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MUON CATALYZED DT FUSION AT LOW TEMPERATURE

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Abstract

The catalytic fusion of hydrogen isotopes induced by muons was discovered in 1956 by the Alvarez group in the Berkeley bubble chamber. The dream of large fusion rates triggered by a single muon did however not materialize, because the rates for the production of mesic molecules were found to be small in terms of the muon life time. This situation changed dramatically in the late 70's due to the discovery of resonance effects in d~d formation and the theoretical prediction of extremely fast rates in the deuterium-tritium (DT) cycle.

Several experimental programs are currently under way to study muon catalyzed DT fusion. The largest efforts were made by Jones et al. at Los Alamos, New Mexico and by Breunlich et al. at the Swiss Institute for Nuclear Research (SIN). First results indeed confirm the existence of a very rapid DT cycle producing multiple fusions of the type d~t + α + n + μ + 17.6 MeV.

This paper presents a systematic experimental investigation of MCF at low temperatures (12-35K) in gas, liquid and solid of various densities and tritium concentrations. Our improved analysis allows a clear separation of intrinsic dt sticking from kinetic effects. Strongly density-dependent cycle rates with values up to 1.93 x 10^{4} s^{-1}, yields of 124 fusions per muon, and ωs lower than theoretically predicted, but with no strong dependence on either tritium concentration or density are reported.

Introduction

A review given by Ponomarev /1/ and the first issue of Muon Catalyzed Fusion /2/ gives the latest comprehensive account of both the theoretical and experimental description of muonic catalyzed fusion (MCF). At the Conference in Gatchina, just concluded in May 87, many new results /3/ were presented including parts of the work presented here. Measurements made at SIN include much new data which has been partially analysed and the other papers presented here deal with some of these new results. Here I shall concentrate on the cycle rate and sticking probability for the gas-liquid-solid data obtained in March/April 1987.

The yield of DT neutrons per muon is given as follows

\[ Y_n = \frac{\phi \lambda_c}{\lambda_0 + w \phi \lambda_c} = \left[ w + \frac{\lambda_0}{\phi \lambda_c} \right]^{-1} \]

where \( \phi \) is the density relative to liquid Hydrogen, \( \lambda_0 \) is the natural decay rate of muons, \( w \) is the effective sticking fraction including losses due to impurities and other hydrogen fusion channels, and \( \lambda_c \) is the normalized cycle rate averaged over the processes in the paths of the muon to the mesonic atom. \( \lambda_c \) has an unexpected density dependence as well as a complex temperature variation. \( w \) however in the range of our measurements is relatively constant for the tritium concentration of 40\% where \( \lambda_c \) is near its maximum and is about 0.60\%.

In 1983 we performed the first series of measurements in DT gases at low densities (\( \phi=1\% \)), in 1984 and 1985 we used DT mixtures of higher densities (gas runs with \( \phi=1\%\text{-}8\% \)) including liquids (\( \phi=120\% \)). These measurements have been extensively reported on recent conferences and are now published /4/.

In parallel, similar but complementary DT experiments were conducted at LAMPF by a collaboration led by S.E. Jones /5/, and more recently at KEK (Japan) K. Nagamine et al. and at LNPI Gatchina by D. Balin et al. /2/.

Indeed, all these experiments have demonstrated the enormous rapidity of the muonic DT cycle, producing at certain conditions every few nsec a DT reaction and yields exceeding 100 fusions per muon.

The purpose of our latest run was to provide additional data using a new target specifically made for studying the cycle rate variations with density near the solid liquid interface. In addition the rates in nonequilibrated mixtures, and a few gas phase density points were explored. The controversy concerning sticking probability was explored. In particular the protonium effects were studied and the nitrogen impurity effect was verified. The new methods of analysis have been applied to the data which are insensitive to various counter efficiencies and changes were made to improve background effects.

Our new results confirm basically the picture about the DT kinetics and about sticking, evaluated from previous experiments /4/. Now a greatly enlarged and improved set of data is available which will eventually lead to much more precise information about MCF.

Setup of the 1987 SIN Experiment

The experimental arrangement of our previous DT measurements, performed at SIN during 1983-85, is well documented in recent literature/4/. For the new series of runs, that were performed in March/April 1987 at SIN, we have included several significant improvements and developments. They allow us, to study in much more detail the density dependence of cycle rates and sticking, the effects of molecular compositions and temperatures and the influence of fusion side
channels. Possible distortions from electronic effects can also be investigated with much better accuracy.

