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Cold Fusion Phenomenon in Open, Nonequilibrium, Multi-component Systems – Self-organization of Optimum Structure

Hideo Kozima

421-1202 Yatsu, Aoi, Shizuoka, 421-1202 Japan

Abstract

The cold fusion phenomenon (CFP) is known to occur only in specific systems (CF materials) satisfying several necessary conditions. One of the most important characteristics, which is sometimes put aside not taken into consideration explicitly, is that the system is open, nonequilibrium, multi-component one where complexity reveal various characteristics including the self-organization to realize an optimum structure favorable to some effects. There have been discovered three empirical laws in the CFP; (1) the stability effect on nuclear transmutation products, (2) the $1/f$ dependence of the frequency of occurrence on the intensity of excess energy production, and (3) bifurcation of the intensity of events (neutron emission and excess energy production) in time. Referring to the suggestion given by the three laws that the cold fusion phenomenon belongs to the complexity induced by nonlinear interactions between agents in CF materials (systems composed of regular arrays of transition metals interlaced with those of hydrogen isotopes), such characteristics of the CFP confirmed by experiments as the threshold value of the ratio of D/Pd or H/Ni about 0.8, favor for higher temperature of the system, occurrence of positive feedback, are explained by self-organization of the superlattice composed of a sublattice of the host element (transition-metal or carbon) and another of the hydrogen isotope (*p* or *d*) where the super-nuclear interaction of lattice nuclei mediated by interstitial hydrogen isotopes works to realize the neutron bands.

1. Introduction

The science of complexity has developed in the last half of the 20th century to give a rather complete perspective of the whole nature including human beings than that depicted by the physical science of simple systems developed since the birth of modern science in the 16th century.

Now, our understanding of nature is not confined to the traditional area of natural science described by linear differential equations but extend to the events determined by nonlinear dynamics without quantitative reproducibility.

As we know from the beginning of the research in the cold fusion phenomenon (CFP), there is no quantitative reproducibility and this fact is used sometimes to denounce the value of the investigation in the CFP. This situation is unreasonable if we recollect the fact that the qualitative reproducibility or probabilistic law is popular in nuclear physics. One of these examples is the α-decay of $^{226}_{88}$Ra nucleus; We can not predict when a specific nucleus $^{226}_{88}$Ra under investigation will decay to $^{222}_{86}$Rn by emission of $^{4}_{2}$He but we know the constant of the decay describing a statistical law for the temporal variation of the number of $^{226}_{88}$Ra nuclei in a system.

It is interesting to notice the existence of several empirical laws or regularities for physical quantities observed in the CFP [Kozima 2006, 2009, 2010, 2012a]. These laws suggest statistical nature of the events in this field. The laws or regularities can be divided into three; (1) the stability effect on nuclear transmutation products, (2) the $1/f$ dependence of the frequency on the intensity of the excess energy production, and (3) the bifurcation of the intensity of events (neutron emission and excess energy production) in time. Recognizing the existence of these laws in this field, we might be able to take a correct point of view for the science of the cold fusion phenomenon (CFP).

In the investigation of events observed in the CFP, we tend to be fascinated by wonderful facts exceeding our common sense in physics and chemistry and to forget fundamental laws in physics underlying the observed events. One of such laws governing the world of the CFP is thermodynamics. Distribution of interstitial atoms (hydrogen isotopes) in an *fcc* (or hcp) lattice of a host metal (e.g. Pd and Ni or Ti) is determined by a condition that the Helmholtz free energy F should be minimum in the equilibrium condition of a closed system. The Helmholtz free energy $F(V, T)$ is expressed by energy E, entropy S and temperature T of the system as

- $F\,(V, T) = E - TS.$ (1.1)

In a closed equilibrium system at a constant volume and temperature, the state with minimum free energy F is the most stable one and realized at last. When temperature T is zero, the most stable state is that the energy E minimum. However, a higher entropy state with a higher energy is favorable if temperature T is finite.

Therefore, the distribution of deuterons in an *fcc* Pd lattice in a PdD system (cf. Fig. 1.1) is not confined only to octahedral sites but also to tetrahedral sites at finite temperature and the regular distribution of deuteron is not realized even when the Pd lattice is perfect. This fact should be remembered to investigate multi-particle reactions for nuclear transmutations in the CFP.

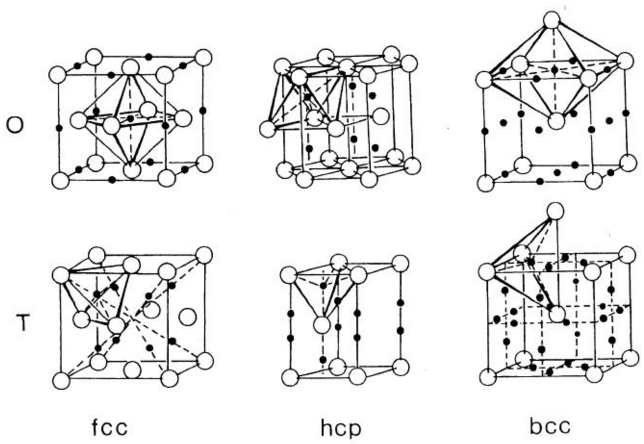


Fig. 1.1 Interstitial sites [octahedral (O) and tetrahedral (T) sites] in *fcc*, *hcp*, and *bcc* lattices [Fukai 2005]

Another fundamental law closely related to events in the CFP is the density dependence of effects in general. It is easy to illustrate several examples of the dependence of an effect *y* on the density *x* of an agent causing the effect with a constant *c* as follows.

1. Single-particle event: $y = cx$ (single-particle events such as the decay of a radioactive nucleus, e.g. α-decay of $^{226}_{88}$Ra)

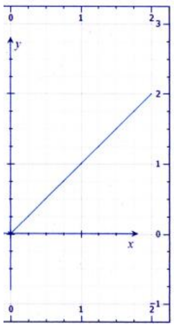


Fig. 1.2 $y = cx$. In the single particle events such as the decay of a radioactive nucleus, the effect *y* of the event (e.g. emission of alpha particle) is proportional to the density *x* of the element (e.g. $^{226}_{88}$Ra) causing the effect with a constant *c*

The effect *y* (e.g. emission of alphas) is proportional to the density *x* of the element (e.g. radium nuclei) in the system.

