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NUCLEAR REACTIONS IN SOLIDS

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NUCLEAR REACTIONS IN SURFACE LAYERS OF DEUTERIUM-LOADED SOLIDS

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Using the concept of the neutron Bloch wave presented previously, the possibility of an effective nuclear reaction of a thermal neutron and a nucleus in a boundary region of crystals is determined. Many experimental data of nuclear transmutations in the surface layer or surface region of solid materials loaded with deuterium supposedly induced by nuclear reactions with a thermal neutron are investigated using the nature of the neutron Bloch wave in solids. The physics of the nuclear reaction phenomena is discussed in the trapped neutron catalyzed fusion model.

I. INTRODUCTION

In the ~10 yr after the discovery[1] of nuclear reactions in solids [the so-called cold fusion (CF) phenomenon] in 1989, we have had many data of various events in this field, from the generation of excess heat, neutrons, and tritium to the generation of ^4He and nuclear transmutations (NTs). These events have shown their characteristics, which are only explained by nuclear reactions in solids including hydrogen isotopes with some other attributes. Furthermore, it has been confirmed that protium is also responsible for the CF phenomenon alternative to deuterium, one of the inevitable key elements supposed in the first stage of research.

One of the characteristics of CF events is the locality of the supposed contributing nuclear reactions. The generated ^4He and tritium have mainly been observed outside the sample solids, showing implicitly that the reaction generating them was in the near-surface region. There is, furthermore, evidence showing explicitly the surface nature of the reactions responsible for these nuclear products.

The detection of ^4He by Morrey et al.[2] and Chien et al.[3] showed a possibility of nuclear reactions generating ^4He in a surface region with a thickness of ~40 μm in the former although the observed rather smaller amount than the excess heat was inconsistent with the initial supposition of a *d-d* reaction. This discrepancy of amounts was resolved by the adequate amount of ^4He detected by Miles et al.[4] in the gas phase in the experiment.

More direct evidence of the surface nature of the CF phenomenon has been given by NTs in surface layers or in near-surface regions. The NTs of electrode metal elements (or metals in electrolyte) to elements with a difference of mass number <5 have been measured by Bush and Eagleton,[5,6] and Bush[7] (Na → Mg, K → Ca, Rb → Sr), Okamoto et al.[8] (Al → Si), Savvatimova et al.[9,10] (Pd → Ag, etc.), Notoya[11] and Notoya et al.[12] ($^{39}K \rightarrow {}^{40}K$, etc.), Dash et al.[13] and Dash[14] (Pd → Ag, Pt → Au), and Yamada et al.[15] (O → C), all in surface layers or on the surface of electrodes. An exception is the data by Passell,[16] where a decrease of boron (B) in the volume of a Pd cathode was detected. We have explained some products in these data as an absorption of a thermal neutron by a preexisting nucleus in the experimental system followed by beta or alpha decay and called the NT by a decay[17,18] and denoted as NT_D.

The NTs of metal elements with a difference of mass number >10 have been measured by Bockris et al.[19] (Pd → Fe, etc.), Ohmori et al.[20] (Au → Fe, etc.), Mizuno et al.[21,22] (Pd → Ru, Cr → Ti, etc.), Qiao et al.[23] (Pd → Zn), Miley et al.,[24] and Miley and Patterson[25] (Pd → Al, Fe, etc.), all in the surface region. Some of these data sets have been explained as an absorption of a thermal neutron by a nucleus preexisting in the system followed by a fission of the composite nucleus and called the NT by a fission[17,18] and denoted as NT_F.

