a single parameter in otherwise identical irradiation experiments. The larger increases at low doses in Nb_3Sn irradiated with fast-neutrons at 6 K versus irradiation at 140 C indicate larger flux pinning by defect cascades than defect clusters or loops. Comparing 6 K irradiations on Nb_3Sn with low and high values of the unirradiated J_c shows flux pinning by cascades can raise low J_{co} material to the values of the high J_{co} material. This is not the case in similar experiments on cold-worked and annealed NbTi which indicate that the cascades are much weaker pins than the cell walls. Furthermore, investigations on 30 GeV p^+ irradiations at 4 K indicate ∿70% recovery in the observed J_c decreases in NbTi, but very little recovery in Nb_3Sn, in agreement with fast-neutron irradiations.

A careful investigation of 27 K fast-neutron irradiation of V_3Ga showed a large sensitivity of T_c, $H_{c2}(T)$, $[dH_{c2}/dT]_{T=T_c}$, and F_p to irradiation, as has been observed in fast-neutron irradiated Nb_3Sn. The interpretation is difficult because the pinning force does not obey a scaling law. In Nb_3Sn H_{c2} was observed to increase at low doses, whereas the V_3Ga studies deduced a decrease in H_{c2}. The role of H_{c2} changes in J_c changes is an area requiring more effort. In connection with the fact that the increase in J_c with irradiation is a function of the unirradiated J_c, a model was discussed that compares the maximum pinning force before irradiation to the mean energy transferred by the irradiation to a lattice atom producing displacements. It was predicted that for samples above an initial J_c value, no further enhancement would be possible with irradiation.

RADIATION CONSIDERATIONS FOR FUSION MAGNETS

The penetration of γ-rays and neutrons from the plasma of a fusion reactor through the blanket and shield make it necessary to consider radiation damage to the magnet components in designs of superconducting magnets. The components that are particularly sensitive to radiation are the superconductor, stabilizer, and insulators. Figure 3 is a cut-away view of a fusion reactor showing the various magnets and shielding. Of particular concern to the designers is the increased flux at portions of the magnet due to the various instrumentation or beam injection ports. A comprehensive calculation has been performed on the shielding-radiation flux trade off for the magnet under various conditions expected for tokamak power reactors. The radiation level is translated into property changes which are then put in terms of total magnet performance and economics. The results show that the superconducting fusion magnets can operate at a much higher radiation level than previously anticipated. There is, however, a need for better data on the effects of radiation at low temperatures on the various magnet components due to the fact that any uncertainties in the data require an overestimation in the shielding requirements with the resulting economic penalties. Since there is no low temperature neutron irradiation results for the insulators, the shield-magnet economics were minimized with no insulator input, which is clearly a serious emission but required by the lack of data.

Calculations were performed on the field obtainable with presently available stabilized superconductors and structural materials. For cryogenically stabilized magnet systems and for neutron doses expected for shielded toroidal magnets, the change of ρ of the stabilizer is the limiting component in the maximum attainable magnetic field. A different calculation shows that this conclusion is changed for Nb_3Sn magnets at high doses, for which the T_c degradation becomes the dominant effect.

A brief description was given of the low temperature irradiation facility of the proposed pulsed neutron source to be built at Argonne National Laboratory. Discussion of this facility is timely since it is anticipated that within a few years the presently existing low temperature neutron irradiation facilities in this country will be shut down.

An experimental paper related to fusion magnets discussed the effect of 14 MeV neutron irradiation at 4 K on T_c of NbTi. Eventhought most of the 14 MeV neutrons will be severely energy degraded after the shield is penetrated, there will be $\sim$5% of the neutrons with energy in this regime. The decrease in J_c with dose agrees with fast neutron results, but the decrease per neutron is a factor of 37 larger. However, more work is required since J_c changes with dose are very sensitive to the unirradiated J_c. Approximately 70% of the J_c decrease recovers after a room temperature anneal, similar to fast-neutron irradiations.