2. Two-body reaction: $y = cx^2$ (e.g. the reaction of two independent particles as (a) the chemical reaction of H and Cl in an aqueous solution of HCl and (b) *d-d* fusion reactions in PdD_x lattice)

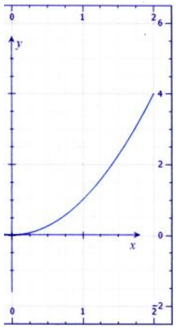


Fig. 1.3 $y = cx^2$. In the cases where the event is caused by two independent elements, the effect y is proportional to the square of the density x of the element with a constant c irrespective of the mechanism causing the event

The effect (e.g. the production of the molecule of HCl in an aqueous solution of HCl or proton and triton pair by a *d-d* fusion reaction) increases as the square (or product) of density (or densities) as shown in Fig. 1.3 without the critical density for the effect. Existence of the critical density in the CFP has been observed in the many experiments and is considered a characteristic of the CFP as illustrated in Fig. 1.7.

3. Four-body reaction: $y = cx^4$ (four-body reaction of independent and equivalent particles in optimum geometrical distribution). Irrespective of the fusion mechanism among four particles (e.g. deuterons) distributed on symmetrical sites, the fusion probability is proportional to x^4 where x is the occupation probability of a particle at the site.

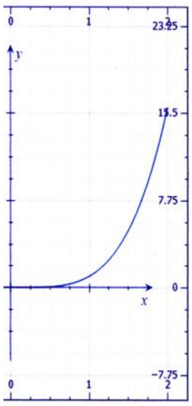


Fig. 1.4 $y = cx^4$. In the cases where the event is caused by four independent elements, the effect y is proportional to the fourth power of the density x of the element with a constant c irrespective of the mechanism causing the event

In this case, the effect remains very small until the density of the agent participating to the reaction reaches at about 0.7 and increases gradually until $x = 1$ and then abruptly. This tendency is rather pronounced in the following cases of six-body and eight-body reactions illustrated only by figures.

4. For six and eight elements (particles) distributed symmetrically, the fusion probabilities are proportional to x^6 and x^8, and are depicted in Figs. 1.5 and 1.6, respectively.

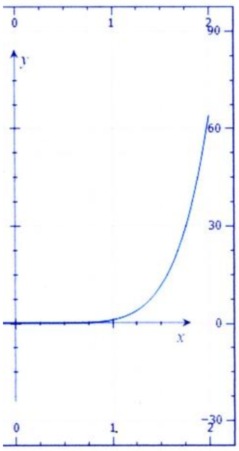


Fig. 1.5 $y = cx^6$. In the cases where the event is caused by six independent elements, the effect y is proportional to the sixth power of the density x of the element with a constant c irrespective of the mechanism causing the event

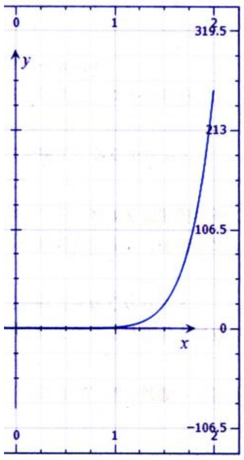


Fig. 1.6 $y = cx^8$. In the cases where the event is caused by eighth independent elements, the effect y is proportional to the eighth power of the density x of the element with a constant c irrespective of the mechanism causing the event

In the cold fusion phenomenon (CDP), we have observed many examples of effect-density relations in this almost a quarter of a century since 1989. The experimental data obtained in this field do not meet with these dependences without criticality shown above in Figs. 1.2 – 1.6 but show the critical behavior that the effect occurs only when the density of H or D in the CF materials exceeds a critical value. This is an obvious indication that the CFP is a cooperative phenomenon but not individual particle one.

An example of typical experimental data of critical behavior showing existence of cooperative phenomenon is the data obtained by an extensive experiment by McKubre et al. [McKubre 1993] shown in Fig. 1.7 where the abscissa is extended to left until D/Pd = 0.77 to show the criticality more clearly. The compound PdD_x has several phases and α' phase stable at room temperature exists from about $x = 0.6$ to x = 1 as seen in Fig. 1.8.

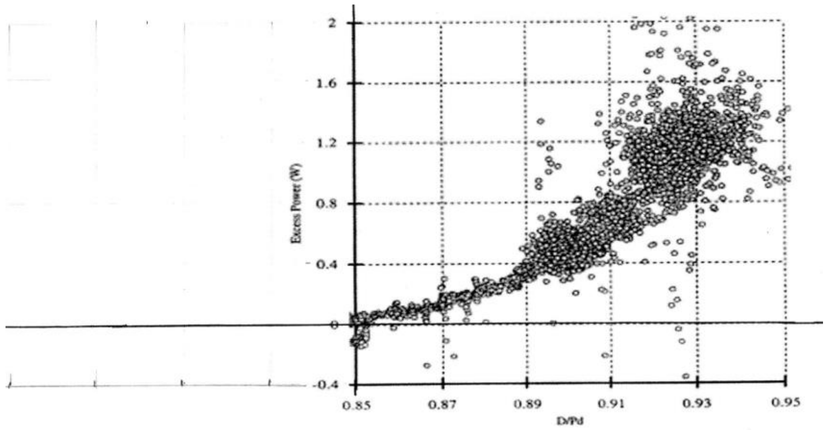


Fig.1.7 Experimental data of excess power vs. D/Pd ratio obtained by McKubre et al. [McKubre 1993]. The abscissa of this figure is extended to left until D/Pd = 0.77 from 0.85 of the original one to show clearly the critical nature of the excess energy generation

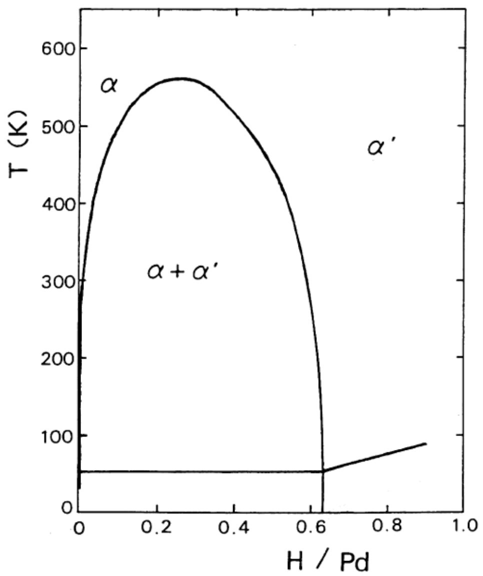


Fig. 1.8 Phase diagram of the Pd-H system which is very similar to that of the Pd-D system [Fukai 2005]

Comparison of Fig. 1.7 with Fig. 1.3 for $y = cx^2$ shows clearly that the reaction resulting in this excess energy generation is not the simple *d-d* fusion reactions often assumed easily in investigations in this field.

