LBL-10831
Preprint c.2

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Materials & Molecular Research Division

Submitted to IEEE Transactions on Magnetics

ELECTRON MICROSCOPY STUDY OF Co/Fe/B/Si AMORPHOUS ALLOYS

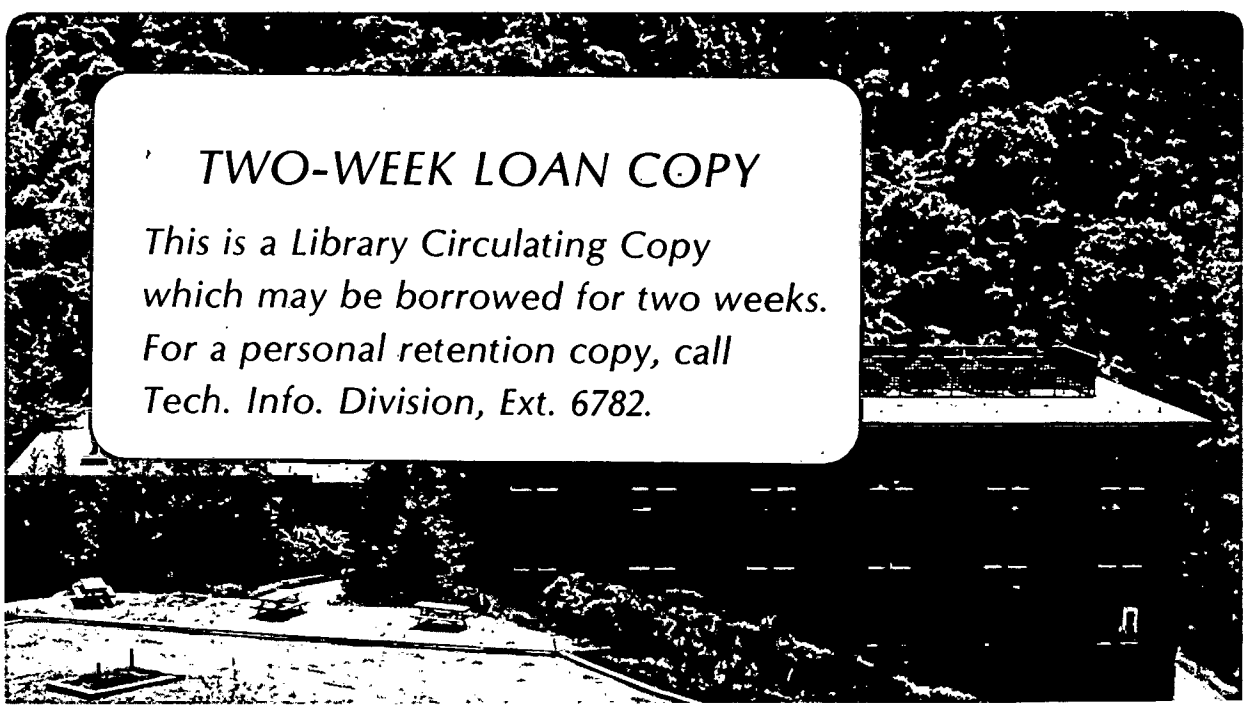
L. Rabenberg, R. K. Mishra, G. Thomas, O. Kohmoto and T. Ojima

May 1980

RECEIVED
LAWRENCE
BERKELEY LABORATORY

JUL 9 1980

LIBRARY AND
DOCUMENTS SECTION



TWO-WEEK LOAN COPY

This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 6782.

LBL-10831 c.2

Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

ELECTRON MICROSCOPY STUDY OF Co/Fe/B/Si AMORPHOUS ALLOYS

L. Rabenberg, R.K. Mishra, G. Thomas
Department of Materials Science & Mineral Engineering
Materials and Molecular Research Division
Lawrence Berkeley Laboratory, University of California
Berkeley, California 94720
and
O. Kohmoto and T. Ojima
TDK Electronics Company, R&D Laboratory
Ichikawa, Chiba, Japan

ABSTRACT

Changes in magnetic structures with annealing are studied using Lorentz electron microscopy and are correlated with changes in magnetic properties for the $Co_{71.4}Fe_{4.6}Si_{9.6}B_{14.4}$ amorphous alloy. Domain wall stabilization is shown to be the dominant factor resulting in decreasing μ and increasing H_c and K during low temperature annealing. Annealing near T_c results in an isotropic magnetic structure due to domain wall relaxation, and annealing above T_{cry} results in magnetically hard crystalline particles. It is concluded that treatments capable of producing a magnetically isotropic structure can produce the best soft magnetic materials.

I. INTRODUCTION

It has now been established that proper annealing treatments improve the permeability and coercivity of rapidly quenched amorphous alloy magnets[1,2,3]. Alloys with $T_c<T_{cry}$ can be annealed above T_c to increase their ac permeability.[4] When these alloys are annealed below T_c, in the absence of any external field, a sort of "magnetic after effect" is observed.[5] Annealing in an external magnetic field generally induces anisotropy and leads to increased Br, reduced H_c[1,6] and reduced ac μ. Also, it has been shown that magnetic field annealing, without inducing any magnetic anisotropy, can lead to higher ac permeabilities.[7,8]

It has been suggested that directional arrangement of atom pairs occurs during field annealing[9,10] and even during annealing below T_c in the absence of an external field. Structural relaxation and stress relief during annealing have been studied in some detail.[10] However, not much is known about the microstructural changes in the amorphous alloys that accompany the changes in the magnetic properties during annealing. In the present work, transmission electron microscopy is used to study the microstructural changes and changes in the magnetic domain configuration in a zero magnetostrictive Fe/Co/B/Si alloy during annealing. These changes are correlated with the changes in the magnetic properties.

