

## IN REMEMBRANCE OF VLADIMIR P. SKRIPOV: SOME PERSONAL REFLECTIONS

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*The present paper is devoted to the memory of an outstanding scientist, organizer of science, and personality, Vladimir P. Skripov, in connection with the celebrations of his 90th birthday. In particular, I would like to address here three directions of his research: the experimental and theoretical analysis of critical phenomena, the properties of metastable liquids, and the kinetics of nucleation and growth processes both with respect to bubble formation and crystallization, which have been of particular importance for my own research in this field. It has been always a great pleasure for the author of the present paper to have the possibility to discuss with Vladimir P. Skripov problems of common scientific interest. Unfortunately, we do not have such opportunity any more. However, even at such conditions, his advice will continue to guide our research and to assist in the solution of a variety of research problems in the future. Beyond these scientific aspects, the remembrance of him, his work, and personality, will always be a not-to-forget source of deep delight and admiration.*

**KEY WORDS:** critical phenomena, nucleation, spinodal and pseudo-spinodal curves, metastable liquids, crystallization, Kauzmann paradox

### 1. INTRODUCTION: HOW WE CAME IN CONTACT

The present paper is devoted to the memory of an outstanding scientist, organizer of science, and personality, Vladimir P. Skripov. A brief overview on his scientific career and the topics of his research is given in an accompanying paper by Baidakov (2017). Here I would like to add some more details on three directions of his research: experimental and theoretical analysis of critical phenomena, the properties of metastable liquids, and the kinetics of nucleation and growth processes, which have been of particular importance for my own research in this field.

As one of the first directions of his research, Vladimir P. Skripov was involved in the analysis of different aspects of the physics of critical phenomena. This highly important scientific topic appeared around 1822–23 as the result of experiments of Cagniard de la Tour and Faraday. It was intensively studied in the 20th century by a large number of scientists including seven Nobel Prize laureates (see Ivanov, 2008, for details). Vladimir Pavlovich made some outstanding contributions to this field, establishing experimentally the existence of a maximum of the specific heat near to the critical point (a liquid–vapor equilibrium state where the bulk state parameters of the liquid and the vapor coincide and the surface tension approaches zero) long before it became treated world-wide as a sensation and performing experimental measurements on the dependence of the correlation length on temperature near to this particular state (Skripov and Kolpakov, 1965a,b; Skripov and Semchenko, 1955). This work was then continued by a comprehensive analysis of metastable systems (systems stable with respect to small but unstable with respect to sufficiently large fluctuations denoted in the description of nucleation as critical clusters), in general, and the kinetics of formation and growth of supercritical nuclei of a newly evolving phase both in application to the formation of bubbles and crystallites of a solid phase (Skripov, 1974; Skripov and Faizullin, 2006; Skripov and Koverda, 1984). In this connection, measurements of the average lifetime of a superheated liquid as a new means of studying nucleation kinetics have been developed. This method has become of considerable importance in recent decades also in

the analysis of nucleation phenomena in molecular dynamics simulations. By this reason, we performed a detailed reconsideration of essential features of this method in a recent paper (Schmelzer et al., 2017b).

For the first time, I came across Vladimir P. Skripov and his work in the course of a stay at the Institute of Physical Chemistry of the Bulgarian Academy of Sciences in Sofia in 1984. Looking through the personal library of Academician Ivan S. Gutzow, I found his book (Skripov, 1974) there as one of the main substantial parts. Being engaged about 10 years later into an industrial research project (Schmelzer et al., 1998) on different aspects of the technology of production of polymeric foams at BASF, Germany, performed in cooperation with my friends and colleagues Ivan S. Gutzow and the Corresponding Member of the Ukrainian Academy of Sciences, Vitali V. Slezov, and coworkers and my oldest son, it was very helpful in the search for the solution of the problems we had to tackle. Of course, not knowing at that time the whole work performed under the guidance of Vladimir Pavlovich, anyway, I already in the introduction to the report noted: *“An extensive analysis - as it seems - the most comprehensive study, at present, of different theoretical and experimental aspects of bubble formation has been performed in the group of Skripov in Yekaterinenburg”* (Schmelzer et al., 1998, page 2).

