SUPERCONDUCTING COATINGS FOR ACCELERATING RF CAVITIES : PAST, PRESENT, FUTURE

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ABSTRACT

About 200 superconducting RF accelerating cavities of 352 MHz frequency, each consisting of four cells, are required to upgrade the LEP energy from an initial value of 55 GeV to about 90 GeV. About 170 of these cavities, which have already been ordered from European industry, will be made of copper and coated internally with a thin layer of niobium. The coating will be carried out by sputtering using a recipe developed at CERN. A brief history of this development which began in 1980 is presented and the main advantages of this solution are illustrated. Some possible lines of further improvement are discussed.

1. WHY NIOBIUM COATED CAVITIES ?

Superconducting RF cavities are usually made of niobium metal sheet by welding together half cells produced either by deep drawing or lathe spinning [1]. At the end of the 70s, the accelerating field which could be obtained in Nb cavities was limited by quench. At high accelerating fields, the enhanced power absorption by localised resistive surface defects may be sufficient to drive the neighbouring superconductor into the normal state, resulting in a sudden dissipation of the stored RF power (quench). The level of power absorption a cavity may withstand without quenching is directly dependent on the thermal conductivity of the cavity wall. Therefore, great efforts have been devoted to improving the Nb purity, in order to increase its thermal conductivity at liquid helium temperature [2].

An alternative solution to this problem consists in replacing Nb with copper as the cavity construction material. High purity copper presents, at 4.2 K, a thermal conductivity an order of magnitude higher than the Nb of high purity (RRR = 300) presently available on the market. Obviously, the cavity must be coated internally with a superconducting film, which will present a lower thermal conductivity. However, the thermal impedance of the film is negligible because only a very thin coating has to be applied. Due to the Meissner shielding currents, the penetration of the electromagnetic field in a superconductor is confined to a very thin superficial layer.

For instance, a 1 μm thick Nb film is largely sufficient to shield completely the underlying more resistive copper.
Another interesting feature of coated cavities is that the independent functions of the coating and of the substrate allow a wider choice of superconducting materials, because the latter does not have to provide the otherwise essential thermal and mechanical properties.
Finally, an advantage which has been fully appreciated when the order for LEP cavities was placed consists in the important saving on the cost of Nb.

2. DEVELOPMENT OF COATED SUPERCONDUCTING CAVITIES

A development programme was started at CERN in 1980 aiming at the production of coated superconducting RF cavities. To start with, Nb was chosen as coating material.
The coating was performed by sputtering initially making use of a bias sputtering configuration [3]. The cathode consisted of three electrodes rotating around the axis of the cavity. The electrode geometry was designed to produce a coating of uniform thickness on the cavity walls (see Fig. 1). The bias voltage was applied to the cathode shields. For a film thickness of about 1 μm at the equator, the duration of the coating process was of the order of 15 hours [3].
About 15 single-cell 500 MHz cavities were produced by the end of 1984, demonstrating that accelerating fields and Qo values comparable to those of bulk Nb cavities could be obtained, together with the expected stability with respect to quenching [4,5]. During this period it was also found that extreme surface cleanliness prior to cavity coating is mandatory to ensure good film to substrate adhesion and that UHV criteria must be rigorously applied to obtain films of good quality, i.e. RRR values of the order of 10 to 15.
In spite of the reasonably good RF performance obtained, the developed sputtering configuration suffered from three main inconveniences. First, the coating time was too long for subsequent industrial production. Second, its extension to a four-cell cavity structure was problematic ; during coating, heating results in cathode elongation which in turn applied large forces on the ceramic insulators which link the cathode to the shield. This problem is obviously aggravated with increasing cathode length, as required for multicell structures and may result in the ceramic breaking and loss of the cathode electric insulation. Finally, rotation of the cathode produced microparticles, which could be buried in the growing film resulting in surface defects.
In addition to these problems, the thickness profile obtained was highly inhomogeneous (film four times thicker at the iris than at the equator) in spite of the ad hoc cathode shaping. This feature, not detrimental for Nb, would severely endanger the feasibility of some

other promising coatings, such as NbN and NbTiN which may be produced by reactive sputtering (see par. 5).

To overcome all these inconveniences, a new electrode design, based on a cylindrical magnetron sputtering configuration, has been developed in 1984. The cathode consists of a cylindrical stainless steel tube inserted along the cavity axis and tightly linked to the cavity via a ceramic insulator (see Fig. 2). This cylinder is surrounded by a Nb liner (which acts as the Nb source during sputtering) and contains a solenoid electromagnet. The stray field of the magnet protrudes into the volume of the cavity and confines the discharge plasma by trapping the ionising electrons. Thanks to the enhanced ionisation efficiency of the trapped electrons, the sputtering rate is increased and the discharge argon pressure may be reduced by about two orders of magnitude (from the high 10^{-2} to the high 10^{-4} mb pressure range). This last feature improves film adhesion by reducing the energy lost by Nb atoms via gas collisions during their transfer to the cavity wall and, furthermore, minimises the risk of argon (and impurities) trapping in the film during growth.

Another advantage of the cylindrical magnetron configuration is that uniform film thickness may be achieved in spite of the very dissimilar geometries of the cathode and of the cavity. This is possible because the emission of sputtered atoms is reduced at low angles (cosine law), i.e. in the direction of the cavity iris which however is closer to the cathode than the equator. By properly choosing the diameter of the cathode and the length of the magnet, the opposing effects of distance and emission angle may compensate each other, resulting in a film of uniform thickness.
Finally, the extension from a single-cell to a four-cell structure is trivial in this case (see Fig. 3) and the coating time of a complete LEP cavity is reduced to 4 hours. A detailed description of the adopted cylindrical magnetron configuration may be found in ref. 6.

Single-cell 500 MHz cavities coated by magnetron sputtering displayed better performance than both diode sputtering coated cavities and bulk Nb cavities (see Fig. 4). A similar improvement has been found [6] on four-cell cavities of the LEP type (see Fig. 5). Furthermore, coated cavities are insensitive to any magnetic field present while cooling (at least as far as fields of the order of magnitude of the earths magnetic field are concerned), a feature which makes magnetic field shielding and/or compensation unnecessary [7]. Both the higher Qo values and the insensitivity to external magnetic fields are important advantages not anticipated when the coating development programme was decided.
In spite of all these good features, coated cavities still suffered from an important degradation of the initial Qo value when increasing the accelerating field. This degradation was such as to considerably reduce, at fields of practical importance, the benefit of the higher Qo at low

field. To eliminate this problem, a careful optimization of the coating parameters was carried out on single-cell 500 MHz cavities.
Up to June 1988, about 20 such cavities were coated by magnetron sputtering under different conditions. It was found [8] that Qo degradation could be reduced by increasing the coating temperature. The best results were obtained by producing the coating at 200ºC (see Fig. 6), a temperature which should not be exceeded in order to avoid the risk of cavity collapse. A similar benefit of the higher coating temperature has also been noticed for LEP type cavities.