II. EXPERIMENT

Amorphous alloys of nominal composition $Co_{71.4}Fe_{4.6}Si_{9.6}B_{14.4}$ were prepared by roller-quenching techniques. Specimens were annealed in argon for an hour at various temperatures and were rapidly quenched. Thin foils for transmission electron microscopy (TEM) were prepared by electropolishing 3mm discs, cut from the annealed and unannealed ribbons.

Conventional TEM and Lorentz electron microscopy (LEM) were done in a Philips EM301 transmission electron microscope. Structural imaging was done using a JEOL 200CX high resolution electron microscope. A Philips P400 analytical electron microscope with a scanning attachment was used for the energy dispersive microanalysis.

6-10mm Φ rings were punched from the samples and the low field magnetic properties were measured for the ring-laminated toroids. Effective permeability was measured in a 10mOe field at 1kHz. Hysteresis loops were obtained by an integrating flux meter. T_c, T_{cry}, B_s and K were measured as described in References 2 and 11.

III. RESULTS AND INTERPRETATION

Electron diffraction patterns, taken from areas ∿1000Å thick and ∿2μ in diameter from an as-quenched alloy specimen show diffuse rings around the transmitted beam, characteristic of amorphous structures, with the first ring diameter corresponding to 1.9Å spacing. High resolution structural images[12] taken from the as-quenched alloys (Fig. 1) show no trace of any crystalline order even in the atomic level. Conventional electron micrographs show no contrast changes.

Fig. is a typical example of the magnetic domain wall structures observed in LEM. The major boundary is a cross-tie wall having Néel type segments with Bloch lines, characteristic of thin films.[13] In addition to the Bloch walls, Fig. 2 shows magnetization "ripples" within the domains. That the contrast is from magnetic fluctuations or "ripples" was confirmed by applying an in-plane field in the microscope using a special specimen holder when these ripples were seen to get aligned. The "ripple" effect has been routinely observed in fine polycrystalline films[13], however, this is the first reported observation of "magnetization

ripple" in amorphous alloys.

The behaviors of the ripples and the Bloch/Néel walls were followed by heating the specimen in-situ. Upon heating, the ripples were observed to relax and align along the lines of force inside the domains as shown in Fig. 3. For temperatures up to ∿300°C, no change in the configuration of the cross-tie walls was observed. At still higher temperatures, (>400°C), the cross-tie walls began to relax to configurations where the angles between the walls were nearly equal and the cross-tie structure was more uniform. Fig. 4 shows the change in thedomain wall configuration (and the accompanying change in the "ripple" alignment) before and after heating the specimen to above 400°C.

In samples thermally treated to produce a few widely scattered crystals (which are known to form in this alloy above 480°C)[14], Lorentz microscopy showed that the domain walls interact strongly with the crystalline particles as shown in Fig. 5. The chemical microanalysis of the crystallites in the STEM showed no significant difference in the Fe, Co and Si concentrations between the crystal and the amorphous matrix.

The measured values of T_c, T_{cry} and B_s of the alloy are 457°C, 488°C, and 9.4kG, respectively. The changes in μ and H_c due to annealing are shown in Figs. 6(a) and (b). Permeability decreases up to ∿400°C and the B-H loop has a waspwaist type shape (like perminvar). But annealing the sample just below and above T_c increases μ. H_c shows the reverse behavior, and increases rapidly above T_c when μ drops drastically. The hevavior of K with annealing is similar to that of H_c. The B_r of the sample increases slowly from ∿1.0kG in the as-quenched alloys to ∿1.5kG in alloys annealed at 400°C, above which B_r increases rapidly and reaches 4.5kG for alloys annealed at 460°C.

IV. DISCUSSION

The observed "magnetization ripples" are due to fluctuations in local anisotropy inside a domain. These fluctuations are a result of the low remanence of the alloy.

The change in the cross-tie domain wall configuration at temperatures above 400°C are due to changes in local magnetic property and hence local chemical composition in the alloy. At these temperatures, the atomic species are more mobile (compared with low temperatures) and thus the structure of the alloys annealed at these temperatures is more homogeneous, allowing the domain walls to relax as observed in Fig. 4. For samples annealed at 460°C, $B_r/B_s \simeq 0.48$, approaching 0.5, expected from an isotropic material and thus it is not surprising that domain wall configuration (Fig. 4) in the samples changes to the one expected for an isotropic material. In these samples, H_c is low and μ is high, as one would expect from isotropic materials.

In the samples heated to still higher temperatures (~480°C), small crystallites begin to form. As seen in Fig. 5, these crystallites act as pinning sites for the domain walls and H_c increases in Fig. 6(b). μ drops for samples containing these crystallites. Since STEM analysis showed no difference in the Fe and Co concentration of the crystals and the matrix, it is suggested that the magnetic properties of the crystallites are influenced strongly by crystalline anisotropy.

