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

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Introduction

Meanings of $\mu$CF experiment with ortho-para controlled D2
- Search for efficient $\mu$CF toward energy break-even
- Understand rich physics of muonic atom and molecule in $\mu$CF

Early studies

$\Rightarrow$ only in solid at 3 K
- Thin solid layer target
- Fusion 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) $\Rightarrow$ Good S/N ratio
- Fusion neutron detection $\Rightarrow$ Long mean free path
- Raman spectroscopy method $\Rightarrow$ Absolute o/p ratio
Ortho-Para Effect

\[ d\mu F=\frac{3}{2} + D_2 K_i=x \ \nu_i=0 \Leftrightarrow [(dd\mu)J=1,\nu=1 \ \text{de}e]K_f=x \ \nu_f=7 \]

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

\[ [M. \ Leon \ and \ J.S. \ Cohen, \ Phys. \ Rev. \ A31 \ (1985) \ 2680] \]

Ortho-Para State of D₂

Parity of total nuclear spin → Parity of rotational state

Energy gap between J=0 and J=1 is 7.4meV (86K)

\begin{itemize}
  \item Resonant energy \sim \text{meV}
  \begin{align*}
    \varepsilon_0 \sim 1.96 \text{eV} \\
    \varepsilon_0 \sim 1.96 \text{meV}
  \end{align*}
\end{itemize}

\begin{tabular}{cccccc}
\hline
\varepsilon_f (meV) & F & K_i & K_f & S \\
\hline
-6.649 & 3 & 1 & 0 & \frac{1}{2} \\
-3.325 & 1 & 1 & 1 & \frac{1}{2} \\
0.8157 & 0 & 0 & \frac{1}{2} & \frac{1}{2} \\
3.312 & 1 & 2 & 0 & \frac{1}{2} \\
4.14 & 0 & 1 & 0 & \frac{1}{2} \\
10.78 & 2 & 0 & 2 & \frac{1}{2} \\
\hline
\end{tabular}
**Condensed Matter Effect**

Theory for condensed-matter


Muonic atom scattering in solid
- Bragg scattering
- Coherent and incoherent phonon
- Internal excitations of D2
  ⇒ Non thermalization of $d\mu$ (2meV)

Resonant $dd\mu$ formation in solid & liquid
  ⇒ Resonant energy shift (-1.8meV)
  Broadening (0.5meV)

Ortho-para effect in solid was not completely understood
  ⇒ Further experimental study under various condition are required!!
Experimental Setup

50 MeV/c $\mu^-$ beam @ M9B TRIUMF

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

Target (<30 litter at STP):
- Phase: solid, liquid, gas
- T=5K-38.5K
- $\phi=0.03-1.4$
- Ortho 38-99%
Target Preparation

Gas Purification and Normal target:
   Pd filter ⇒ 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 (Al₂O₃: p.s.~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 ortho-para composition. (20 mW 473nm blue laser: Class 3B)
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.
The ortho-para dependence of the neutron time spectra was observed

\[ \frac{\lambda_3}{2} \] and \[ \frac{\lambda_{31}}{22} \] were obtained from fittings with the static model

\[
F_{\text{res}}(t, t_0) + F_{\text{nr}}(t, t_0)
\]

\[
F_{\text{res}}(t, t_0) = \frac{1}{3} \phi \beta_3 \frac{\lambda_3}{2} \exp\left(\frac{(\lambda_{3}^{\text{res}})^2 \sigma^2}{2}\right) \exp\left\{\lambda_{3}^{\text{res}} (t - t_0)\right\} \text{erfc}\left(-\frac{t - t_0 - \lambda_{3}^{\text{res}} \sigma^2}{\sqrt{2} \sigma}\right)
\]

\[
F_{\text{nr}}(t, t_0) = \frac{1}{6} \phi \beta_2 \frac{\lambda_2}{2} \exp\left(\frac{(\lambda_{2}^{\text{nr}})^2 \sigma_A^2}{2}\right) \exp\left\{\lambda_{2}^{\text{nr}} (t - t_0)\right\} \text{erfc}\left(-\frac{t - t_0 - \lambda_{2}^{\text{nr}} \sigma_A^2}{\sqrt{2} \sigma_A}\right)
\]
Obtained Rates in Condensed Matter

