

MESIC MOLECULES AND MUON CATALYSED FUSION

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NORTH-HOLLAND PUBLISHING COMPANY - AMSTERDAM

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Received 6 December 1981

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Abstract:

We discuss the energy spectrum of mesic molecules, reviewing the recent advances in the methods of calculations and in the measurements of the energy levels.

We also present recent ideas concerning the possibility of using the muon catalysed fusion for energy production.

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PHYSICS REPORTS (Review Section of Physics Letters) 86, No. 4 (1982) 169–216.

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1. Introduction

Muon catalysed fusion is the name given to a series of reactions resulting in the fusion of two hydrogen isotopes nuclei which are kept close together by a negative muon.

This process, firstly observed by Alvarez et al. (1957), is particularly interesting for the study of some peculiar molecular systems. Also it could offer, at least in principle, a nonconventional approach to energy production.

The interplay of different disciplines is a characteristic feature of the study of muon catalysed fusion (m.c.f.) and, more generally, of the physics of exotic atoms*. In m.c.f. atomic and molecular physics are interwoven with nuclear physics and, perhaps, with energetics.

A bubble chamber picture of m.c.f. is useful for the discussion of the several reactions—atomic, molecular and nuclear—which are involved in the process. Fig. 1 exhibits a picture taken in a bubble chamber filled with hydrogen and a small (2.2×10^{-5}) deuterium contamination. A negative muon entering into the chamber is slowed down and eventually stopped by ionising collisions at point A. The tracks (B–C) and (D–F) correspond to the motion of a muon with an energy of 5.4 MeV. The track originated at E represents the electron produced in muon decay. The history of the muon while travelling between points A and B (or C and D) can be traced through several steps.

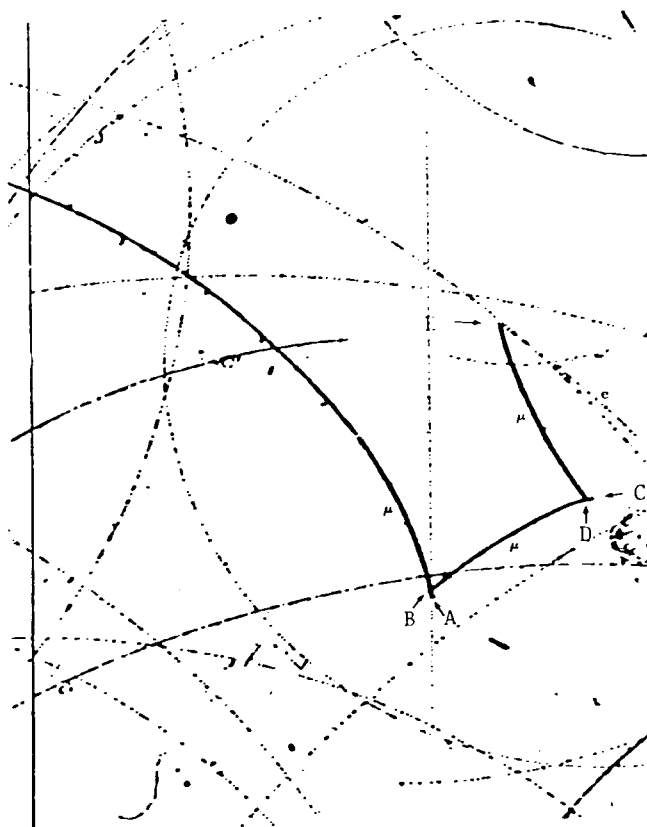


Fig. 1. A picture of muon catalysed fusion, showing an example of two successive fusion events. (Photograph by courtesy of Prof. R.H. Hildebrand.)

* For general reviews on the field of exotic atoms see for example Hughes and Wu (1975), Fiorentini and Torelli (1977) and Crowe et al. (1979).

Once the muon has been slowed down to an energy of some eV a $(p\mu)$ atom is formed through the Auger process:



The muonic hydrogen atom $(p\mu)$ is electrically neutral and small, its Bohr radius being

$$a_\mu = a_0 m_e / m_\mu \approx 250 \text{ fm} , \quad (1.2)$$

where a_0 is the electron Bohr radius. Consequently, it has many of the properties of the neutron and can diffuse freely through the liquid hydrogen (L.H.). When it comes close to the deuteron in an HD molecule the muon is transferred to the deuteron[†]:



Subsequently a $(pd\mu)$ molecular ion is formed when the $(d\mu)$ atom hits a hydrogen atom in the target[‡]:



In the literature the term “mesomolecule” is currently used for systems like $(pd\mu)$. This term is twice wrong since the muon is not a meson and the $(pd\mu)$ system is electrically charged. The expression “muonic molecular ion” would be more appropriate. Nevertheless we will adhere to the common usage for the sake of brevity and simplicity.

Mesomolecules, being composed by two heavy particles bound together by a lighter particle of opposite charge, resemble the H_2^+ ion in the same way as muonic atoms resemble ordinary atoms. It follows immediately from dimensional considerations that the scales of length and energy are respectively inversely and directly proportional to the mass of the light particle. Hence, one expects that mesomolecules are extremely small objects and are characterised by much higher energies than ordinary molecules.

Indeed, one finds that the internuclear equilibrium distance R_0 and the vibrational energy E_{vib} are of the order

$$R_0 \approx 500 \text{ fm} , \quad (1.5)$$

$$E_{\text{vib}} \approx 250 \text{ eV} . \quad (1.6)$$

The above figures show that in a mesomolecule the nuclei are kept quite close and with high relative kinetic energy. The equivalent conditions in a plasma require a density*:

$$D \approx R_0^{-3} \approx 10^7 \text{ L.H.D.} \quad (1.7)$$

and a temperature:

[†] Of course the rate for reaction (1.4) is proportional to the deuterium concentration, but it is generally so high that, after the $(p\mu)$ is formed, the transfer reaction is the most likely process (see in the next section the discussion of the Berkeley experiment for a proof of this statement).

[‡] It is worth observing that the probability of forming a $(dd\mu)$ system is negligible at such a small deuterium concentration.

* L.H.D. = liquid hydrogen density.

$$T \approx E_{\text{vib}}/k \approx 3 \times 10^6 \text{ K} . \quad (1.8)$$

This situation is the same as in the interior of a white dwarf. It is intuitively clear that under such extreme conditions nuclear synthesis reactions will occur quite rapidly.

A proton and a deuteron fuse through an electromagnetic interaction:



In the mesomolecule a virtual photon can be absorbed by the muon (see fig. 2) thus giving:



The energy of the muon is $E_{\mu} = 5.4 \text{ MeV}$, precisely what was measured in the picture.

In conclusion, this is the mechanism which produces the muon tracks (B–C) and (D–E). The chain breaks at the point E where the muon decays.

The muon acts as a catalyst of a chain of nuclear reactions in that (a) it keeps the two nuclei close to each other and with high relative velocity and (b) once the two nuclei fuse the muon is ready to start a new reaction. Hence the name “muon catalysed fusion” is given to the whole series of reactions†.

The reaction we analysed is just an example of a wide class. Other fusion processes, in which different hydrogen isotopes are involved, proceed along similar lines. They are listed in table 1, first column.

The complexity and the richness of the field is clearly shown in the above analysis. Indeed, it results that many problems of atomic, molecular and nuclear physics have to be studied in order to get a complete understanding of the m.c.f.

