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Generation of DD – Reactions in a Ferroelectric KD₂PO₄ Single Crystal During Transition Through Curie Point ($T_c = 220$ K)

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A new approach to develop a source of 2.45 MeV neutrons caused by polarization reversal in KD₂PO₄ single crystal (DKDP) during its passage through the Curie point ($T_c = 220$ K) is presented. The background of this approach is referred to observation of neutron/proton emission in DKDP during paraelectric - ferroelectric phase transition to spontaneous polarization state (and vice versa) upon the heating/cooling of crystal through Curie point $T_c = 220$ K. The proposed source is based on earlier established proof of deuteron acceleration and neutron generation in the crystalline lattice of ferroelectrics during their transition to spontaneously polarized state (polarization reversal). In order to obtain neutron yield for practical application, the proposed solution foresees a separate DKDP crystals serving as cathode and anode and undergo to simultaneous ferroelectric phase transition in low pressure deuterium atmosphere.

1. Introduction

In their Letter to Nature, B. Naranjo et al [1] presented a desktop neutron generator based on deuteron beam ionized and accelerated up to 100 keV by electric field arising from the spontaneous polarization of ferro/pyroelectric LiTaO₃ crystal at a low pressure deuterium gas adsorption. The results of this work are appeared to be solid with respect to both experimental and theoretical aspects of 2.45 MeV DD-neutron production. However, a certain early history exists on a deuteron fusion driven by a strong crystalline electric field.

The fracto-fusion caused by a weak neutron emission has been observed during the fracture of deuterated dielectric crystals (i.e. induced polarization), where cracks serve as tiny accelerators [2,3]. Such crack propagation in crystalline dielectrics is accompanied by high-energy electron and X-ray emissions up to 100 keV (see [2,3] and references therein), similarly to that emitted by pyroelectrics with a large spontaneous polarization [1]. The secondary deuterons generated by fast electron emission can be accelerated up to keV energy range inside this cracks by a strong electric field ($> 10^8$ V/m) released on the rapture of chemical bonds in these crystals. The DD-reaction in such crack is prompted by deuteron bombardment of “deuterated target” (i.e. crack’s wall surface).

The idea to use spontaneous polarization in deuterated ferroelectrics to obtain DD-fusion was first explored at the Institute of Physical Chemistry, The Russian Academy of Sciences, Moscow [4,5]. The reason was a strongest electric field in the lattice ($E \sim 10^{10}$ V/m) that arises in the course of spontaneous polarization during heating or cooling of KD₂PO₄ (DKDP) single crystal through the Curie point $T_c = 222$ K (i.e. exactly the same presupposition that is considered in [1], when explaining benefits of the spontaneous polarization for deuteron acceleration). In these articles, the neutron emission has been detected during DKDP crystal passage through the Curie point resulting in spontaneous polarization. KH₂PO₄ (KDP) single crystal (similar to DKDP one, but containing just hydrogen, instead) did not demonstrate neutron emission above the background level during transition through its particular Curie point ($T_c = 123$ K due to isotopic effect.). In contrast to [1], in the prior experiments [4,5] a deuterium atmosphere was not used because the host-deuterium in ionic form is presented in the lattice. In such a case the DKDP crystal serves both as a source of accelerated deuterons (suggesting no need for their field ionization) and the deuterated target, simultaneously. These DKDP crystals have generated reproducible neutron emission, though three orders of magnitude lower than that in [1]. The cause of a high neutron yield in [1] is referred to both full deuterium ionization provided by tungsten tip and deuteron source-target separation, resulting in significantly higher D⁺ energy and current compared to those inside the cracks.

Furthermore, later a team from Tokyo Metropolitan University, inspired by fracto-fusion works [2-5], performed experiment on neutron generation during the fracture of piezoelectric LiNbO₃ crystals (with the structure similar to that of the LiTaO₃) in a D₂ atmosphere with pressure $p \geq 30$ kPa [6,7]. In this particular experiment the ionization of deuterium gas occurred at the surface of the charged piezoelectric particles, similarly to [1], so the source of deuterium was a D₂ gas. Since the surface of the LiNbO₃ particles with adsorbed deuterium served as a neutron target (contrary to the separate ErD₃ target in [1]), the detected neutron yield was also much lower than that obtained by Naranjo et al.

Here we demonstrate our results on DD-reaction (neutrons and tritium) in DKDP during transition through Curie point that are enhanced by recent measurements of in-situ 3 MeV proton emission using CR-39 track

detectors. We also discuss a new approach to develop a source of 2.45 MeV neutrons caused by polarization reversal in KD_2PO_4 single crystal (DKDP) during its passage through the Curie point ($T_c = 220 \text{ K}$).

