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In Memory of Professor Włodzimierz Żuk

Pamięci Profesora Włodzimierza Żuka

К памяти профессора Влодзимежа Жука

In the memory of Professor dr Włodzimierz Żuk I find arguments to recall his contribution to the initial stage of the work on single crystal growth.

The need for single crystals came forme the research on neutron diffraction. Professor Żuk had been familiar with these problems since in the 50 He was the co-organizer of the Department of Elementary Particles in Warsaw. In 1954 Professor Żuk proposed several problems for the M. S. degree in the Department of Physics the Maria Curie--Skłodowska University in Lublin. One of the problems was the growth of large single crystals, KCl crystals being most desirable.

I was one of the students in physics who was to decide about the choice of the work for the M. S. degree. Discussions with colleagues on single crystal growth were not much fruitful since we had no notion how to grow crystals. What was known was the idea of single crystal as a material of special properties.

That was the clue to the decision: to do something new of required properties. The charm of personal success which was the building of the mass spectrograph by Professor Żuk was not meaningless for a young student.

That is how I decided to grow crystals. With the help of Mieczysław Subotowicz, assistant in the Department of Physics, who gathered introductory bibliography on crystal growth. I presented to Professor Żuk a program and a schedule of the work. We decided to start with metallic crystals, e. g. bismuth and zinc.

During my research I recognized the two features of Professor Żuk: more official countenance at lectures and wery friendly and helpful assistance in individual student work. This man had a great experience in the laboratory work. Therefore, instead of many words some short remarks were enough to correct mistakes in the work. A discrete smile on his face, also not easy to be found, had one meaning - acceptance of results and encouragement for further work. It was always a pleasure to inform him that something new had been achieved in the experiment since these results were as much important for him a satisfactory execution of the student's theoretical knowledge.

A great care that Professor Żuk took of experimental results shortly proved very fruitful twice in my work. The first time when I was awarded with on IAEA fellowship by Professor Bronisław Buras for the growth of large metallic single crystals suitable for neutron diffraction studies and the second time when I began my post-graduate stay at MIT in 1959. My sponsor, Professor R. E. Ogilvie, asked me about the results of my work being done before I came to the United States. I was very much ashamed when I showed two reports only and informed about the grown crystals. Have you done it by yourself? - was the question. Yes, I answerd. O. K., you will have a talk on that on the seminar this week, decided Professor. It seems that the presented results on crystal growth opened to me the door of that laboratory. I understood what is the estimate of concrete results in good laboratories. Professor Żuk pointed out a correct way in laboratory work.

I dare to write some memorial remarks about Professor Żuk not only because of my personal motives but also because of the nearly 30-years-old continuous work on metallic single crystal growth in this country. This development initiated in Lublin could not be predicted

