Cold nuclear fusion induced in KD_2PO_4 single crystals by a ferroelectric phase transition

A. G. Lipson, D. M. Sakov, E. I. Saunin, V. B. Kalinin, M. A. Kolovov, B. V. Deryagin, and A. A. Khodyakov

Institute of Physical Chemistry, Russian Academy of Sciences
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A reproducible generation of neutrons and tritium—the products of cold nuclear fusion—has been observed during temperature cycling of ferroelectric KD_2PO_4 (DKDP) single crystals near their Curie point (T_c=222 K). A substantial asymmetry in the yield of cold-fusion products in the neutron (n) and tritium (T) channels was found experimentally ($n/T \approx 10^{-7}$). It was found by a thermal depolarization method that the neutron emission occurs in the same temperature interval as the switching of domain walls in the crystal. It is suggested that the cold fusion in the DKDP crystals stems from a nonzero probability for the simultaneous occupation of two neighboring D sites by deuterons near the surface of the crystal in the course of the ferroelectric phase transition and a possible displacement of deuterons substantial distances, up to 3.5 Å, in strong electric fields.

INTRODUCTION

Nuclear effects in deuterated solids are currently attracting active experimental and theoretical research. It has been solidly established that neutrons are emitted when titanium is saturated by deuterium from the gas phase and also at palladium and titanium cathodes during electrolysis in D_2O (Refs. 1–4). In some cases, the generation of a significant amount of tritium, exceeding the neutron yield by seven to nine orders of magnitude, has also been detected during electrolysis.[5,6]

The overwhelming majority of studies on the initiation of "cold fusion" reactions which have been published to date have been carried out on metal deuterides. It has been suggested[7] that these systems present the conditions required for the occurrence of *dd* reactions in the crystal lattices of the samples. The reasoning here is that when group IV and V metals of the periodic table and also Pd are saturated with deuterium under nonequilibrium conditions the following events may occur: (a) Regions with a superstoichiometric deuterium concentration arise. (b) Regions with a high concentration of elastic energy arise, causing deformation and cracking of the samples. (c) Phase transitions of various types occur and are another source of elastic energy. (d) Regions appear in which the Coulomb potential of the deuterium nuclei might be screened by electrons, so the energy barrier for *dd* reactions might be substantially lowered. Assumptions (a)–(d) (or one of them) are usually invoked to describe cold fusion processes in metal deuterides.[7] Conditions (a)–(d) by no means hold in all cases (whether they do depends on the concentration and purity of the deuterium, the temperature, the defectiveness of the sample, the surface state of the sample, etc.) Accordingly, since it is not possible to monitor the properties of the samples throughout their volume during the experiments, the reproducibility of the results on cold fusion in such systems leaves something to be desired.

Nevertheless, deuterated entities with controllable properties of this type do exit. Examples are deuterated single crystals with a high deuterium concentration in which reversible phase transition occur: ferroelectric–paraelectric, ferroelectric–(ionic conductor), etc. Such crystals have the further advantage over metal deuterides that strong electric fields, on the order of 10^8 V/cm, arise in them in the course of phase transitions.[8] In the course of cracking,[9] such fields are sufficient to accelerate free deuterons to energies on the order of 1 keV, or to several tens of eV over distances comparable to the interatomic distances (i.e., with a single unit cell).

As one of the present authors has shown,[10–13] a splitting of cation sites is observed in most such crystals. This splitting leads to anomalies in physical properties: a dipole order in low-temperature phases, an ionic conductivity in high-temperature phases, an ultrafast nuclear relaxation,[14] etc. We accordingly selected the ferroelectric crystal KD_2PO_4 (DKDP) as an entity of potential interest for research on the cold fusion problem. The structure of both known modifications of this crystal have split D sites separated by a distance of 0.45 Å.

For the sake of objectivity we should point out that the use of deuterated ferroelectrics to initiate a fracture-related fusion was first proposed by Golubnichiĭ *et al.* in Ref. 15. However, they did not link the possible realization of cold fusion with a ferroelectric phase transition in that paper.