The resonant rates in \( \text{o-D}_2 \) is 15\% lower than that in \( \text{n-D}_2 \)

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

36.2 K
\( \phi = 0.17 \)

Normal \( \Rightarrow \) reproduced by the static model

\[ \frac{\lambda^\text{Normal}_3}{2} = 4.5(2)(3) \]
\[ \frac{\lambda^\text{Normal}_{31}}{22} = 40(1)(2) \]

Neutron yield in o-D2 is 12% enhanced
consistent with theory for dilute gas
Delayed Structure

Fusion neutron spectrum in o-D\textsubscript{2} deviates from simple exponential function around 150 ns ($>>1/\phi$ ns=6 ns)

The delayed structure cannot be described by static formation rate assuming rapid d\mu thermalization

Slow d\mu thermalization

The probable explanation for the structure is the transient dd\mu formation during slow d\mu 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 ($\phi=0.03-1.3$) were obtained in May 2007!

- **para 100%**
- **ortho 100%**
Surprisingly, it is not simple linear density dependence!

Target density (= intermolecular interaction of D2) is an essential parameter in d–d μCF.
Resonant Condition in High Density

Possible explanations of density dependence

1. Level shift on excited state of \[(dd\mu)\text{dee}\] 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 \(\mu\text{CF}\)
   - \(dt-\mu\text{CF}\)  -14 meV (for ortho) –11 meV (for para)
   - \(dd\mu\) 0.8 meV (for ortho) -3.3 meV (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 $\mu$CF

- Competition between $d\mu$ thermalization and $dd\mu$ formation
- Resonant $dd\mu$ formation with intermolecular interaction of D$_2$
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

To observe 2.45-MeV d-d fusion neutrons

Gamma-rays:
Pulse shape discrimination method
n-γ discrimination by using NE213 scintillator + PSA & DLA modules

Muon nuclear capture neutrons:
Delayed electron condition
Demand on a electron event after the neutron event
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 an electron event after the neutron event
Target Preparation

Gas Purification:
- Pd filter ⇒ Make pure (< 1ppb) & normal (66% ortho) target

Make ortho-rich target:
- 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 $\mu$CF to determine the OP ratio of our target.

Raman Laser System

Placed near experimental area for in-situ measurement
Hyperfine Transition Rate \(1\)

\[
\begin{align*}
\lambda_{3\frac{1}{2} 2\frac{1}{2}}^{SC}, \quad \lambda_{3\frac{1}{2} 2\frac{1}{2}}^{BD} \quad \text{and} \quad \Gamma_{1\frac{1}{2} 2\frac{1}{2}} \quad \text{were derived by using the ortho-para dependence of} \quad \lambda_{3\frac{1}{2}} \quad \text{and} \quad \lambda_{3\frac{1}{2} 2\frac{1}{2}}. \\
\text{No ortho-para dependence of} \quad \lambda_{3\frac{1}{2} 2\frac{1}{2}}^{SC} \quad \text{and} \quad \Gamma_{1\frac{1}{2} 2\frac{1}{2}} \quad \text{was assumed}
\end{align*}
\]

\[
\begin{align*}
\lambda_{3\frac{1}{2} 2\frac{1}{2}}^{SC} &= \frac{\tilde{\lambda}^{\text{ortho}}_{3\frac{1}{2} 2\frac{1}{2}} (\tilde{\lambda}^{\text{normal}}_{3\frac{1}{2}} - \lambda_{nr}) - \tilde{\lambda}^{\text{normal}}_{3\frac{1}{2}} (\tilde{\lambda}^{\text{ortho}}_{3\frac{1}{2}} - \lambda_{nr})}{\tilde{\lambda}^{\text{normal}}_{3\frac{1}{2}} - \tilde{\lambda}^{\text{ortho}}_{3\frac{1}{2}}} \\
\lambda_{3\frac{1}{2} 2\frac{1}{2}}^{BD} &= \tilde{\lambda}^{\text{ortho}}_{3\frac{1}{2} 2\frac{1}{2}} - \lambda_{3\frac{1}{2} 2\frac{1}{2}}^{SC} \\
\Gamma_{1\frac{1}{2} 2\frac{1}{2}} &= \frac{\tilde{\lambda}_{f}^{\text{ortho}}_{3\frac{1}{2} 2\frac{1}{2}} (\tilde{\lambda}^{\text{normal}}_{3\frac{1}{2} 2\frac{1}{2}} - \tilde{\lambda}^{\text{ortho}}_{3\frac{1}{2}})}{\tilde{\lambda}^{\text{normal}}_{3\frac{1}{2}} - \tilde{\lambda}^{\text{ortho}}_{3\frac{1}{2}}}
\end{align*}
\]
Hyperfine Transition Rate 2