Really, the different processes are of different relevance in the determination of the fusion yield. It comes out that the atomic capture, the transfer reaction and the nuclear reactions in mesomolecules occur in times which are generally shorter than the muon lifetime

$$\tau_{\mu} = 1/\lambda_{\mu} \approx 2.2 \times 10^{-6} \text{ s} . \quad (1.11)$$

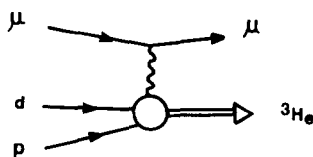


Fig. 2. Feynman graph of reaction $(pd\mu) \rightarrow {}^3\text{He} + \gamma + \mu$.

Table 1. List of nuclear reactions between hydrogen isotopes.

Fusion reaction	Q value
$p + p \rightarrow d + e^+ + \nu$	2.2 MeV
$p + d \rightarrow {}^3\text{He} + \gamma$	5.4 MeV
$t + p$ (50%)	4 MeV
$d + d \rightarrow {}^3\text{He} + n$ (50%)	3.3 MeV
${}^4\text{He} + \gamma$ (~0%)	24 MeV
$d + t \rightarrow {}^4\text{He} + n$	17.6 MeV
$p + t \rightarrow {}^4\text{He} + \gamma$	20 MeV
$t + t \rightarrow {}^4\text{He} + n + n$	10 MeV

† In principle nuclear reactions can occur in ordinary molecules, but the average nuclear distance is too large to yield an appreciable rate. One computes that the probability of nuclear reactions in the HD molecule is 10^{-24} per year and per cubic meter of liquid HD [Gershtein and Zel'dovich (1961)]. This means that in the water of all the oceans a nuclear reaction occurs every some 10^7 years.

The formation of the mesomolecule is the process which requires the longest time. For this reason the fusion yield is essentially connected with the process of mesomolecular formation. Also, it has been shown that the mesomolecular formation rate is particularly sensitive to the mesomolecular energy spectrum. One concludes that experiments on muon catalysed fusion are particularly relevant for the study of mesomolecule formation and of mesomolecular spectroscopy.

As mentioned at the beginning, this is one of the several reasons of interest in the study of m.c.f. Recently, by means of experiments on the m.c.f. it was possible to determine some energy levels of mesomolecules with an accuracy ~ 1 meV on an energy scale of ~ 1 keV. To appreciate the value of such an accuracy it is worth recalling that the vacuum polarization shift of these states is of the order of several meV. This indicates that accurate spectroscopic studies of mesomolecules are now possible.

Since mesomolecules are a clean three body system bound by the Coulomb interaction, the above results allow refined tests of methods of solution of the Schroedinger equation.

Also, these measurements have some implications on the calculation of the energy levels of ordinary molecules. In ordinary molecules one can apply successfully the so called adiabatic (Born–Oppenheimer) approximation, whose validity is based on the smallness of the ratio of the electron mass to the nucleus mass. Only to a sophisticated level of precision it is necessary to take into account corrections to this approximation. On the other hand, in mesomolecules nonadiabatic effects are quite important due to the finite value of the ratio of the muon to nucleus mass, η . Consequently nonadiabatic effects are extremely enhanced. An experiment with an accuracy 10^{-6} allows to check a theoretical calculation up to terms of order η^5 included. The corresponding term in the calculation of ordinary molecules would account for a relative energy shift of some 10^{-17} !

Another reason of interest in the m.c.f. lies in the speculation of using this process for the purpose of energy production. The idea is very simple. As shown in fig. 1, muons can induce fusions of hydrogen nuclei, and an energy of a few MeV is released in any reaction (see table 1 for the Q -value of the various reactions). One can then speculate about reaction mechanisms and experimental arrangements such that the muons can catalyse so many fusions that the released energy exceeds the energy cost of muon production. This hope, frequently expressed at the time of the experimental discovery of m.c.f., was soon frustrated by quantitative analyses of the energy balance. In the last few years, however, some new facts provided support to the idea of practical applications of the m.c.f. It was shown that in a suitable deuterium-tritium mixture a muon can catalyse hundreds of fusions, a number which exceeds by two orders of magnitude what was previously expected. Furthermore, an ingenious scheme was devised in order to minimise the energy cost of muon production and increase the energy output of the reaction chain. These recent investigations have brought m.c.f. closer to practical applications, although the dream is still a dream.

Several aspects of the m.c.f. have been extensively discussed in many review papers [Gershtein and Zel'dovich (1961), Alvarez (1972), Massey et al. (1974), Gershtein and Ponomarev (1975), Ponomarev (1977), Ponomarev (1978), Meyer-ter-Vehn (1979), Rafelski (1979), Breunlich (1981), Fiorentini (1981)]. Particularly we would like to recommend Gershtein and Zel'dovich (1961) and Alvarez (1972) as extremely enjoyable introductions to the subject. This paper will be concerned in the developments of the field in the last six years. In this period there have been significant advances both in theory and experiment, which however have not yet been comprehensively reviewed. Particularly we want to discuss how this new, accurate spectroscopy of mesic molecules has become possible and the recent developments in the idea of using m.c.f. for energy production.

Before entering this central part of the paper, we present in section 2 a short historical survey of m.c.f. The subject is now about 35 years old and there has been a continuous development in theory and

experiment, stimulated by each other. We feel the reader can be interested in getting a view of the evolution of the field.

Next, in sections 3 and 4, we present the methods of calculation of the energy levels of mesomolecules and show the most recent results. The formation processes of mesic molecules are discussed in section 5, where we also show the close connection between the energy spectrum and the formation rate*.

This is a rather important point. Indeed, the accurate experimental information on the energy levels arise from this connection.

A discussion on the possibility of using m.c.f. for energy production is given in section 7. In this respect it is important to understand the causes by which the catalytic chain comes to an end. Beside muon decay, another effect can occur: this is the process in which the muon sticks to one of the charged products of the fusion reaction. The sticking process can be really the final bottle-neck of the m.c.f. Recent theoretical evaluations of this effect are reported in sect. 6.

The conclusions of the paper are summarized in section 8.

As we mentioned at the beginning, the m.c.f. can be interesting to physicists working in quite different fields. Having this in mind, we tried to write down a paper which is readable to a wide audience. The cost to be paid is that any reader will find tedious some section and naive some other. We cannot but apologise to all our readers.

2. Historical outline

The possibility of nuclear reactions in mesomolecules was pointed out since the very identification of the muon and of the π^- meson. In their famous experiment Lattes, Occhialini and Powell (1947a,b) obtained several photographs in which a secondary track, belonging to a charged particle with an energy of about 4 MeV, emerged from the stopping point of a meson. The process was correctly interpreted by the authors as the decay of the—yet undiscovered— π meson:

$$\pi^- \rightarrow \mu^- + \nu. \quad (2.1)$$

Clearly, the experiment was so important that any alternative explanation of the result should be carefully considered. In this spirit Frank (1947) discussed the possibility that the secondary track originated from the fusion reaction:

$$(pd\mu) \rightarrow {}^3\text{He} + \mu. \quad (2.2)$$

Frank himself showed that this interpretation was hardly tenable, due to the too small amount of deuterium present in the photographic emulsion. However, the idea of m.c.f. was born.

Soon after, in a very frequently quoted paper, which today is no longer available but by oral tradition, A. Sakharov (1948) discussed the subject with a scope and a foresight which were ahead of his time. In fact, Sakharov realized that, once a mesomolecule was formed, nuclear fusion was to occur almost immediately and, more important, he conceived the possibility of using the m.c.f. for energy production.

* All the collisional rates are normalized to liquid hydrogen density, $D_0 = 4.2 \times 10^{22}$ atoms/cm.