ION IMPLANTATION AND SUPERCONDUCTIVITY

Due to the metastable character of many superconductors, ion implantation has proven to be advantageous for producing various types of superconductors. Ion implantation is especially appropriate for investigating concentration dependent properties, since the concentration can be systematically varied on the same sample and the measured changes attributed to the implanted atom. By this method it is possible to exceed the equilibrium solubility limits, and a number of new superconductors have been made by ion implantation.

The introduction of lattice disorder by the implanting ion or even the possibility of driving the sample amorphous must be considered in analyzing the experiments. In this connection, lattice disorder was attributed to the increase in transition temperature to 4.54 K for In implanted in ions at 4.2 K. Also, the greatly enhanced T_c's in Al films resulting from disordering is attributed to a softening of the phonon spectrum. The ion implantation experiments frequently yield similar results to quench condensation and splatt cooling experiments, since disorder can be introduced by these methods also. However, ion implantation offers the distinct advantage of being able to carefully control and vary the level of damage or impurities.

Experiments have not been very successful in trying to enhance T_c of A-15 compound films by ion implantation due, in part, because the ion induced damage hinders the formation of the metastable A-15 phase. In Nb-Ge films, a T_c of only 8 K has been attained compared to the chemically vapor deposited value of 23 K.

Channeling was used to investigate the role of C ion irradiation in the transition metal carbides with the B1 structure. The damage produced during the irradiation was analyzed and it was found that T_c can be enhanced by annealing samples that were made stoichiometric by ion implantation. Also, T_c was found to be fairly insensitive to extended defects such as stacking faults and twins. Similar investigations in Nb and V indicate that the T_c changes are caused by disorder introduced by the implanting ion and stabilized by chemically active impurities (O or N).

SUMMARY

The purpose of organizing a discussion meeting on radiation effects on superconductivity was to bring researchers in the field together for the first time in order to actively discuss the present status and future direction of our field. The organizers feel and many of the participants have indicated, that this purpose has been very satisfactorily fulfilled. The stimulating discussions among contributors and scientists from Argonne involved in radiation damage research hopefully has brought closer together the two somewhat diverse areas of radiation damage and superconductivity. However, it is felt that many of the ideas and models in the highly developed field of radiation damage are not being utilized, as they should be, in analyzing radiation damage in superconductors.

A very encouraging aspect of the meeting was the discussion that centered around the research tools being applied to this area for the first time. Whereas in the past the measurements have primarily been J_c, T_c, ρ, magnetic susceptibility, ac losses, and x-ray diffraction, talks were given on the recent application to this area of channeling techniques, Mössbauer spectroscopy, specific heat, and more detailed x-ray measurements. It was also very evident that the most meaningful experiments were those that measured two or more properties on the same sample as a function of irradiation dose and post-irradiation annealing. It was pointed out that annealing experiments should be helpful in trying to understand the defects responsible for the changes in the various superconducting properties.

The areas requiring the most extensive future research are:

(1) The nature of the defect responsible for the dramatic T_c decreases in A-15 compounds (specifically disorder versus large static displacements) and the role of these defects vis-a-vis other defects present in determining flux pinning changes.

(2) The summation problem, i.e., the proper way to sum individual defect fluxoid interactions (primarily in elemental superconductors) in order to compare to the experimentally determined volume pinning force density.

(3) More accurate data on the effect of liquid helium temperature neutron irradiaion (fission and 14 MeV) on J_c of commercially important composite superconductors (primarily NbTi, but also Nb_3Sn and V_3Ga).

(4) The application of ion implantation in producing superconductors with enhanced properties.