We have given a phenomenologically consistent explanation of selected data sets in the whole event of the

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CF phenomenon from the excess heat to the NT using a model, the trapped neutron catalyzed fusion (TNCF) model, which we proposed,[26,27] where the existence of trapped thermal neutrons in solids was assumed as a fundamental premise with the density n_n as an adjustable parameter. The fundamental premise is based on the two facts confirmed in experiments that (a) the CF phenomenon has not been observed if there are no background neutrons as shown by Jones et al.,[28] Ishida,[29] and Forseley et al.[30] and (b) it has been enhanced by artificial irradiation of thermal neutrons as shown by Shani et al.,[31] Celani et al.,[32] Stella et al.,[33] and Lipson et al.[34]

Those evidences of nuclear reactions in the surface region introduced earlier have also been interpreted by the TNCF model consistently with other events using assumptions of an effective reaction of the trapped neutron with nuclei in the surface layer and of characteristic properties of nuclei there:

1. The reaction cross sections have been assumed to be the same as those in a vacuum.

2. There is lifetime shortening of radioactive nuclei.

3. There is lowering of the threshold energy of the heavy nuclei for fission reactions with the trapped neutron in the surface layer.

Then, the number of nuclear reactions responsible for these nuclear products (e.g., N_{He} for ^4He generation) was comparable to the number of nuclear reactions N_Q generating Q, the amounts of the excess heat, defined by a relation $N_Q \simeq Q\,(\mathrm{MeV})/5(\mathrm{MeV})$: $N_Q \sim N_{He}, N_t, N_{NT}$. In these analyses, the adjustable parameters n_n have been determined to be 10^8 to 10^{12} cm^{-3}, depending on the experimental situation.

This result of phenomological analysis of the experimental data on the TNCF model tells us that there are nuclear reactions in the surface layer of solids showing CF phenomenon while there are a few reactions in volume.

It should, however, be emphasized here that there are very many experimental data out of the range of explanation by the TNCF model especially in the data of NT_F.

There are many theories (or models) with characteristic premises to explain some events in the CF phenomenon. In those theories, there are works by Bush,[7] Chubb and Chubb,[35] Hagelstein,[36] Preparata,[37] Fisher,[38] and Stoppini.[39]

Bush[7] was the first one who noticed the important role of alkali metals in the electrolytic system while he could not overcome Coulomb repulsion between charged particles (the proton and nuclei of alkaline atoms in his case). Chubb and Chubb,[35] Hagelstein,[36] Preparata,[37] and others not cited earlier also have tried to overcome this difficulty in every manner with little persuasive success in qualitative, even if not quantitative, explanation of concrete data in the events of the CF phenomenon.

On the other hand, Fisher[38] has given an explanation of the mass spectrum of reaction products generated in NT_F using the liquid-drop model of the nucleus assuming the existence of neutron-rich nuclei in the sample. This is on the same line of the TNCF model but using the hypothetical polyneutron as a key agent to form the neutron-rich nuclei to explain the observed data of NT_F. A similar but somewhat different approach was taken by Stoppini[39] assuming a high rate of capture of orbital electrons and consequently almost instantaneous multiple $p \rightarrow n$ transition to form neutron-rich nuclei. Once the neutron-rich nucleus is formed by one of these mechanisms, the following reaction producing nuclear products is almost independent of the mechanisms.

These theories have tried to explain a part of events in the CF phenomenon, and they have had definite success but have not explained the characteristic surface nature of the reactions; the main object of this paper is to explain it by the characteristics of neutron wave functions at the crystal boundary.

In this paper, we discuss the local nature of nuclear reactions inducing NT, one of characteristics of CF phenomenon, revealed by various events from a quantum mechanical point of view. About the irreproducibility (or poor reproducibility) of the CF phenomenon, we notice here only the explanation given by the TNCF model based on the stochastic nature of atomic processes[17,18,26,27] in forming an optimum situation for trapping of thermal neutrons and the reactions in samples.