Five years later the m.c.f. was independently rediscovered by Ya. Zel'dovich (1954), who also made the first detailed estimates of the associated phenomena.

The first observation of m.c.f. was again a re-discovery of the process and a fully unexpected result [Alvarez et al. (1957)]. The Alvarez group were analysing the first hydrogen bubble chamber photographs in Berkeley, with the aim of studying the kaon interactions. The quality of the kaon beam was poor by modern standards, since there was a substantial contamination of muons. In fact, this undesired feature was the source of a very interesting result. The authors observed plenty of "anomalous" pion decays, in which the tracks of the muons had an energy of 5.4 MeV, instead of the well known value of 4.1 MeV corresponding to pion decay at rest. Also, the hypothesis of pion decay in flight could hardly be reconciled with the fact that the muon tracks were remarkably equal. The right interpretation, i.e. reaction (2.2), was soon put forth.

The Berkeley experiment rekindled the interest on the subject and prompted new experimental and theoretical work. Of course the first question to be answered was the dependence of the fusion yield Y_μ on the deuterium concentration C . In a subsequent series of exposures [Alvarez (1957), note added in proof] the Berkeley group observed that the fusion yield first increased by increasing C and then reached a limit for deuterium concentration of a few percent:

$$Y_\mu(C) = Y_0/(1 + k/C), \quad Y_0 = 0.025, \quad k = 0.0004. \quad (2.3)$$

This behaviour shows that, once the $(p\mu)$ atom is formed, the transfer reaction (1.3) is the most likely process, its rate being so fast that already at C about a few percent all the muons are transferred to the deuterium nuclei. If the fusion reaction would occur in flight:



then Y_μ should increase indefinitely with C .

From eq. (2.3) one sees that even for the most favourable deuterium concentration the fusion yield is rather small. Consequently it looks that there is no chance of getting energy out of reaction (1.10).

Soon after, a Liverpool group [Ashmore et al. (1958)] studied the reaction:



in a hydrogen target enriched with deuterium. The time distribution of photons showed the typical parent-daughter shape which one expects from a two step process:

$$dN_\gamma/dt \propto \exp(-at) - \exp(-bt). \quad (2.6)$$

The experiment could only provide constraints for the two parameters a and b :

$$2 \times 10^5 \text{ s}^{-1} < a < 9 \times 10^5 \text{ s}^{-1}, \quad 5.5 \times 10^5 \text{ s}^{-1} < b < 2.2 \times 10^7 \text{ s}^{-1}. \quad (2.7)$$

The group also measured the integrated photon yield, obtaining:

$$Y_\gamma = 0.34 \pm 0.06. \quad (2.8)$$

By comparing this value with the result of the Berkeley experiment one gets for the branching ratio B :

$$B = Y_{\mu}/Y_{\gamma} = 0.066 \pm 0.014. \quad (2.9)$$

It looks funny that the experimental discovery of the m.c.f. occurred through the observation of a relatively rare process. Most of the fusions which occurred in the bubble chamber were undetectable in the Berkeley experiment.

For several years the Berkeley and Liverpool experiments were the only source of data.

In the period 1957–1960 the various processes involved in m.c.f. were investigated in a large series of theoretical papers, particularly by Zel'dovich and Gershtein. The emerging picture turned out to be consistent with the existing data. Also, it resulted that, independently of the hydrogen isotopes mixture used, one could not expect more than a few fusions per stopped muon, since the mesomolecular formation rate was at most of order 10^6 s^{-1} . Clearly, this estimate did freeze any enthusiasm about the possibility of energy production from m.c.f. and more generally weakened the interest in the subject.

The status of affairs, as it was at the end of the fifties, is extensively reviewed in Zel'dovich and Gershtein (1961).

In the subsequent ten years the formation rates of ($\text{pp}\mu$), ($\text{pd}\mu$) and ($\text{dd}\mu$) were actually measured. In the case of ($\text{pp}\mu$) and ($\text{pd}\mu$) measurements at liquid hydrogen temperature (L.H.T.) and at room temperature (R.T.) gave the same results within the experimental errors. This is typical of the Auger formation process,



if the collision energy is much smaller than the Rydberg energy and the binding energy of the mesomolecule, according to the Bethe $1/V$ law for low energy inelastic cross sections [Landau and Lifshitz (1958)].

On the other hand, the formation rate of ($\text{dd}\mu$) showed a marked temperature dependence, increasing by an order of magnitude while going from L.H.T. to R.T. In 1967 Vesman, then a student of Gershtein, proposed that, besides the Auger process, the formation of mesomolecules could also occur by transfer of energy to the vibrational levels of ordinary molecules [Vesman (1967)]. Clearly, this process is possible only if there are very weakly bound states of mesomolecules, with a binding energy smaller than the dissociation energy of ordinary hydrogen molecules, a few eV. Due to the quantized structure of the molecular excitations, this process will have a resonant behaviour and thus the formation rate will be very sensitive to the kinetic energy of the mesoatoms*.

Vesman's conclusions could only be qualitative for the poorness of the experimental data and for the lack of precise calculations of the binding energy of mesomolecules.

The situation, as it was in the mid seventies, is reviewed by Gershtein and Ponomarev (1975).

Only in the last six years it has become clear that Vesman's idea can quantitatively explain the ($\text{dd}\mu$) mesomolecule formation rate. On the theoretical side, Ponomarev's group were able to calculate the energy levels of mesomolecules with an accuracy of order 0.1 eV. In this way they could prove the existence of a weakly bound state of the ($\text{dd}\mu$) mesomolecule with a binding energy of about 2 eV, thus

* It is worth noticing that the first mention of this possibility can be found in Gershtein and Zel'dovich (1961).

showing the effectiveness of the resonant formation process proposed by Vesman. Also, new experiments provided accurate measurements of the $(dd\mu)$ formation rate in a wide temperature range. It comes out that these data can be nicely interpreted within Vesman's scheme.

Furthermore, it was realised that the resonant formation process can be used as a powerful tool for accurate studies of the energy levels of mesomolecules. In this context it is worth mentioning the work in progress on the study of the hyperfine structure (H.S.) of the $(dd\mu)$ mesomolecule by a Vienna-SIN group [Breunlich (1981) and Breunlich et al. (1981)]. In conclusion, it appears that in the study of the $(dd\mu)$ system we are now at the second generation of experiments and we are close to a full understanding of the mesomolecule formation.

In the same theoretical framework which successfully explains the $(dd\mu)$ formation process it was predicted that the $(dt\mu)$ formation rate is particularly large [Gershtein and Ponomarev (1977)]. The prediction was confirmed by experiments [Bystritsky et al. (1980a,b,c)], which set a lower limit:

$$\lambda_{dt\mu} \geq 2 \times 10^8 \text{ s}^{-1}. \quad (2.11)$$

This supports the idea that in a suitable d-t mixture a muon can catalyse hundreds of nuclear reactions during its lifetime. Of course this has revived the dream of using m.c.f. for energy production. In this spirit an ingenious scheme was devised by Petrov (1978 and 1980) in order to minimise the energy cost of muon production and to increase the energy output of the reaction chain.

One has to observe that all this matter is in a rather speculative stage. In fact, new devoted experiments are needed in order to clarify the formation process of the $(dt\mu)$ mesomolecule and it is also necessary to investigate more deeply the feasibility of possible approaches to energy production by means of m.c.f.

3. Energy levels of 3-body Coulomb problem

3.1. Generalities

As previously mentioned, it is now possible to measure the energy of some mesomolecular levels with quite high accuracy, the error being at the level of 1 meV. One can therefore verify the validity of the theoretical calculations to a very high degree.