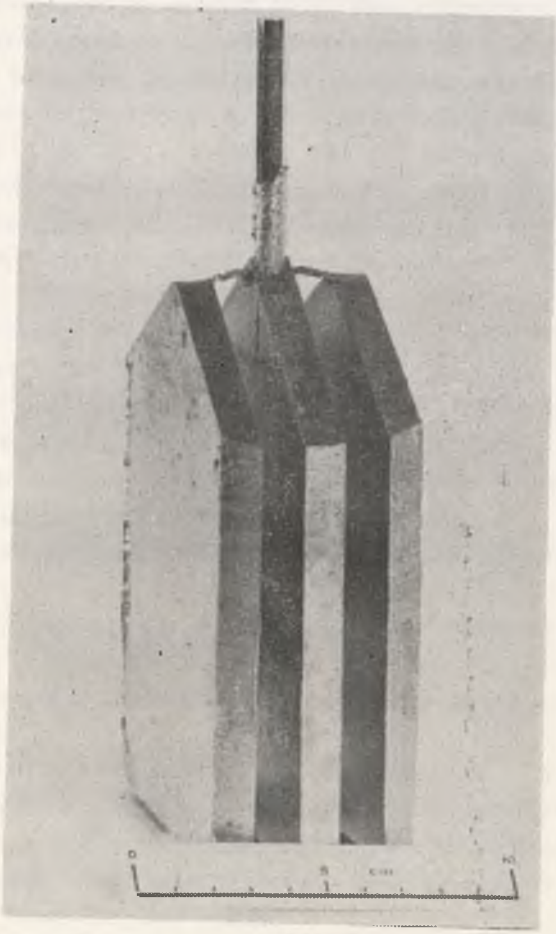

Fig. 1. Three single crystal plates of copper grown from one crystal seed

by Professor Żuk. An immediate need for single crystals resulted in the foundation of the laboratory of crystal research and technology in Świerk. Therefore, I will present some characteristic domains and results of the investigations carried on in this laboratory..

The work has been developed in two directions: research and production. Also the main domain is the technology of crystal growth; considerable research has been done on the characterization and use of the grown crystals. In general, different technological and research problems arose from the need or order of special materials.

One can distinguish the following domains of the work:

1) growth of large single crystals of metals and alloys by the Bridgman method,

2) preparation of these crystals for neutron diffraction purposed, mainly for neutron monochromators,

3) projects and realization of new types of monochromators, e. g. focusing monochromators,

4) thorough study on the nature and formation of low angle boundaries (lineage structure) during crystal growth,

5) investigations of the micromosaic structure in as-grown crystals,

6) computer calculations of the temperature fields in crystal growth system,

7) use of numerical methods for the interpretation of X-ray diffraction patterns,

8) directional solidification of eutectic materials, e. g. InSb-NiSb,

9) preparation of polycrystalline samples and growth of sizable single crystals of reactive materials, e.g. Y-Co compounds.

As an example, I will briefly present the progress of the work on the as-grown single crystal plates.

The plates used as neutron monochromators were usually cut from large cylindrical single crystals grown by the Bridgman method. To eliminate crystal cutting the Bridgman method was adapted for growing large oriented single crystal plates of copper and aluminium [1]. Fig. 1. shows three single crystal plates of copper grown from one crystal seed in a special graphite mould. However, these plates have not been used as neutron monochromators. The reflectivity of the cut plates was better - in general, both, the peak intensity and the half-width of neutron rocking curves were smaller in the case of as-grown plates. One could say that the mosaic spread of these plates was smaller than that of the cut crystals. More accurate neutron measurements made in reflection for several as-grown copper plates at intervals 2,5 cm along the axis of crystal growth have shown (fig. 2) that the peak intensity decreases as the plate is translated from first to last to freeze parts of the plate [2]; the half-width of the rocking curves was near to that measured for the cut plates. These effects were not observed in the plates cut from cylinders. Similar neutron measurements made in transmission confirmed the decrease in peak intensity; the half-width of the rocking curves measured for as-grown plates was smaller than for the cut plates.

These observations show that the mosaic structure of the as-grown plates is non-homogeneous. It depends on the position of the studied volume element of the plate, both, along and across the crystal plate.

To find a correlation between the crystal quality and growth conditions the numerical caluculations of the temperature fields for definite heating conditions during crystal growth were carried on. The enthalpy method was used to calculate the temperature distribution at large surface of the plate [3] and across the plates [4]. For each (i, j) two dimensional element the relation between the enthalpy H and the temperature T is given by [3]

$$H(T_{i,j}) = T_{i,j}(0) + \Theta + \frac{a}{(\Delta x)^2}\int_0^t \left[T_{i+1,j} - 2T_{i,j} + T_{i-1,j} + \left(\frac{\Delta x}{\Delta z}\right)^2 \left(T_{i,j+1} - 2T_{i,j} + T_{i,j-1}\right)\right] dt$$

where $a = \frac{\lambda}{\chi \cdot \rho}$; λ, χ, ρ, Θ are thermal conductivity, specific heat capacity, density of the metal and the ratio of latent heat to specific heat capacity, respectively. The obtained system of integral equations yields, together with the initial condition and the boundary condition the discrete solution of the problem.