Measurements on samples produced inside a single-cell 500 MHz cavity revealed a close correlation between cavity performance and RRR values of coatings, which may reach 25 when produced at 200ºC and 34 at 400ºC [8].

3. FROM LABORATORY TO INDUSTRIAL SCALE PRODUCTION

Since 1988, the main effort has been directed to ascertain the industrial feasibility of Nb coated cavities. This implies a precise definition of all production steps such as the insure reproducibility of quality. A decisive move in this direction has been the decision by LEP management to produce at CERN a small series of coated LEP cavities (12 in total).

This decision served many purposes, namely :

a) to install coated cavities on a CERN accelerator, such as to obtain information on the long term performance in a real operating environment ;
b) to check the reproducibility of performance on a statistical basis ;
c) to allow interested Companies to follow the complete production cycle and in so doing to learn enough about this new technology to make a reasonable cost estimate.

The results obtained are summarised in Fig. 7. They have confirmed the expected quality level and show that a reasonable reproducibility may be obtained, whenever the standardised procedures are strictly applied [9].
After testing, 8 cavities have been assembled in two cryomodules each consisting of four cavities. A first cryomodule has been installed in LEP at the beginning of 1990 and the second one during the summer 1991 [9].
On the ground of these results it has been decided to proceed with the industrial production of coated cavities for LEP 200, according to more stringent specifications (Qo = 4×10^9 at 6 MV/m, instead of Qo = 3×10^9 at 5 MV/m as specified for bulk Nb cavities). Three orders for about 50 LEP cavities each have been placed with three different European

Companies at the end of 1990. Testing of prototypes produced by these Companies is planned for the end of 1991.

4. IS A FURTHER QUALITY IMPROVEMENT POSSIBLE ?

Operating LEP cavities at accelerating fields even higher than 6 MV/m would be highly desirable. However, since the power dissipation in the cavity increases at least as the square of the field, operation at higher field would only be feasible if accompanied by a corresponding improvement in Qo. A practical limit would be imposed otherwise by the available cooling power. Therefore, both E max. and Qo must be increased to achieve a real benefit for the LEP 200 project.

4.1. Field limitations

As stated in the previous paragraphs, coated cavities do not quench at least up to the maximum accelerating fields obtained so far (about 15 MV/m). In the absence of quenching, the field limitation is a consequence of electron field emission.

To the best of our present knowledge, field emission seems to result from a variety of accidental events rather than to be a fundamental limit [2]. Consequently, it should be possible to postpone it at higher fields by taking more stringent precautions concerning cleanliness at all steps of the production process. This conclusion is valid for all types of cavities and this aspect of the problem will not be further discussed here.

A peculiar aspect of coated cavities is instead the absolute cleanliness of its surface after coating. This feature is so obvious that initially no rinsing was foreseen after coating. However, the very first few cavities produced presented poor performance, which improved remarkably after rinsing. Therefore this treatment has been standardised for all cavities produced subsequently. However, meanwhile the coating conditions have changed. On the one hand, more precautions are taken during handling of the cavities and, on the other hand, the fixed magnetron cathode has removed the risk, experienced with diode sputtering, of film contamination by metal particles produced during or after coating (see par. 2).

The usefulness of rinsing after coating has therefore again been questioned and some cavities have been measured without rinsing. The first results have shown that very high fields may be reached in this case (up to 15 MV/m for single-cell 500 MHz cavities) provided that after coating the cavity is not exposed to air outside a class 100 clean room. This implies demounting the cavity from the coating system with its insulation valve closed and demounting both the cathode and the valve in the clean room.

In other cases, when this precaution has not been taken, poor Qo values and low field were obtained which have improved to standard values after rinsing.

A similar test (no rinsing and clean demounting conditions) carried out on LEP cavities did not provide conclusive evidence for or against rinsing. Larger statistics are probably required before clear conclusions may be drawn on this point.

4.2. Improvement of Qo

The RRR values measured on samples produced inside a single-cell 500 MHz cavity are systematically higher than those of samples coated in LEP type cavities. Typically, in the first case RRR is about 25, while in the second case it hardly reaches 17. Furthermore, the RRR of samples coated inside the two central cells (2 and 3) of the LEP cavities are systematically higher than for the two extremity cells (1 and 4). In this latter case, RRR values have never exceeded 14. During the summer 1991, the first samples produced by Industry in LEP cavities also showed the same RRR values and the same pattern observed at CERN (see Table I).

Table I

Comparison of the RRR values obtained in the various cavity cells by CERN and by two different European Companies.

RRR Values			
Cell Nº	CERN	Company I	Company II
1	12.1	14.0	12.3
2	17.9	15.6	16.1
3	15.2	17.5	16.2
4	13.5	13.7	12.3

On the basis of the increased experimental evidence, a clear correlation emerged between the quality of the samples and the order of cell coating, which is carried out according to the cell sequence 1-4-2-3. This correlation has been tentatively ascribed to contamination of the growing film by surface degassing. During coating, all surfaces inside the cavity are bombarded by a continuous flux of UV photons and, depending on polarity, by electrons (cavity walls) or ions (cathode). In spite of the bakeout carried out before coating, these surfaces act as a gas source until they are either cleaned by ion bombardment (cathode) or covered with a clean layer of Nb (cavity wall). Consequently, degassing should decrease resulting in films of higher purity at the end of the coating process.

To elucidate this point, a short cleaning discharge has been carried out in each cell prior to coating samples in the usual order. A RRR improvement has been observed, but the values were still below those expected and the usual quality pattern was still noticeable.

It must be recalled that in the usual coating situation the magnets are kept at fixed positions in the middle of the cells (see Fig. 3). Therefore the cathode surfaces between magnets are only partially cleaned during the standard coating process. In a further experiment a cleaning discharge has been applied by moving a magnet all along the cathode length to avoid this inconvenience. High and uniform RRR values (above 20) have been achieved in this case for the first time. Only samples from cell 4 were slightly worse, probably due to gas backstreaming from the adjacent turbomolecular pump.

For the time being, no cavity has been produced according to this new procedure. However, all improvements on RRR have resulted so far in better Qo (E) curves. Furthermore, the Qo values from single-cell 500 MHz cavities are usually superior to those obtained on LEP cavities (after frequency scaling). Therefore some improvement in cavity performance may be expected by implementing this new coating procedure.