A bit later, looking through the book by Debenedetti (1996), I found there a widely similar high evaluation of Skripov's monograph. In particular, Debenedetti noted: *“More than twenty years ago, Skripov's Metastable Liquids presented a masterful synthesis of knowledge about superheated liquids...”*. In a personal letter to Vladimir Pavlovich, Debenedetti even noted that he considered Vladimir Pavlovich as his teacher. And further Debenedetti wrote in his monograph: *“Twenty years ago, the study of metastable liquids was mostly that of superheated liquids. Skripov's Metastable Liquids is the best example of that viewpoint. Today many of the most interesting questions and all of the promising applications in the field involve supercooled liquids...”* as one of the motivations of writing his monograph. I am not going to discuss here whether such statement is true or not. In any case, Debenedetti was obviously not aware of a similarly important book by Skripov and Koverda (1984), where the thermodynamics and kinetics of crystallization processes of supercooled liquids was discussed in detail at a level considerably exceeding even a number of presently published studies on this topic, however, published exclusively in Russian language.

From this and similar experience with the work of other Russian and Ukrainian colleagues and friends, we considered it as highly important to prepare overviews on the work published so far mainly in Russian language in English. But this was, of course, not the only driving force. These activities resulted finally in a series of book publications including the monographs of Skripov and Faizullin (2006) and Baidakov (2007) and the proceedings of Schmelzer (2005, 2014) containing several chapters written by colleagues from Yekaterinburg (V.P. Skripov and M.Z. Faizullin: *Solid-Liquid and Liquid-Vapor Phase Transitions: Similarities and Differences*; V.G. Baidakov: *Boiling-Up Kinetics of Solutions of Cryogenic Liquids* in (2005) and *Crystallization of Undercooled Liquids: Results of Molecular Dynamics Simulations* in (2014); V.Ya. Shur: *Correlated Nucleation and Self-Organized Kinetics of Ferroelectric Domains*; J.W.P. Schmelzer, G.Sh. Boltachev, and V.G. Baidakov: *Is Gibbs Thermodynamic Theory of Heterogeneous Systems Really Perfect?*).

Going back to the beginning, when we started to organize Research Workshops on Nucleation Theory and Applications at the Bogoliubov Laboratory of Theoretical Physics of the Joint Institute of Nuclear Research in Dubna starting in 1997, I considered it, consequently, as highly important to try to establish also direct contacts and to invite Vladimir Pavlovich and colleagues to take part in the meetings organized there every year as a rule in April. Another good friend and colleague, Vladimir M. Fokin, being engaged mainly in crystallization (e.g., Fokin et al., 2006), having both Skripov's books (Skripov, 1974; Skripov and Koverda, 1984) in his library as well, supplied me with the address. Vladimir Pavlovich answered immediately to my letter but already had other plans for the respective period in 1998, so he asked Vladimir G. Baidakov to follow the invitation. In this way, Vladimir Georgievich took part for the first time in the second workshop in Dubna in 1998. In the same year, I had then the pleasure to visit for the first time the Institute of Thermal Physics (ITP) in Yekaterinburg. It was the beginning of a very fruitful and pleasant cooperation continuing till now.

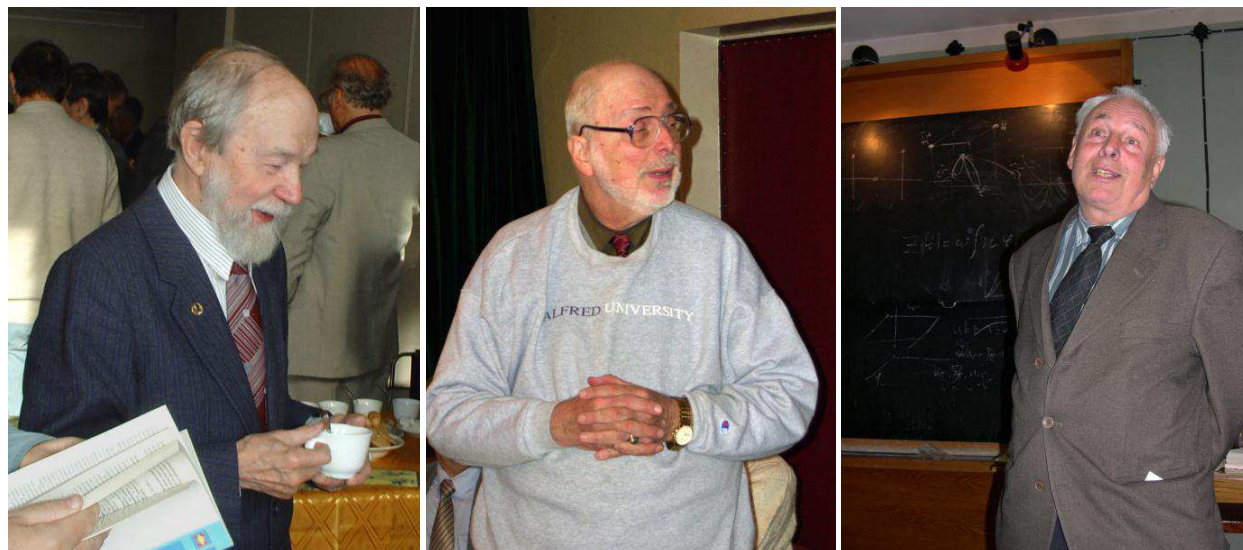
## 2. MEETINGS IN DUBNA AND YEKATERINBURG