A difficulty which is not met in the study of ordinary molecules is the relatively large value of the ratio m_μ/M_N , where M_N is the mass of the lighter of the hydrogen isotopes constituting the nuclei of the mesomolecule. This implies that the Born–Oppenheimer method is a poor approximation for mesomolecules. Moreover, variational calculations are of limited applicability when excited states (like those whose energy can be accurately determined by experiment) are considered. As a consequence, peculiar methods are to be devised in order to achieve a sufficiently sharp estimate of mesomolecular levels.

The most relevant contributions of the last few years are due to the Dubna group, whose work we will mainly outline in the following. Essentially, their approach consists in a refined application of the adiabatic representation for the Coulomb three-body problem, where the drawbacks, often ascribed to this approximation in the literature, are overcome.

In this section we will discuss the pure Coulomb three-body problem. One should note that for the calculation of the actual mesomolecular levels to the desired degree of accuracy (~ 1 meV) one has to

take into account also other effects, like relativistic corrections, electron screening, nuclear size effects and nuclear interactions. This is deferred to the next section.

In conclusion, the Hamiltonian whose levels we will investigate in this section is the following:

$$\mathcal{H} = -\frac{1}{2M_a} \Delta_{\mathbf{R}_a} - \frac{1}{2M_b} \Delta_{\mathbf{R}_b} - \frac{1}{2m_\mu} \Delta_{\mathbf{R}_\mu} - \frac{1}{|\mathbf{r}_\mu - \mathbf{R}_a|} - \frac{1}{|\mathbf{r}_\mu - \mathbf{R}_b|} + \frac{1}{|\mathbf{R}_a - \mathbf{R}_b|}, \quad (3.1)$$

where the labels a and b are referred to the two nuclei, and units with $\hbar = e = 1$ have been chosen. In terms of the center of mass coordinate

$$\mathbf{R} = (m_\mu \mathbf{r}_\mu + M_a \mathbf{R}_a + M_b \mathbf{R}_b) / (m_\mu + M_a + M_b), \quad (3.2)$$

the internuclear coordinate

$$\mathcal{R} = \mathbf{R}_b - \mathbf{R}_a \quad (3.3)$$

and the coordinate

$$\mathbf{r} = \mathbf{r}_\mu - (\mathbf{R}_a + \mathbf{R}_b) / 2, \quad (3.4)$$

the Hamiltonian reads:

$$\mathcal{H} = -\frac{1}{2M_T} \Delta_{\mathcal{R}} - \frac{1}{2M_0} \left(\nabla_{\mathbf{R}} + \frac{\kappa}{2} \nabla_{\mathbf{r}} \right)^2 - \frac{1}{2m_0} \Delta_{\mathbf{r}} - \frac{1}{r_a} - \frac{1}{r_b} + \frac{1}{R}, \quad (3.5)$$

where

$$\begin{aligned} M_T &= m_\mu + M_a + M_b; & M_0^{-1} &= M_a^{-1} + M_b^{-1}; & m_0^{-1} &= m_\mu^{-1} + (M_a + M_b)^{-1} \\ \kappa &= (M_b - M_a) / (M_b + M_a); & r_a &= r_\mu - \mathbf{R}_a; & r_b &= r_\mu - \mathbf{R}_b. \end{aligned} \quad (3.6)$$

We have supposed $M_a \geq M_b$.

After separating the motion of the center of mass, one has the following Schroedinger equation:

$$H\Psi(\mathbf{r}, \mathbf{R}) = E\Psi(\mathbf{r}, \mathbf{R}), \quad (3.7)$$

with

$$H = -\frac{1}{2M_0} \left(\nabla_{\mathbf{R}} + \frac{\kappa}{2} \nabla_{\mathbf{r}} \right)^2 - \frac{1}{2m_0} \Delta_{\mathbf{r}} - \frac{1}{r_a} - \frac{1}{r_b} + \frac{1}{R}. \quad (3.8)$$

In the following we will tackle the problem of solving eq. (3.7) by several approaches of increasing complexity and precision. In so doing we will follow, to some extent, a chronological route, which will make clear the progress and improvements achieved at any step. The whole section is rather lengthy, and some parts of it might be skipped by the reader not interested in some technical details of the calculation, so we think it useful to indicate how the section is organised.

In section 3.2 a brief sketch of the Born–Oppenheimer (B.O.) approximation is given, since this approximation yields a qualitative picture of the mesomolecular spectrum. In section 3.3 the adiabatic basis is introduced, and in section 3.4 the perturbative approach to the solution of the system of equations obtained is described. Sections 3.5 and 3.6 discuss the subject of the dissociation limit, which has vexed people for a long time, and expose its solution in a general frame (3.5) and for the special case of a two-level approximation (3.6). Section 3.7 is devoted to the description of the most recent calculations, performed by a truncation of the whole system of equations. Finally, in section 3.8 the results of the various calculations are summarized, and compared with each other and with the results obtained by other means.

3.2. Born–Oppenheimer approximation

The simplest approach to the solution of eq. (3.7) is the Born–Oppenheimer approximation [Born and Oppenheimer (1927)]. Although it is inadequate for the problem under discussion, we think it may be useful to show how it works in this case, since it is widely known and can give some insight into some peculiar aspects of mesomolecules, striking the difference existing with the twin electronic problem, i.e. the H_2^+ ion.

As is well known, the B.O. approximation starts from considering the nuclei as infinitely heavy with respect to the negatively charged particle. Neglecting terms as small as m_0/M_0 in eq. (3.7), one has for the muon motion the Schroedinger equation:

$$\left(-\frac{1}{2m_0}\Delta_r - \frac{1}{r_a} - \frac{1}{r_b} + \frac{1}{R}\right)\varphi_R(\mathbf{r}) = E(R)\varphi_R(\mathbf{r}), \quad (3.9)$$

where R appears as a parameter. The solutions of the above equation are classified according to the principal quantum number n , the projection of the angular momentum over the internuclear axis m and their symmetry with respect to inversion in the midpoint between the nuclei, the uneven and odd states being labeled with g (gerade) and u (ungerade) respectively.

Since the scale of length for the solutions of eq. (3.9) is $1/m_0$, and the scale of energy is m_0 , one can guess the curve $E(R)$ by recalling the analog for the H_2^+ ion. In fig. 3 one finds the curve $E(R)$ for the

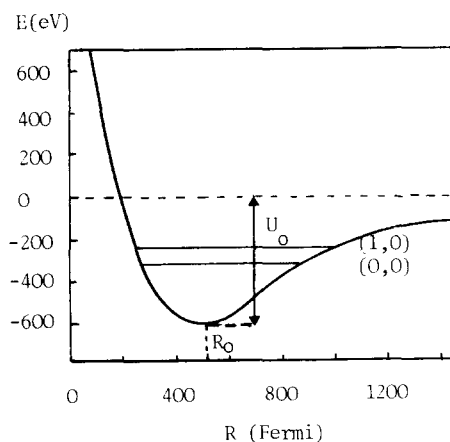


Fig. 3. Internuclear potential energy and spectrum for the $(dd\mu)$ mesomolecule in the Born–Oppenheimer approximation, described in section 3.2.

ground state $1\sigma g$ (spectroscopic notation for $n = 1$, $m = 0$, even parity) of the $(dd\mu)$ mesomolecule.