A second possible line of improvement consists in using Nb of higher purity for the cathode. So far we used Nb of RRR = 130. Since the coatings present much lower values, it had been assumed that nothing could be gained by using Nb of higher purity which is now readibly available on the market. However, samples produced by one Company using a cathode made of RRR = 300 Nb have been found to be better than those produced at CERN under the same conditions. These results have been reproduced at CERN, showing that RRR = 32 (instead of about 25) could be obtained inside a single-cell 500 MHz cavity. Although this improvement must be confirmed by further testing, a cavity will soon be coated in this way. Niobium of even higher purity has been ordered (RRR = 800) to look for any possible further improvement along this line.

A third approach to LEP cavities improvement consists in increasing the coating temperature. As already mentioned (see par. 2), a substantial benefit has been obtained by increasing the coating temperature to 200ºC. The decision not to exceed this temperature was based on mechanical stability calculations which took into account the progressive cavity wall thickness reduction consequent to the many chemical treatments applied to laboratory cavities between coatings. During its life, a cavity may be coated and chemically cleaned about 10 times. However, new cavities of nominal wall thickness should safely withstand 300ºC heating for the short duration of the coating. Coating of a cavity at 300ºC is planned for the near future.

Any clear indication deriving from the programme described in par. 4 may lead to a modification of the ongoing production of the LEP cavities. Therefore this programme is given at present high priority. Coatings of other superconducting materials, potentially superior to Nb, are also being studied, but with reduced effort because it is unlikely that the benefits they could offer will materialise in time for the LEP project.

5. OTHER COATING MATERIALS

The highest value of the BCS Qo obtained so far on Nb coated cavities of LEP type is 1.2×10^{10} at low field. The intrinsic properties of Nb do not leave room for further improving this limit. Accordingly, when assuming that this value may be systematically reproduced, that the surface residual resistance is zero and that the Qo (E) curve is perfectly flat, an improvement by a factor of three could be achieved with respect to the Qo value specified for the LEP cavities. Taking into account the quadratic dependence of the power dissipation on the accelerating field, in this idealised case the LEP cavities could be operated at 10 MV/m with the same cryogenic cooling capacity required for cavities performing as specified ($Qo = 4 \times 10^9$ at 6 MV/m). This means on the one hand that there is reasonable hope that some improvement may be achieved following the programme described in par. 4, but also, on the other hand, that Nb becomes unsuitable for operation at 4.2 K whenever accelerating fields much higher than 10 MV/m are required.
A substantial Qo increase at 4.2 K may only be achieved by replacing Nb with a superconductor of higher Tc. According to the BCS theory of superconductivity, the dependence of the RF surface resistance at a given frequency on Tc is expressed by the formula :

$$R_{BCS} \propto \rho^{1/2} e^{-\alpha T_c/T}$$

where T is the operating temperature, ρ is the normal state resistivity at operating temperature and α is half of the strong coupling constant. This formula shows the overwhelming importance of Tc, but also that the normal state resistivity should be as low as possible to minimise the surface resistance.

Critical temperatures above 17 K are reported in the literature [10,11] for NbN and (NbTi)N deposited by reactive magnetron sputtering. This technique consists in adding nitrogen gas to the argon used to produce the sputtering discharge, and using Nb or NbTi cathodes. The critical temperatures of these nitrides are very near to the maximum values available if the new high Tc superconductors, which for the time being are not mature for the application envisaged here, are disregarded. With respect to the A15 compounds which present slightly higher Tc, the envisaged nitrides are less sensitive to radiation damage and to crystalline disorder. Finally, cathodes of Nb and NbTi can be produced easily and the sputtering technique is very similar to that used to produce Nb coatings.

Among all the possible NbTi alloy compositions, we have chosen $Nb_{0.40}\,Ti_{0.60}$ and $Nb_{0.55}\,Ti_{0.45}$, the first because it is easily available (this alloy is used for producing superconducting cables), the second because it represents the best compromise between critical temperature and normal state resistivity [11]. In the latter case, a Qo improvement by about a factor of 30 with respect to Nb could be hoped for.

The results so far obtained along this line have been published in ref. 12. The best Qo (E) curve is presented in Fig. 8. In this case, and at low field, the BCS contribution to the surface resistance is about 4 times lower than for Nb coatings, but due to the high residual resistance this improvement is reduced to a factor 2. However, the fast Qo degradation with increasing accelerating field results in values lower than for Nb above 2.5 MV/m. The high residual resistance and the fast Qo degradation are the major obstacles towards the obtention of the theoretical performance of these coatings. But the result shown in Fig. 8 is not very far from that specified for bulk Nb cavities.

It should be noted that for coatings obtained by reactive sputtering the thickness uniformity of the film is of vital importance. Film quality relies on the achievement of its right stoichiometric composition, which may only be obtained if the rates of arrival (on the surfaces to be coated) of metal atoms and gas molecules are properly adjusted. In order to fulfil this condition, the deposition rate of metal atoms must be uniform, because the nitrogen pressure (and hence the impinging rate of gas molecules) is the same at any point of a cavity cell. At present our coatings are 20% thicker at the iris than at the equator of the cavity. The nitrogen pressure is chosen such as to provide best quality at the cavity equator, where the sample holder is connected to the cavity. As a result, temperature mapping shows enhanced thermal losses at the irises of the cavity [12] where the right film stoichiometry is not obtained. To improve on this point the sputtering configuration must be modified and connection ports must be added on the iris of the test cavity to allow for a more adequate quality control on samples.

6. CONCLUSIONS

The industrial production of LEP cavities is a milestone which marks the end of the first phase of the coated cavities development programme. Although some further improvements are desirable and still possible, it may be reasonably hoped that Nb coatings will be brought to perfection under the combined effect of recent better understanding and large scale production.

This development, carried out since 1980, has shown that Nb coated cavities are superior to bulk Nb cavities with respect to thermal stability, surface resistance, influence of applied magnetic field and cost. Under the convincing evidence of these benefits, many other laboratories all around the world have entered this game in recent years.

However, a major potential advantage of this method still remains to be proven. Accomplishing the thermal and mechanical functions by means of a copper substrate opens the door to using better superconducting materials which could provide a major further improvement in the cavities RF performance.

Although the initial efforts have revealed some additional technical difficulties inherent in this approach, which for the time being provides results still below the level of Nb coatings, the increased participation of other laboratories and the stringent performance requirements for future linear particle colliders will certainly contribute to speed up progress in this direction.