2. Experimental

- Transitions of KD_2PO_4 (98 % D purity) single crystals through Curie point $T_c = 220 \text{ K}$ by heating-cooling cycle using LN2 cryostate. Control of transition with Thermally stimulated depolarization (TSD).
- Neutron measurements with $\text{BF}_3 - 7$ counter proportional neutron detector.
- Tritium measurements with liquid scintillator technique with dissolving of DKDP crystals in H_2O .
- Recently done: 3.0 MeV proton measurement with CR-39 (open and filtered with Al foils) attached to DKDP plates cycled through Curie point.

3. Experimental results

The amplitude-differential spectra of neutron emission are presented in Fig. 1. As seen, the sum of spectra for DKDP exposed at the neutron detector during ferroelectric phase transition near 220 K are in good agreement with Cf-252 amplitude spectrum.

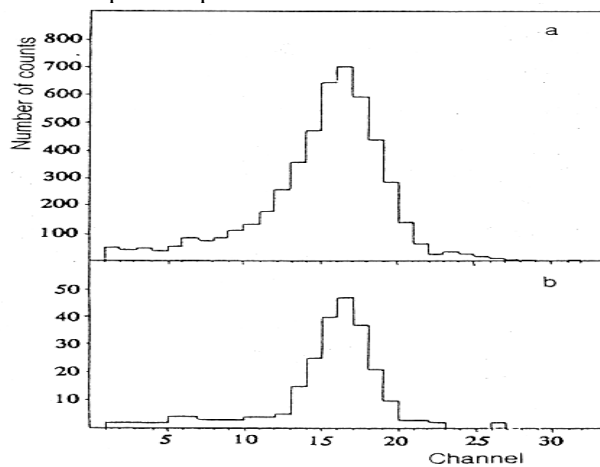


FIG. 1. Distribution of the pulses arriving at the neutron detector among channels of the pulse height analyzer. a—From the Cf^{252} neutron source; b—during temperature cycling of DKDP crystals near T_c .

Figure 1

This picture unambiguously confirms presence of neutron emission during transition through Curie point.

The neutron counts are consistent with spontaneous polarization occurring in DKDP crystals during ferroelectric phase transition as it is shown in Fig. 2.

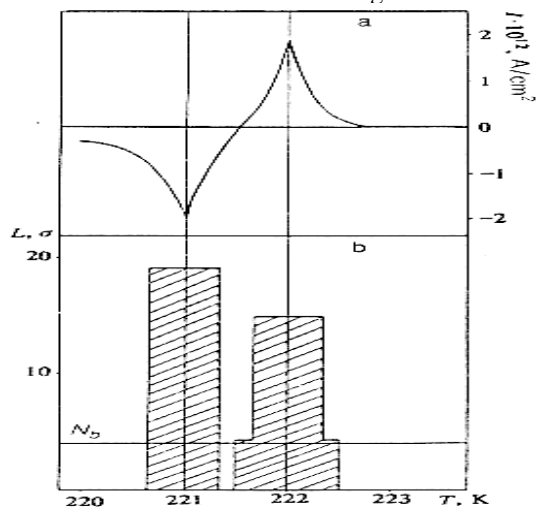


FIG. 4. a—Spectrum of thermal depolarization during the heating of DKDP sample near T_c ; b—corresponding histograms of neutron bursts. Here L, σ , is the confidence level of the observed neutron bursts, and N_0 is the average background level.

Figure 2

Parameters of neutron emission, including flux intensity in DKDP crystals near T_c , in comparison with KDP (KH₂PO₄) and those obtained outside the T_c temperature interval are shown in Table 1.

Table 1

Parameters of Neutron emission in KH₂PO₄ and KD₂PO₄ single crystals in various temperature intervals

Crystal	Temperature interval: ΔT , K	Foreground-Background, cps	Neutron emission Φ_n , n/s
KH ₂ PO ₄ (m=0.8 g)	121-125	0.000±0.005	-
KH ₂ PO ₄ (m=0.8 g)	219-223	0.001±0.005	-
KD ₂ PO ₄ (m=0.8 g)	210-215	0.001±0.005	-
KD ₂ PO ₄ (m=0.5 g)	219-223	0.012±0.004	0.40±0.13
KD ₂ PO ₄ (m=0.8 g)	219-223	0.020±0.004	0.61±0.16
KD ₂ PO ₄ (m=0.8 g)	221.0±0.5; 222.0±0.5	0.025±0.004	0.75±0.15

As seen from the Table 1 the neutron emission is occurred only in DKDP crystals (not in KDP) and strictly in the temperature range corresponding to spontaneous polarization (219-223 K).