The next step is that of taking into account the internuclear motion in the potential $E(R)$. This is done by looking for a wave function which is the product of an internuclear motion wave function $u(\mathbf{R})$ times $\varphi_{\mathbf{R}}(\mathbf{r})$:

$$\Psi(\mathbf{R}, \mathbf{r}) = u(\mathbf{R})\varphi_{\mathbf{R}}(\mathbf{r}). \quad (3.10)$$

The justification for this procedure in the study of ordinary molecules is that the contribution of the vibrational and rotational energies to the total energy are expected to be small. Indeed, if m_l is the mass of the lepton (e or μ) in the field of two hydrogen isotopes, one can estimate

$$E_{\text{vib}} \approx (\hbar^4/a_1^4 m_l m_N)^{1/2} \approx (m_l/m_N)^{1/2} E_{\text{mol}}, \quad (3.11)$$

where E_{vib} is the vibrational energy, a_1 is the Bohr radius of the leptonic atom, m_N is the nucleus mass and E_{mol} is the value of a typical molecular term. Similarly, for the rotational energy E_{rot} one estimates

$$E_{\text{rot}} \approx \hbar^2/m_N a_1^2 \approx (m_l/m_N) E_{\text{mol}}. \quad (3.12)$$

If the lepton is an electron, its motion is much faster than the internuclear motion, and one can neglect the R dependence of $\varphi_{\mathbf{R}}(\mathbf{r})$. Then, averaging the exact Schroedinger equation over \mathbf{r} yields a Schroedinger equation for the internuclear motion.

In the case of mesomolecules, E_{vib} and E_{rot} cannot be considered as tiny corrections, as is clear from eqs. (3.11) and (3.12). In spite of this, the naive B.O. approximation can give an idea of the spectrum one can expect for mesomolecules. The $(dd\mu)$ case is particularly simple, since one can use the well known results referring to the H_2^+ ion in order to have a gross picture of the mesomolecular levels.

The potential $E(R)$ is sketched in fig. 3. Its minimum lies at $R_0 = 512$ fm, its depth being 577 eV. The simplest approach consists in approximating $E(R)$ with a harmonic potential around R_0 , and neglecting the R dependence of the centrifugal barrier. The effective potential for the internuclear motion is then

$$V_{\text{eff}} = E(R_0) + \frac{1}{2}M_0\omega^2(R - R_0)^2 + J^2/2M_0R_0^2,$$

where M_0 is the reduced mass of the nuclei and ω can be derived by an appropriate scaling of the analogous parameter for the H_2^+ ion, $\omega = 9.1 \times 10^{17} \text{ s}^{-1}$. Accordingly, the energy levels are:

$$E_{Jv} = E(R_0) + \omega(v + 1/2) + J(J + 1)/2M_0R_0^2.$$

One sees that bound states, i.e. states with $E_{Jv} < 0$, exist only for $v = 0$ and $J = 0, 1$: $E_{00} = -278$ eV, $E_{10} = -143$ eV. This situation is in sharp opposition to the case of ordinary molecules, where a rich vibrational and rotational spectrum exists.

Features similar to those just outlined (very small number of vibrational and rotational states) are indeed found after performing more accurate calculations. However, in order to do definite statements one has to go beyond the crude B.O. approximation. This can be done in a systematical way by using the so called adiabatic representation.

3.3. The adiabatic representation

The inadequacy of the B.O. approximation is substantially connected with the impossibility of describing the three-body wave function as the product of a muonic wave function times a nuclear wave function. The way-out from this difficulty is in principle simple, and consists in expanding the wave function over a complete set of solutions of the two-center Coulomb problem*.

These are the eigenfunctions of the Hamiltonian

$$h_0(\mathbf{R}) = -(1/2m_0)\Delta_r - 1/r_a - 1/r_b, \quad (3.13)$$

the distance between the centers being R . The complete set of these functions is the so called adiabatic basis. They can be indexed as $|j\rangle = |n_1 n_2 m\rangle$ when they belong to the discrete spectrum, and as $|k, s\rangle = |k, n_2, m\rangle$ for the continuous spectrum†.

As mentioned above, for equal nuclear charges one has the additional quantum number corresponding to the symmetry of the solution with respect to the midpoint of R (g and u solutions).

In spheroidal coordinates $\mathbf{r} = (\xi, \eta, \varphi)$, where

$$\xi = (r_a + r_b)/R, \quad \eta = (r_a - r_b)/R$$

and φ is the azimuthal angle around the internuclear axis, one has ($p = g, u$):

$$\langle \mathbf{r} | jp \rangle = \varphi_{jp}(\mathbf{r}, \mathbf{R}) = \varphi_{jp}(\xi, \eta; R) \cdot \exp(im\varphi) (2\pi)^{-1/2} \times \begin{cases} (-1)^m & m > 0 \\ 1 & m < 0 \end{cases} \quad (3.14a)$$

$$\langle \mathbf{r} | ksp \rangle = \varphi_{ksp}(\mathbf{r}, \mathbf{R}) = \varphi_{sp}(\xi, \eta; k, R) \cdot \exp(im\varphi) (2\pi)^{-1/2} \times \begin{cases} (-1)^m & m > 0 \\ 1 & m < 0 \end{cases}. \quad (3.14b)$$

The normalization is as follows;

$$\langle jp | j'p' \rangle = \delta_{pp'} \delta_{mm'} \delta_{n_1 n_1'} \delta_{n_2 n_2'}, \quad (3.15a)$$

$$\langle ksp | k's'p' \rangle = \delta_{pp'} \delta_{mm'} \delta_{n_2 n_2'} \delta(k - k'). \quad (3.15b)$$

As a matter of fact, as a basis for the expansion of the total wave function it is convenient to choose the linear combinations

$$|ja\rangle = (2)^{-1/2}(|jg\rangle - |ju\rangle), \quad (3.16a)$$

$$|jb\rangle = (2)^{-1/2}(|jg\rangle + |ju\rangle), \quad (3.16b)$$

$$|ksa\rangle = (2)^{-1/2}(|ksg\rangle - |ksu\rangle), \quad (3.16c)$$

$$|ksb\rangle = (2)^{-1/2}(|ksg\rangle + |ksu\rangle). \quad (3.16d)$$

* This approach is by no means new, and its origin can be traced back up to the thirties [e.g. Kronig (1930)]. The method has been used several times in the literature, in spite of the alleged difficulties, and in recent years it has been exhaustively investigated by the Dubna group in a long series of papers. We refer to Ponomarev and Vinitsky (1977), Ponomarev and Vinitsky (1979) and Ponomarev et al. (1980) for bibliography on the subject.

† n_1, n_2 and m are the parabolic quantum numbers, and k is the momentum corresponding to energy $E = k^2/2m_0$.

In the limit of infinite internuclear distance the combinations $|j, a\rangle$ and $|j, b\rangle$ tend to the wave functions describing isolated systems ($a\mu$) and ($b\mu$) respectively*.

The wave functions of the total Hamiltonian are labelled by a set of quantum numbers $\{n\}$ referring to the muonic motion in the field of the nuclei, and by a set $\{\tau\}$ describing the internuclear motion. One can choose as $\{n\}$ the set of parabolic quantum numbers, and as $\{\tau\}$ the set (J, m_J, v, P) , where J and m_J are the total angular momentum of the three-body system and its projection onto the Z axis of the laboratory system, v is the vibrational quantum number and P is the total parity of the system. As will be clear below, a wave function labeled by $\{n\}$ and $\{\tau\}$ differs, for any given value of $\{\tau\}$, from a Born–Oppenheimer wave function with quantum numbers $\{n\}$ by terms of the order m_μ/M_0 at least.