In the present study we have demonstrated that a reproducible generation of neutrons and tritium occurs in single crystals of this type, during temperature cycling near the Curie point.

EXPERIMENTAL METHODS

In the experiments we used DKDP samples with masses of 0.5 and 0.8 g, deuterated to an extent of 98.5%, cut from a single-crystal plate in the (11) direction. The single crystals were grown from solution and had an impurity concentration $\sim 10^{-3}$ mole %. The test samples

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used in these experiments had no visible defects or cracks. The single crystals with $T_c = 222 \pm 1$ K exhibited a resistivity $\rho_V = 1.1 \cdot 10^{11}$ $\Omega \cdot$ cm at a constant voltage $V = 100$ V with a calculated deuterium concentration $N_D = 2.2 \cdot 10^{22}$ cm^{-3}. As control samples we used single-crystal samples of KH_2PO_4 (KDP), with $T_c = 123$ K and the same mass as that of the DKDP samples. For the neutron measurements, the samples were placed in an isolated, thin-walled brass cryostat equipped with a spring-loaded clamp and a thermocouple.[16] After the cryostat was cooled to ~ 100 K in liquid nitrogen, a linear heating was carried out by passing liquid nitrogen through the system. The heating rate was $2 \cdot 10^{-2}$ K/s in all cases. The error of the temperature measurements over the temperature intervals studied was ± 0.3 K.

To determine the change in the tritium concentration during the temperature cycling of DKDP near T_c, we placed samples with a mass ~ 0.5 g in molybdenum-glass cells with a volume ~ 3 cm^3. Half the cells were evacuated beforehand to $\sim 10^{-4}$ torr. The temperature cycling of the cells which were evacuated and of those which were not evacuated ($p = 760$ torr), containing DKDP samples, was carried out by cooling them to liquid-nitrogen temperature and then heating them to room temperature (for a total of 100 cycles). As control samples we used DKDP samples of the same mass which were sealed off in cells (one was evacuated, while the other was at atmosphere pressure), which were not subjected to temperature cycling.

The neutrons were detected by an array of eight NWJ-62 proportional counters ($^{10}BF_3$), positioned in a steel block with silicone oil. The cryostat was placed directly beside the array of counters (which was covered with sheet cadmium 1 mm thick), at a distance of 3 ± 1 cm. The entire apparatus was equipped with passive polyethylene shielding, 10 cm thick. The efficiency of a neutron detector was $3.0 \pm 0.5\%$ in this geometry, according to calibration measurements with a Cf^{252} neutron source with an intensity of $(5 \pm 1) \cdot 10^2$ n/s ($E_{max} \approx 2.3$ MeV). The source was put in the position of the cryostat for these calibration measurements. The natural neutron background measured over long time intervals (before, between, and after the experiments with DKDP and KDP), was 0.012 ± 0.004 count/s (the variations of the background did not exceed 2σ). While measuring the neutron flux, we also monitored the shape and length of the neutron pulses arriving at the detector, and we compared them with the neutron pulses from the Cf^{252} source. All these measurements were carried out at a fixed relative humidity, $\sim 50\%$, with the help of exhaust ventilation and an independent grounding circuit. The apparatus used to detect the neutrons through pulse-height analysis of the pulses arriving at the detector is described in detail in Refs. 3 and 17.

To improve the reliability of the neutron flux measurements, we carried out some control experiments which included measuring the neutron flux densities in the following cases: (1) empty cryostat, thermal cycling over the temperature interval 219–223 K; (2) KH_2PO_4 (KDP) single crystals, thermal cycling over the interval 121–125 K (near $T_c = 123$ K); (3) KDP single crystals, thermal cycling over the interval 219–223 K; (4) DKDP single crystal, thermal cycling over the intervals 210–215 K and 225–230 K (i.e., away from the phase transition). At least 100 heating-cooling cycles were carried out for each type of control experiment.

