Muon Catalyzed Fusion in Gas/Liquid/Solid D2 under Controlled Ortho-Para Ratio

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Introduction

Meanings of μ CF experiment with ortho-para controlled D2

- •Search for efficient μ CF toward energy break-even
- -Understand rich physics of muonic atom and molecule in μCF

Early studies

\Rightarrow only in solid at 3 K

[A.Toyoda et al. Phys.Rev.Lett.24 243401(2003)]

- Thin solid layer targetFusion proton detection
- •Thermal conductivity

The results are disagree with theory

 \Rightarrow condensed-matter effects in solid?



We studied ortho-para effect in solid, liquid, and gas

- •Large volume target (<30 litter at STP)
- Fusion neutron detection
- Raman spectroscopy method

- \Rightarrow Good S/N ratio
- \Rightarrow Long mean free path
- \Rightarrow Absolute o/p ratio

Ortho-Para Effect

$d\mu F=3/2 + \mathbf{D}_{2} K_{i}=x v_{i}=0 \Leftrightarrow [(dd\mu)J=1, v=1 dee]K_{f}=x v_{f}=7$

Ortho-para effect was firstly pointed out by Leon and Cohen



Condensed Matter Effect





@M9B TRIUMF

Detection System:

- NE213 scintillators (N1-N4)
- Beam counters (B1, B2)
- Electron counters (E1-E8)

<u>Target (<30 litter at STP):</u> **Phase: solid, liquid, gas T=5K-38.5K** φ=0.03-1.4 **Ortho 38-99%**

Picture of Experiment



Target Preparation

Gas Purification and Normal target:

Pd filter \Rightarrow Make pure (< 1ppb) & normal(66% ortho) target Ortho-rich target (66-100% ortho):

Paramagnetic-catalyst method (Al₂O₃:Cr₂O₃=2:1)

Spin-flip induced by the magnetic gradient of paramagnetic catalyst Hold 1 day in converter at 10K filled with the catalyst

Para-rich target (21-66% ortho):

- Preferential-adsorption method (Al₂O_{3 : p.s. \sim 3mm)}
- Para-D2 preferentially adsorbed on Al2O3 due to the surface potential Flow D2 gas to Al2O3 under the equilibrium adsorption condition at 25K

Raman spectroscopy:

The Raman spectroscopy method was employed to measure the orthopara composition. (20 mW 473nm blue laser: Class 3B)

Raman Spectra



Targets with 21%-99% o-D2 were produced !!!

Currently in all field of science, we are the only group that can control the ortho concentration in such a wide range.

Fusion Neutron Spectra in Solid and Liquid



The ortho-para dependence of the neutron time spectra was observed

$$\begin{split} \lambda_{\frac{3}{2}} & \text{and} \quad \lambda_{\frac{3}{22}} & \text{were obtained from fittings with the static model} \\ & F_{res}(t,t_0) + F_{nr}(t,t_0) \\ & F_{res}(t,t_0) = \frac{1}{3}\phi\beta_{\frac{3}{2}}\tilde{\lambda}_{\frac{3}{2}}\exp(\frac{(\lambda_n^{res})^2\sigma^2}{2})\exp\{\lambda_n^{res}(t-t_0)\}\operatorname{erfc}(-\frac{t-t_0-\lambda_n^{res}\sigma^2}{\sqrt{2}\sigma}) \\ & F_{nr}(t,t_0) = \frac{1}{6}\phi\beta_{\frac{1}{2}}\tilde{\lambda}_{\frac{1}{2}}\exp(\frac{(\lambda_n^{nr})^2\sigma_A^2}{2})\exp\{\lambda_n^{nr}(t-t_0)\}\operatorname{erfc}(-\frac{t-t_0-\lambda_n^{nr}\sigma_A^2}{\sqrt{2}\sigma_A}) \end{split}$$



The resonant rates in o-D₂ is 15% lower than that in n-D₂ 8% decrease of the HF transition rates in ortho-rich targets

Even in liquid phase, the effect is disagree with the theory High-density effect

Neutron Time Spectra in Gas



Delayed Structure



Fusion neutron spectrum in o-D2 deviates from simple exponential function around 150 ns (>>1/\$\$\$ ns=6 ns)

The delayed structure cannot be described by static formation rate assuming rapid dµ thermalization

Slow $d\mu$ thermalization

The probable explanation for the structure is the **transient ddµ** formation during slow dµ thermalization \Rightarrow epithermal effect

Density Effect (Preliminary Data)

How are the liquid data and the gas data connected? The complete data set of the density dependence with ortho-para D2 (ϕ =0.03-1.3) were obtained in May 2007!





Surprisingly, it is not simple linear density dependence!

Target density (= intermolecular interaction of D₂) is an essential parameter in d-d μ CF.

Resonant Condition in High Density

Possible explanations of **density dependence**

1. Level shift on excited state of [(ddµ)dee] in a high density

High precision (0.05 meV) in isolated system

Study of the excited D2 in a high density

2. The "three body effect" in d-d μCF

- $dt-\mu CF$ -14meV(for ortho) -11meV(for para)
- $dd\mu$ 0.8meV(for ortho) -3.3meV(for para)



Further theoretical investigation of a high-density effect will be important

Summary

We performed the ortho-para experiment in various condition. The ortho-para ratio of D_2 was perfectly controlled (21-99%).

- •Ortho-para effects in solid were consistent with the previous experiments.
- •Ortho-para effects in liquid were similar to those in solid.
- Yield of fusion neutrons was enhanced in ortho D_2 gas.
- Neutron time-spectra in ortho D_2 gas had a delayed structure.
- Ortho-para effects were dependent on the target density.