7. ACKNOWLEDGEMENTS

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Finally, we are indebted to R. Guarnieri (IBM-Yorktown) for having suggested the use of magnetron sputtering and for his help in designing the adopted cathode geometry.

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Figure Captions

Fig. 1 Schematic view of a 500 MHz cavity with the diode sputtering cathode. Three identical rotating cathodes are used. The bias voltage is applied to the shield.

Fig. 2 Schematic view of the magnetron sputtering configuration for single-cell 500 MHz cavities.

Fig. 3 Extension of the magetron sputtering configuration to the case of 4-cell LEP cavities.

Fig. 4 Variation of Qo as a function of the accelerating voltage Ea for 500 MHz single-cell cavities produced in different ways (best values). Measurements carried out at 4.2 K.

Fig. 5 Comparison of bulk Nb (best result) and coated LEP cavities measured at 4.2 K. Curves 1 and 2 represent the RF performance of the first two cavities coated by magnetron sputtering.

Fig. 6 Comparison of the RF performance measured at 4.2 K on 500 MHz single-cell cavities coated by magnetron sputtering at room temperature (curve A), at 80ºC (curve B) and at about 200ºC (curve C).

Fig. 8 Qo (Ea) of single-cell 500 MHz cavities measured at 4.2 K for the best Nb coated (Δ) and (NbTi)N coated (O) cavities. The X represents the value specified for the bulk Nb LEP cavities to be produced by Industry (Qo value scaled according to the square of frequencies).

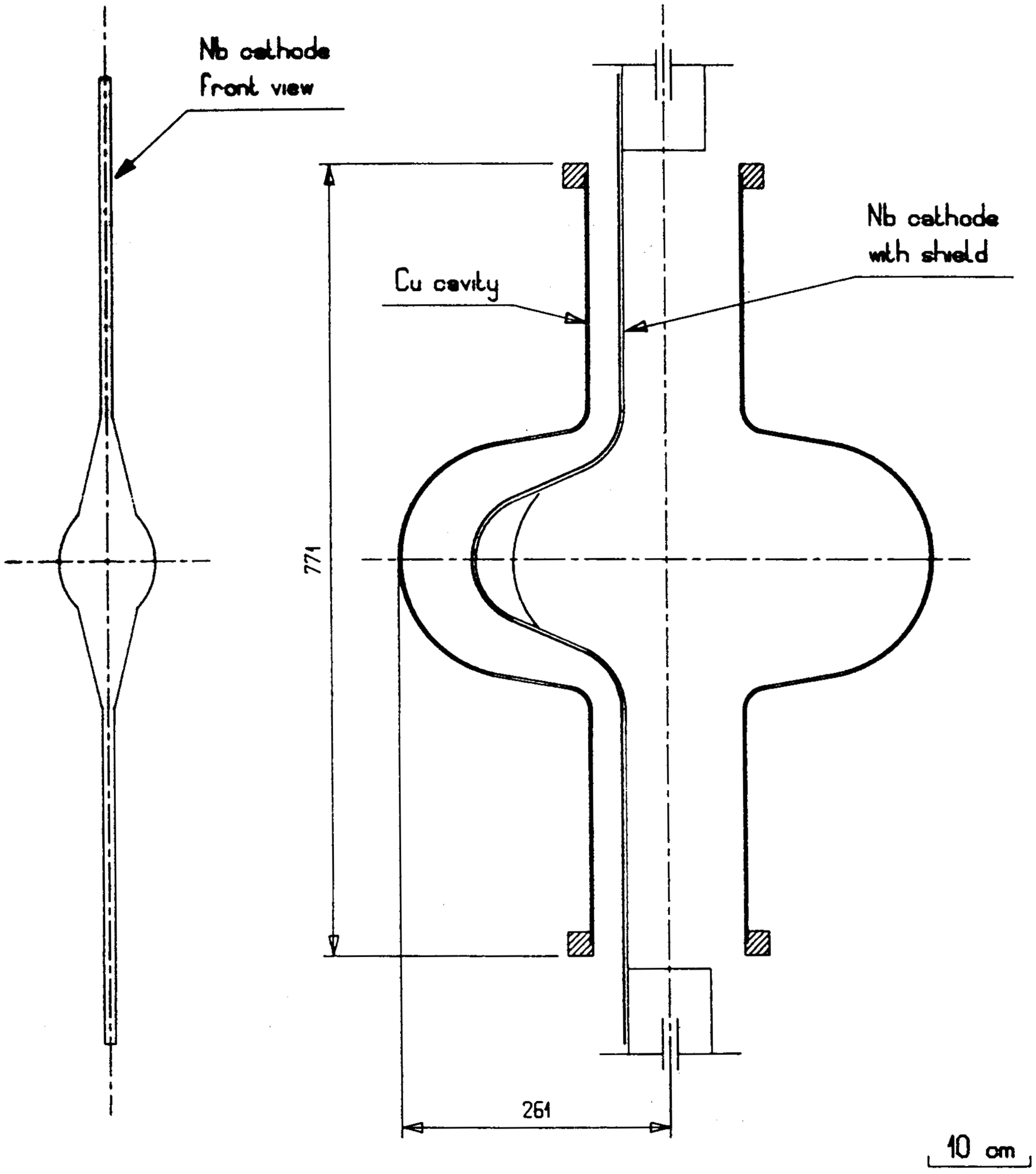
Nb cathode
front view
Cu cavity
Nb cathode
with shield
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Fig. 1