The computer program written using the described model [3, 4] considers at initial time the temperature of the melt T_o and four parameters describing the growth conditions; the temperature of the furnace, T_f, the temperature gradient below the crucible, G_f, the lowering rate of the crucible, V, and the heat transfer coefficient, . The program will print: (a) the temperature fields in the coordinate system (x, z) for the required growth times, (b) the coordinate values of the isotherms for the required temperatures.

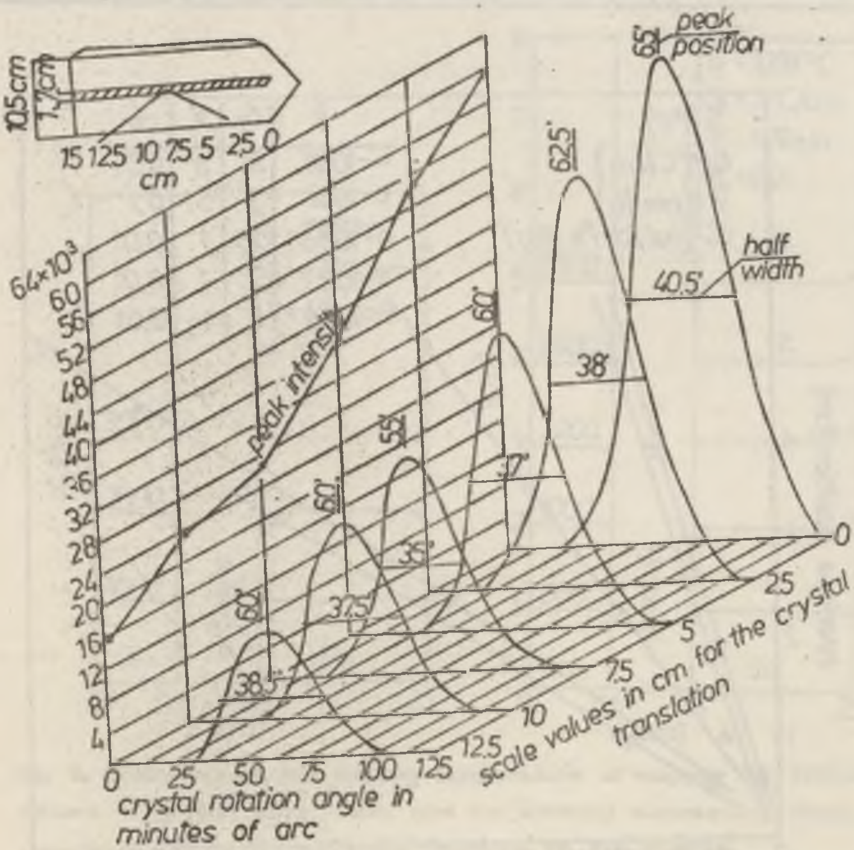

Fig. 2. Neutron rocking curves measured in reflection for as-grown copper plate at intervals 2,5 cm along the axis of crystal growth. Plate orientation (110), 5° 111

The results of the calculation concerning the growth problems comprise the interface position, the temperature gradients and the interface shape, i. e., the interface isotherms and the temperature profiles for succeeding growth times at planar positions, x, in the region of the solid-liquid interface. Fig. 3 shows, for example, the dependence of the interface position on time, calculated in the middle of the central copper plate for different growth parameters. The change of the interface position depends mainly on the lowering rate of the crucible - the higher the lowering rate, the larger changes in the interface position. Fig. 4 presents the isotherms at the melting temperature of copper for different values of growth parameters and for several succeeding times. Due to the symmetry of the system a half