Since 1999, in the period 1999–2005, Vladimir Pavlovich also directly took part in the workshops in Dubna and gave a comprehensive description of the work performed at ITP. He presented in the Dubna meetings the following

talks: (i) Melting at Negative Pressures, (ii) Binary Solutions and Generalized Equations for Phase Equilibria, (iii) Bubble Formation in Liquids: The Trouble with Water and Water-Gas Solutions, (iv) Thermodynamics of Melting and Simon's Equation (in 1999); (v) On the Surface Tension of Simple Substances at the Crystal-Liquid Boundary (with Mars Z. Faizullin), (vi) Extension of the  $\lambda$ -curve of  $^4\text{He}$  into the Region of Metastable States of Liquid Helium (in 2000); (vii) The Account of the Metastable State of Molecular Systems in the Determination of the Phase Diagrams (in 2001); (viii) Physico-Mechanical Properties of Gas-saturated Polymethylmethacrylate and Nucleation Kinetics (in 2002); (ix) Some Thoughts about Scientific Research Arising from the Own Experience in this Field, (x) Is there a spinodal in (one-component) melt crystallization? (in 2003); (xi) On the Second Crossover Near the Critical Point (with Dmitry Yu. Ivanov) (in 2004); (xii) On the Role of the Internal Pressure in the Phase Transformation Kinetics (with Mars Z. Faizullin) (in 2005). These talks were supplemented by contributions of coworkers of Vladimir Pavlovich giving in this way an overview on the enormous work done or initiated by him and his students and colleagues.

Here I would like to mention also the contributions given by the Corresponding Members of the Russian Academy of Sciences, Vladimir P. Koverda, V.G. Baidakov, M.Z. Faizullin, V.N. Skokov, G.Sh. Boltachev, E.D. Nikitin, A.V. Reshetnikov, S.P. Protsenko, V.E. Vinogradov, A.V. Vinogradov, A.E. Galashev, D.V. Volosnikov, and A.O. Tipeev. These discussions have been continued at the 3rd, 4th, and 5th Russian workshops on *Metastable States and Fluctuation Phenomena* in 2005, 2007, and 2017, correspondingly, all of them organized at the ITP in Yekaterinburg. These results are reflected also to a considerable degree in the proceedings of the workshops *Nucleation Theory and Applications*. The pdf-files of the respective volumes can be obtained from the website (<http://theor.jinr.ru/meetings/2016/>) of the Bogoliubov Laboratory of Theoretical Physics in Dubna or directly from the author of the present contribution. By the way, one of the participants of one of these meetings mentioned in a personal conversation that, provided he would have known in advance the broad spectrum of problems tackled and results obtained by Vladimir Pavlovich and coworkers, he never would have even started to work on nucleation and related phenomena.