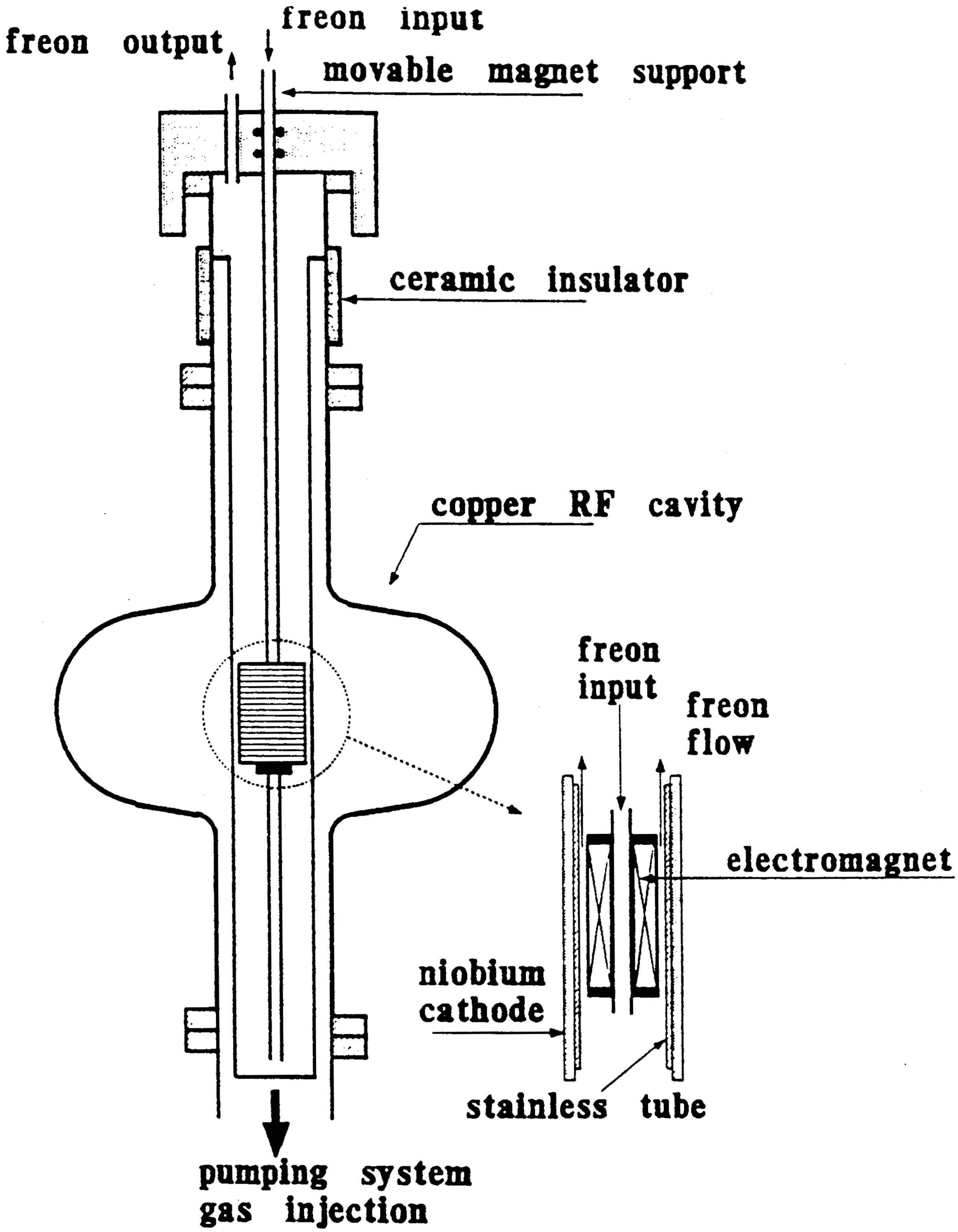
freon input
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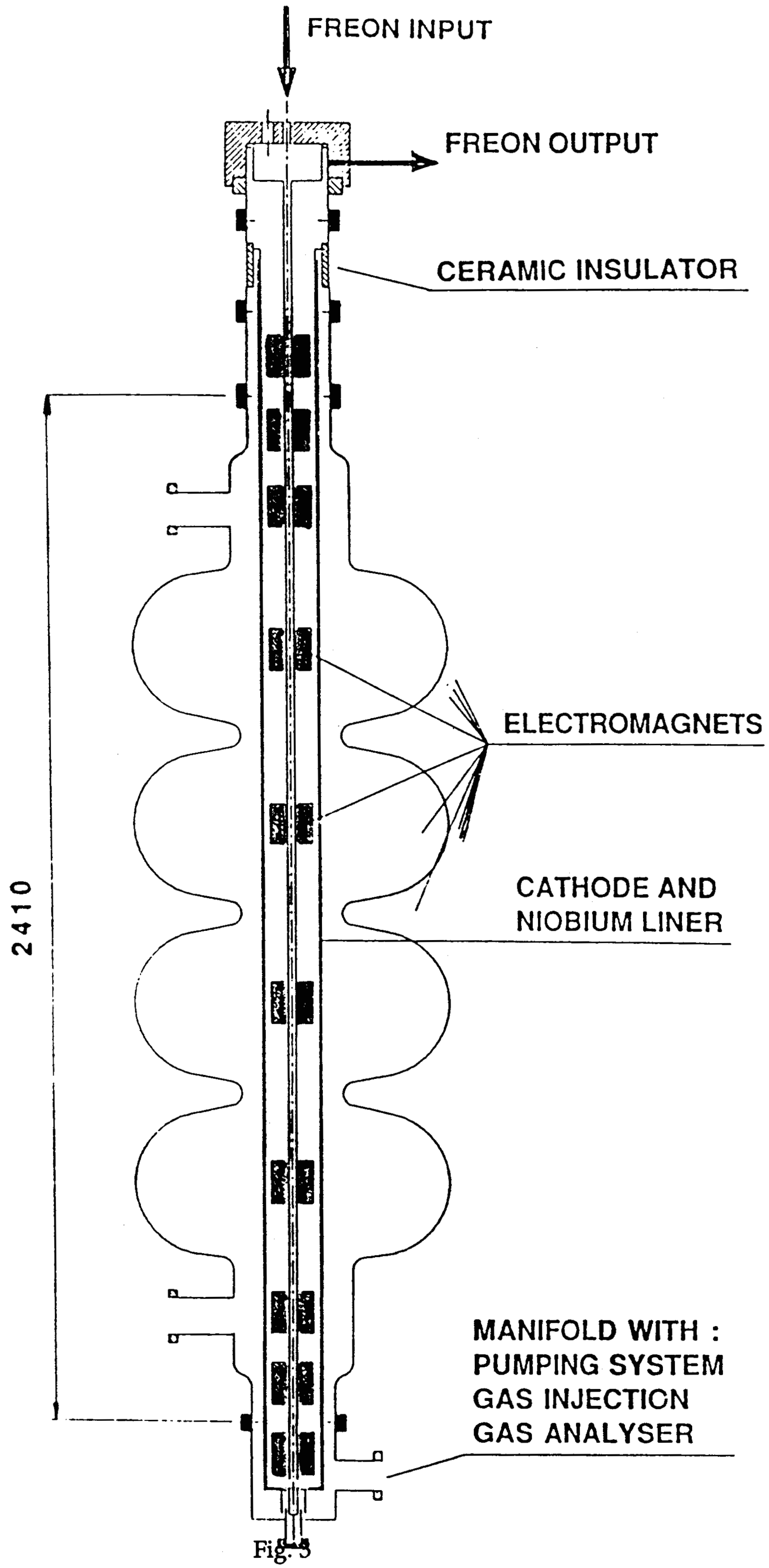
Fig. 2