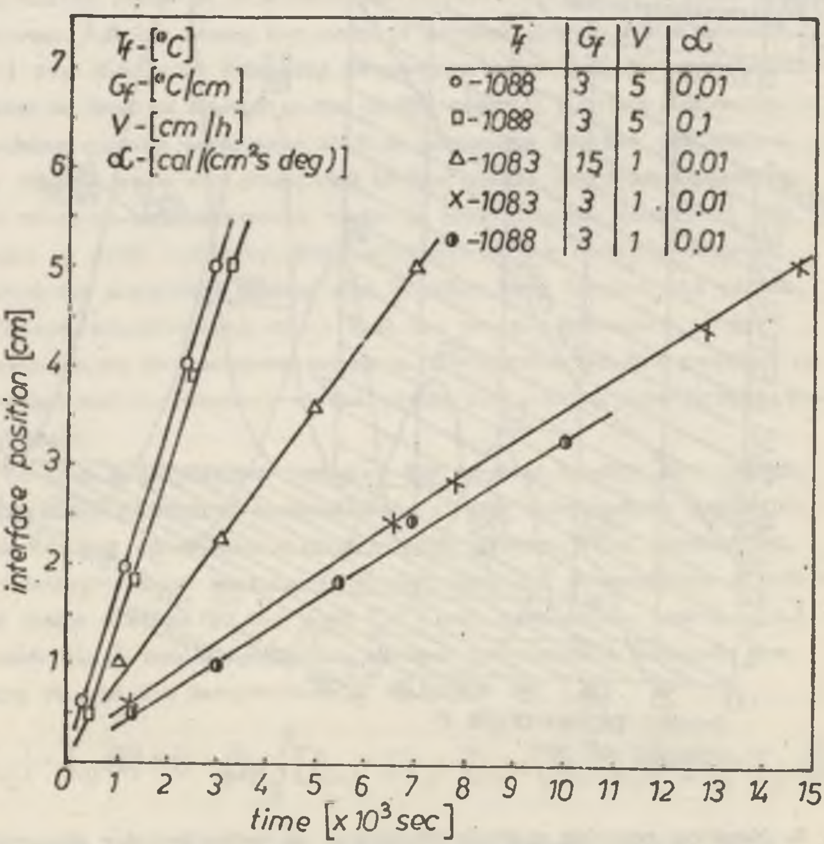

Fig. 3. Dependence of the interface position on time, calculated in the middle of the central copper plate different growth parameters

of the central plate and one of the side plates are considered. It is shown that the interface isotherms in the plates are curved at the edge of the plate: they are convex for small lowering rate, 1 cm/h, and are concave for higher lowering rate, 5 cm/h. It is assumed that in as-grown metallic single crystal plates these effects can influence the mosaic structure. Since the growth direction is normal to the interface, the size and the spread of mosaic block can be much influenced by the interface curvature near the side surfaces of the plates.

Since 1960 grown single crystals have been prepared for sale. Thousands of crystals have been sent abroad and to many laborato-