II. BEHAVIOR OF NEUTRON BLOCH WAVE AT BOUNDARY

We have given a numerical calculation of a neutron energy band[40] on the simple one-dimensional periodic potential of the Kronig-Penny type with a period a and showed that the energy minimum E_B of the lowest band could be at the edge ($k \sim k_0 \equiv \pi/a$) of the Brillouin zone, depending on the interaction between the neutron and lattice nuclei. In such a case, energy E_k of a neutron changes little from E_B when wave number k shifts from the edge $k_0 = \pi/a$. Magnetic interaction between neutrons results in two similar energy bands each for neutrons with spin up (the z component of the spin is $+\frac{1}{2}\hbar$) and down (the z component of the spin is $-\frac{1}{2}\hbar$).

The band structure of the neutron energy (neutron band) in solids has important influence on the properties of the neutron. First of all, a neutron in a lowest-allowed band in a solid (such as A) surrounded by another solid (such as B) may be trapped in A if the allowed band of A corresponds in energy to a forbidden band of B, and its thickness is enough to prevent tunneling of the neutron. Second, the trapped neutron in a Bloch wave state of the lowest-allowed band, interacting coherently with lattice nuclei in A, can be in a lower energy state to prevent its automatic transmutation into a proton by beta decay if the interaction is an appropriate one. This possibility is enhanced by formation of a Cooper pair of two neutrons with opposite spins and opposite quasi-momenta $\boldsymbol{k}$ and $-\boldsymbol{k}$ in the band. Third, the trapped neutron in A has a large probability density at the boundary between A and B. This feature is shown numerically in this section. The large density expected in the boundary region might be responsible in appropriate condition to form the polyneutron assumed by Fisher in his explanation[38] of the mass spectra observed in NT_F.

To investigate characteristics of the trapped neutron at a boundary between two crystals A and B mentioned earlier, we take the Bloch wave of the lowest-allowed band in A as a plane wave and the boundary as a potential wall linearly increasing with decrease of the coordinate (say x) perpendicular to the boundary (at $x = x_0$) given below in zeroth approximation:

$$V(x) = \alpha(x_0 - x) \qquad (x \leq x_0) \ , \tag{1}$$

$$= 0 \qquad (x_0 < x) \ . \tag{2}$$

Classically, a particle with a mass m_n and an energy E moving leftward to the boundary wall loses kinetic energy gradually from $x = x_0$ and reaches 0, a classical turning point, determined by a relation $E = V(0)$ to be reflected there. The classical probability of existence at $x (\propto 1/v(x))$ is proportional to a quantity $\sqrt{m_n/(E - V(x))}$, which is shown by a dotted line in Fig. 1 for the boundary region near $x \sim 0$ with arbitrary units for the coordinates.

Quantum mechanically, the corresponding quantity, the probability density $\rho(x) \equiv |\psi(x)|^2$, is calculated numerically with the Airy function for the wave function $\psi(x)$ and shown by a solid line in Fig. 1 for the case where $\alpha = 1$. As is well known in quantum mechanics, e.g., for the case of a harmonic oscillator, the quantum behavior of a microscopic object approaches a classical one with the increase of the quantum number, and the qualitative coincidence of two curves in Fig. 1 is an example of this property. Physical investigation of a quantum object at a boundary region is therefore partly given by behavior of a classical particle as done in our previous work.[17] There is, however, characteristic coherence of wave functions determined by their phase.

Two neutron Bloch waves with opposite spins and wave numbers will form a Cooper pair with spin zero as pointed out before.[17,41] Wave functions of the neutron Cooper pairs interfere to form composite wave intensification or destruction of each other. If the wave functions of the neutron Cooper pairs can be expressed in a similar manner to one of the neutron Bloch waves, they add their amplitude coherently to a definite distance x_{coh} from the boundary in the crystal A. This feature may be called local coherence of neutron Bloch waves.[42] This distance x_{coh} of the local coherence is given as $1/\Delta k$ when the difference of wave numbers of neutrons are Δk.