Fig. 1A shows the new target cell, developed at Lawrence Berkeley Lab and built at SIN, which was used throughout the 1987 MCF series. The inside of the copper cell is silver coated to detect possible muon transfer to walls and the cell has a useable volume of 20 ccm. It is designed for operating pressures up to 20 bar, so as to run DT mixtures in all 3 thermodynamical phases gas - liquid - solid. The cooling to cryogenic temperatures is provided from the top by a piston operated refrigeration unit (10W at 20K) and from the bottom by a liquid Helium transfer system and reservoir at 4K. The heat switches are 2 Copper plates separated by thin gaps that can be evacuated or flushed with He gas and are operated to regulate the heat flows according to needs. With a 10W heating resistor, controlled by temperature sensors, the target temperature is stabilized to the desired values.

The main filling line acts also for evacuation to reach ultra clean target conditions. A residual pressure of $3 \cdot 10^{-8}$ Torr was reached. In the top part 2 sniffing lines were installed to circulate DT gas which evaporates directly from the liquid phase into a new automatically operated mass spectrometer system for online analysis of the molecular hydrogen compositions.

This target system was operated between 10K and 50K at stable conditions, using DT mixtures in liquid and solid phase at densities $90\% < \phi < 150\%$ of liquid protium and in gaseous phase ($\phi < 30\%$).

Fig. 1B shows the detector arrangement installed around the new DT target. A narrow muon beam of 59 - 63 MeV/c (depending on the target density) of 25,000 muons/sec and 2% FWHM momentum resolution was used in SIN's $\mu E 4$ area. Electronic muon stops were identified by a $2 \cdot 3 \cdot 3a \cdot 4$ coincidence. Veto counter 3a, having a 15mm bore, defined the beam size. A ratio of "good" to electronic muon stops of about 40% was obtained for liquid/solid mixtures and 10% to 25% for most gas runs. A pileup circuitry rejected events when another particle passed the beam telescopes (2 3 or 2 3a) 0-8 µsec prior to the muon stop. Pileup events for second particles after the muon stop were registered into the computer for later offline study.

There are 2 pairs of plastic counters (30\(\times\)30 cm\(^2\)), acting as charged particle detectors, which measured the electrons from muon decay.

For absolute calibration and high resolution measurements (recoil energies and rise times) 2 NE213 liquid scintillation detectors were mounted (areas 127 cm\(^2\), thickness 2.5 cm and 10 cm each,) with pulse shaping circuits for n-gamma discrimination.
Fig. 1a: New DT Target. The copper cylinder is embedded between 2 heat switches. Below is the Helium refrigeration system.

Fig. 1b: Experimental set-up of detectors (plane view). B1-B4 are fast plastic bar scintillators run in the multiple hit mode. NE 213 I und II are absolutely calibrated neutron detectors with n-γ pulse shape discrimination. Ge is a Germanium diode to view X-rays from muon transfer to walls or impurities. Plastic counters 2 and 3 form the muon telescope, in anticoincidence to 3a and to 4. EL and ER are pairs of charged particle detectors, to measure electrons from muon decay.
There are 4 plastic bar detectors (each 5x5x25 cem, \(\varepsilon_n=0.3-0.4\%\)) used as fast counters for 14 MeV neutrons in the routing mode for recording of up to 4 neutron hits and corresponding energy signals per muon per detector.

A Ge detector was placed behind the target vacuum vessel to monitor X-rays from muon transfers to impurities or target walls. The energy and time spectra were registered. The analysis of the Ge spectra provide the upper limits for impurities during the experimental runs, indispensible for the evaluation of the lower limits of sticking in muon catalyzed DT fusion.

Below the target, a BGO detector (9 cm diameter x 9cm depth) was positioned for the measurement of high energy gamma radiation, e.g. produced in \(p+d\) fusion (5.4 MeV) or \(p+t\) fusion (19 MeV). During the runs with pure PT mixtures a NaI detector was installed additionally.