Brief survey of the experimental data obtained in the CFP in relation to fundamental laws of physics given above shows us necessity of cooperative consideration for the physics of nuclear reactions in transition metal hydride and deuteride.

We have used phenomenological approach to the CFP for almost more than 20 years using the TNCF model [Kozima 1994] and have given several semi-quantitative explanations for events in this field [Kozima 1998, 2006]. The fundamental premises of the model have been assumed based on the experimental data. The premises on the other hand have been investigated quantum mechanically using such novel knowledge of nuclear physics as exotic nuclei and also of transition metal hydrides as non-local wavefunction of hydrogen isotopes obtained in the last half of 20th century.

One of the fundamental premises of the TNCF model, existence of the trapped neutrons in the metal hydrides, is explained by the neutron band formed as a result of the super-nuclear interaction between lattice nuclei mediated by interstitial protons or deuterons [Kozima 2004, 2006]. As was shown in the trials for neutron band formation, the super-nuclear interaction between neutrons in different lattice nuclei mediated by interstitial protons or deuterons gives an essential effect in the CFP. The super-nuclear interaction, the key concept of the calculation is realized when the superlattice is formed where the sublattice of host metals is interlaced by a sublattice of proton (deuteron) on interstitials. This optimum structure may be realized by self-organization mechanism in the open, non-equilibrium system locally in the CF material but not as a whole. The localization of nuclear reactions observed in the CFP will be related to the localized formation of the optimum superlattice by the self-organization.

We will show indirectly the possible formation of the superlattice by self-organization mechanism in the CFP using known facts related to the complexity.

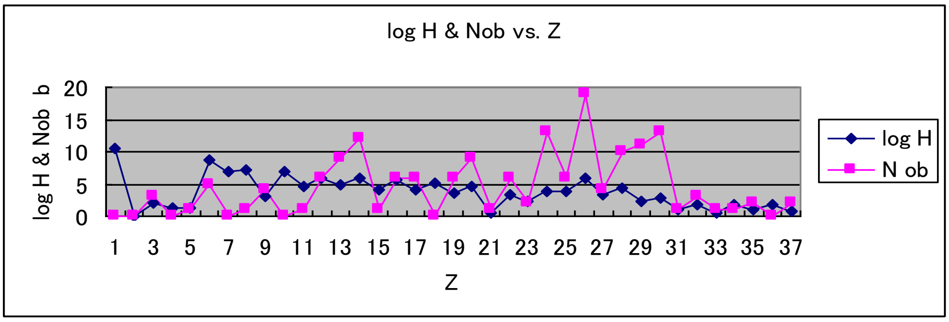
2. Three Empirical Laws in the CFP deduced from Experimental Data and Their Explanation by Nonlinear Dynamics

In the vast amount of information about events of the CFP obtained in these more than 20 years since 1989, we can recognize several regularities or laws between observables in the CFP. The three laws we have figured out are specified as follows [Kozima 2006, 2008a, 2009, 2010, 2012a]; (1) The stability effect on nuclear transmutation products, (2) the $1/f$ dependence of the frequency of observation on the intensity of the excess energy production, and (3) the bifurcation of the intensity of events (neutron emission and excess energy production) in time. We give a brief explanation for them in this section.

2.1 Stability Law on Nuclear Transmutation Products

If we survey numbers of a specific element produced by the nuclear transmutation in the CFP, we notice that the frequency obtaining the element corresponds to the amount of the element in the universe (e.g. [Suess 1956]). Plotting (i) the number of experiments $N_{ob}(Z)$ where observed an elements ${}_{Z}X$ according to its proton number Z together with (ii) the relative amount $H(Z)$ of the element in the universe in logarithmic scale, we obtain a diagram shown in Fig. 2.1 [Kozima 2006, 2012a]. The coincidence of the numbers (i) and (ii) gives the stability effect on nuclear transmutation products. We may call this regularity the “stability law” for nuclear transmutation in the CFP.

(a)



(b)

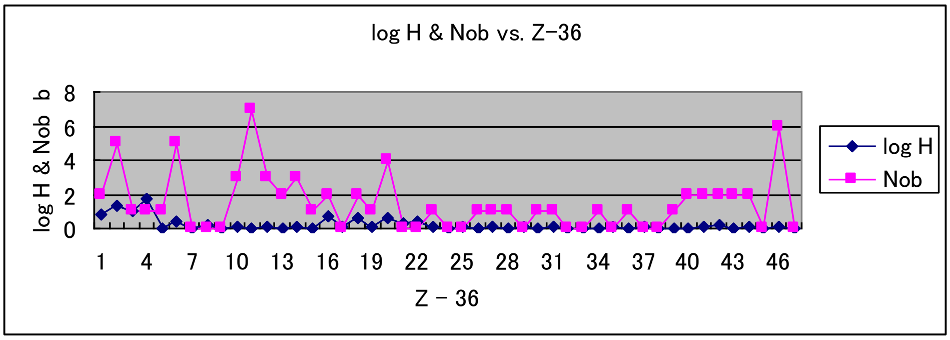


Fig. 2.1. Correspondence between the frequency $N_{ob}(Z)$ observing elements in the CFP and the relative abundances $\log_{10}H(Z)$ of elements in the universe [Suess 1956]: (a) Z = 3 – 38 and (b) Z = 39 – 83 [Kozima 2006]

Another example of this law has been observed by several researchers including Hora et al. [Hora 1998] as shown in Fig. 2.2.

The maxima of $N(Z)$ in many experimental data sets including the data shown in Fig. 2.2 [Hora 1998] rather agree with the magic numbers with exception of the magic number 20 where a clear minimum of $N(Z)$ was observed in all cases. This coincidence of the maxima of $N(Z)$ and the magic numbers is another example of the stability effect on the nuclear transmutation in the CFP. The exceptional case of Z = 20 needs another factor for explanation.