FREON INPUT
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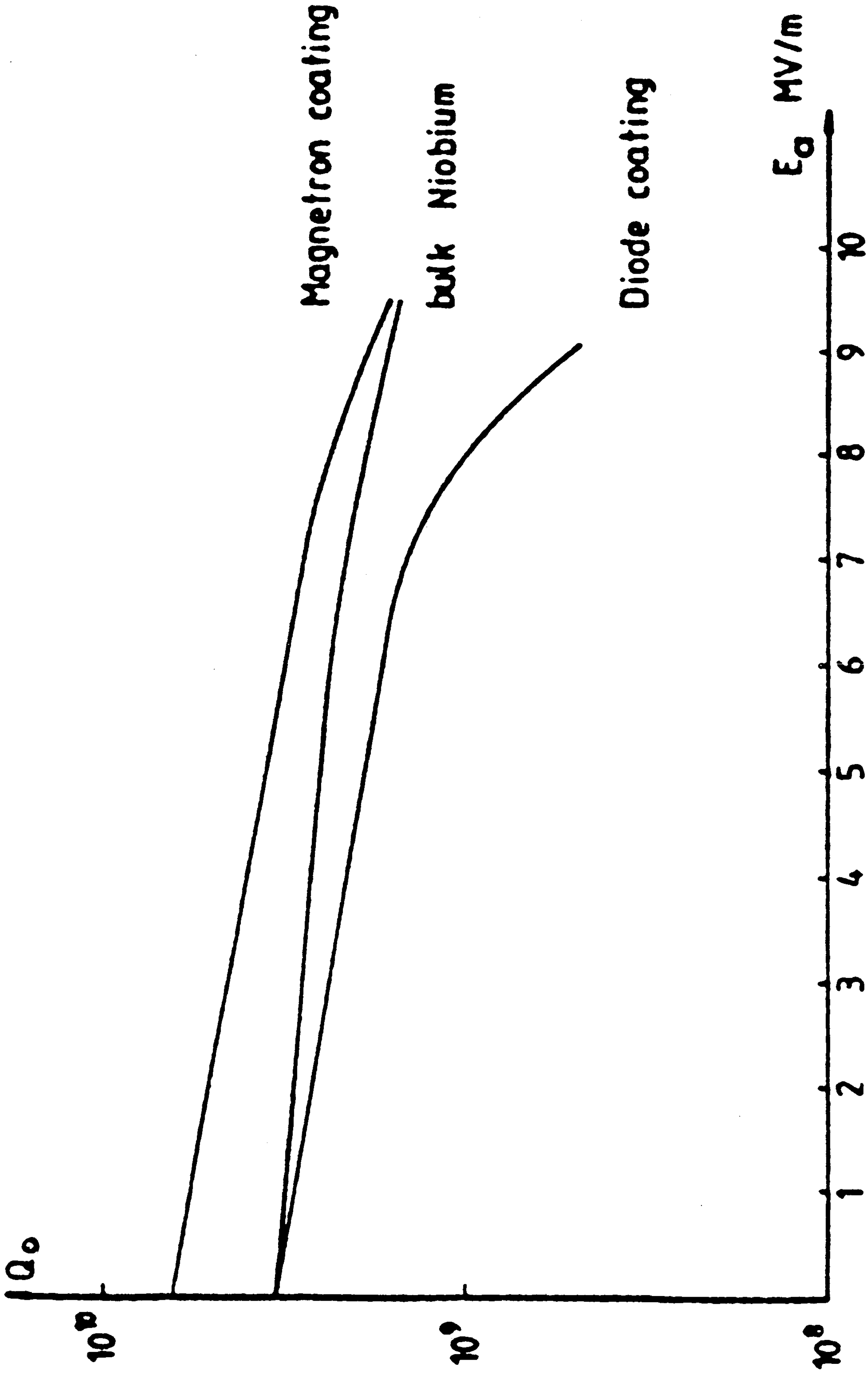
GAS ANALYSER