Time distributions were measured using CAMAC TDC's with 8 usec wide time ranges and 1 nsec time width. The trigger definition was made electronically simple (e.g. any detector event within an 8 usec time gate with respect to the muon stop signal), so as to produce undistorted raw time spectra.

A new "Starburst-\(\mu\)VAXII" system was installed increasing the data handling capacity 10 fold as compared to previous \(\mu\)CF runs. During the data runs online sampling and analysis was possible allowing inspection of the data quality.

**New results on DT fusion**

In a straight series of successive data runs we have observed all existing fusion reactions induced by muons. We have first used very pure target fillings of deuterium \((c_d = 99.6\%)\) and of tritium \((c_t = 99.7\%)\) at several densities. The main DT measurements were then done with DT mixtures at about 25 different atomic and molecular compositions, temperatures and densities.

In this work only a first and preliminary analysis of new DT data is presented and comparison with our previous experiments is made. The delayed coincidence method and the method of analyzing the time distribution of successive neutron hits was applied. This method was successfully used for the analysis of 1984 measurements in liquid DT.

This method is based on the assumption, that in a fast cycling mesomolecular system the time distribution of fusion events is simply described by

\[
N_f(t) = \phi \lambda_c e^{-\lambda t}, \quad \lambda_n = \lambda_0 + \nu \lambda_c
\]

where \(\lambda_c\) is the DT cycle rate normalized to unit density of liquid H\(_2\) \((\phi = 1 = 4.25 \cdot 10^{22} \text{ atoms/ccm})\) and \(\nu\) is the average loss per cycle ("raw sticking" coefficient) apart from muon decay \((\lambda_0)\).

For a neutron counter of efficiency \(\varepsilon_n\) using multiple hit routing (deadtime \(\Delta t=50\text{ns}\)), expressions (2) leads to observable time spectra as follows:
Fig. 2 shows the multiple hit time distributions of one of the bar
detectors, measured in liquid DT with \( c_T = 40\% \). The drawn line is a fit according
to expressions (3) and (4) by additional inclusion of a small component of
accidental events (which was also measured separately). The resulting values for
\( \epsilon_n \phi \lambda_c \) and \( \nu \phi \lambda_c \) are in very good agreement with our data measured 1984 at nearly
the same conditions (\( \phi = 1.2 \), \( c_T = 0.36 \) and 0.42). Using the absolute normalisation
of 1984 the observed cycle rate \( \phi \lambda_c \) in liquid equilibrated DT (\( c_T = 0.4 \)) is
130 Me
and we present in the following figures a set of new results on \( \lambda_c \) and \( \nu \).

Fig. 3a shows a summary of all measured DT cycle rates plotted versus
tritium concentration \( c_T \), normalized to unit density. The 1983/84 data and the
fit according to our kinetic model/4/ are included also. Evidently, 2 strong
effects are dominating this presentation, a) the density dependence of \( \lambda_c \) b) the
effect of non equilibration of molecular mixtures. For the first time, data
are now available (at \( c_T = 40\% \) and \( c_T = 90\% \)), which were taken at exactly the same
experimental conditions, except that the mixture was first filled without
equilibration and was then equilibrated between the runs by warming it up to
300K. This effect of equilibration thus demonstrates directly the dominance of
\( \lambda D_2/d\mu t \), the rate of mesomolecule formation on the D_2 molecule!

The largest cycle rate was observed in solid non equilibrated DT at \( c_T = 40\% \)
to be 193 Mc with a sticking factor \( \nu = 0.57\% \), see Fig. 3b. This condition
produced the highest neutron yield per muon \( Y_n = 124 \pm 10 \), seen at SIN!

The combination of our previous with the new data allows for the first time
to present the density dependence of DT cycle rates over a range of more than 2
orders of magnitude. This is shown in Figure 3b for \( c_T = 40\% \) and 90\%. The low \( \phi \)
(83/84) data are interpolated between results obtained at \( c_T = 34, 46, 50 \) and 88\%).