Simultaneously, Vladimir Pavlovich took a very active part in the discussion of the contributions of other participants. In joint effort with Ivan S. Gutzow and Vitali V. Slezov he acted as one of our “aksakals” (see Fig. 1) promoting by their evaluation the search for the correct solutions of partly highly complex problems and redirecting the research, if required, to more perspective problems or ways of solving them. This was always done by him in a very correct



**FIG. 1:** Academician Vladimir P. Skripov at the 3rd Russian workshop on *Metastable States and Fluctuation Phenomena*, Yekaterinburg, 2005 (left), Academician of the Bulgarian Academy of Sciences, Ivan S. Gutzow (middle part), and the Corresponding Member of the Ukrainian Academy of Sciences, Vitali V. Slezov, at the research workshop on *Nucleation Theory and Applications*, Dubna, 2004 (right)

form and promoted significantly the constructive and friendly atmosphere at the meetings in Dubna. Some photos of Vladimir Pavlovich made in the course of the Dubna meetings are shown in Fig. 2.

It was also always a pleasure to see him acting in organizing science at the ITP, in the highly accurate manner he was responding to proposals to act as a referee or to promote somebody for different purposes. In addition, it was also always a pleasure to discuss with him problems not directly related to science but anyway of huge importance like different modifications to prepare tea correctly in Uzbek-like style, the most proper selection of shoes in order to cope effectively with certain problems of Russian streets or the choice of the most appropriate types of beer available at that time in Dubna. His general outstanding attitude to his colleagues is, as it seems to me, clearly expressed in one of his poems entitled by the name of a small flower (“nesabudka” in Russian or “forget-me-not” in English translation) which was prepared by Ira G. Polyakova, St. Petersburg. It was used by me for the first time in the cover page when editing the 3rd volume of the Dubna-Proceedings. The cover art from those proceedings can be seen in Fig. 3. Both his scientific achievements and his attitude to his coworkers and colleagues are, in this way, appreciated there.

Quite often, the activities in Dubna and Yekaterinburg went considerably beyond the current problems analyzed. For example, a whole spectrum of topics, Vladimir Pavlovich discussed with Dmitry Yu. Ivanov, found their reflection in the monograph written by D.Yu. Ivanov (2008). With Rainer Feistel, different aspects were discussed concerning the possible incorporation of the work of Vladimir Pavlovich and his coworkers into the activities of the International Association for the Properties of Water and Steam. As one consequence, the results of common work and an overview on the work in Yekaterinburg on the properties of water have been prepared as contributions to the 15th International Conference on the Properties of Water and Steam in Berlin, Germany, in September 2008 (Baidakov, 2008; Schmelzer and Baidakov, 2008).

Much too early these discussions, pleasant meetings, and common work with Vladimir Pavlovich were interrupted. As I already noted in one of the Dubna-Proceedings in Schmelzer (2008): *“It has been always a great pleasure for the author of the present paper to have the possibility to discuss with Vladimir P. Skripov problems of common scientific interest either in Yekaterinburg or . . . at our meetings on Nucleation Theory and Applications at the Bogoliubov Laboratory of Theoretical Physics at the Joint Institute for Nuclear Research in Dubna near Moscow. Unfortunately, we do not have such opportunity any more. However, even at such conditions . . . his advice will continue to guide our research and to assist in the solution of a variety of research problems also in future.”* Below I will briefly discuss some of the directions of research which highly benefitted from the work and discussions with Vladimir Pavlovich and/or performed in direct cooperation with his colleagues.