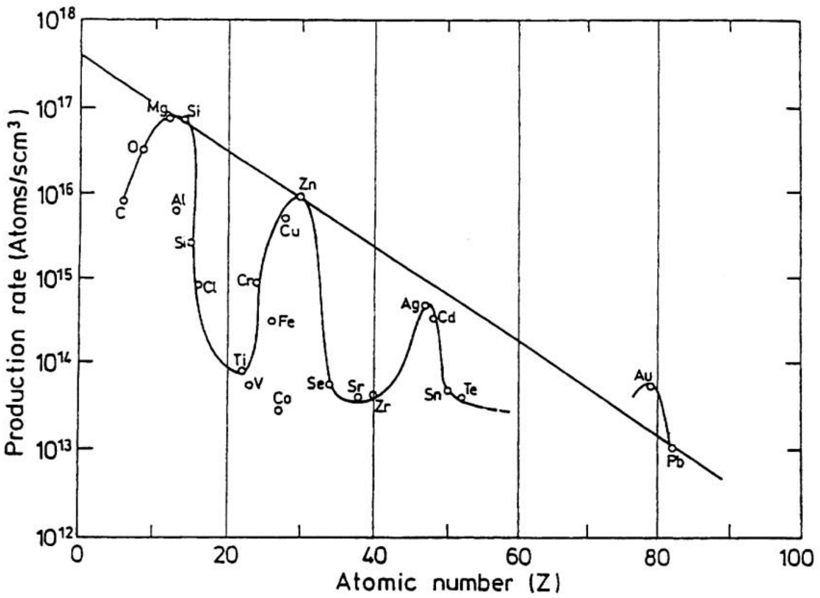


Fig. 2.2 Measured production rate $N(Z)$ of the atom with a proton number Z for the nuclear transmutation vs. the atomic number Z in PdH_x where exponential decay of the maxima on Z follows a relation of an equation $N(Z) = N'\exp(-Z/7.86)$ with $N' = 3.56 \times 10^{17}$ atoms/cm^{3}s [Hora 1998 (Fig. 1)]. The maxima of $N(Z)$ agree with the magic numbers except $Z = 20$.

This law (stability law) shows that the stability of a nucleus reveals its nature in the cf-material composed of high density neutrons in the neutron valence band [Kozima 2006 (Sec. 2.4.2)] and spring out as a nucleus just as in the case of nuclear transformation in stars

2.2 Inverse Power Dependence of the Frequency of Observations on the Intensity of Excess Energy Production

In several experimental data sets, we are able to count numbers N_Q of experiments where observed a specific amount Q of excess energy per a definite number of host atoms in CF materials. When we plot them as a function of Q, we obtain N_Q vs. Q plots [Kozima 2006 (Sec. 2.12), 2008a, 2008b, 2012a]. The first plot was obtained for the data by McKubre et al. [McKubre 1993] as shown in Fig. 2.3. This plot clearly shows that there is an inverse power relation of frequency vs. intensity with an exponent of 1.0 famous in the field of complexity [Milotti]. This regularity may be called the "inverse power law" for the frequency on the intensity of the excess energy production.

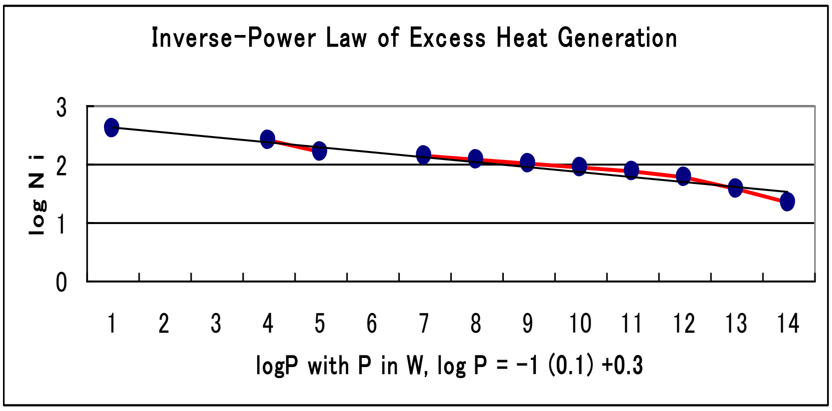


Fig. 2.3. Inverse power law revealed by excess power generation measured by McKubre et al. [McKubre 1993]

Another example of this law is obtained for the data obtained by Kozima et al. [Kozima 2008c] as depicted in Fig. 2.4. In this case, the exponent of the dependence is 2.

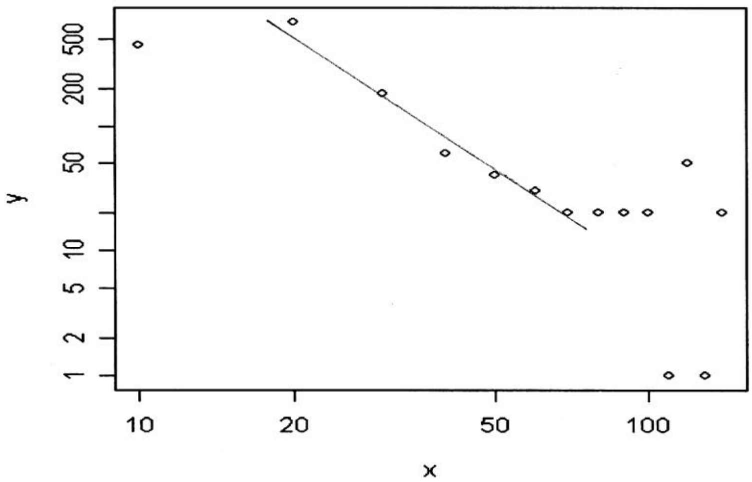


Fig. 2.4. Distribution of the frequency N_p (= y) producing excess power P_{ex} (= x). To depict log-log curve, values of N_p and P_{ex} were arbitrarily multiplied by 10^n (x = 100 in this figure corresponds to P_{ex} = 1 W) [Kozima 2008c]

On the other hand, H. Lietz [Lietz 2008] tried to check the inverse power law using the data accumulated by E. Storms in his book [Storms 2007]. The resulting plot by H. Lietz is given in Fig. 2.5 which shows the exponent of 1.0.

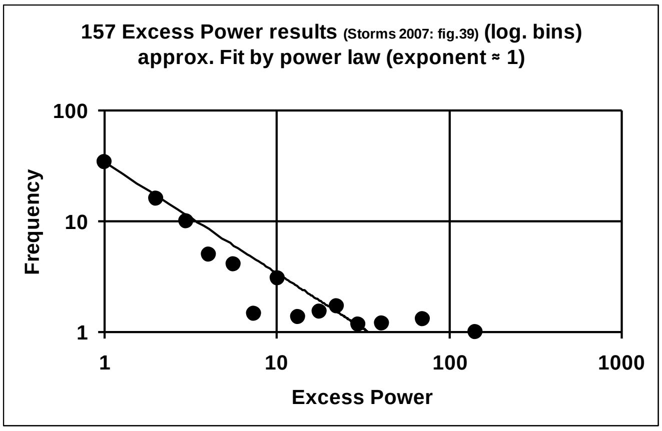


Fig. 2.5. Distribution of 157 excess energy results by Lietz [Lietz 2008] using the data collected by Storms [Storms 2007]. Values have been stored in bins of size 10. The line shows a power-law fit to the binned data with an exponent of 1.0 (r^2 = 90%) (Fig. 3 of [Lietz 2008])

Therefore, we may conclude that the excess energy generation in the CFP satisfies the inverse power law and is governed by a statistical law popular in complexity.