In samples heated to temperatures below 300°C, the cross-tie wall configuration remains unchanged, while the ripples become more aligned. This indicates that the effect of annealing at such temperatures is a sort of magnetic ageing effect and the small fluctuations become aligned in the presence of the internal magnetic field inside the domain[5] (similar

to magnetic field annealing). This leads to an increase in the anisotropy constant K and decrease in μ. This continues to ~400°C (Fig. 6) until the atomic mobility becomes high enough for the material to become isotropic. The domain wall stabilization is also responsible for the waspwaist type BH loop in materials annealed below 400°C.[5] The observations confirm experimentally the model suggested by Shiiki et al.[15]

It may be concluded from these observations that the Fe/Co/B/Si amorphous alloys consisting of a microstructure free from crystals and heat-treated to produce an isotropic magnetic configuration will show the best soft magnetic properties. These predictions agree well with the results reported in references 7 and 8.

ACKNOWLEDGEMENTS

L. Rabenberg, R.K. Mishra and G. Thomas wish to acknowledge the financial support of the Department of Energy under Contract No. W-7405-ENG-48. Structural images were taken by JEOL Corporation, Japan.

REFERENCES

1. F.E. Luborsky, J.J. Becker and R.C. McCary, IEEE Trans. Magnetics, MAG-11, 1644 (1975).
2. O. Kohmoto, N. Yamaguchi, K. Ohya, H. Fujishima and T. Ojima, IEEE Trans. Magn. MAG-14, 949 (1978).
3. R. Hasegawa and R.C. O'Handley, J. Appl. Phys., 50, 1551 (1979).
4. E.M. Gyorgy, H.J. Leamy, R.C. Sherwood and H.S. Chen, in AIP Conference Proceedings, Vol. 29, 198, (1976).
5. H. Fujimori, S. Ohta, T. Masumoto and K. Nakamoto, in Rapidly Quenched Metals III, (The Metals Society 1978), Vol. 2, p.232.
6. H.S. Chen, S.D. Ferris, E.M. Gyorgy, H.J. Leamy and R.C. Sherwood, Appl. Phys. Lett. 26. 405 (1975).
7. R.C. O'Handley and M.C. Narasimhan, IEEE Trans. Magn. MAG-15, 970 (1979).
8. O. Kohmoto, H. Fujishima and T. Ohima, IEEE Trans. Mag., March 1980, (in press).
9. H. Fujimori, H. Morita, Y. Obi and S. Ohta, in Amorphous Magnetism II, R.A. Levy and R. Hasegawa, eds., (Plenum Press, 1976), 393.
10. F.E. Luborsky and J.L. Walter, IEEE Trans. Magn. MAG-13, 953 (1977).
11. O. Kohmoto, K. Ohya, N. Yamaguchi, H. Fujishima and T. Ojima, J. Appl. Phys. 50, 5054 (1979).
12. J. A. Allpress and J.V. Sanders, J. Appl. Cryst. 6, 165 (1973).
13. P.J. Grundy and R.S. Tebble, Advan. Phys. 17, 153 (1968).
14. A. Inoue, T. Masumoto, M. Kikuchi and T. Minemura, Sci. Rep. RITU, A, 27, 127 (1979).
15. K. Shiiki, T. Watanabe and M. Kudo, J. Appl. Phys. 50, 5419 (1979).

FIGURE CAPTIONS

Fig. 1 High resolution structural image and selected area electron diffraction pattern (in the insert) taken from as-quenched alloy showing no crystalline periodicity even at an atomic level. XBB 804 4573

Fig. 2 Cross-tie wall configuration in a thin foil specimen, imaged in the LEM mode showing 180° Bloch wall (AA), Néel segments (B), and Bloch lines (C). Note the background "ripple" contrast inside the domains. XBB 804 4578