Acknowledgments

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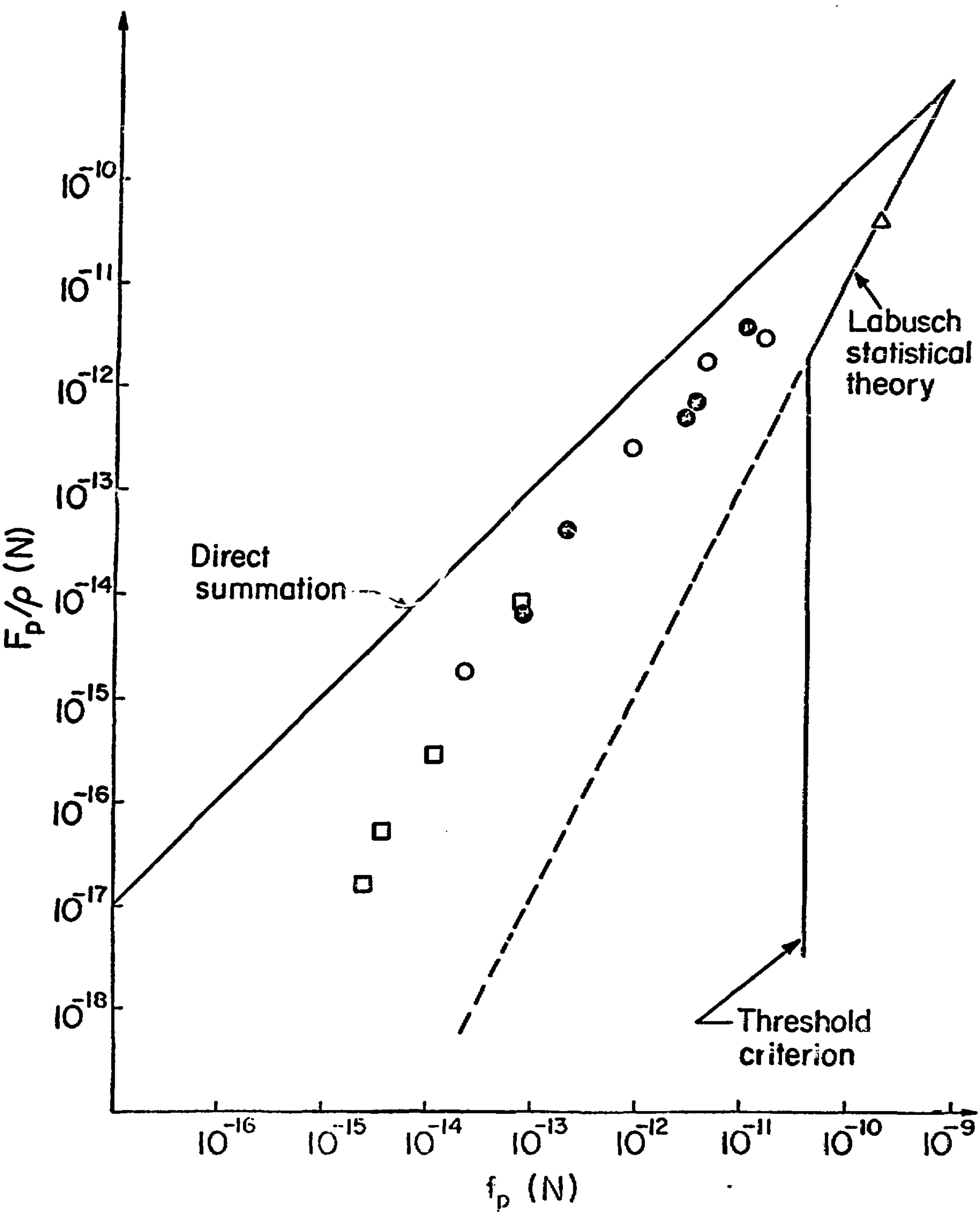
Figure Captions