Experiments to determine the tritium concentration in the DKDP samples during thermal cycling over the temperature interval 78–290 K were carried out on a Beta-1 apparatus by a liquid-scintillation method (three photomultipliers were positioned at angles of 120° and were connected in a majority-coincidence circuit). The cells holding the test samples were opened up in a special apparatus. The gas which was released was captured by a flowing carrier gas (Ar or He), which was then bubbled through distilled water. The DKDP crystals were dissolved in a known amount of H_2O, and the solution was then neutralized to pH ≈ 7. The change in the volume of the samples did not exceed 10%. The glass of the cells and also the glass of the tube of the comparison sample were placed in a reactor which was heated to 450°. The carrier gas (Ar) was then blown through this reactor and then bubbled through distilled water. All the solutions obtained in the manner were used for scintillation counts after the samples to be analyzed were mixed with a liquid scintillator (ZhS-8I). The Beta-1 apparatus was calibrated with the help of a standard metrological source of tritium water (T_2O).

To study the ferroelectric phase transition in DKDP, to determine T_c, and to coordinate the scales for the temperature dependence of the neutron emission and that of the spontaneous polarization, we used the method of thermal depolarization. These experiments were carried out in a vacuum of 10^{-6} torr in a copper cryostat, which served as a grounding electrode, over the range 80–300 K (Ref. 18). Samples with a surface area of 1.0 ± 0.2 cm^2 were heated (or cooled) linearly at a rate of 0.1 K/s. Graphite paste was used as an ohmic contact.

EXPERIMENTAL RESULTS

Table I shows the data obtained in the experiments carried out to detect neutrons. In control experiments (1–4), the neutron count rate did not exceed the natural neutron background, within the confidence limits. For the DKDP samples near T_c (219–223 K), in contrast, we observed a significant increase above the natural background level, by $(8–15)\sigma$. Figure 1 shows histograms of the pulse height spectra obtained (a) from the Cf^{252} source and (b) during temperature cycling of the DKDP. The pulse height spectrum of the DKDP agrees well with that of the source (the peaks occurs in channels 17 and 18). This agreement is evidence that neutrons with an energy close to that of neutrons from Cf^{252} fission ($E = 2.3$ MeV), which have essentially the same energy as the neutrons from the *dd* reaction ($E = 2.45$ MeV), are generated during temperature cycling of DKDP crystals near T_c. The shape and length of the pulses which reach the detector from the DKDP ($\tau = 1$ ms, height of 0.2–0.4 V) are close to those of the pulses from the Cf^{252} source. This agreement can serve as yet another piece of evidence that a generation of neutrons actually does occur in single-crystal DKDP samples

TABLE I. Characteristics of the neutron emission in KD_2PO_4 and KH_2PO_4 crystals during temperature cycling.

№	System	ΔT, K	$\bar{N}$, count/h	n_D, n/s	n, n/s	L, σ
1	Empty cryostat	219 — 223	0,012 ± 0,005	0,14	—	—
2	KH_2PO_4	121 — 125	0,011 ± 0,004	0,14	—	—
3	KH_2PO_4	219 — 223	0,012 ± 0,005	0,14	—	—
4	KD_2PO_4	210 — 215	0,013 ± 0,004	0,14	0,03 ± 0,04	< 1
5	KD_2PO_4	225 — 230	0,012 ± 0,005	0,14	—	—
6	KD_2PO_4 (m = 0,5 r)	219 — 223	0,024 ± 0,005	0,10	0,40 ± 0,18	8
7	KD_2PO_4 (m = 0,8 r)	219 — 223	0,031 ± 0,006	0,10	0,61 ± 0,19	11
8	KD_2PO_4 (m = 0,5 r)	221 ± 0,3 222 ± 0,3	0,035 ± 0,006	0,10	0,75 ± 0,16	15

Here $\bar{N}$ is the average number of counts over a measurement time $t \approx 6 \cdot 10^3$ s (this is the effect before subtraction of the background); $n_D = 2n_b/E^2t$ ($n_b = 0.012 \pm 0.004$ count/s; E is the detector efficiency); $n = (\bar{N} - n_b)/E$ is the magnitude of the effect after subtraction of the background; and $L(\sigma)$ is the extent to which the background is exceeded, in units of σ (the number of standard deviations above the background level).