2.3 Bifurcation of the Intensity of Events (Neutron Emission and Excess Energy Production) in Time

The third law in the CFP is a little subtle compared with the former two [Kozima 2012a].

Even if the number of examples is scarce, we have several fortunate data sets of temporal evolution of effects in the CFP. The first one is that of neutron emission from TiD_x obtained by De Ninno et al. in 1989 [De Ninno 1989]. The data are shown in Figs. 2.6 and 2.7.

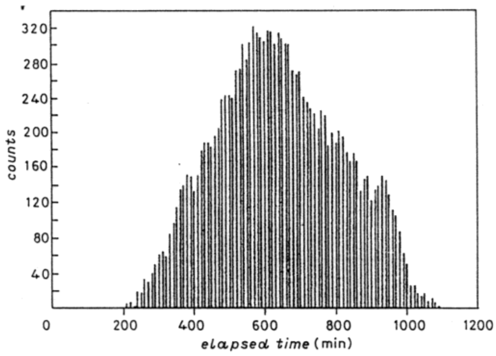


Fig. 2.6. Diagram showing the time evolution of the neutron emission from TiD_x sample during the run *A* (April 15-16, 1989). The values indicated are integral counts over periods of ten minutes [De Ninno 1989]

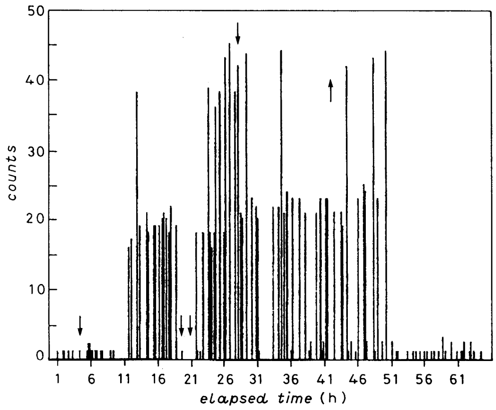


Fig. 2.7 Diagram showing the time evolution of the neutron emission counts (ordinate) during the run B (7-10 April, 1989) by De Ninno et al. The values indicated are integral counts over periods of 10 minutes [De Ninno 1989]

Another data set is the excess energy generation observed by McKubre et al. as shown in Fig. 2.8 [McKubre 1993].

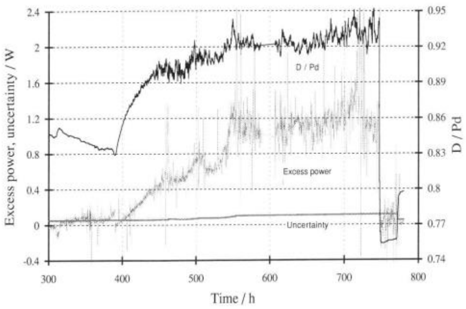


Fig. 2.8 Variations of Excess Power, Uncertainty and Loading ratio [McKubre 1993].

Furthermore, we can cite another example of the temporal evolution of excess energy generation measured by Kozima et al. in Fig. 2.9 [Kozima 2008c].

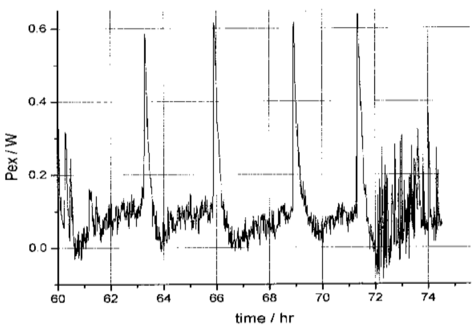


Fig. 2.9 Excess power pulses during a 14 hour period of an experiment (070108) of Kozima et al. which lasted 12 days as a whole [Kozima 2008c]

By the nature of events in complexity, we can give only qualitative explanation of experimental result [Kozima 2008a, 2008b] in analogy to the mathematical results of numerical calculations using the logistic difference equation [Gleick 1987]. The analogical explanations of the laws observed in the CFP will be given in the next section using the nature of an equation of nonlinear dynamics and Feigenbaum's theorem [Feigenbaum 1978].

3. Cold Fusion Material as Open, Nonequilibrium, Multi-component System

We have had enough experience to deduce a few general conclusions on the behavior of cold fusion materials where occurs the cold fusion phenomenon (CFP), i.e. phenomenon including nuclear reactions and accompanying events occurring in solids with high densities of hydrogen isotopes (H and/or D) in ambient radiation belonging to Solid-State Nuclear Physics (SSNP) or Condensed Material Nuclear Science (CMNS).

Several of their characteristics may be listed up as (1) co-existence of host transition metals (or noble metals or carbon) and hydrogen isotopes (protium and/or deuterium), (2) higher ratio of hydrogen to host concentrations, (3) nonequilibrium situation, (4) existence of thermal neutrons, (5) higher temperature of the sample, (6) positive feedback to enhance effects [Kozima 2012b].

The similarity of some behaviors of events in the CFP to those of the nonlinear dynamics shown in the preceding section gives us temptation to investigate the cold fusion materials from a viewpoint related to nonlinear dynamics resulting in complexity.

Some characteristics of complexity we have met in natural phenomena are listed up as follows in relation to the CFP. (i) Self-organization in open, nonequilibrium systems (cf. Sec. 3.3), (ii) bifurcation of possible states (cf. Sec. 3.2), (iii) chaotic behavior of events lacking quantitative reproducibility (cf. Sec. 3.2). The well-known examples of the first are the convection (Bénard) cells in fluid dynamics and Zhabotinsky reaction in chemical reaction. An example of the bifurcation is given by a simple logistic difference equation (l.d.e.) as shown in Fig. 3.4 below. A typical example of the third characteristic is the so-called "butterfly effect" showing uncontrollable effects of a minor change of a part of a system on the tremendous change on the long-range behavior of the system.

We investigate the characteristics of CF systems from the viewpoint of complexity.

3.1 High D/Pd and H/Ni Ratios and High Temperature of Samples

It is fairly well known that it is necessary to have a higher D/Pd (or H/Ni) ratio than a critical (or threshold) value around 0.8. One of the best known examples was given very early in this field by McKubre et al. (cf. Fig. 1.7) [McKubre 1993]. On the other hand, as we know from many experimental data, it is desirable to have higher temperature of samples to realize pronounced effects in the CFP (e.g. [McKubre 1993]). One of the recent data is given by Celani et al. [Celani 2010].