Fig. 5


Q_o
10^{10}
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Diode coating

Fig. 4

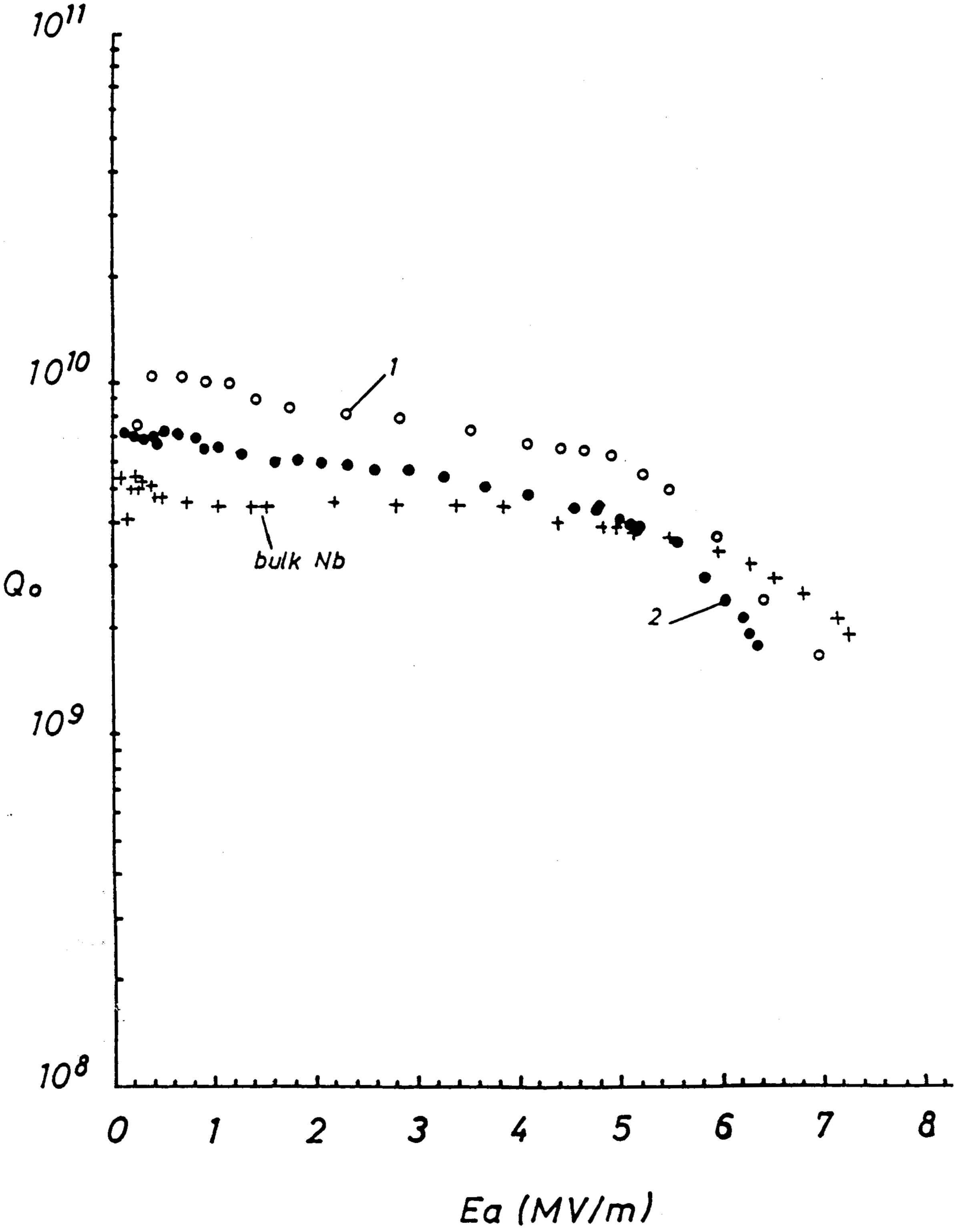

Fig. 5