The wave function is expanded in the adiabatic basis as follows:

$$\begin{aligned} \Psi_{n\tau}(r, R) = R^{-1} \sum_{m=0}^J \mathcal{D}_{mm_J}^J(\Phi, \Theta, \varphi) \\ \times \sum_{i=a,b} \sum_{n_2=0}^{\infty} \left\{ \sum_{n_1=0}^{\infty} \varphi_{ji}(\xi, \eta; R) \chi_{ji}^{n\tau}(R) + \int_0^{\infty} dk \varphi_{si}(\xi, \eta; k, R) \cdot \chi_{si}^{n\tau}(k, R) \right\}, \end{aligned} \quad (3.17)$$

where Θ and Φ are the polar angles of the vector \mathbf{R} in the laboratory system, and $\mathcal{D}_{mm_J}^J$ is the proper combination of normalized Wigner D functions:

$$\mathcal{D}_{mm_J}^J = (4\pi(1 + \delta_{0m}))^{-1/2} [(-1)^m \exp(im\varphi) D_{mm_J}^J(\Phi, \Theta, 0) + \exp(-im\varphi) D_{-mm_J}^J(\Phi, \Theta, 0)], \quad (3.18)$$

which assures the wave function to have a given value of J and m_J .

Substituting expansion (3.17) into the Schroedinger equation (3.7) and averaging over the coordinates r , Θ and Φ , we obtain an infinite system of integrodifferential equations for the functions $\chi_{ji}^{n\tau}$ and $\chi_{si}^{n\tau} \ddagger$:

$$\begin{cases} I \left\{ \frac{d^2}{dR^2} - \frac{J(J+1) - 2m_J^2}{R^2} - \frac{2M_0}{m_0 R} + \frac{2M_0}{m_0} \epsilon_{jv} \right\} \chi_i(k, R) = \sum_j U_{ij} \chi_j + \sum_{s=1}^{\infty} \int_0^{\infty} dk U_{is}(k, R) \chi_s(k, R) \\ I \left\{ \frac{d^2}{dR^2} - \frac{J(J+1) - 2m_J^2}{R^2} - \frac{2M_0}{m_0 R} + \frac{2M_0}{m_0} \epsilon_{jv} \right\} \chi_s(k, R) = \sum_i U_{si} \chi_i + \sum_{s=1}^{\infty} \int_0^{\infty} dk' U_{ss'}(k, k', R) \chi_{s'}(k, R). \end{cases} \quad (3.19)$$

One should keep in mind that χ is in fact a two-component vector and correspondingly I and U are 2×2 matrices, I being the identity and U the effective potential, whose expression can be found in Pomarev and Vinitsky (1979) and Faifman et al. (1976). The quantity \ddagger

* Actually, as discussed below, the dissociation limit is not exact, in that the isolated systems obtained for infinite internuclear distance do not possess the correct reduced mass and energy.

† The procedure is sometimes termed as method of perturbed stationary states [Mott and Massey (1965)]. We adhere to the usage of the Dubna group by calling adiabatic basis the set of eigenfunctions of the two-center Coulomb problem, and adiabatic representation the consequent expansion of the wave function, since these names seem to us more suggestive of the physical ideas underlying the method.

‡ $E_{n\tau}$ is the eigenvalue in eq. (3.7).

If the wave functions $|\psi^{(i)}\rangle$ are normalized according to the condition

$$\langle \psi^{(0)} | \psi^{(i)} \rangle = \delta_{0i} \quad (3.25)$$

(intermediate normalization), the corrections $E^{(s)}$ to the energy are given by the following formulae [Dalgarno and Stewart (1956)]:

$$E^{(2s)} = \langle \psi^{(s)} | V | \psi^{(s-1)} \rangle - \sum_{i=1}^s \sum_{j=1}^{s-1} E^{(2s-i-j)} \langle \psi^{(i)} | \psi^{(j)} \rangle, \quad (3.26a)$$

$$E^{(2s+1)} = \langle \psi^{(s)} | V | \psi^{(s)} \rangle - \sum_{i=1}^s \sum_{j=1}^s E^{(2s+1-i-j)} \langle \psi^{(i)} | \psi^{(j)} \rangle. \quad (3.26b)$$

It is worth observing that, in order to have the energy correction of order $2s$ or $2s+1$ one needs to know the correction to the wave function only up to order s . Moreover, apart from the equation for $E^{(0)}$ and $\psi^{(0)}$, eqs. (3.24) do not describe eigenvalue problems, but are simply nonhomogeneous differential equations.

In this frame, the perturbative term for the system (3.19) is represented by the off diagonal part of U_{ij} plus the value $U_{n_a, n_a}(\infty)$, which corresponds* to the value of the energy of the isolated ($a\mu$) atom in state n in this form of the adiabatic representation. This value, however, differs from the exact value of the energy of the ($a\mu$) atom by a term of the order m_0/M_0 . This is due to the splitting adopted for the total Hamiltonian into a two-centre Coulomb Hamiltonian and a kinetic term. Obviously, the total energy of the three-body system is independent of the way it is broken up into the sum of subsystem energies. However, the fact that the dissociation limit does not yield the correct value of the isolated atom energy was often regarded as a drawback of the adiabatic representation, and will deserve some comments below.

According to what was said above, the complete system of equations is written [Ponomarev and Vinitsky (1977)], for $n = 1$, in the following way:

$$I \left(\frac{d^2}{dR^2} + \frac{2M_0}{m_0} \epsilon \right) \chi_i = \sum U_{ij}^{(0)} \chi_j + \sum U_{ij}^{(1)} \chi_j,$$

with

$$\epsilon = E_{1\tau} - (m_0/2M_0)U_{1a,1a}(\infty), \quad (3.27)$$

$$U_{ij}^{(0)} = (U_{ij} - IU_{1a,1a}(\infty))\delta_{ij}, \quad (3.28)$$

$$U_{ij}^{(1)} = U_{ij} - U_{ij}^{(0)}. \quad (3.29)$$

The off-diagonal terms $U_{ij}^{(1)}$, which couple different internuclear states, are smaller than the diagonal part U_{ii} by a factor of order m_0/M_0 .

Defining the operator

$$L_i = I \frac{d^2}{dR^2} - U_{ii} \quad (3.30)$$

* The actual value in the dissociation limit is $(m_0/2M_0)U_{n_a, n_a}(\infty)$.

$$\left(L_j + \frac{2M_0}{m_0} \epsilon^{(0)}\right) \chi_{ja}^{(1)} - U_{ja,jb}^{(0)} \chi_{jb}^{(1)} = U_{ja,1a}^{(1)} \chi_{1a}^{(0)} + U_{ja,1b}^{(1)} \chi_{1b}^{(0)}, \quad (3.33a)$$

$$\left(L_j + \frac{2M_0}{m_0} \epsilon^{(0)}\right) \chi_{jb}^{(1)} - U_{jb,ja}^{(0)} \chi_{ja}^{(1)} = U_{jb,1a}^{(1)} \chi_{1a}^{(0)} + U_{jb,1b}^{(1)} \chi_{1b}^{(0)}. \quad (3.33b)$$

Eqs. (3.32) and (3.33) were first used to calculate the energy of the ground state of the $(pd\mu)$ mesomolecule [Ponomarev and Vinitsky (1977)]. The equations were solved by a continuous analog of Newton's method, which is described in the appendix.

The calculated energy levels are reported in table 2a. One sees that the spectrum is qualitatively similar to that discussed in sect. 3.2. The error for the calculated levels is about 0.1 eV. Within such accuracy one can be confident in the existence of the $(J, v) = (1, 1)$ weakly bound states of the $(dd\mu)$ and $(dt\mu)$ mesomolecules. In fact, the existence of weakly bound states was not proved until 1976, when the Dubna group performed the first calculations in the above frame.