### 3. SOME PARTICULAR DIRECTIONS OF COMMON RESEARCH INTERESTS

#### 3.1 Classical Nucleation Theory and Possible Generalizations

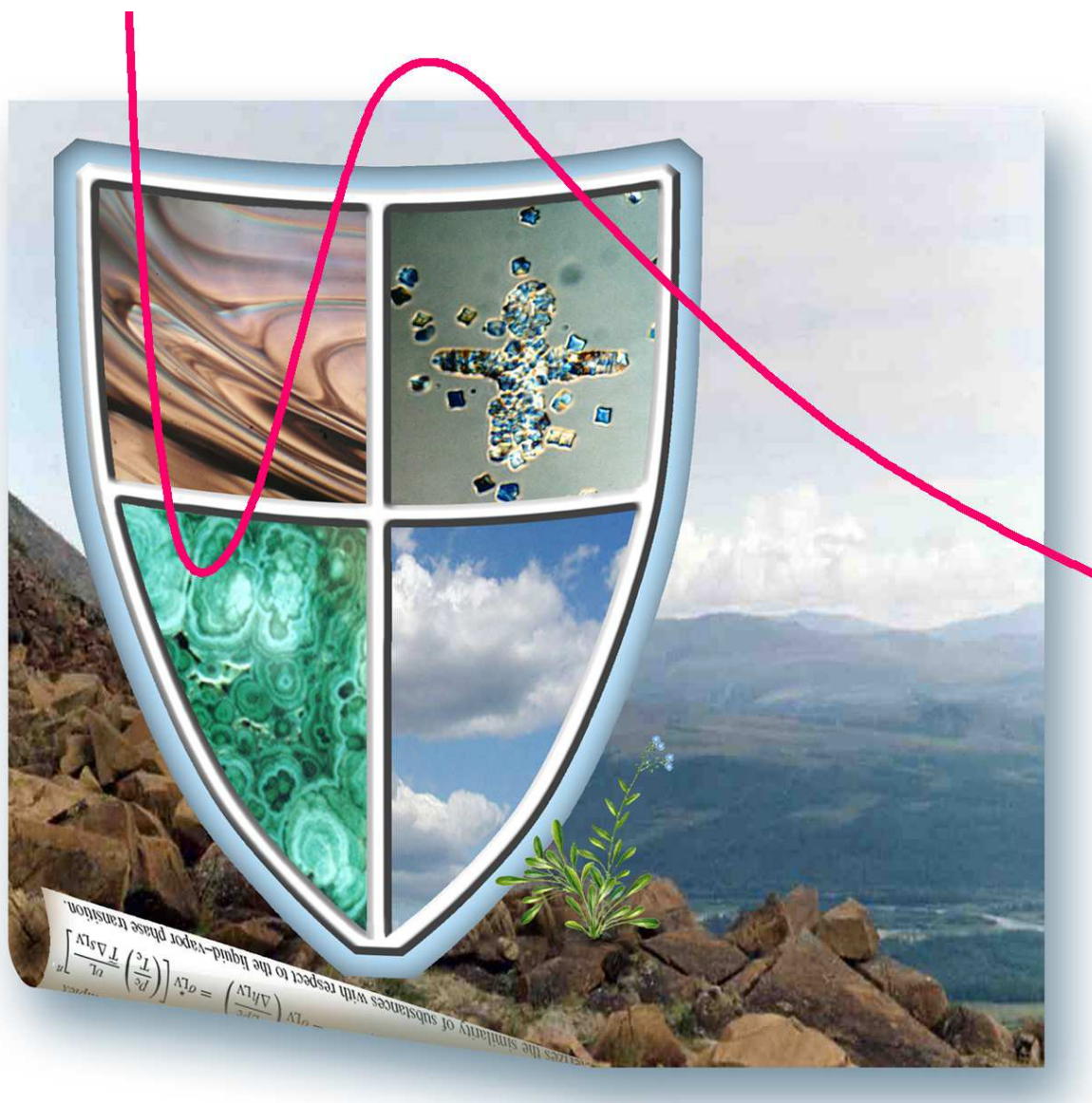
Having completed at the time of our first meetings the work on foam formation in polymeric liquids, it was, of course, highly interesting to learn what could have been added knowing more about the research performed in such direction in Yekaterinburg. But since this project was completed, some of the common activities were concentrated soon onto another problem resulting from the already-mentioned BASF project. As it turned out in this analysis, the standard methods of correction of the theoretical results on steady-state nucleation rates in order to make them comparable with experimental data did not work satisfactorily. A principally new approach was needed we denoted later as the generalized Gibbs approach. Its essence is described in detail in Gutzow and Schmelzer (2013), it is a generalization of the classical Gibbs’ approach. The development of such generalization is not an easy task as evident, for example, by a statement of Lord Rayleigh noting that *“Gibbs’ theory is too condensed and too difficult for most, I might say all readers.”* Fortunately, this does not hold for the colleagues in Yekaterinburg, which led us finally to a successful completion of this task formulated in a comprehensive form first in Schmelzer et al. (2006) in a common paper with G.Sh. Boltachev and V.G. Baidakov.