Generation of DD-reaction in DKDP crystals during transition through Curie point is confirmed by measurement of 3.0 MeV protons emission (d(d, p)t reaction) during similar transitions (Figure 3):

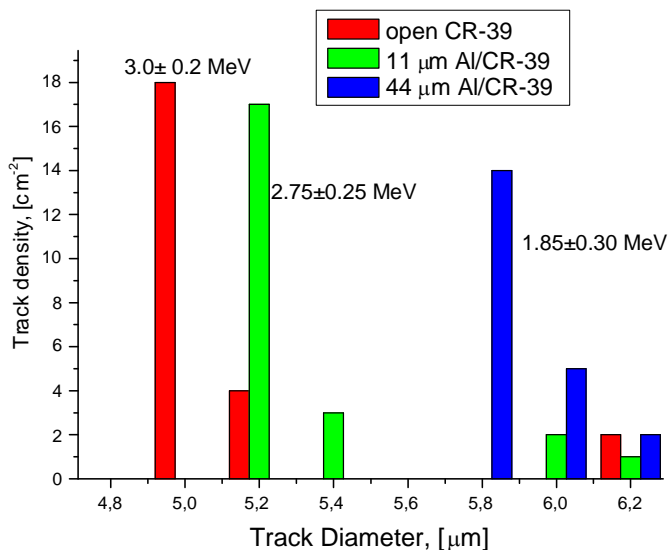


Figure 3: 3 MeV Proton Emission: 40 transitions through Curie point in a row with attached open and filtered CR-39 detectors: The detected track diameters are consistent with 3 MeV proton energy losses in Al foils [8].

Tritium concentration in DKDP has been measured by liquid scintillation technique. The sample were 100 times cycled through Curie point in a glass capsule. After the cycling the crystals, glass and gas atmosphere of the ampule were analyzed. The results (in units of 10^9 T-atoms per gram of DKDP) are presented in Table 2

Table 2

Tritium concentration measurements by liquid scintillator technique (in units of 10^9 T-atoms per gram of DKDP)

# sample	DKDP crystal	Cell glass	Cell atmosphere	Total
control	5.0 ± 0.2			5.0 ± 0.2
*100 cycles through Tc	5.9 ± 1.1	8.3 ± 1.3	9.2 ± 1.5	23.4 ± 1.8

*The DKDP sample $m = 0.5$ g was subjected to 100 heating –cooling cycles in isolated glass cell at atmospheric pressure. Accordingly to this measurement the yield of tritium was found to be $Y_t = (1.82 \pm 0.25) \times 10^8$ [t/transition]. Neutron and proton emission Yields per transition through Curie point:

- Neutron yield from $d(d, n)He3$ reaction was found to be $Y_n = 20 \pm 4$ [n/transition-g] in the range of 219-223 K.
- Branching ratio between neutrons and tritium should be: $Y_n/Y_t \sim 10^{-7}$.
- Recently measured (with CR-39) 3.0 MeV proton yield from $d(d,p)t$ reaction, taking into account efficiency of CR-39 detection of 3.0 MeV protons ($\epsilon_p = 0.076$) at $\langle N_p \rangle = 0.5 \pm 0.1$ count/transition was found to be

$$Y_p = 12 \pm 3 \text{ [p/transition-g]},$$

Thus, there is no significant difference between DD-reaction channels when 3.0 MeV proton and neutron yields are compared. That is why, the 7 order enhancement of tritium channel compared to neutron and proton yield observed during DKDP cycling through Curie point cannot be explained by conventional DD-reaction.

4. Novel neutron source based on ferroelectric transition in DKDP

Taking into account both results of works [1] and [4,5] we are presenting novel type of neutron source based on transition of DKDP single crystal through Curie point at $T_c = 220$ K (polarization reversal) and employed on principles of a high voltage glow discharge in a low pressure deuterium atmosphere (**Fig. 4**). In order to increase deuteron kinetic energy the “paired DKDP crystal system” suggesting both ferroelectric electrode pair [9] operating as cathode and anode is proposed.