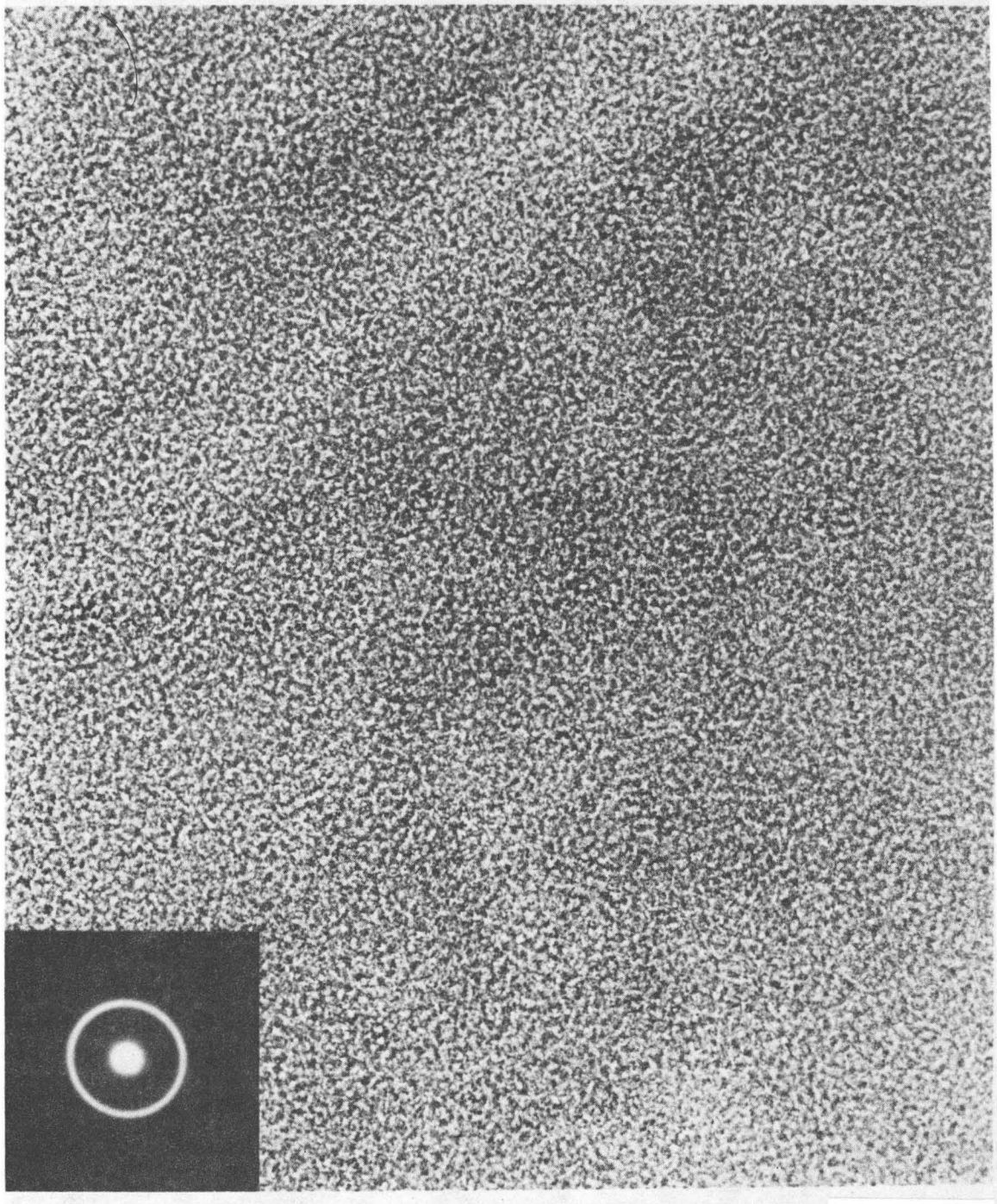
Fig. 3 LEM images taken from the same area showing the alignment of "ripples" upon aging the sample. The Bloch walls in (a), taken from as-quenched sample are unchanged in (b) when the sample is heated. XBB 804 4577

Fig. 4 LEM images showin the relaxation of doamin walls upon aging the sample at temperatures above 400°C. The wall configuration in B evolves from that in (a) which is from the as-quenched sample. XBB 804 4575

Fig. 5 LEM image showing the interaction of domain walls AA by the crystalline phase B. XBB 804 4579

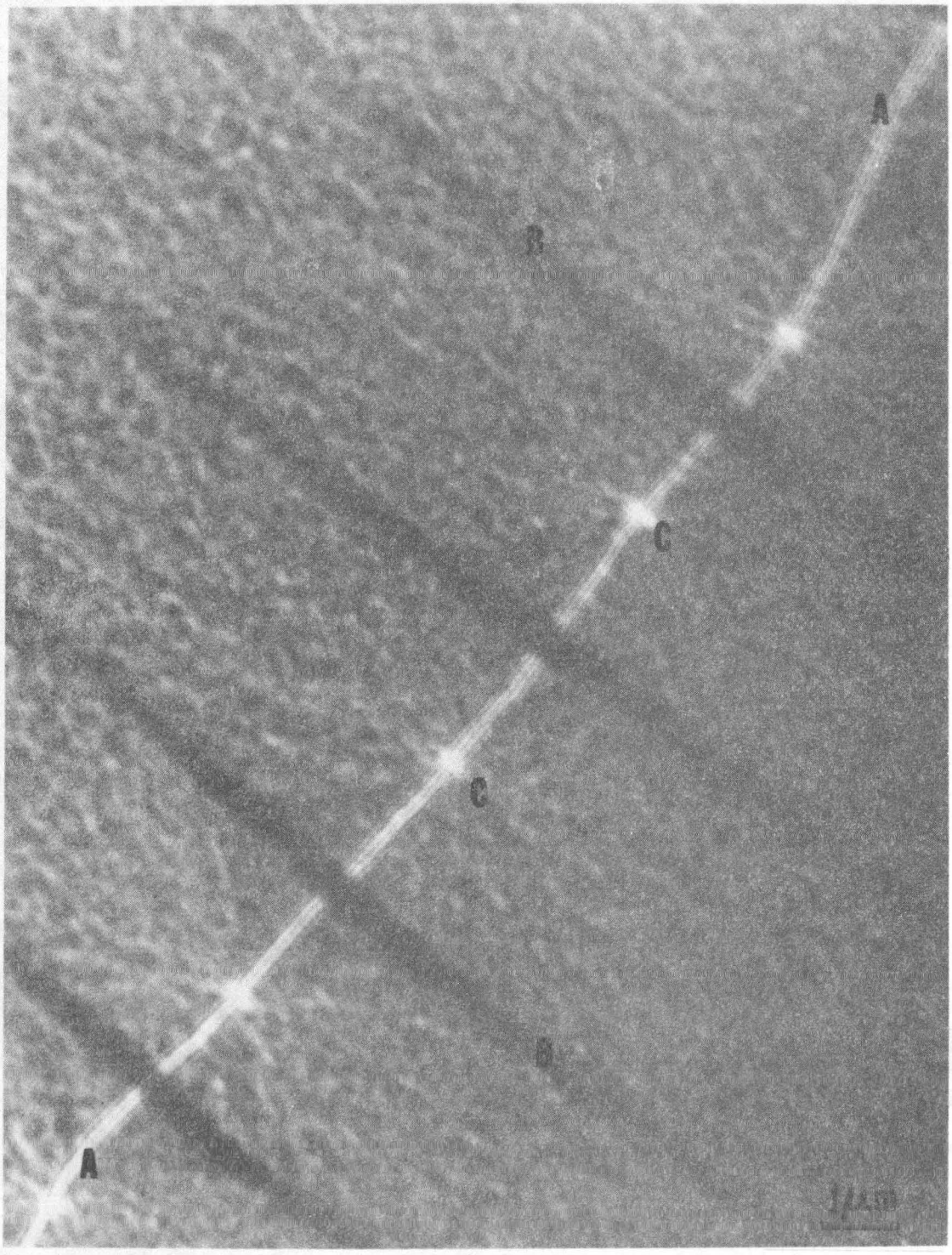
Fig. 6(a) Plot of μ vs. annealing temperature. XBL 802 4641