To show the local coherence, one of the quantum mechanical characteristics of the wave function of the Cooper pair approximated by the one given in Fig. 1, we made a calculation of wave functions for $\alpha = \infty$ and sinusoidal waves for the wave function, for simplicity. In this case, the exponential part in $x \leq 0$ becomes zero, and the transient region between x_0 and 0 disappears. Illustrated in Fig. 2 are (a) a single wave function with $k_0 = \pi/a$, (b) the probability densities of 11 locally coherent waves with wave numbers between $k = 0.9k_0$ and $1.0k_0$, and (c) with wave numbers between $k = 0.95k_0$ and $1.00k_0$. The figures in Fig. 2 show clearly the dependence of x_{coh} on the Δk; $x_{coh} \sim 1/\Delta k$.

If reactions between a neutron and another nucleon (nucleus) occur in this region where there is the local coherence of the trapped neutrons, the reaction probability proportional to the probability density of neutrons becomes extremely large compared with a case where there

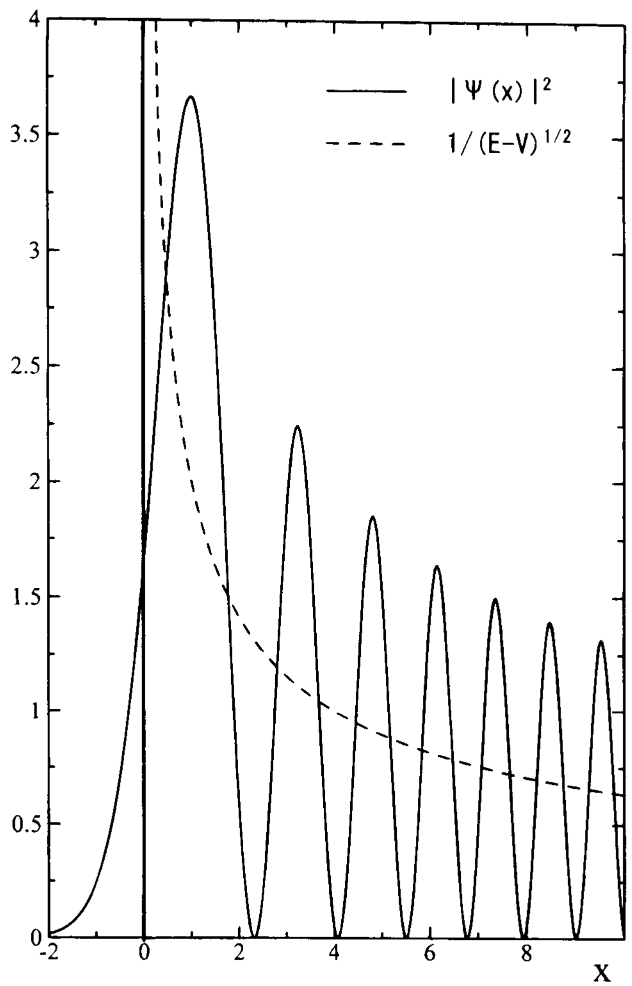

Fig. 1. Classical probability of existence (dotted line) and quantum mechanical probability density (solid line) of a particle with a mass m_n and an energy E (in arbitrary units) in the boundary region $x \sim 0$ determined by a condition $E = V(0)$ with a potential $V(x) = 0\,(x_0 \leq x)$, $= \alpha(x_0 - x)\,(x \leq x_0)$ for $\alpha = 1$.