Furthermore, it is noticed that there is a preferable combination of host metals and hydrogen isotopes; Pd and D, Ni and H, Ti and D are best combinations of elements for the CFP [Kozima 2000].

In the experimental data obtained in metal-hydrogen systems, we have vast number of experimental results concerning properties of hydrogen isotopes in transition metals. We can cite here data on the diffusion of D and H in Ni and Pd as shown in Figs. 3.1 and 3.2, respectively [Birnbaum 1972].

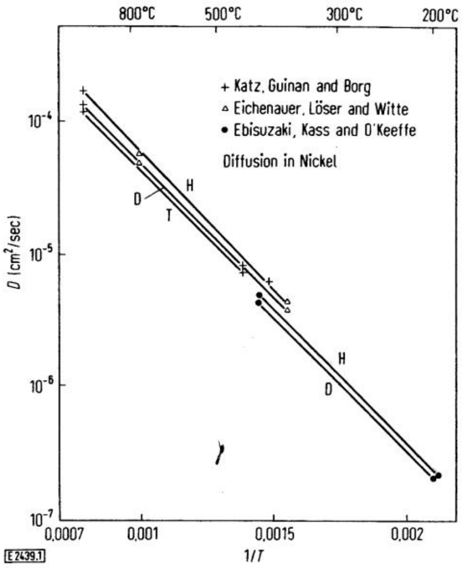


Fig. 3.1 Diffusion coefficients of hydrogen, deuterium and tritium in nickel [Birnbaum 1972]

It is seen in Fig. 3.1 that protium diffuses faster than deuterium in Ni as usually expected by the lighter mass of the former in the whole temperature range from 200 to about 1000 ^{o}C. On the other hand, as we see in Fig. 3.2, the situation in the case of Pd is very different from that in Ni and other ordinary metals. In this case, deuterium is more diffusive than protium in the room temperature range from –73 up to about 200 ^{o}C but is normal in higher temperature range than 200 ^{o}C.

This peculiar behavior of deuterium in Pd is closely related to the CFP in palladium hydrides and deuterides from our point of view. Usually, the CFP is observed in the latter and the former is sometimes used as the blank for the latter. In the higher temperatures than 200 ºC, however, we can expect the CFP in the former as well as latter which will be checked easily. This point has been partially confirmed by Hioki et al. by experiments using Pd nano-particles in zeolite and FSM (Folded Sheet meso-porous Material) [Hioki 2013]. They observed excess energy production in both hydride and deuteride specimens a little higher value in the latter at room temperature. This fact clearly discards the *d-d* fusion reactions as the principal cause of excess energy generation in the CFP which are not effective in transition-metal hydrides.

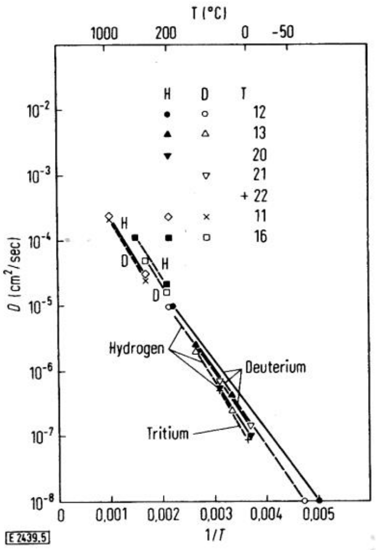


Fig. 3.2 Diffusion of isotopes of hydrogen in palladium [Birnbaum 1972]

Therefore, we have to rely on another mechanism effective for explanation of various events both in deuterium and protium systems such as proposed by us [Kozima 1998, 2006]. The proposed mechanism becomes effective when there is a regular superlattice composed of interlaced sublattices of the host metal (e.g. Pd or Ni) and the hydrogen isotope (D or H). The self-organization of Pd-D or Ni-H lattice in an open, nonequilibrium condition of gas-contact or electrolytic systems serves to realize the superlattice favorable for the mechanism proposed in our papers for the CFP [Kozima 2006 (Sec. 3.7)]. The investigation of the condition to make the self-organization process effective to obtain more complete superlattice structure of interlaced sublattices of host and hydrogen isotope is inevitably qualitative due to the nature of complexity as we know well.

3.2 Positive Feedback for Nuclear Reactions

The change of surface morphology and rare experiences of explosion in CF experiments discussed in other papers [Kozima 2007, 2010, 2011, Smedley 1993] may be an evidence of positive feedback of a parameter (or parameters) for the CFP.

It is possible to contemplate a mechanism of positive feedback of nuclear reactions, if we can identify the parameter $n_{\rm n}$ in the TNCF model with the variable x_n of the logistic difference equation (l.d.e.);

$$x_{n+1} = b\, x_n(1 - x_n) \quad (n = 0, 1, 2, ---) \qquad (3.1)$$

used to describe the bifurcation diagram by J. Gleick [Gleick 1987] (cf. Fig. 3.4).

The scenario goes as follows. By any chance, we have a situation in a CF material in which cf-material is formed in a localized region at surface layers. An alien nucleus in one of the regions interacts with a neutron or a neutron-proton cluster $^A_Z\delta$ and liberate a definite energy [Kozima 2011]. The liberated energy heats up the temperature of the region increasing the value of $n_{\rm n}$. The elevated value of $n_{\rm n}$ increases the reaction probability and the temperature is elevated further. However, this feedback process does not continue without limit. If the temperature becomes too high, the favorable superlattice of the interlaced sublattices of host and hydrogen isotope is destroyed by thermal motion of the nuclei. This destroys the super-nuclear interaction between adjacent lattice nuclei and decreases $n_{\rm n}$. This variation of the parameter $n_{\rm n}$ should be similar to the variation of the recursion function $f(p)$ depicted in Fig. 3.3 (cf. Eq. (3.3)).