Fig. 6(b) Plot of H_c vs. annealing temperature. XBL 804 4959



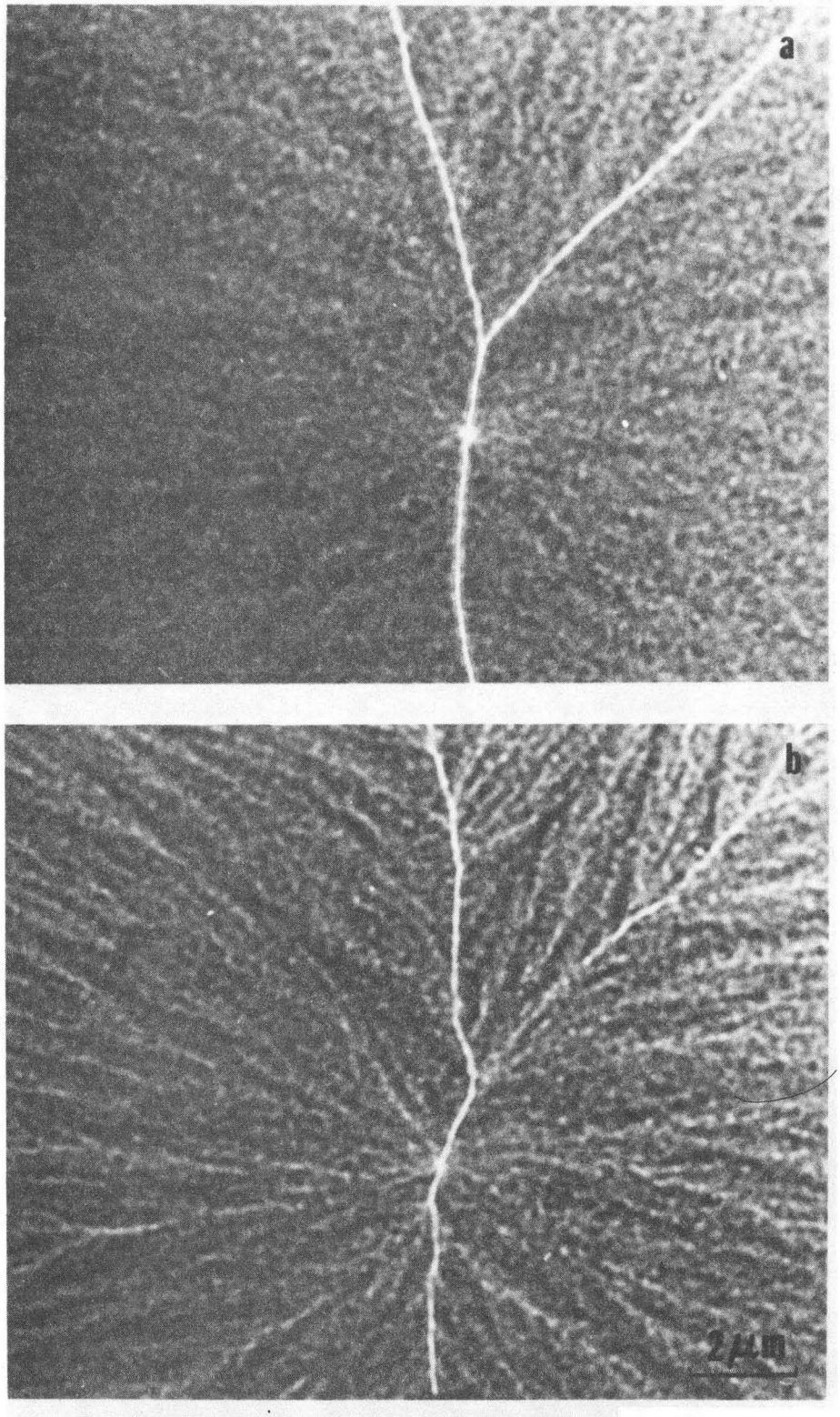
XBB 804 4573

Fig. 1



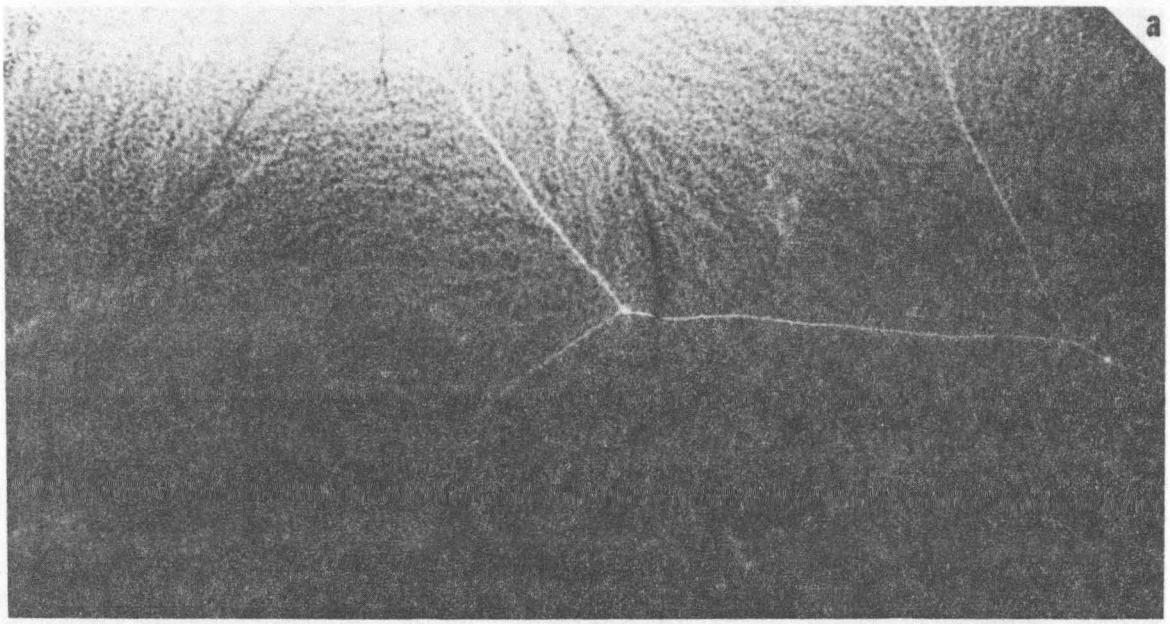