during temperature cycling near T_c (Ref. 4). Most of the neutron events (which were monitored in terms of the pulse height, shape, and length simultaneously) were detected near temperature positions 221 and 222 K. Over the time corresponding to 100 heating-cooling cycles the resultant effect for the temperature interval 221 ± 0.3 K is 0.75 ± 0.16 n/s (the natural neutron background has been subtracted, and the detector efficiency has been taken into account). This result is three times the neutron background and has a confidence level of 15σ (Table I). The neutrons are emitted in sporadic bursts (30–100 neutrons per burst), which are observed during passage through the temperature interval $(221–222) \pm 0.63$ K.

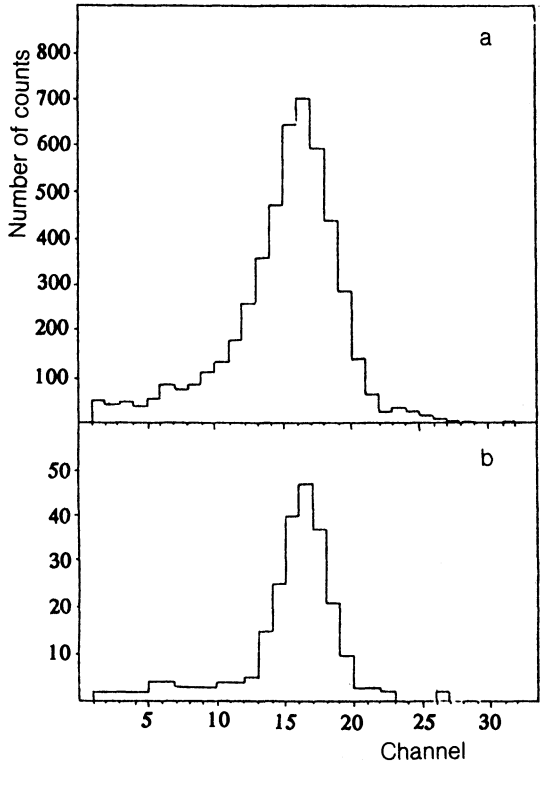

FIG. 1. Distribution of the pulses arriving at the neutron detector among channels of the pulse height analyzer. a—From the Cf^{252} neutron source; b—during temperature cycling of DKDP crystals near T_c.

Note that during the first 20–30 cycles the reproducibility of the neutron bursts is essentially 100%. Thereafter, the reproducibility worsens: It falls off to 60% after 80–90 cycles. After 80–90 cycles the crystal is completely covered by a network of cracks. As we will show below, this circumstance significantly reduces the spontaneous polarization of DKDP and shifts T_c downward. For small, defect-free samples of DKDP, we again see a proportionality in the increase in the neutron count rate as the sample mass is increased by a factor of 1.6, while the intensity of the neutron count rate increases by a factor ~ 1.5 (Table I).

Figure 2 shows the measurements of the neutron emission for the DKDP and KDP single crystal near T_c for heating time intervals $\Delta\tau = 60$ s. The distribution of the number of events n_i is described well as a function of the number of pulses in the neutron channels of the pulse-height analyzer (channels 13–31; curve *1* in Fig. 2a) by a Poisson distribution

$$n_i = Nm^i e^{-m}/i!,$$

where N is the number of intervals, m is the average number of pulses per interval, and $i = 0, 1, 2, 3, \ldots$. This circumstance is evidence that background values of the neutron emission which satisfy a Poisson distribution of the random quantity i are observed near T_c (121–125 K) for the KDP samples. For the DKDP single crystals (curve *1* in Fig. 2b), for the same $N = 100$, we observe near T_c (219–223 K) a significant deviation from the Poisson distribution found for KDP (curve *2* in Fig. 2b). The appearance of n_i events for $i = 4$ in KDP (or in the background count)