Epithermal effect and **High-density effect** are important even in d-d μCF

•Competition between $d\mu$ thermalization and $dd\mu$ formation

•Resonant dd μ formation with intermolecular interaction of D2

Proposed Condition

Experimental condition in E968 and E1061



Observed ortho-para effect in high-density region($\phi > 0.88$) was drastically different from that in low-density region($\phi < 0.17$). We propose the measurement at middle density !! Why the measurement in the proposed conditions are important?

-Details of E968 and E1061 will be explained in the following slides-

Neutron Detection



Neutron Detection

To observe 2.45 MeV d-d fusion neutrons

Gamma-rays:

n- γ discrimination by using NE213 + PSA & DLA modules

Muon nuclear capture neutrons:

Demand on a electron event after the neutron event



Target Preparation

Gas Purification:

Pd filter \Rightarrow Make pure (< 1ppb) & normal(66% ortho) target

<u>Make ortho-rich target</u>:

Catalyst method

Hold 1 day in converter at 10K filled with paramag. catalyst





Raman Spectroscopy

For the complete controlling of OP state, absolute measurement of the composition of ortho-para mixture is essential.

The Raman spectroscopy method was employed for the first time in μCF to determine the OP ratio of our target.



Hyperfine Transition Rate 1

 $\begin{array}{l}\lambda_{31}^{SC} \\ \frac{\lambda_{31}}{22}, \ \lambda_{22}^{BD} \\ \frac{\lambda_{31}}{22} \\ \text{and} \ \Gamma_{\frac{1}{22}} \\ \frac{\lambda_{31}}{2} \\ \frac{\lambda_{31}}{2} \\ \text{No ortho-para dependence of} \end{array} \begin{array}{l} \text{were derived by using the ortho-para} \\ \lambda_{31} \\ \frac{\lambda_{31}}{22} \\ \frac{\lambda_{31}}{22$





The reason for the discrepancy has not yet been explained

Back-Decay Rate



The back-decay rates $\Gamma_{\frac{1}{2}\frac{1}{2}}$ are consistent with the previously observed rates and the theoretical rate (1.5 ns⁻¹)

Epithermal effect



Epithermal effect in d-t μCF [*Jeitler et al. Phys. Rev. A51 (1983) 1757*] Strong resonance ~hundreds eV

Acceleration mechanism

•muon transfer $d\mu \rightarrow t\mu$

•spin flip

In our case, the structure is clearly depends on the ortho-para ratio!

Lowest resonance ~meV

It is difficult to extract the exact rates of $\frac{\lambda_3}{\frac{1}{2}}$ and $\frac{\lambda_{31}}{\frac{1}{22}}$

Competition between $d\mu$ thermalization and $dd\mu$ formation



Origin of Bump Structure



•Molecular formation from epithermal $d\mu$ (F=1/2)

× Small OP dependence of the resonant condition for $d\mu$ with F=1/2

•Para heating

× Small OP dependence of $d\mu$ decelerating process

•Molecular formation from epithermal d μ (F=3/2)

slow thermalization

Preliminary Calculations by Adamczak



<u>Time-Dependent ddµ Formation Rate 1</u>



We calculated the "timedependent" ddµ formation rates

Time evolution of overlap integral between Maxwellian $d\mu$ energy distribution and the resonant condition

$$\lambda_{\frac{3}{2}}(t) = \int \mathrm{d}\epsilon f(\epsilon, \epsilon_{avg}(t)) \lambda_{\frac{3}{2}}(\epsilon)$$

Mean energy has time dependence

<u>Time-Dependent ddµ Formation Rate 2</u>



•The "time-dependent" rates in ortho-, normal-, and para-D2 were calculated

•The time dependence of the hyperfine transition rate was considered as the following;

$$\tilde{\lambda}_{\frac{3}{2}\frac{1}{2}}(t) = \lambda_{\frac{3}{2}\frac{1}{2}}^{SC} + \frac{\Gamma_{\frac{1}{2}\frac{1}{2}}}{\tilde{\lambda}_{f}}(\tilde{\lambda}_{\frac{3}{2}}(t) - \lambda_{nr}) \qquad \lambda_{\frac{3}{2}\frac{1}{2}}^{SC} = 24 \ \mu s^{-1} \text{ and } \Gamma_{\frac{1}{2}\frac{1}{2}} = 1.5 \ ns^{-1}$$

Time-Dependent ddµ Formation Rate 3



The calculated neutron time spectra for o-D2 and n-D2 reproduce the experimentally observed spectra

All previously observed rates in normal targets should be reviewed taking the time-dependence into account ???

Stronger resonance in o-D2 is consistent with theory

Condensed Matter Effect



Condensed Matter Effect by Adamczak



- slow $d\boldsymbol{\mu}$ thermalization in solid
- Phonon effect \Rightarrow small
- Resonance energy shift \Rightarrow -1.8 meV
- Resonance broadening $\Rightarrow 0.5 \text{ meV}$



The effect of epithermal $d\mu$ atoms is too strong in theory <u>Slope</u>

Exchange resonance energy in o-D2 for that in n-D2

Further effect in the condensed-matter is required to reproduce experiment

Resonant Fusion Neutrons Yield per Muon



Difference of neutrons yield between ortho (Y^on) and para (Y^pn) targets are defined as

$$\delta Y_n = \frac{1}{c_o^o - c_o^p} \frac{Y_n^o - Y_n^p}{Y_n^o + Y_n^p}$$

C^po: Ortho concentration for para target C^oo: Ortho concentration for ortho target

Even at the same T, OP effect is inversion between gas and liquid.

Strong density dependence is clearly observed.

Effect of the surrounding D2 molecule on the resonance condition is strong.