The polarization reversal in both single crystal DKDP cathode (having a conic shape) and rounded shape anode separated by a spacing of ~ 1 cm is caused by heating/cooling through the $T_c = 220$ K. To this goal, both cathode and anode inserted inside metallic Cu frames that are attached with the dielectric epoxy composition to the cryostat wall. The liquid nitrogen (LN) cryostat is supplied with a heater allowing change in DKDP temperature near 220 K. In order to keep a constant positive electric charge at the cathode surface, the back face of the DKDP cathode is grounded via an ohmic contact (silver film) sputtered on top of the crystal attached to the epoxy layer. To keep negative electric charge at the surface of the anodic DKDP piece, a small positive potential is applied to its back face. The sealed cryostat chamber is filled with D_2 gas at pressure about 10 mtorr. During simultaneous transition through Curie point both cathode and anode are triggered to a spontaneous polarization state. In such a case, the strength of electric field in the gap between their surface would reach a value $E \sim 10^8$ V/cm due to a large value of spontaneous polarization in DKDP ($P_s \sim 5.0 \times 10^{-6}$ C/cm²). This electric field will stimulate an intensive field emission of energetic electrons (up to hundreds keV range) from the cathode surface and X-ray generation during anode electron bombardment. The electron emission will produce mostly secondary D^+ ions from the anode because the binding energy of deuterons in DKDP lattice is very small (hydrogen bonds). The deuterons escaping from the anode will be accelerated in the electrostatic field in the interelectrode gap toward the surface of DKDP cathode and bombard it with energy about 100 keV in the laboratory system. At realistic time of the passage through T_c chosen as ~ 10 s (e.g., $\Delta T = 222-220$ K and $dT/dt \sim 0.2$ K/s, comparable with Ref. [5]), the expected electron current density would be $J_e \sim 1.0$ μ A/cm². Assuming a secondary ion emission coefficient ~ 1 and taking into account possibility of direct deuteron field emission from the DKDP anode tip, we suppose that the D^+ current density would be the same order of magnitude as an electron emission one. Thus, at $E_d \sim 100$ keV and $J_d \sim 1$ μ A/cm², the expected neutron yield can be estimated as 10^5-10^6 n/s in 4π ster. Notice that the deuterons escaping from the anode surface would be fully substituted by deuterium ion adsorption at the DKDP surface and its ferroelectric properties could be recovered, resulting in multiple usage of the source (passage through T_c) to generate neutrons.

The proposed neutron source can be further modified resulting in production of enhanced stationary neutron yield by application of pulsed voltage (several kV) between cathode and anode at constant temperature $T <$

T_c (e.g. $T = 77$ K). This application provides polarization reversal at spontaneously polarized ferroelectric state by triggering electric dipole moment orientation by AC electric field.

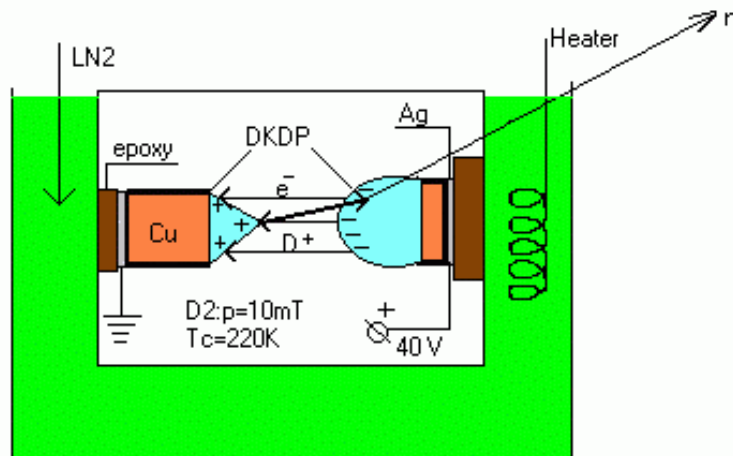


Figure 4: Diagram of cryo-set up of 2.45 MeV neutron source based on a high voltage discharge between DKDP single crystalline cathode and anode during their passage through Curie point [10].

Conclusions

- Generation of DD-reaction resulting in neutron and 3 meV proton emissions in DKDP ferroelectric crystal during passage through Curie point has been established.
- It was shown that neutron and proton channels in DKDP crystal give comparable nuclear yields. Large amount of tritium production ($\sim 2 \times 10^8 T^3$ at./transition) cannot be referred to usual DD-reaction.
- The factor of spatial separation of deuteron source and target in deuterated ferroelectrics can be used to obtain large neutron yield during transition of these ferroelectrics to spontaneously polarized state.
- New type of neutron source based on electric discharge between two ferroelectric KD_2PO_4 (DKDP) crystals during their polarization reversal at $T = 220$ K in D_2/T_2 atmosphere is proposed. No high voltage power supply. Small size. Projected intensity $Y_n = 10^6(D_2) \cdot 10^8(T_2)$ n/s.
- Potential applications include Homeland security and oil exploration as a bore hole source

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