Two components are clearly emerging at \( c_T = 40\% \), a strong density dependence at
\( \phi \leq 20\% \) which may be attributed to rapidly changing fast muon transfer \( q_{1S}/4,7/ \)
and a rather weak (quadratic) dependence at \( \phi > 20\% \) which may be explained by
triple collisions/8/. Figs. 3a/3b also demonstrate the effect of non
equilibration (increase of D_2 and T_2 over DT concentrations from equilibrium
(\( c_{D_2} : c_{DT} : c_{T_2} = c_2^2 : 2c_{D_2}c_T : c_T^2 \)) which is most significant at \( c_T = 90\% \). In
addition a small, but statistically significant temperature effect of cycle
rates in solid is found at \( c_T = 40\% \): \( \lambda_c(T=12K) : \lambda_c(T=17K) = 1.024 \pm 0.008 \)
suggesting a slight increase with the temperature approaching the 0 point.
Fig. 2:
Time distribution of fusion neutrons in one of the bar detectors (1st to 4th hit) for the condition of highest observed yield of 124 n/μ (c_t=40.5%, solid phase, non equilibrated). The fit shows agreement with the theoretical formulas (3)/(4).
Fig. 3a:
Normalized DT cycle rates $\lambda_c$ versus tritium concentration $c_t$ (summary plot of all measurements 1983 - 1987). The most pronounced effect is the density dependence of $\lambda_c$ and the effect of non equilibration of the DT mixture. The drawn curve is a fit to the 1984 data in liquid equilibrated DT.

Fig. 3b:
Density dependence of the normalized cycle rates $\lambda_c$ at $c_t = 40\%$ (dots) and $c_t = 90\%$ (triangles, preliminary data). The 1983/84 data are given as squares ($c_t = 0.4$) and inverse triangle ($c_t = 0.9$). The results at $\phi \lesssim 0.2$ are very striking. Also shown are the effects of non equilibration, which become as expected very large at high $c_t$. The quoted error bars are almost exclusively due to systematic errors, estimated by using different analysis methods.
Figures 4a and 4b display our new results of sticking \( w \) in comparison with previous data, plotted versus tritium concentration \( c_t \) and versus density \( \phi \), respectively. Only "raw stickings" of the new measurements are given, since the correction for other channels (especially \( dud \) and \( ttt \) fusion) will require a detailed analysis of the mixture compositions and of the whole DT kinetics. Nevertheless, some conclusions can already be drawn from the raw data:

1. The most carefully measured points (at \( c_t=40\% \)) agree well with our previous results. Raw sticking from data with larger cycle rates (e.g. taken in solid phase or in non equilibrated mixtures) are somewhat smaller, which is mainly due to smaller contributions from the \( ttt \) channel. This is most strikingly seen in liquid DT at \( c_t=90\% \), where we observe \( \lambda_c=31.6 \text{ Me} - w =1.07\% \) in the non equilibrated state, versus \( \lambda_c=7.8 \text{ Me} - w =2.5\% \) in the equilibrated mixture.

2. At one condition 2.3\% protium was added to the \( c_t=40\% \) (liquid) mixture, which increased the raw sticking by \( 0.086 \pm 0.021\% \). This results allows us to determine the sticking correction due to the presence of protium with good accuracy.

3. Apart from factors discussed in points 1 and 2, we have no significant variance of sticking with density \( \phi \) at a level of less than 5\% (\( 0.9<\phi<1.5 \)). Our complete set of data is consistent with DT sticking \( \omega_s \) being constant at large densities, with \( \omega_s \approx 0.45\% \) the error from absolute calibration being \( \pm 8\% \).

4. The precise evaluation of sticking at low densities requires a more refined analysis than the one presented in this work, since background components become relatively large and need to be properly eliminated by coincidence requirements. Thus our 1984 analysis yielding \( \omega_s \) (low \( \phi \)) = \( (0.5 \pm 0.1)\% \), is still the most precise assertion.

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References
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Fig. 4a: Sticking results plotted versus tritium concentration $c_t$, at high densities (liquids-solids, all SIN data, dots = 1987 results). The point marked with "P" shows the effect of a 2.3% protium admixture. The points measured in non-equilibrated mixture are lower (especially at high $c_t$), which is due to a smaller contribution of $tut$ sticking. The open squares show pure DT sticking, after correction for other fusion channels (1984 data only).

Fig. 4b: Raw sticking $w$ plotted versus density $\phi$. The corrections for obtaining DT sticking $\omega_s$ are only given for the 1984 SIN data and include the ± 8% uncertainly from absolute calibration. As can be seen, the density dependence is rather weak. The error bars include the systematic errors.