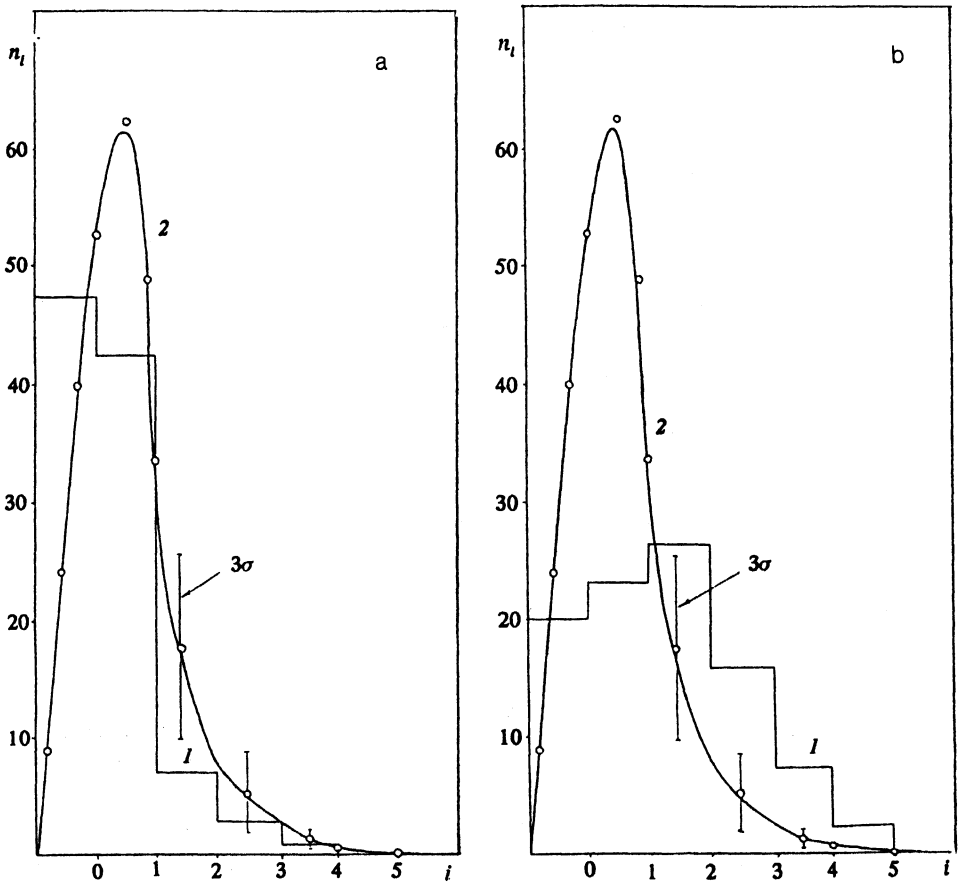

FIG. 2. a: Curve *1*—Distribution of the number of neutron events (n_i) with respect to the number of neutron pulses (i) for a KDP single crystal, recorded for sample heating time intervals $\Delta\tau=60$ s near T_c (121–125 K) over 100 heating-cooling cycles; curve *2*—Poisson distribution for KDP ($N=100$, $m=0.63$). b: Curve *1*—Distribution of the number of neutron events (n_i) with respect to the number of neutron pulses (i) for a DKDP single crystal, recorded for sample-heating time intervals $\Delta\tau=60$ s near T_c (219–223 K) over 100 heating-cooling cycles; curve *2*—Poisson distribution in control experiments with KDP.

has a probability $p=2\cdot10^{-3}$, while $i=5$ the probability is $p\sim2.5\cdot10^{-4}$. For DKDP, on the other hand, we have $n_4=7$ and $n_5=2$. The data in Fig. 3 are fairly convincing evidence that the neutron count rates observed near T_c for the DKDP single crystal are not random fluctuations of background values.

The experimental results obtained from a large number of measurements thus allow us to conclude that a generation of neutrons is observed during temperature cycling of fairly high-quality DKDP single crystals near T_c.