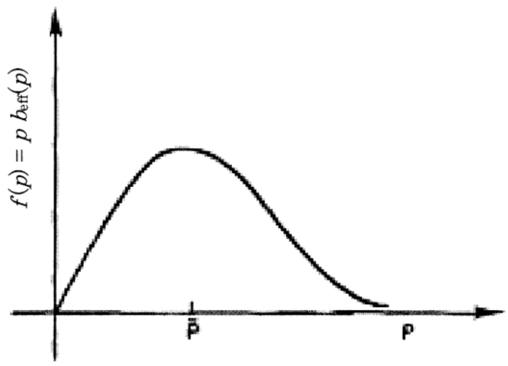


Fig. 3.3 Dependence of the recursion function f in the recursion relations $f(p) = p \cdot b_{\rm eff}(p)$ on the variable p [Gleick 1987]

The relation

$$f(p) = p \cdot b_{\rm eff}(p) \qquad (3.2)$$

depicted in Fig. 3.3 reduces to the l.d.e. (3.1) $x_{n+1} = b\, x_n(1 - x_n)$, if we take

$$b_{\rm eff}(p) = b(1 - p). \qquad (3.3)$$

Then, the Feigenbaum's theorem says that the bifurcation diagram depicted in Fig. 3.4 describes possible behavior of the CFP assimilated by Eq. (3.1). The parameter b in the l.d.e. (3.1) may be determined by atomic and nuclear states of the surface region of CF materials. The appearance of different features in the CFP as shown in Figs. 2.6 –

2.9 corresponds to different values of the parameter b depending on each situation. It will be the next step of our future work to clarify nature of the parameter b in the CFP and to control the phenomenon as freely as possible.

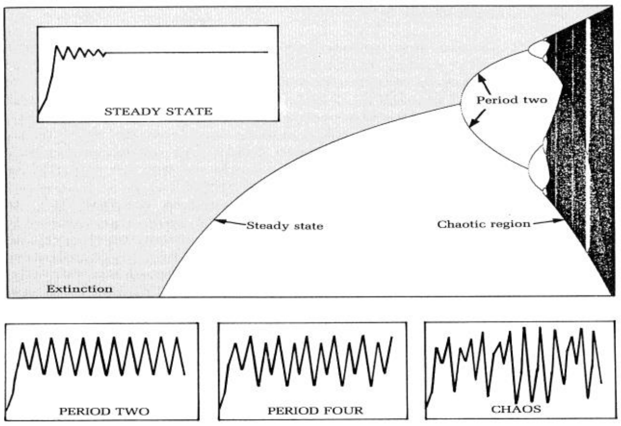


Fig. 3.4 Bifurcation diagrams to show period-doubling and chaos [Gleick 1987]. The main figure depicts x_∞ vs. parameter b, x_∞ on the ordinate (x_∞ is x_n at $n = \infty$) and b of the logistic difference equation, $x_{n+1} = b\, x_n (1 - x_n)$ $(0 < x_0 < 1)$, on the abscissa. The inserted figures, a) Steady state, b) Period two, c) period four, and d) chaos, depict variations of x_n with increase of suffix n (temporal variation if n increases with time) for four values of b; a) $1 < b < 3$, b) $3 < b < 3.4$, c) $b \simeq 3.7$, d) $4 < b$ The region a), b) and d) correspond to “Steady state”, “Period two” and “Chaotic region” in the main figure, respectively [Kozima 2012a]

3.3 Self-organization of Optimum Structure (Superlattice) of CF Materials

In the equilibrium state at $T \neq 0$, a state with the entropy $S > 0$ is favorable even if the energy E of the state is higher than the state with $S = 0$ because the equilibrium condition demands that the Helmholtz free energy $F(V, T)$ is minimum at constant temperature and volume (Eq. (1.1) recited as Eq. (3.5)) :

$$dF = -p\, dV - S dT = 0, \tag{3.4}$$

$$F(V, T) = E - TS. \tag{3.5}$$

In an open, nonequilibrium condition, it is possible to realize a state with $S = 0$, or a state with perfectly regular array of atoms by self-organization at finite temperature. There are several examples of self-organization in physics, chemistry, biology, and others as illustrated below.

In the open and non-equilibrium dynamical system where energy and component materials are fed from outside, there appears patterns energetically unstable compared to the homogeneous structure without any structure. We can give several examples observed in nature as illustrated in Figs. 3.5 – 3.7.

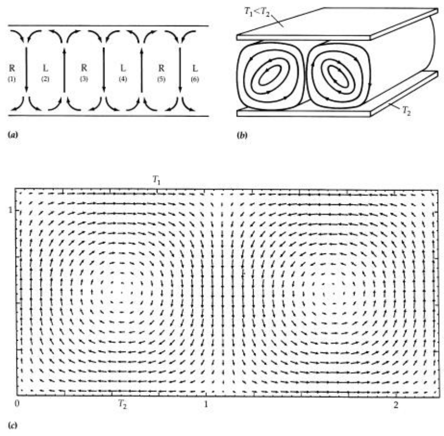


Fig. 3.5 Two views of convection (Benard) cells (a) and (b) and complexity and long-range order out of molecular chaos in a system under nonequilibrium constraint (c) [Nicolis 1989]

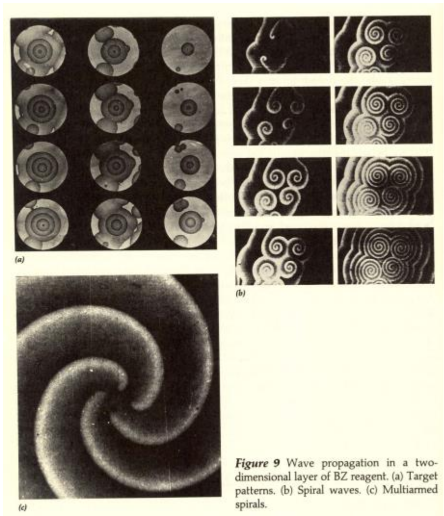


Fig. 3.6 Belousov-Zhabotinski (BZ) reaction [Nicolis 1989]

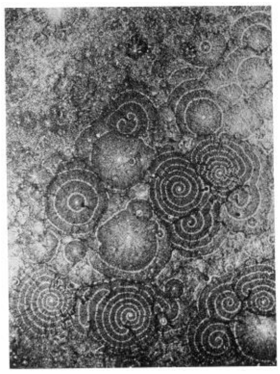


Fig. 3.7 Cell populations of Dictyostelium discoideum on an agar surface [Nicolis 1989]

In the case of the CFP, the optimum superlattice of the host lattice nuclei (e.g. Pd) and hydrogen isotopes (e.g. *d*) is apt to be formed in localized regions at surface layers with a width of about several microns where the number ratio $\eta = N_d/N_{\mathrm{Pd}}$ of the hydrogen isotope *d* and the host metal Pd is expected higher than other parts of the sample. Experimental data show that the formation of superlattice is possible when η becomes larger than ~ 0.8. The superlattice may be localized in small region with a diameter of about several micrometers as experimental facts show.

We can give a schematic structure of the superlattice in Fig. 3.8. In reality, the wavefunctions of hydrogen isotopes (*d* or *p*) centered at interstitials overlap with nuclear wavefunctions of lattice nuclei Pd (Ni) centered at lattice points. Then, the contact nuclear interaction between them makes possible the super-nuclear interaction between neutrons in the lattice nuclei on lattice points as explained below in Fig. 3.9 [Kozima 2006 (Sec. 3.7)].