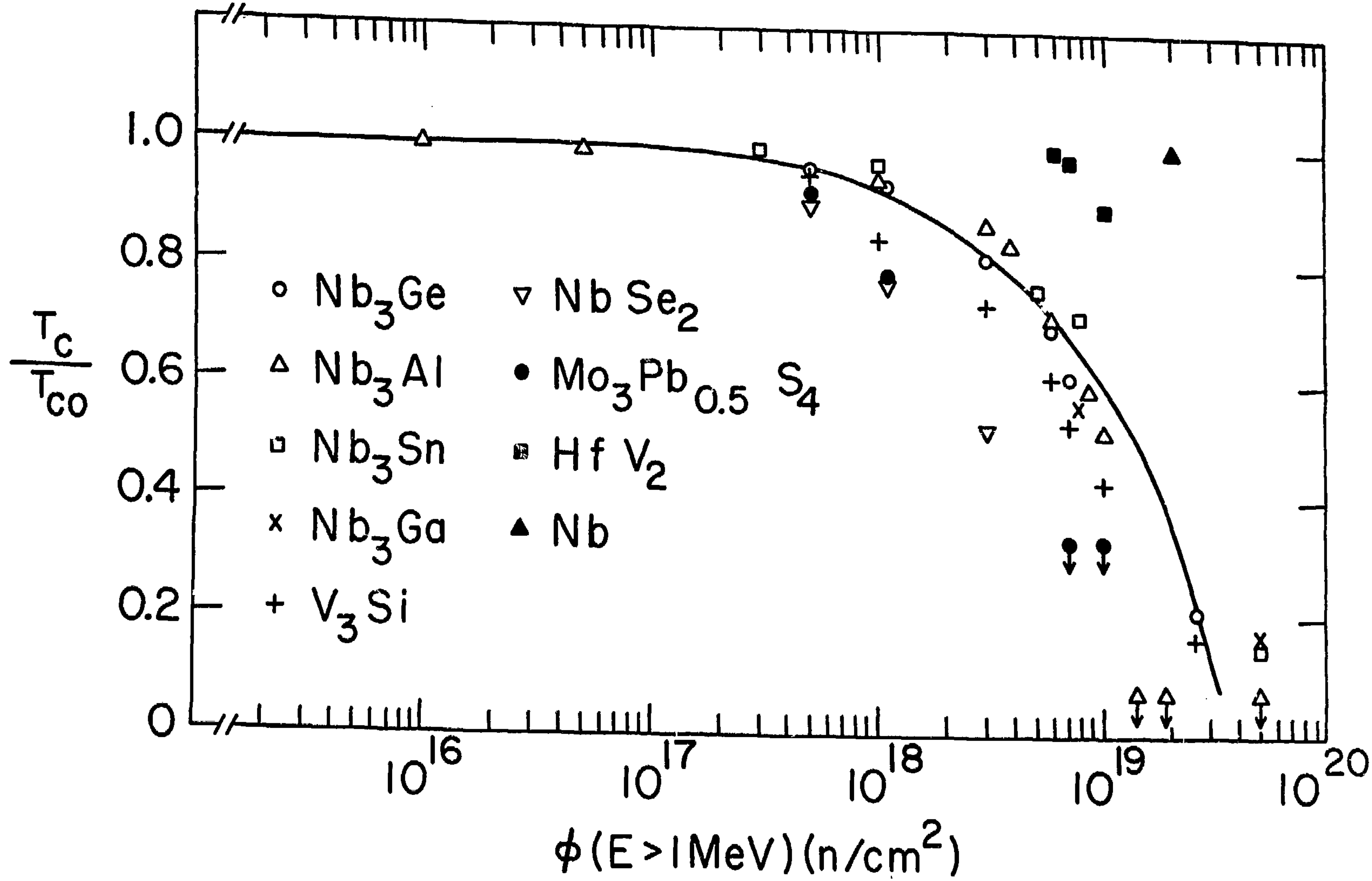
FIG. 1. The specific pinning force $F_p/\rho(N)$ (defect density) vs elementary interaction force F_p for defects in Nb. Data are for pinning by dislocation loops (□), voids due to irradiation by neutrons (O) and Ni^+(●), and Nb_2N precipitates (Δ). Also shown are the predictions of the Labusch statistical summation and a direct summation. (Figure from Kramer).

FIG. 2. Reduced transition temperature as a function of fast neutron dose for various superconductors [6].