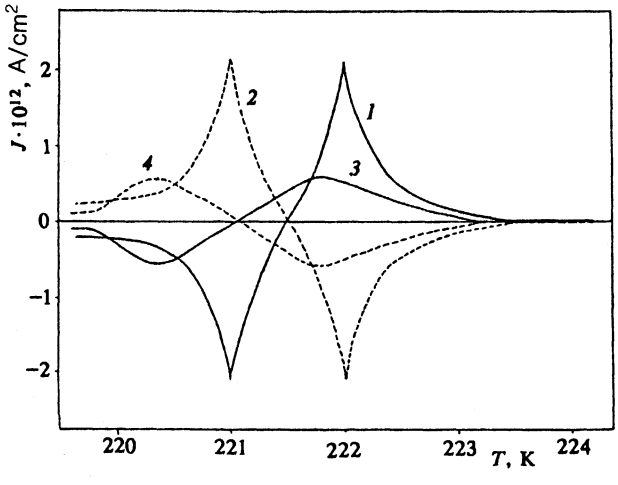

FIG. 3. Spectra of the thermal depolarization of DKDP crystals during temperature cycling in vacuum near T_c. *1*—Fresh sample, heating; *2* (dashed curve)—fresh sample, cooling; *3,4*—sample after ten heating-cooling cycles; *3*—heating; *4* (dashed curve)—cooling.

Measurements of the thermal depolarization carried out for a temperature identification of the neutron bursts yielded the following results. The spectrum of the thermal depolarization of "defect-free" DKDP single crystals which had not been subjected to temperature cycling consists of two peaks, differing in polarity (Fig. 3). When the heating of the samples is replaced by a cooling, the sign of the thermally stimulated currents changes. The extrema of these currents correspond to temperatures of 221 and 222 K, respectively. The data of Refs. 19 and 20 suggest that this behavior of the thermal-depolarization currents can probably be attributed to a change in the sign of the polarization in the course of a polarization reversal of the domain walls near T_c.

After multiple temperature cycling (curves *3* and *4* in Fig. 3), we see a significant decrease in the intensity of the thermal-depolarization spectra. These spectra also become blurred and shift toward lower temperatures. These results indicate that the quality of the single-crystal sample is degraded during the temperature cycling because of the formation of a system of small cracks.[21] There is the further implication of a significant elimination of deuterium from the surface layer, into vacuum. These factors lead (on the one hand) to a substantial decrease in the spontaneous polarization of the DKDP and (on the other) to a decrease in the deuterium concentration in the surface layer of the sample. As a result, there should probably be an effect on

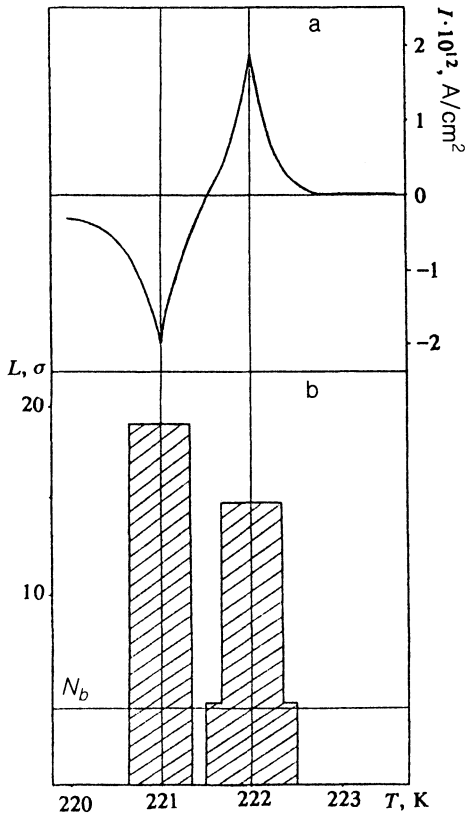

FIG. 4. a—Spectrum of thermal depolarization during the heating of DKDP sample near T_c; b—corresponding histograms of neutron bursts. Here L, σ, is the confidence level of the observed neutron bursts, and N_0 is the average background level.

the ability of the deuterium to initiate cold fusion reactions.