XBB 804 4578

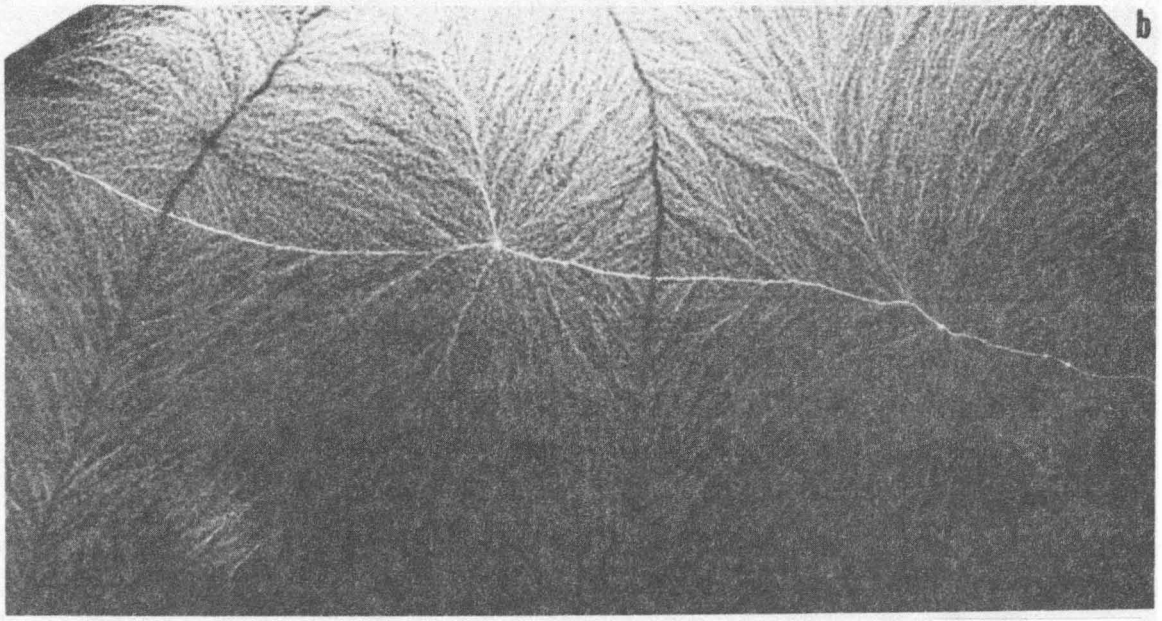
Fig. 2



XBB 804 4577

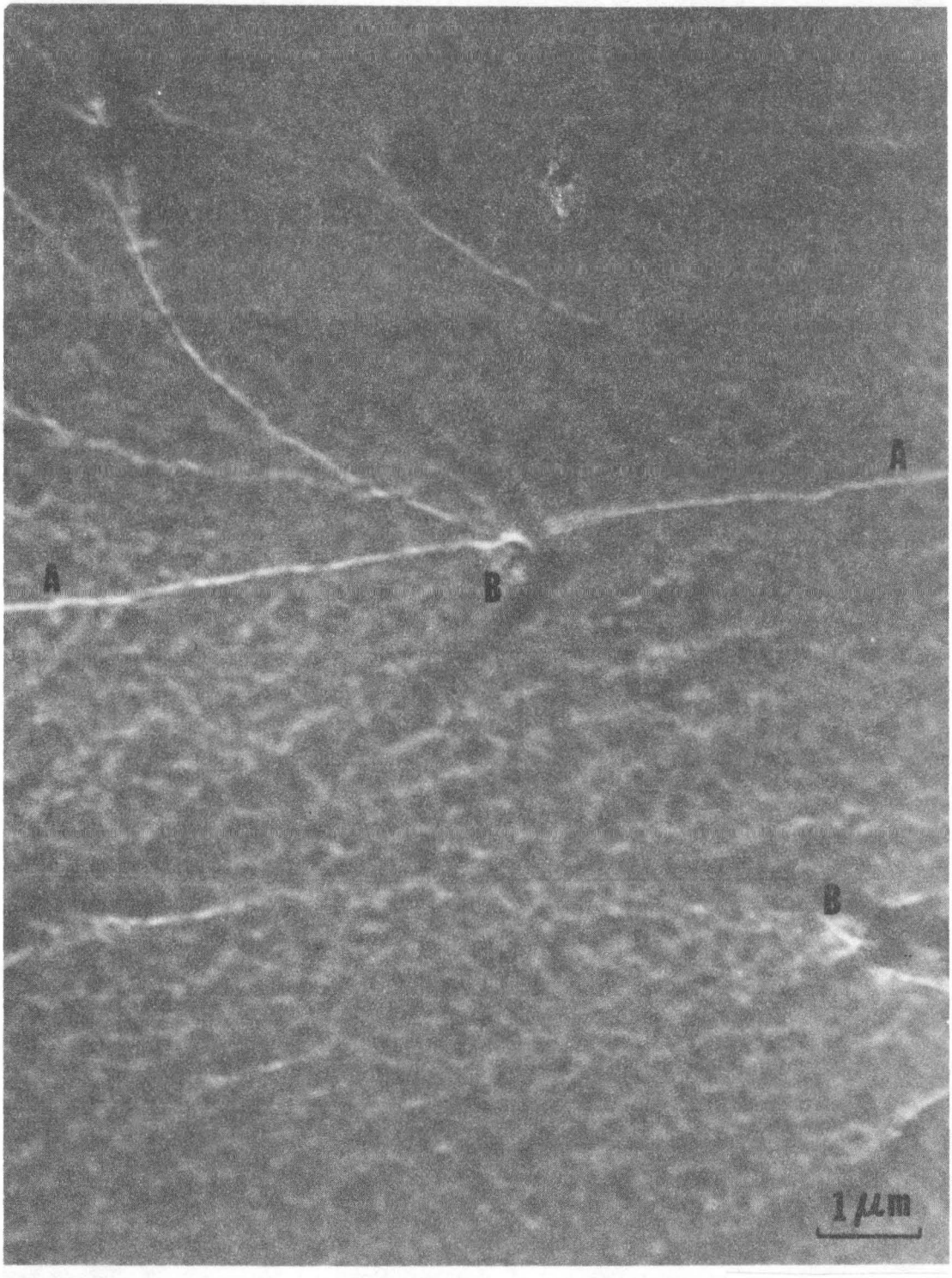
Fig. 3





XBB 804 4575

Fig. 4



XBB 804 4579