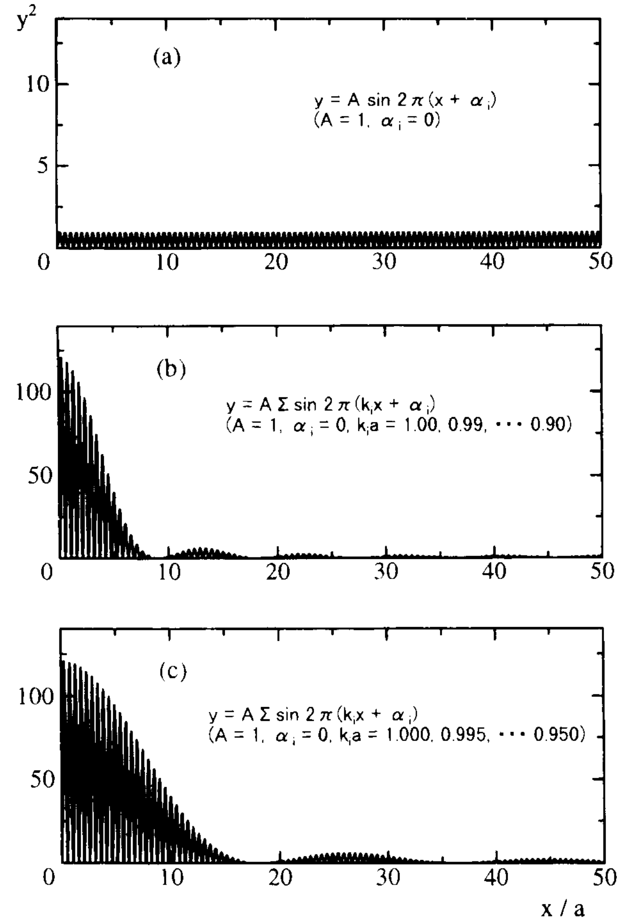

Fig. 2. Illustration of local coherence of neutron Bloch waves near the boundary region by sinusoidal waves in phase at $x = 0$. Probability amplitudes of (a) a single sinusoidal wave with $k = k_0 \equiv \pi/a$, (b) eleven sinusoidal waves with different wave numbers between $k = 0.90\pi/a$ and $k_0 \equiv 1.00\pi/a$, and (c) eleven sinusoidal waves with different wave numbers between $k = 0.95\pi/a$ and $k_0 \equiv 1.00\pi/a$, where a is the period of the Kronig-Penny potential.

is no coherence by a factor N of the neutron number in the band. In our previous analyses on the TNCF model,[17,18,26,27] where we assumed the same reaction cross section in the surface layer as in free space, the arbitrary parameter n_n was determined from experimental data as $n_{n,free} = 10^8$ to 10^{12} cm^{-3}. When we take the local coherence of the neutron wave function into consideration, the parameter should be replaced by taking the square root of these values to $n_n = 10^4$ to 10^6 cm^{-3}.

III. INTERACTION OF A NEUTRON BLOCH WAVE AND A NUCLEUS IN SURFACE LAYERS

As shown in Sec. II, trapped neutrons have a large probability density near the boundary region of the trapped region A and surrounding region B if the boundary is expressed by a potential wall $V(x)$ in Eq. (1). In the boundary region between A and B discussed in Sec. II, the atomic arrangement and composition vary gradually from those of A to those of B, in reality. If the thickness of the crystal B is enough to prevent tunneling of the neutron in an allowed band in A, which corresponds energetically to a forbidden band in B, the potential wall against the neutron may be approximated by an increasing function of x, and the potential Eq. (1) is considered as its zeroth-order approximation.

The fusion probability of a neutron and a nucleus is known to be proportional to an interaction time, i.e., a time they stay in a range where the nuclear force is working except resonances due to virtual levels. This property results in the well-known general energy dependence of the fusion cross section of $1/\sqrt{E}$ for the reaction of a neutron with a target nucleus. In the case of interaction between the trapped neutron and lattice nuclei in a surface layer, the interaction time is reduced to the probability density of the neutron in it.

Therefore, the trapped neutron, which is quasi-stable unless it suffers a large perturbation, fuses with a nucleus at a periodic position seen from the periodic lattice where the neutron is trapped. The local coherence of neutron wave functions intensifies the reaction probability to N^2 times that of a single neutron. It is conceivable a simultaneous reaction of several neutrons and a nucleus occurs in the region where the local coherence intensifies the probability density up to N^2 times that of a single neutron. However, we confine this paper to only reactions of a neutron and a nucleus.