Comparison of the data on the thermal depolarization with the measurements of the neutron flux lends support to the suggestion that the neutron emission in DKDP is due to a polarization reversal of domains near T_c. These two parallel processes near T_c are plotted in Fig. 4. The confidence level (L) of the neutron events detected is plotted along the ordinate here.

Table II shows data on the tritium concentration which builds up in the DKDP samples over 100 heating-cooling cycles and also on the concentration in control samples. Neither the glass from which the cells were made nor the atmosphere in the cells which were not subjected to temperature cycling exhibit tritium in excess of the background count rate. In the original DKDP samples, on the other hand, there are measurable amounts of tritium ($\sim 5 \cdot 10^9$ tritium atoms per gram). After 100 heating-cooling cycles, the tritium concentration in the evacuated DKDP samples decreases about 45% from its original level, while there is essentially no change in this concentration in the samples which were initially at atmospheric pressure. Most of the excess tritium is observed in the atmosphere of the cells and in the cell glass, according to these measurements. The tritium content in the glass of the evacuated cells is slightly higher, although the content is identical in the gaseous atmosphere of the cells which were and were not evacuated. The complete set of results for the samples in the evacuated and unevacuated cells leads to essentially identical values, exceeding the corresponding sum for the original samples by a factor of about 4.5; here we allow for the tritium content in the sample, in the glass, and in the gaseous atmosphere.

It follows from these results that tritium builds up at a rate of about $2.4 \cdot 10^8$ tritium atoms per gram per cycle in the samples subjected to thermal cycling. The significant T content in the glass of the cells is somewhat surprising. It might be due to either a selective absorption of tritium by the glass surface or the circumstance that the glass is bombarded by T atoms with fairly high energies (a few MeV).

It has thus been established experimentally that a generation of products of cold fusion, i.e., neutrons and tritium, is observed during temperature cycling of DKDP single crystals near T_c.

DISCUSSION OF RESULTS

In discussing possible models for the initiation of *dd* reactions in the course of a ferroelectric phase transition in DKDP crystals, we must bear in mind that DKDP exists in two phases, according to the data of Refs. 19–23: a noncentrally symmetric paraelectric phase (space group $I\bar{4}2d$) above the phase transition temperature (T_c) and a polar ferroelectric phase (space group $Fdd2$ below T_c. The D sites in each phase are split to a distance 0.45 Å. In the paraelectric phase, the two sites are filled equiprobably, while in the ferroelectric phase one site is filled preferentially (95%). This circumstance leads to the polarity and serves as the microscopic reason for the dipole order of the ferroelectric type. One might suggest that at the

TABLE II. Tritium concentration (in units of 10^9 tritium atoms per gram of the KD_2PO_4 crystal) for samples subjected to temperature cycling.

№	DKDP crystal	Cell glass	Cell atmosphere	Σ
1	5,0 ∓ 1,0	—	—	5,0 ∓ 1,0
2	4,5 ∓ 1,1	—	0,7 ∓ 0,2	5,2 ∓ 1,0
3	5,9 ∓ 1,1	8,3 ∓ 1,5	9,2 ∓ 1,6	23,4 ∓ 1,4
4	3,7 ∓ 0,8	10,8 ∓ 1,8	9,2 ∓ 1,6	23,7 ∓ 1,6

1—Control sample, not subjected to temperature cycling, in a glass cell at $p=760$ torr;
2—control sample in a glass cell at $p=10^{-4}$ torr;
3—sample subjected to 100 heating-cooling cycles, $p=760$ torr;
4—sample subjected to 100 heating-cooling cycles, $p=10^{-4}$ torr.

ferroelectric–paraelectric phase transition, at 221 K, a nonzero probability for simultaneous filling of the two crystallographic sites by the deuterium atoms arises in some part of the crystal (probably near its surface). At least two factors would act in this direction.