Fig. 5

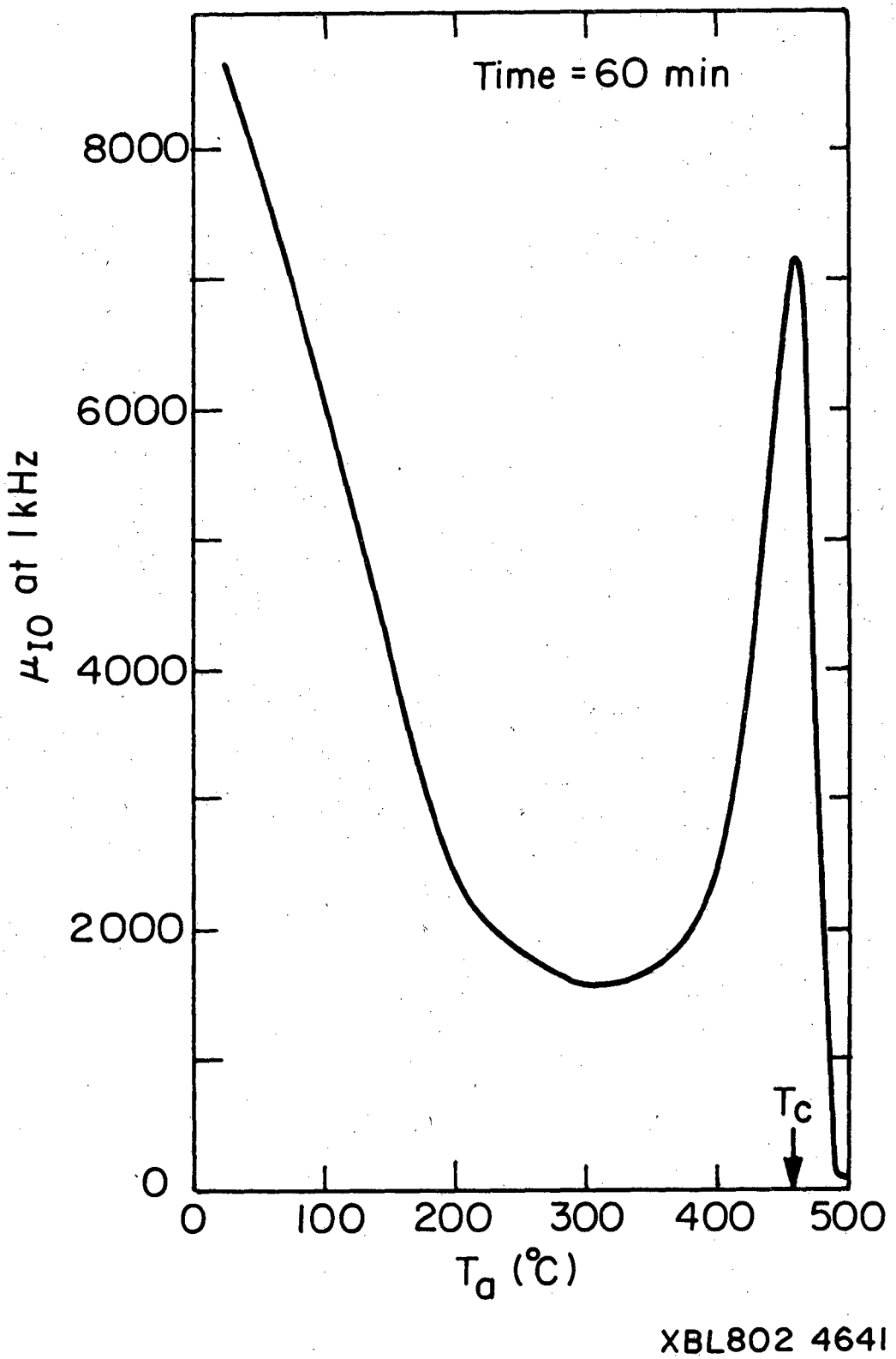

Time = 60 min
8000
6000
4000
2000
0
μ_{10} at 1 kHz
T_c
0
100
200
300
400
500
T_a (°C)
XBL802 4641

Fig. 3(a)