In line with a growing amount of experimental data, results of computer modeling, and density functional approaches the generalized Gibbs’ approach leads to the following conclusion: The critical clusters, corresponding to the saddle point of the thermodynamic potential surface determined via the generalized Gibbs’ approach, have, in





**FIG. 2:** The author remembers with great pleasure and admiration the talks of Vladimir Pavlovich and the discussions with him at the Research Workshops on *Nucleation Theory and Applications* at the Bogoliubov Laboratory of Theoretical Physics of the Joint Institute for Nuclear Research in Dubna, Russia, in the years 1999–2005. The photos above are made by Alexander S. Abyzov in 2004 showing Academician Vladimir P. Skripov with Dmitry Yu. Ivanov (St. Petersburg, Russia), Rainer Feistel (Rostock, Germany), and Jörn W.P. Schmelzer (Rostock, Germany and Dubna, Russia) (from left top to right bottom) and with the participants of one of the workshops in Dubna in 2001 (bottom)



**FIG. 3:** Artwork of the cover page of the 3rd volume of the conference proceedings *Nucleation Theory and Applications* honoring the work of Academician Vladimir P. Skripov and coworkers “somewhere” in the Ural mountains

general, bulk properties significantly different from the properties of the newly evolving macroscopic phase. One particularly interesting result is the following: According to the classical Gibbs’ approach, the temperature in the critical cluster is equal to the temperature of the surrounding ambient phase. The generalized Gibbs’ approach predicts a different result, the temperature in the critical cluster is, as a rule, different from the temperature of the ambient phase. The problem, which of the mentioned predictions is the correct one, has been intensively discussed, among other occasions, at the research workshop on *Nucleation Theory and Applications* in Dubna, Russia, in 2004 (for details, see Schmelzer, 2008). In the course of this discussion, Vladimir P. Skripov made the following remark: “*I suppose it to be quite possible that equality of temperature is not a necessary condition of thermal equilibrium*” (of course, in application to critical cluster formation, i.e., to clusters of nanometer sizes governing nucleation). This point of view was met by a negative response by Vitali V. Slezov and also Ivan S. Gutzow expressed some critical comments.

So, also our “aksakals” could have from time to time different opinions and the question then is, how to resolve this dispute?

As one of the possible paths of resolution, one could apply general considerations like the first Clarke’s law. It states: “*If an elderly and very distinguished scientist says that something is possible he is almost certainly right, but if he says that it is impossible he is very probably wrong.*” So, Clarke’s law favors the point of view of Vladimir P. Skripov in support of the mentioned consequences of the generalized Gibbs approach. Of course, a more direct proof would be highly desirable. Such analysis was performed in Boltachev and Schmelzer (2010); Schmelzer et al. (2013) in cooperation with A.S. Abyzov. At the time when these topics were analyzed first in detail, it was very helpful to get some response in support. Meanwhile the generalized Gibbs’ approach is already entering not only my own (Gutzow and Schmelzer, 2013) but also modern textbooks of other authors like Neuville et al. (2017). To repeat, without the cooperation with Vladimir G. Baidakov and G.Sh. Boltachev these developments could not have been advanced. By the way, in Boltachev and Schmelzer (2010) we discussed also possible definitions of temperature for small systems and the intensively and controversially analyzed question whether temperature fluctuations may exist or not. Somewhat later our point of view was reconfirmed by experimental investigations by Chua et al. (2017).

### 3.2 Pressure Dependence of the Viscosity

In 2004, in connection with experimental investigations on pressure-induced nucleation, I was asked by experimentalists to supply them with a most suitable for their purposes relation for the description of the dependence of the Newtonian viscosity on pressure. This is a highly interesting problem of high relevance for the understanding of pressure induced nucleation and glass transition caused by pressure variations which may proceed similarly to vitrification by cooling of glass-forming liquids.

As well-known, a variety of investigations exists concerning the temperature dependence of the viscosity. Despite quantitative differences in the details, all of them show clearly that viscosity always increases with decreasing temperature. The situation with respect to the pressure dependence of viscosity is much more complex and much less studied. Consequently, we tried to develop our own equation allowing one to understand the basic general trends and also peculiarities like the decrease of viscosity with increasing pressure observed at certain conditions. Based on such considerations, in Schmelzer et al. (2005) a relation was derived for the description of the interrelation between variations of the viscosity,  $\eta$ , in dependence on temperature,  $T$ , and on pressure,  $p$ . The work described in Schmelzer et al. (2005) and continued in Schmelzer and Abyzov (2017b) in the present volume was significantly facilitated by discussions with Vladimir P. Skripov and the acquaintance with his work, in particular, with the monograph written by Skripov and Faizullin (2006). The way how their ideas were employed are described in mentioned papers.