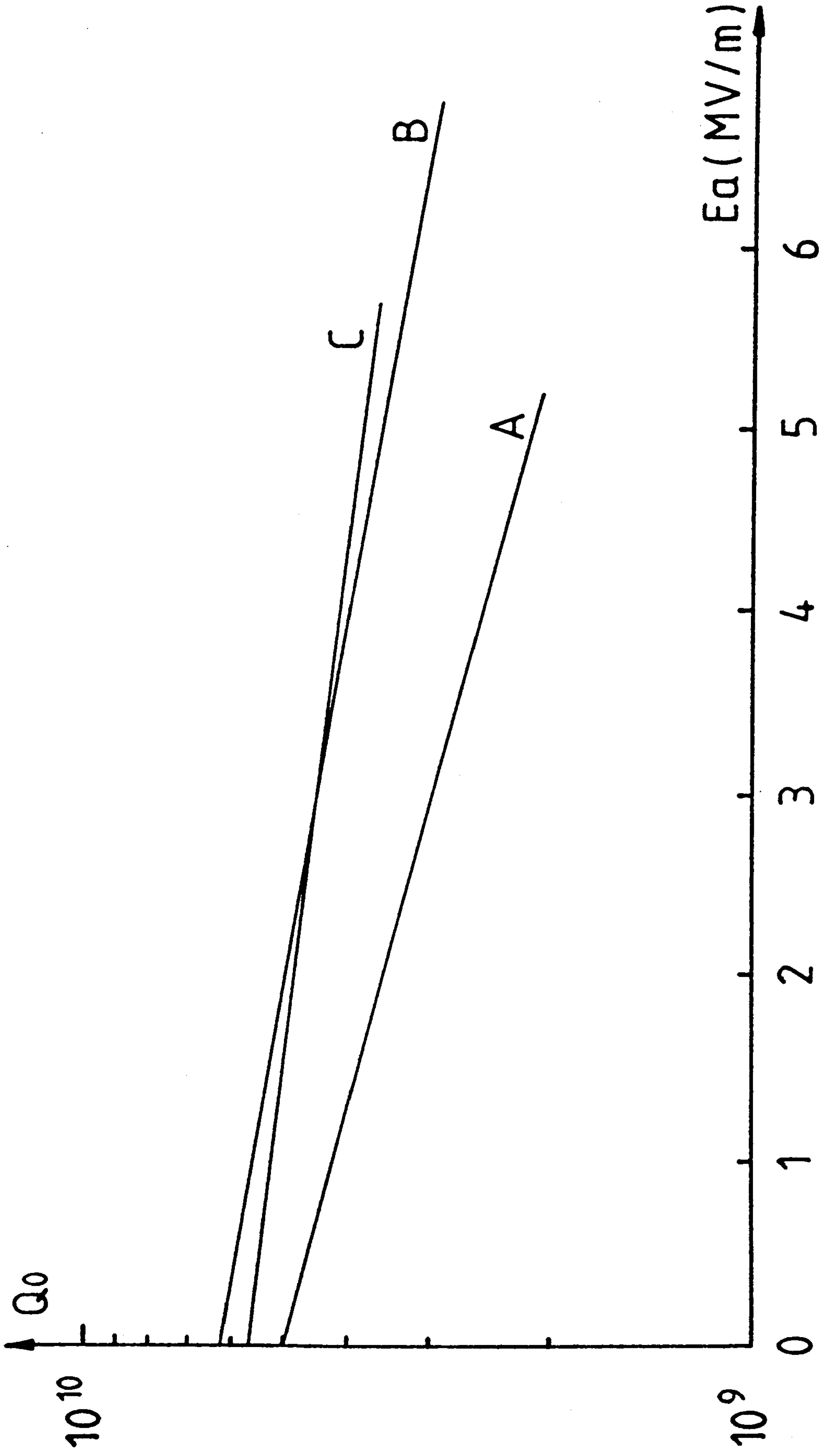

Fig. 6

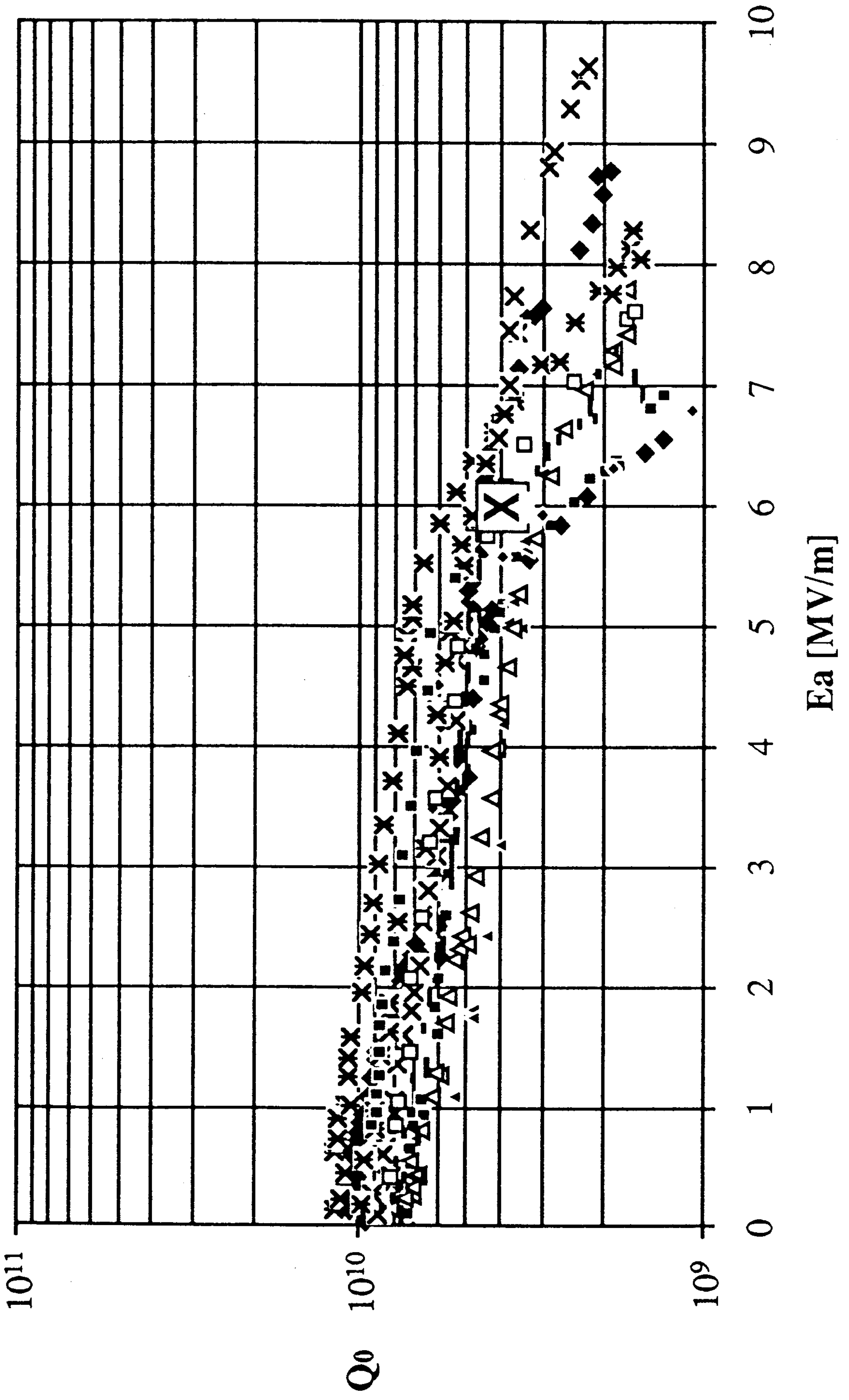

Fig. 7

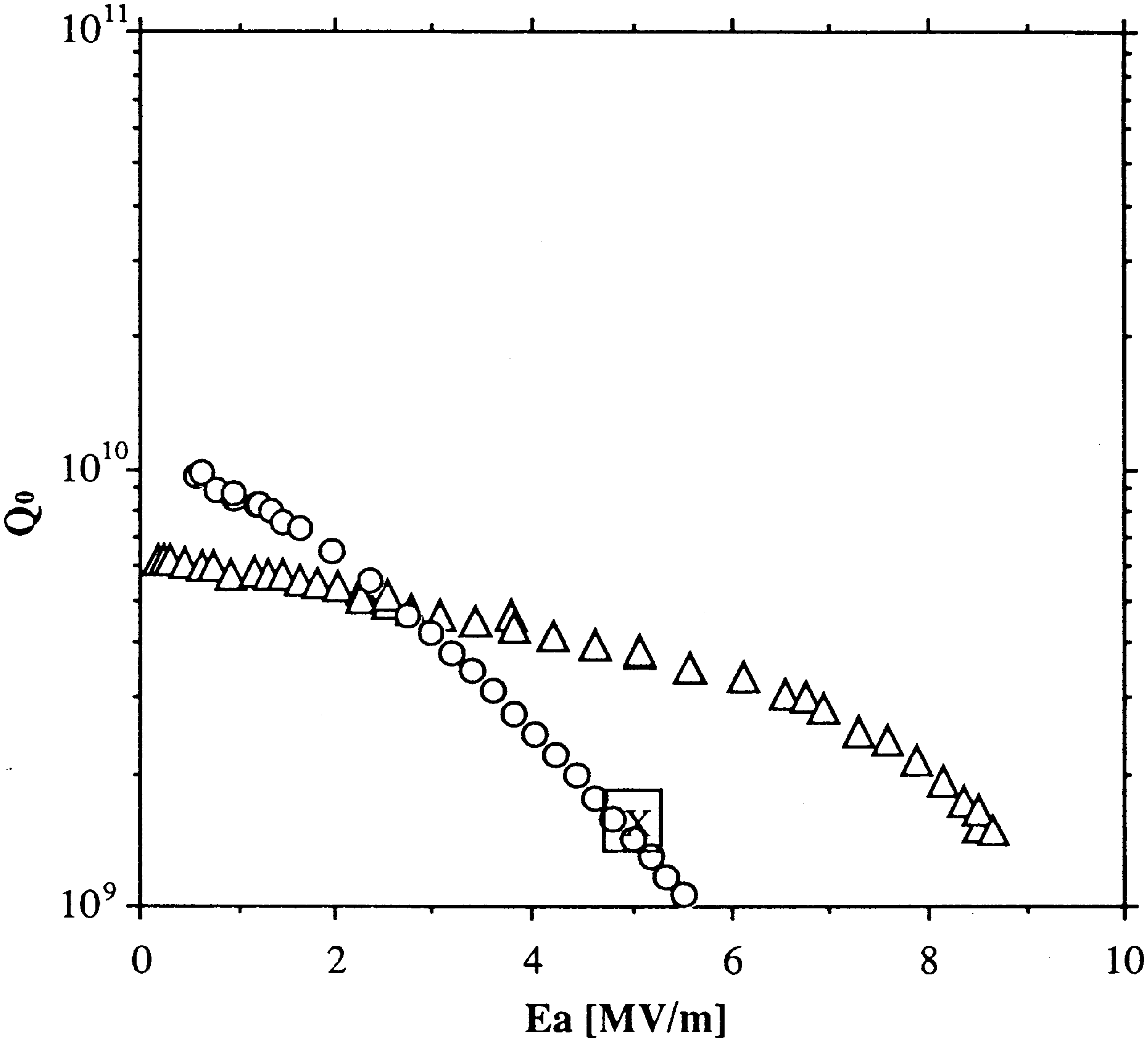

Fig. 8