First, because of the large spontaneous polarization of DKDP ($p_s \sim 5 \cdot 10^{-6}$ C/cm^2), strong electric fields ($\sim 10^8$ V/cm) arise in the crystal, especially near its surface.[24] These fields accelerate free electrons to energies on the order of several tens of eV over distances comparable to interatomic distances[1)] (5–10 Å). A deuteron with this energy would be able to overcome the potential barrier between the two sites (which is on the order of a few times kT), or it could undergo a tunneling.

Second, in the course of the ferroelectric transition an excess elastic energy arises and reaches $\Delta W_s = 40$ erg/cm^3 (Ref. 20) at the domain walls (the dimensions of a domain are a few tens of unit cells). This event could also stimulate the filling of both of the split sites.[18] Furthermore, the motion of deuterons in DKDP crystals in the course of an order-disorder phase transition crystals in the model of a proton-lattice coupling is strongly coupled to optical phonons.[19] If the energy of the optical phonons is concentrated at domains $\sim$1500–2000 Å in size,[20] the conditions in the DKDP may also allow a coherent *dd* reaction stimulated by multiphonon excitations.[24,25]

When deuterons come within a distance $\sim$0.45 Å of each other, which is typical of metallic deuterium (e.g., at the core of Jupiter, according to the data of Ref. 26), and also if such deuterons are accelerated to an energy $\sim$10 eV by the electric fields which arise in the course of the spontaneous polarization of a ferroelectric crystal, the rate constant for the *dd* reaction by the neutron channel may reach $\lambda_{dd} \sim 10^{-20}$–$10^{-23}$ s^{-1} per deuteron pair.[26,27] This is just the neutron yield, 10^{-20}–10^{-21} s^{-1}, which is observed in our case. If we assume that the "coherent" *dd* reactions, which in practice would occur in the "neutron-free" channel under the influence of multiphonon excitations, occur primarily in the $d(d,\mathrm{T})p$ channel,[25,27] then we can also find an explanation for the predominance of the buildup of tritium in the case of DKDP ($n/\mathrm{T} \sim 10^{-7}$). One process which might lead to an accumulation of tritium is the hypothetical "semicoherent" process for which a mechanism was described in Ref. 25. As a result of this process, tritons with an energy $\sim$5.5 MeV should have been generated. In this case there is the possibility of an implantation of fast tritons in the glass of the cells; this effect would help explain the presence of an anomalously high concentration of tritium absorbed by the glass of the cells during the temperature cycling (Table II).

It should also be noted that, because of the significant cracking and an intense desorption of deuterium in the DKDP crystals as they are temperature cycled near T_c, the acceleration mechanism for the *dd* reaction which we proposed in Ref. 9 might be extremely appropriate for explaining the observed effects. This mechanism involves the charged walls of cracks, which are bombarded by deuterons with energies on the order of 1–10 keV. However, this mechanism involves an equiprobable emission of cold-fusion products through the neutron and tritium channels, and it is totally incapable of explaining the appearance of the colossal excess amounts of tritium, which are also observed in other experiments on cold fusion.[5,6] The *dd*-reaction mechanism in DKDP crystals should thus apparently be thought of as an "acceleration-coherent" mechanism, since the conditions in these crystals are such that both processes could occur and could complement each other.[28]

Further research, including research on the tritium accumulation kinetics and on the neutron emission from large-mass single crystals, is being planned in an effort to find a more detailed explanation of the mechanism for the cold fusion reactions in DKDP single crystals.

In conclusion we wish to stress that, regardless of the actual mechanism for the cold fusion in deuterated ferroelectrics, this study has provided the first experimental observation of a reproducible emission of cold-fusion products (neutrons and tritium) in ferroelectric DKDP single crystals with controllable properties. This emission is observed near the Curie point.

We wish to thank A. A. Volkov and V. B. Kozin for furnishing the single crystals, S. Yu. Stefanovich for stimulating discussions, and A. V. Strelkov for interest in this study.

[1)] According to Ref. 8, such strong electric fields are possible in ferroelectric crystals only at the surface, since in the interior they would be screened by field ionization of deuterium atoms.

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