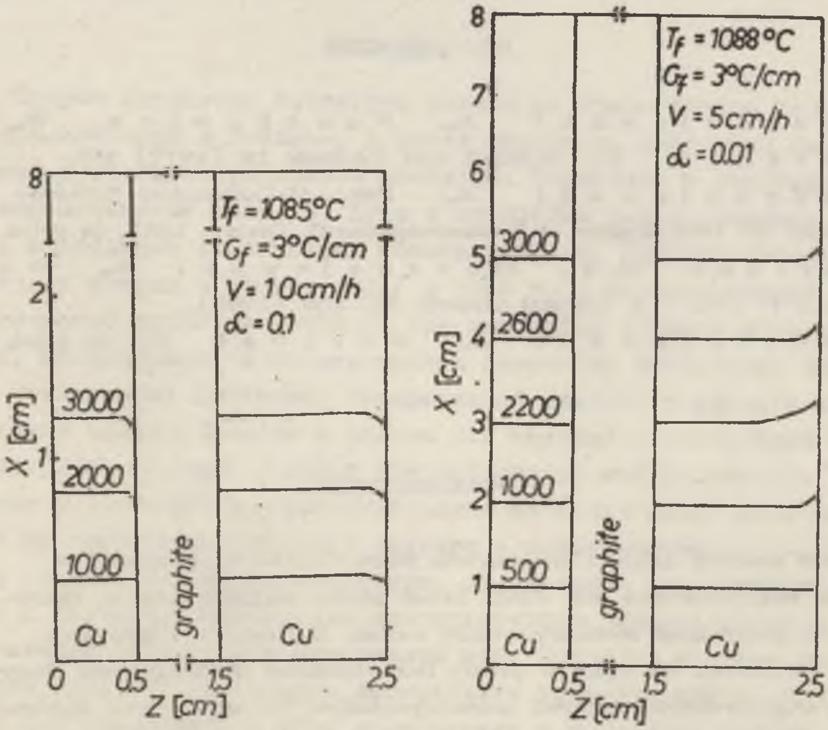

Fig. 4. Isotherms at the melting temperature of copper for different values of growth parameters and for several succeeding times. Times (in seconds) marked on the curves

ries in the country for use in various investigations. Most of these crystals were prepared as neutron monochromators. Świerk gained a reliable opinion as a center of investment of single crystals.

As a member of a group involved in crystal research and technology I can say that a small share in these achievements comes from Professor W. Żuk not only because of his "first word" in this domain but mainly because of the fudication of proper way in the laboratory work. He remains in my mind as a good teacher and a good man.

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STRESZCZENIE

Jako student fizyki Uniwersytetu Marii Curie-Skłodowskiej w Lublinie zdecydowałem się wziąć temat pracy magisterskiej z zakresu fizyki hodowania monokryształów metali. Serdeczna i życzliwa pomoc Profesora W. Żuka w pracy indywidualnej studenta była decydująca o powodzeniu hodowli monokryształów. Po otrzymaniu dyplomu magistra rozpocząłem moje studia pomagisterskie w MIT w r. 1959, hodując duże monokryształy metalu, przydatne do badań dyfrakcji neutronów. Był to początek blisko 30-letniej ciągłej pracy nad hodowlą monokryształów metali w Polsce, przeznaczonych do badań naukowych i na sprzedaż, głównie jako monochromatorów neutronowych. W związku z hodowaniem monokryształów wykonano sporo prac naukowych: natura i tworzenie się granic niskokątowych, struktura mikromozaikowa, obliczenia pól temperaturowych w układach do hodowania monokryształów, kierunkowe zestalanie się materiałów eutektycznych itd. Instytut Badań Jądrowych w Świerku uzyskał opinię poważnego ośrodka hodowli monokryształów. Pewien udział w tych osiągnięciach miał także Profesor W. Żuk, wytyczając właściwe kierunki pracy laboratoryjnej.

РЕЗЮМЕ

Будучи студентом Института физики на Университете Марии Склодовской-Кюри в Люблине, я писал дипломную работу по физике на тему роста монокристаллов металлов. Сердечное и доброжелательное отношение проф. В. Жука к студентам имело решающее значение в процессе выращивания монокристаллов. Защитив дипломную работу, я поехал в Бостон /США/ в 1959 г. в Массачусетский технологический институт / МТИ /, где выращивал большие монокристаллы, использованы в исследованиях дифракции нейтронов. Это было начало моей 30-летней беспрерывной работы в области выращивания монокристаллов в Польше для научных исследований и для продажи, главным образом для нейтронных монохроматоров.
В связи с выращиванием монокристаллов написано много научных работ по следующим вопросам: природа и возникновение низкоугольных границ, мозаичная структура, определение температурных полей с системах выращивания монокристаллов, направленная кристаллизация евтектичных материалов идр. Институт ядерных исследований стал крупным цетром выращивания монокристаллов. Профессор В.Жук участвовал в этих достижениях и руководил лабораторными работами.

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