This is the physical basis of the premise of surface reaction of the trapped neutron proposed in the TNCF model giving the consistent explanation of those events, the excess heat, tritium, ^{4}He generations, and NT, in electrolytic CF systems.[17,18,26,27] The nuclei inducing trigger reactions with the trapped neutron were ^{6}Li and ^{2}H in the Pd-D-Li system where those events had been observed and relevant quantities had been measured extensively.

The detection of a small amount of ^{4}He in a surface region with a thickness of 40 μm by Morrey et al.[2] and a sufficient amount of ^{4}He in gas by Miles et al.,[4] introduced in Sec. I, is consistently interpreted by the surface nature of trigger reactions of a neutron with ^{6}Li and with ^{2}H $(= d)$:

$$n + {}^6_3\mathrm{Li} = {}^4_2\mathrm{He}(2.1\ \mathrm{MeV}) + t(2.7\ \mathrm{MeV}) \quad (3)$$

and

$$n + d = t(6.98\ \mathrm{keV}) + \gamma(6.25\ \mathrm{MeV}) \ . \quad (4)$$

The generation of energetic neutrons has been explained by the following breeding reactions, induced by the energetic tritons generated in these trigger reactions, producing first a 14.1-MeV neutron by the t-d reaction:

$$t(\varepsilon) + d = {}^4_2\text{He}(3.5\ \text{MeV}) + n(14.1\ \text{MeV}) \ , \quad (5)$$

$$n(\varepsilon) + d = n + d(\varepsilon') \ , \quad (6)$$

and

$$d(\varepsilon) + d = {}^3_2\text{He}(0.82\ \text{MeV}) + n(2.45\ \text{MeV}) \ , \quad (7)$$

$$= t(1.01\ \text{MeV}) + p(3.02\ \text{MeV}) \ . \quad (8)$$

In the cases of NT_D introduced briefly in Sec. I, the processes responsible were assumed as follows:

$$n + {}^{23}_{11}\text{Na} = {}^{24}_{11}\text{Na}^* = {}^{24}_{12}\text{Mg} + e^- + \bar{\nu}_e + 1.39\ \text{MeV} \ , \quad (9)$$

$$n + {}^{27}_{13}\text{Al} = {}^{28}_{13}\text{Al}^* = {}^{28}_{14}\text{Si} + e^- + \bar{\nu}_e + 2.88\ \text{MeV} \ , \quad (10)$$

$$n + {}^{39}_{19}\text{K} = {}^{40}_{19}\text{K}^* = {}^{40}_{20}\text{Ca} + e^- + \bar{\nu}_e + 1.32\ \text{MeV} \ , \quad (11)$$

$$n + {}^{85}_{37}\text{Rb} = {}^{86}_{37}\text{Rb}^* = {}^{86}_{38}\text{Sr} + e^- + \bar{\nu}_e + 1.77\ \text{MeV} \ , \quad (12)$$

$$n + {}^{106}_{46}\text{Pd} = {}^{107}_{46}\text{Pd}^* = {}^{107}_{47}\text{Ag} + e^- + \bar{\nu}_e + 1.17\ \text{MeV} \ , \quad (13)$$

and

$$n + {}^{196}_{78}\text{Pt} = {}^{197}_{78}\text{Pt}^* = {}^{197}_{79}\text{Au} + e^- + \bar{\nu}_e + 0.719\ \text{MeV} \ . \quad (14)$$

On the other hand, a part of the data of NT_F has been explained by fusion of a nucleus and a thermal neutron followed by a fission. Reactions of several neutrons with a nucleus are left for future work, which will give a similar result to that obtained by Fisher[38] but with a different source of neutrons to form neutron-rich nuclei. Some new elements observed in experiments introduced earlier in Sec. I have been explained by the following reactions:

$$n + {}^{A}_{24}\text{Cr} = {}^{A-3}_{22}\text{Ti} + {}^4_2\text{He} - Q \ , \quad (15)$$

$$n + {}^{A}_{46}\text{Pd} = {}^{A-3}_{44}\text{Ru} + {}^4_2\text{He} - Q \ , \quad (16)$$