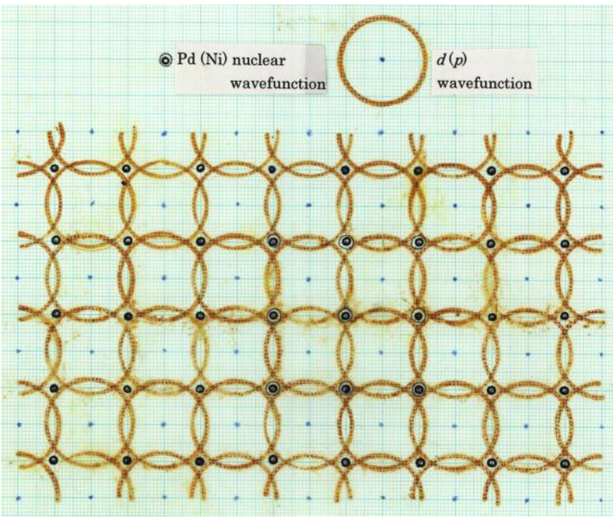


Fig. 3.8 Schematic diagram showing the optimum superlattice of host nuclei (Pd or Ni) at lattice points and hydrogen isotopes (*d* or *p*) at interstitials formed by self-organization in the open, non-equilibrium condition. Nuclear wavefunctions with extension of only a few femtometers of lattice nuclei are exaggerated largely to be seen on the figure. The extension of deuteron (proton) wavefunctions centered at interstitials is represented by a single circle in contact with nuclear wavefunctions at nearest lattice points

When the superlattice is formed at localized regions in the surface layer, it is possible to appear the neutron valence band by the super-nuclear interaction between neutrons in different lattice nuclei mediated by interstitial hydrogen isotopes as illustrated in Fig. 3.9 [Kozima 2006].

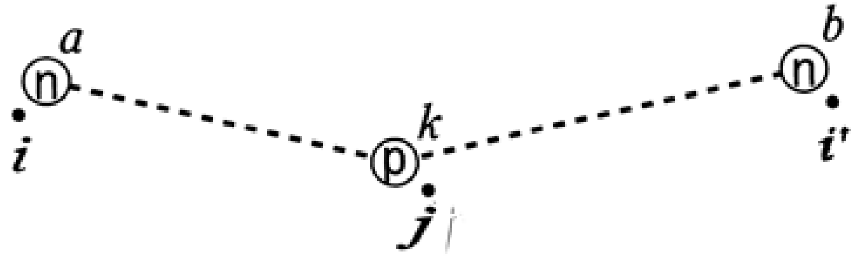


Fig. 3.9 Super-nuclear interaction of two neutrons in different lattice nuclei at site ***i*** an ***i'*** mediated by interstitial protons at sites ***j***'s [Kozima 2006]

The number of states in a neutron valence band formed in a localized cube with side lengths of 10 μm (10^{-5} cm) is about 10^9. So, the neutron state density N_n in the localized superlattice is ~ $10^9 \times 1/(10^{-5})^3 = 10^{24}$ cm^{-3} when the band is fully occupied by neutrons. The TNCF model comparison with experimental data sets has given the neutron number density n_n of about 10^7 ~ 10^{10} cm^{-3} for the CFP [Kozima 2006 (Table 2.2)] which seems not inadequate in view of the number of states in the neutron valence band formed at the localized superlattice.

This ratio $n_n/N_n = 10^{-17}$ ~ 10^{-12} is consistent with the assumption of the TNCF model that the trapped neutron behaves as thermal neutrons in the nuclear reactions with other nuclei (cf. electrons in a conduction band of intrinsic semiconductors).

The choice of the side length of 10 μm (10^{-3} cm) for the localized superlattice is based on the experimental facts that nuclear products observed in the CFP are localized in areas of width about a few micrometers in surface layer of a few micrometer depth. Recent experimental data obtained by Hioki et al. suggest Pd nanoparticles with diameters of a few nanometers occluding H or D atoms have shown excess energy generation [Hioki 2013]. This data may suggest existence of the superlattice of Pd-D and Pd-H in surface regions with rather small diameters of about a few nanometers.

When the optimum superlattice is formed at least locally by the self-organization mechanism as observed in the complexity, the super-nuclear interaction illustrated in Fig. 3.9 generates the neutron balance band and realizes the trapped neutrons in the band as assumed in the TNCF model for the CFP.

4. Conclusion

The non-equilibrium condition, one of the necessary conditions for the CFP in the transition-metal hydrides and deuterides, has been investigated from our point of view that the CFP is a phenomenon realized as a complexity consistently with other knowledge of physics of the CFP related to the situation where occurs various nuclear events in the CF materials.

The three empirical laws [Kozima 2012a] explained in this paper suggest that the cold fusion phenomenon (CFP) is characterized by complexity [Gleick 1987]. One of characteristics of the complexity is qualitative reproducibility which is also recognized as a characteristic of the CFP.

The characteristics of complexity include the self-organization, the bifurcation and the chaos. We have shown examples of experimental events observed in the CFP for the latter two characteristics in our papers [e.g. Kozima 2012a] and also in Sec. 2 of this paper. The first one, however, is not observed directly in the CFP by experiment.

Formation of favorable structure for the CFP in the open and nonequilibrium systems (CF materials) may be closely related to the self-organization of a superlattice (double regular arrays) composed of sublattices of host nuclei and hydrogen isotopes, e.g. PdD or NiH, from our point of view. If this is the true scenario for the CFP, it is understandable that the occurrence of the events in this field is subtle and is only qualitatively reproducible as our experience in this field tells us.

It might be possible to show possible realization of super-nuclear interaction [Kozima 2004, 2006] between neighboring lattice nuclei by computer simulation. Simplified schedule is realized by assuming an *fcc* sublattice of a host nuclei ${}^{A}_{Z}X$ interlaced by another sublattice composed of protons (or deuterons) at octahedral interstitials of the former sublattice. The nucleus ${}^{A}_{Z}X$ has a neutron in the evaporation level with a wavefunction ψ_n extended out beyond ordinary nuclear radius R_0. The proton (or deuteron) centered at an interstitial point is assumed in a level with a wavefunction φ_p (or φ_d) extending out to overlap with the neutron wavefunction ψ_n. The super-nuclear interaction between adjacent lattice nuclei mediated by interstitial proton (or deuteron) is a key factor for the explanation of the CFP common in protium and deuterium systems. It is desirable to check this possibility by simulation to develop the physics of the CFP and will be one of our future projects.

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