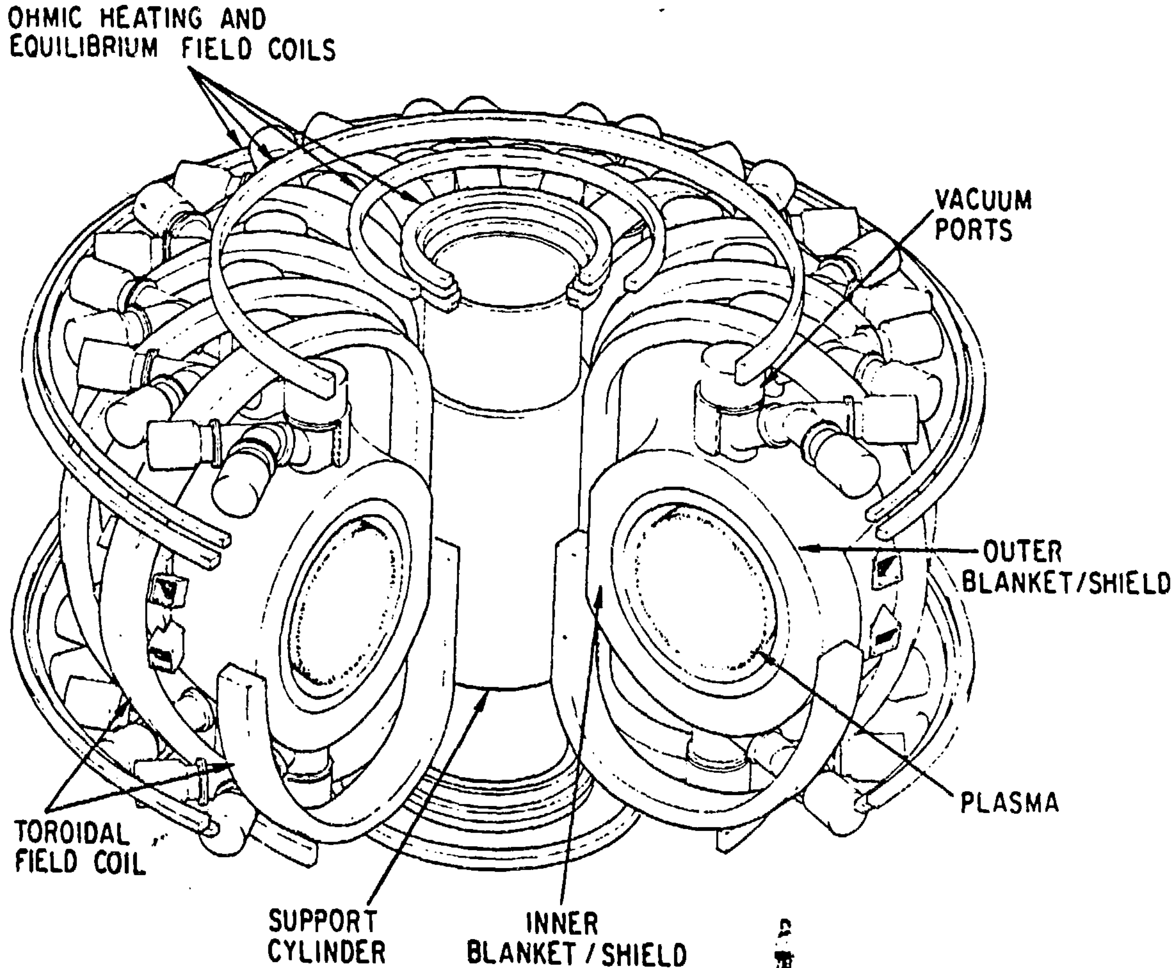
FIG. 3. Cut-away view of tokamak fusion reactor (Figure from Abdou).



F_p/ρ (N)
10^{-10}
10^{-11}
10^{-12}
10^{-13}
10^{-14}
10^{-15}
10^{-16}
10^{-17}
10^{-18}
Direct summation
Labusch statistical theory
Threshold criterion
10^{-16}
10^{-15}
10^{-14}
10^{-13}
10^{-12}
10^{-11}
10^{-10}
10^{-9}
f_p (N)



1.0
0.8
0.6
0.4
0.2
0
Tc/Tco
Nb3Ge
Nb3Al
Nb3Sn
Nb3Ga
V3Si
Nb Se2
Mo3Pb0.5 S4
Hf V2
Nb
10^16
10^17
10^18
10^19
10^20
φ(E>1MeV)(n/cm^2)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