\[ \lambda^{SC}_{31/22} \] is about 40% of the theoretical one

The reason for the discrepancy has not yet been explained
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

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

Lowest resonance $\sim \text{meV}$

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

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

Epithermal effect in $d$-$t$ $\mu$CF


Strong resonance $\sim \text{hundreds eV}$

Acceleration mechanism

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

• spin flip
Bump Structure 2

(a) 36.2 K \( \varphi = 0.17 \)
- Ortho (99%) [\( /10\text{ns} \)]
- Normal (67%) [\( /10\text{ns} \)]
- Difference [\( /40\text{ns} \)]

(b) 36.2 K \( \varphi = 0.17 \)
- Ortho (90%) [\( /10\text{ns} \)]
- Normal (67%) [\( /10\text{ns} \)]
- Difference [\( /40\text{ns} \)]

(c) 36.2 K \( \varphi = 0.17 \)
- Ortho (84%) [\( /10\text{ns} \)]
- Normal (67%) [\( /10\text{ns} \)]
- Difference [\( /40\text{ns} \)]

(d) 36.2 K \( \varphi = 0.085 \)
- Ortho (99%) [\( /20\text{ns} \)]
- Normal (67%) [\( /20\text{ns} \)]
- Difference [\( /80\text{ns} \)]
**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 $d\mu$ (F=3/2)
  - Slow thermalization
Preliminary Calculations by Adamczak

- Bump structure for ortho
- Neutron yield
- Bump structure for para

The calculation does not reproduce correctly our experimental results concerning neutron time spectra.

Condensed-matter effects in the dense gaseous deuterium of our target (0.17LHD)?
Time-Dependent $d\text{d}\mu$ Formation Rate 1

We calculated the “time-dependent” $d\text{d}\mu$ formation rates

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

$$\lambda_{\frac{3}{2}}(t) = \int d\varepsilon f(\varepsilon, \varepsilon_{avg}(t)) \lambda_{\frac{3}{2}}(\varepsilon)$$

Mean energy has time dependence.
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;

\[ \tilde{\lambda}_{3/2}^{1/2}(t) = \lambda_{3/2}^{SC} + \frac{\Gamma_{1/2}}{\tilde{\lambda}_f} (\tilde{\lambda}_{3/2}^{1/2}(t) - \lambda_{nr}) \]

\[ \lambda_{3/2}^{SC} = 24 \, \mu s^{-1} \text{ and } \Gamma_{1/2} = 1.5 \, ns^{-1} \]
The calculated neutron time spectra for o-D$_2$ and n-D$_2$ 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-D$_2$ is consistent with theory.
The effect is obviously different between the condensed target and gas target.
Condensed Matter Effect by Adamczak

\[ d\mu(F = \frac{3}{2}) + nD_2, \quad T = 3 \text{ K} \]

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

[A. Adamczak et al., PRA 64 (2001)]
Comparison with theory by Adamczak

Prompt

The effect of epithermal dμ atoms is too strong in theory

Slope

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 ($Y_\text{o}n$) and para ($Y_\text{p}n$) targets are defined as

$$\delta Y_n = \frac{1}{C_{\text{p,o}} - C_{\text{o,o}}} \frac{Y_\text{o} - Y_\text{p}}{Y_\text{o} + Y_\text{p}}$$

$C_{\text{p,o}}$: Ortho concentration for para target
$C_{\text{o,o}}$: 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.