3.5. The dissociation limit in the adiabatic representation

Having outlined the main features of the application of the adiabatic representation, we think that, before describing further developments, it is convenient to discuss briefly those aspects that, up to recent times, have discredited the method as hardly applicable for practical calculations. As mentioned earlier, the most striking drawback of the method appears to be the incorrect dissociation limit. In order to clarify this point we recall that the definition of the adiabatic basis was strictly connected with the identification of a two-center part of the total Hamiltonian, whose eigenfunctions are the adiabatic basis. The choice previously adopted was:

$$H = T_0 + h_0 + 1/R, \quad (3.34)$$

$$T_0 = -(1/2M_0)(\nabla_{\mathbf{R}} + \frac{1}{2}\kappa\nabla_r)^2, \quad (3.35)$$

$$h_0 = -(1/2m_0)\Delta_r - 1/r_a - 1/r_b. \quad (3.36)$$

The eigenfunctions of h_0 obviously depend on the value of the mass m_0 , as do the eigenvalues $E(\mathbf{R})$ and the system of equations obtained by projection over the basis.

In particular, for infinite internuclear distance, the functions

$$\varphi_{ja}^{(0)}(\mathbf{r}, \mathbf{R}) = (2)^{-1/2}[\varphi_{ju}^{(0)} + \varphi_{jg}^{(0)}],$$

and

$$\varphi_{jb}^{(0)}(\mathbf{r}, \mathbf{R}) = (2)^{-1/2}[\varphi_{ju}^{(0)} - \varphi_{jg}^{(0)}],$$

where $\varphi_{u,g}^{(0)}$ are eigenfunctions of h_0 , tend to the wave functions $\psi_j(m_0r_a)$ and $\psi_j(m_0r_b)$ of isolated $(a\mu)$ and $(b\mu)$ atoms in states $j = (n_1, n_2, m)$ and $j' = (n'_1, n'_2, m')$ respectively, the energies being $-m_0/2n^2$ and $-m_0/2n'^2$. Since m_0 differs from the reduced mass of both the $(a\mu)$ and $(b\mu)$ atoms, these values do not coincide with the energy of the isolated atoms. Also, the wave function does not show the expected behaviour. Indeed, the asymptotic behaviour of the χ functions* is governed by the equation

* We show explicitly the m_0 dependence of the internuclear wave functions χ .

$$\frac{d^2}{d(m_0 R)^2} \chi_j(m_0 R) + \left(\frac{2M_0}{m_0^2} E_{n\tau} - U_{ji}(\infty) \right) \chi_j = 0, \quad (3.37)$$

which yields for the wave function $\Psi_{n\tau}(\mathbf{r}, \mathbf{R})$ the asymptotes

$$\Psi_{n\tau}(\mathbf{r}, \mathbf{R}) \xrightarrow{R \rightarrow \infty} \begin{cases} \psi_n(m_0 r_a) D_{mm_j}^J(\Theta, \Phi, 0) \exp[-\tilde{k}_{na} R]: r_a \ll R, \\ \psi_n(m_0 r_b) D_{mm_j}^J(\Theta, \Phi, 0) \exp[-\tilde{k}_{nb} R]: r_b \ll R, \end{cases} \quad (3.38)$$

where:

$$\tilde{k}_{na}^2 = 2M_0(E_{n\tau} - \tilde{E}_{na}), \quad (3.39a)$$

$$\tilde{k}_{nb}^2 = 2M_0(E_{n\tau} - \tilde{E}_{nb}), \quad (3.39b)$$

$$\tilde{E}_{na} = -(m_0/2n^2)(1 - m_0(1 + \kappa)^2/4M_0), \quad (3.40a)$$

$$\tilde{E}_{nb} = -(m_0/2n^2)(1 - m_0(1 - \kappa)^2/4M_0). \quad (3.40b)$$

These expressions should be compared with the exact asymptotic behaviour:

$$\Psi_{n\tau} = \begin{cases} \psi_n(m_a r_a) D_{mm_j}^J(\Theta, \Phi, 0) \exp[-k_{na} R], & r_a \ll R \\ \psi_n(m_b r_b) D_{mm_j}^J(\Theta, \Phi, 0) \exp[-k_{nb} R], & r_b \ll R \end{cases} \quad (3.41)$$

with

$$k_{na}^2 = (E_{n\tau} - E_{na})[(M_a + m_\mu)^{-1} + M_b^{-1}]; \quad 1/m_a = 1/m_\mu + 1/M_a$$

and

$$k_{nb}^2 = (E_{n\tau} - E_{nb})[(M_b + m_\mu)^{-1} + M_a^{-1}]; \quad 1/m_b = 1/m_\mu + 1/M_a.$$

The asymptotic behaviour and the binding energy

$$E_b = E_{n\tau} - E_a,$$

where E_a is the ground state energy of the $(a\mu)$ atom, nucleus a being heavier than nucleus b , are different in the two cases. Moreover, the diagonal term

$$(m_0/M_0)U_{na,na}(\infty) = \tilde{E}_{na}(\infty)$$

—and similarly for $U_{nb,nb}(\infty)$ —is not simply $-m_0/2n^2$, as could be expected according to the splitting of the Hamiltonian, the difference being of the order of m_0/M_0 :

$$\tilde{E}_{na} = -m_0(1 - (1 + \kappa)m_0/4M_0)/2n^2. \quad (3.42)$$

The root of the difficulty outlined above can be traced back to the fact that, with usual splitting of the Hamiltonian into a kinetic part T_0 and a potential part $h_0 + 1/R$, the limit of infinite R does not yield an

isolated atom Hamiltonian plus a commuting term describing the internuclear motion. Indeed, recalling that ∇_R tends to $\frac{1}{2}\nabla_{r_a}$ for $r_a \ll r_b$, whereas it tends to $-\frac{1}{2}\nabla_{r_b}$ for $r_a \gg r_b$, one can check the aforementioned behaviour and find the way-out from the above difficulty in the same time [Ponomarev and Vinitisky (1979)]. It suffices to define new kinetic and potential terms by adding and subtracting the limit for $R \rightarrow \infty$ of the kinetic part T_0 .

Depending on the condition $r_a \ll r_b$ or $r_b \ll r_a$ one has respectively:

$$H = T_a + h_a + 1/R, \quad (3.43a)$$

or

$$H = T_b + h_b + 1/R, \quad (3.43b)$$

where

$$T_{a,b} = -(1/2M_0)[\nabla_R + \frac{1}{2}\kappa\nabla_r]^2 - \frac{1}{2}(1 \pm \kappa)\Delta_R, \quad (3.44)$$

$$h_{a,b} = -(1/2m_{a,b})\Delta_r - 1/r_a - 1/r_b. \quad (3.45)$$

One can choose as adiabatic basis the set of the eigenfunctions of either h_a or h_b . These sets of functions are simply connected, just like the eigenvalues, by a scaling factor γ . If

$$h_{a,b}\varphi_j^{a,b}(\xi, \eta; R) = E_j^{a,b}(R)\varphi_j^{a,b}(\xi, \eta; R), \quad (3.46)$$

one has:

$$\varphi_j^b(\xi, \eta; R) = \varphi_j^a(\xi, \eta; \gamma R), \quad (3.47a)$$

$$E_j^b(R) = \gamma E_j^a(\gamma R); \quad \gamma = m_b/m_a. \quad (3.47b)$$

It is obvious that, according to the foregoing discussion, the combinations

$$\varphi_{a,i}^a = (2)^{-1/2}(\varphi_{i,g}^a - \varphi_{i,u}^a), \quad (3.48a)$$

$$\varphi_{b,i}^b = (2)^{-1/2}(\varphi_{i,g}^b + \varphi_{i,u}^b) \quad (3.48b)$$

tend to the wave functions of isolated atoms ($a\mu$) and ($b\mu$) respectively, the eigenvalues being the correct ones:

$$E_n^a(R) \xrightarrow{R \rightarrow \infty} -m_a/2n^2, \quad (3.49a)$$

$$E_n^b(R) \xrightarrow{R \rightarrow \infty} -m_b/2n^2. \quad (3.49b)$$

In view of the above considerations, the most convenient choice for the adiabatic basis seems to be that consisting of the functions $\{\varphi_{a,i}^a; \varphi_{b,j}^b\}$ (basis 1). One should notice that, contrary to what happens for the basis $\{\varphi_{i,a}^a, \varphi_{j,b}^b\}$ (basis 2) or $\{\varphi_{i,a}^b, \varphi_{j,b}^a\}$ (basis 3), this basis is not orthonormal. In Ponomarev and