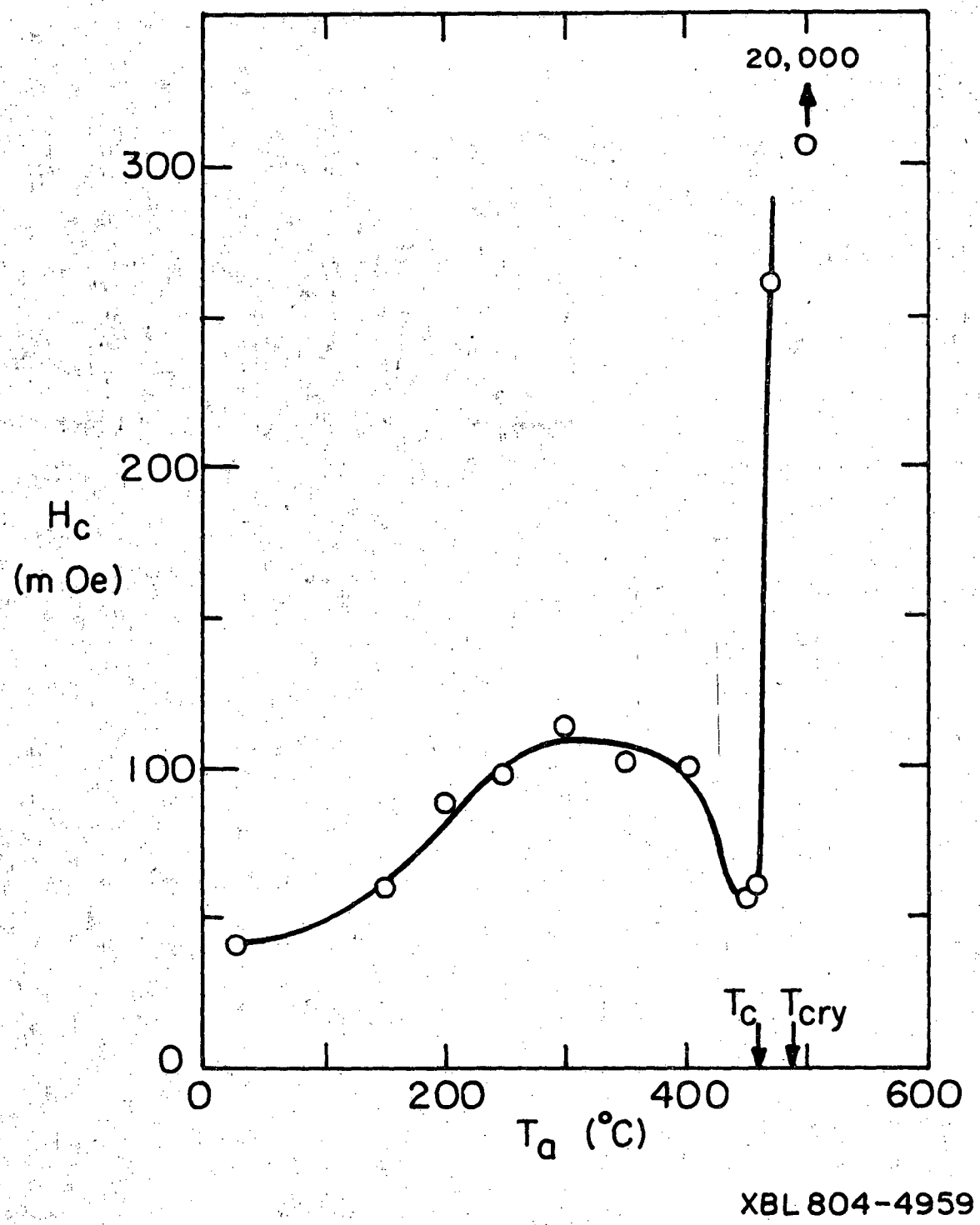


Fig. 6(b)

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable.

TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720