$$n + {}^{197}_{79}\text{Au} = {}^{57}_{26}\text{Fe} + {}^{141}_{53}\text{I} + Q \ , \quad (17)$$

$$n + {}^{A}_{46}\text{Pd} = {}^{A+1}_{46}\text{Pd}^* \ , \quad (18)$$

and

$${}^{A+1}_{46}\text{Pd}^* = {}^{A'}_{13}\text{Al} + {}^{A+1-A'}_{33}\text{As} \ , \quad (19)$$

$$= {}^{A'}_{30}\text{Zn} + {}^{A+1-A'}_{16}\text{S} \ . \quad (20)$$

In the case of Cr, A = 50 to 54, and in the case of Pd, A = 102 to 110 and A' = 64 to 70, and therefore

$$A + 1 - A' \equiv A'' \geq 32 \ .$$

Other possible fission reactions of ${}^{A+1}_{46}\text{Pd}$ formed by absorption of a neutron are assumed as follows:

$${}^{A+1}_{46}\text{Pd}^* = {}^{A'}_{29}\text{Cu} + {}^{A''}_{17}\text{Cl} \ , \quad (21)$$

$$= {}^{A'}_{23}\text{V} + {}^{A''}_{23}\text{V} \ , \quad (22)$$

$$= {}^{A'}_{28}\text{Ni} + {}^{A''}_{18}\text{Ar} \ , \quad (23)$$

$$= {}^{A'}_{26}\text{Fe} + {}^{A''}_{20}\text{Ca} \ , \quad (24)$$

$$= {}^{A'}_{27}\text{Co} + {}^{A''}_{19}\text{K} \ , \quad (25)$$

$$= {}^{A'}_{24}\text{Cr} + {}^{A''}_{22}\text{Ti} \ , \quad (26)$$

$$= {}^{A'}_{30}\text{Zn} + {}^{A''}_{16}\text{S} \ , \quad (27)$$

and

$$= {}^{A+1}_{47}\text{Ag} + e^- + \bar{\nu}_e \ . \quad (28)$$

Using these reactions based on the premise of existence of the trapped neutron in the TNCF model, the experimental data gave the value of the parameter n_n between 10^8 to 10^{12} cm^{-3}, which should be interpreted as values at high times of the system, giving optimum values of the excess heat, tritium, ^{4}He, neutrons, and NTs starting from rather small values of n_n.

IV. INTERACTION OF NEUTRON BLOCH WAVES AND A NUCLEUS IN SURFACE LAYERS

The same investigation given earlier for the interaction of a trapped neutron and a nucleus in surface layers is applicable to the interaction of a nucleus and many trapped neutrons in surface layers where various nuclear transmutations have been observed.

If there are many trapped neutrons in the lowest-allowed band in a crystal, they are in different Bloch states with phase factors characterized by $\boldsymbol{k}$ but with almost the same energy at the minimum of the band. Then, the interaction between trapped neutrons with almost the same energy and a nucleus in the boundary region can induce a drastic change of properties of the nucleus, e.g., change of its decay characteristics, by a coherent addition of the individual interaction due to almost the same phases of the wave functions at $x = 0$ (see Fig. 1).

In other words, all trapped neutrons in an allowed band interact with a nucleus in the surface layer coherently, i.e., they interact with a nucleus like a single particle with enormous probability density at the position of the nucleus. This is an appearance of the wave nature of a microscopic object, a neutron, i.e., a characteristic of quantum mechanics.

A nucleus interacting with many neutrons can fuse with only one neutron at a time, however. Therefore, there appears the particle nature of a microscopic object, a neutron, in a fusion reaction of the neutron and a nucleus. The latter nature has been shown in the explanation of elimination of radioactivity of radioactive elements by electrolysis in CF cells may be interpreted as decay time shortening or fission by threshold energy lowering of those elements deposited on the cathode surface by the trapped neutrons in the cathode.

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