### 3.3 Existence and/or Absence of Spinodal and Pseudo-Spinodal Curves

Several decades ago, the question was intensively discussed concerning the precision of the localization of the spinodal curve in liquid–gas phase transformations or segregation processes in solutions. This question was motivated by problems in its statistical–mechanical determination and the absence of specific features in the phase transformation kinetics in the vicinity of the spinodal curve when modeling it by Monte-Carlo simulations. In this respect, intensive experimental investigations of the thermodynamic properties of metastable systems and their extension to higher degrees of deviation from the respective binodal curves I consider as highly important. As a result of such experimental studies and their analysis it was well-established that the spinodal does have a well-defined physical meaning and can be determined uniquely (Baidakov, 1994).

Another very important result in the analysis of melt crystallization is the establishment of the absence of a spinodal in one-component melt crystallization as found out for the first time by Skripov and Baidakov (1972). I came in direct contact with these problems in the recent analysis of a paper by Walter Kauzmann (Kauzmann, 1948) intensively discussed till now in glass science. In this paper, Kauzmann wrote: “*Suppose that when the temperature is lowered a point is eventually reached at which the free energy barrier to crystal nucleation becomes reduced to the same height as the barriers to the simpler motions. . . At such temperatures the liquid would be expected to crystallize just as rapidly as it changed its typically liquid structure to conform to a temperature or pressure change*”



*in its surroundings. . . There are good theoretical reasons for believing in the existence of such a ‘pseudo-critical temperature’.*” Kauzmann distinguishes such pseudo-critical states from critical points or states along the spinodal curve noting: *“In the past there has been a considerable amount of speculation concerning the existence of a critical point between crystalline and liquid states analogous to the critical point between liquids and gases. No experimental evidence for or against such a critical point has ever been found Simon (1937), though there is reason to believe that none is possible (Bernal, 1937); but see Frenkel (1946). . . . It is apparent, however, that the behavior with which we are here concerned has a certain similarity to the behavior at a critical point in that here, as at a true critical point, the free energy barrier between the crystal and the liquid disappears. On the other hand, there is a fundamental difference in that the two states do not really merge and their free energies are decidedly different . . . , so that one cannot go reversibly from the one state to the other without a normal phase change.”*

Kauzmann states here that the problem of existence or absence of a spinodal is a very important one having, however, at the time when he performed his analysis (1948) no definite solution. This statement underlines the importance of the mentioned above result established by Skripov and Baidakov (1972). He supposes the possibility of existence of a curve with partly similar properties denoting it as the pseudo-spinodal. In our analysis (Schmelzer and Abyzov, 2017a; Schmelzer et al., 2017a) it was shown that (i) a spinodal in melt crystallization does not exist also in any multi-component system as far as one basic assumption of classical nucleation theory is fulfilled, i.e., when the crystallites have the composition of the newly evolving macroscopic phase. Further it was shown that (ii) a pseudo-spinodal with properties as supposed by Kauzmann generally does not exist. And, finally, it was demonstrated that (iii) the so-called Kauzmann paradox is not in conflict with basic laws of nature, such conflicts are prevented either by (normal) crystallization or by a (conventional) glass transition.

#### 4. CONCLUDING REMARKS

Science plays a well-defined role in society fulfilling well-known tasks. But this is not the only motivation for scientific research. As noted by Henri Poincaré: *“The Scientist does not study nature because it is useful to do so. He studies it because he takes pleasure in it; and he takes pleasure in it because it is beautiful. If nature were not beautiful, it would not be worth knowing and life would not be worth living.”* But there also exists another source of motivation to deal with scientific problems, the possibility to come into direct contact with people like Vladimir Pavlovich. The remembrance of him, his work, and personality, is always a not-to-forget source of deep delight and admiration.

#### ACKNOWLEDGMENTS

Finally, I would like to express my deep gratitude to the colleagues and friends from the Institute of Thermal Physics in Yekaterinburg for their kind hospitality in the course of my visits for now about twenty years. Hopefully, our cooperation can be continued also in the future. I would like to express also my gratitude to Dmitry Yu. Ivanov (St. Petersburg, Russia) for his support in the preparation of the present paper.

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