Vinitsky (1979) one can find the expression of the linear transformation $S(R)$ which connects basis 1 with basis 2, and the form the effective potential takes when basis 1 is adopted.

One should also observe that, in order to have the correct dissociation limit, one can choose the basis obtained via the asymptotic transformation of basis 2. We mean thereby that one can choose the basis obtained by applying the linear transformation $\bar{S} = \lim_{R \rightarrow \infty} S(R)$ to basis 2 in order to ensure the correct dissociation limit. A discussion of the properties of \bar{S} can be found in Ponomarev and Vinitsky (1979).

Separation (3.43a) was used for the calculation of the ground state of the (pp μ) and (dd μ) mesomolecules, according to the technique outlined in subsection 3.4. The results obtained (252.79 eV and 324.99 eV respectively), with an error of some tenths of eV, are in fair agreement with the results of other methods of calculation (see section 3.8).

The binding energy

$$\epsilon_{J\nu} = E_{1\tau} - E_{1a}$$

is represented as*

$$\epsilon_{J\nu} = \epsilon_{\text{disc}}^{(0)} + \epsilon_{\text{disc}}^{(2)} + \epsilon_{\text{cont}}^{(0)} + \epsilon_{\text{cont}}^{(2)}. \quad (3.50)$$

The several contributions can be found in Ponomarev and Vinitsky (1979). To give an idea of the complexity of the task of calculating the energy levels, it suffices to say that one hundred thirty pairs of equations were considered, ten corresponding to ten pairs of states (n_1, n_2, m) with $n_1 + n_2 + m \leq 2$, and the rest to states of the continuous spectrum with k in the range (0.2–10).

3.6. The effective two-level approximation

The most severe obstacle to an easy use of the adiabatic representation for the calculation of the mesomolecular energies is the large number of coupled equations one has to consider in order to achieve a satisfying accuracy. One can of course limit to taking into account for any level only the coupling between the states which, for infinite internuclear distance, give the (a μ) and the (b μ) isolated atoms respectively. This two-level approximation is an improvement of the crude B.O. approximation, but it neglects contributions of the order of $(m_\mu/M_0)^2$.

However, calculations were made within this frame for several mesomolecules (see section 3.8 for a list of the results obtained in this way, and Gershtein and Ponomarev (1975) for an extended review of the work performed with this method). An important result was the hint of a weakly bound state of the (dd μ) mesomolecule [Ponomarev et al. (1973, 1974)], whose existence had been first suggested by Belyaev et al. (1960).

The error in the calculation being at least of the order of $(m_\mu/M_0)^2$ over an energy of some hundreds eV, it was hard to regard these results as ultimate. On the other hand, going beyond the two-level approximation implied solving a large number of equations. The difficulty was to a certain extent circumvented by a clever trick devised by the Dubna group [Ponomarev et al. (1980)]. The method amounts to a diagonalization, up to order $(m_\mu/M_0)^2$ included, of the system of equations

* One should observe that in this frame the energy E_{1a} is the true energy of the (a μ) isolated atom, which is different from the value obtained in previous applications of the adiabatic representation, because of the incorrect dissociation limit one had in that circumstance.

$$\left[I \frac{d^2}{dR^2} + 2M(IE_{nr} - \bar{U}_{ij}/2M) \right] \bar{\chi}_i(R) = \sum_{j \neq i} \bar{U}_{ij}(R) \bar{\chi}_j(R), \quad (3.51)$$

where

$$M = (M_a^{-1} + m_\mu^{-1}) / (M_a^{-1} + M_b^{-1}),$$

which is obtained by projecting the Schroedinger equation over the basis obtained by applying the transformation \bar{S} to basis 2. The functions $\bar{\chi}_i$ are the internuclear motion wave functions in that representation.

In this way, for any given level i , one has a system of two coupled equations which is considerably simpler than a large array of equations, the dissociation limit remaining correct, in opposition to what happens for the ordinary two-level approximation. It is necessary to realise, however, that the expression of the effective potential in the effective two-level approximation requires the evaluation of matrix elements between the wave functions belonging to the selected level and – in principle – all the other levels. It is in this respect that some truncation procedure is needed. On the other hand, the procedure devised in Ponomarev et al. (1980) in principle allows diagonalization of the system (3.19) to any desired order in $(m_0/2M)$.

The method of diagonalization starts with a truncation of the complete system of equations, and consists in the application of a succession of unitary transformations $\exp[iT_k]$, each T_k removing the nondiagonal part of the effective potential up to order k in $(m_\mu/2M_0)$. The procedure is rather involved and has been worked only for the second order. A detailed discussion can be found in Ponomarev et al. (1980).

After performing the appropriate transformation, one has to solve the following system of two coupled equations:

$$\left[\frac{m_0}{2M_0} Q \frac{d^2}{dR^2} + IE_{nr} - \tilde{U}_{ij} \right] \tilde{\chi}_j = 0 \quad (3.52)$$

where the matrices Q and \tilde{U} and the vector $\tilde{\chi}$ are the transformed, up to order $(m_\mu/2M_0)^2$, of the kinetic energy, effective potential and wave function respectively. The asymptotic value of the Q matrix is:

$$\frac{m_a}{M_0} Q(\infty) = \begin{pmatrix} M^{-1} - (m_a/M_a)^2 & 0 \\ 0 & M^{-1} - \gamma^{-1}(m_b/M_b)^2 \end{pmatrix} = \begin{pmatrix} \mu_a^{-1} & 0 \\ 0 & \mu_b^{-1} \end{pmatrix}, \quad (3.53)$$

where μ_a and μ_b are the reduced masses of the system consisting of the $(a\mu)$ atom plus nucleus b, or $(b\mu)$ atom plus nucleus a. In the same time, the \tilde{U} matrix has the limit

$$(\tilde{U}_{ii})_{ab} = \begin{pmatrix} E_{ia} & 0 \\ 0 & E_{ib} \end{pmatrix}, \quad (3.54)$$

so that the correct momenta in channels (a) and (b) – corresponding to isolated $(a\mu)$ atom and $(b\mu)$ atom for infinite separation – are obtained.

As previously noticed, the calculation of the matrix Q and of the effective potential \tilde{U} in the

two-level effective approximation requires the knowledge of several matrix elements connecting the state $|i\rangle$, for which a two-level approximation is required, with all the other states, at least in principle. Calculations were made for the ground state, in which matrix elements between the ground state and the first twenty excited states of the discrete spectrum, and coupling of the ground state with states $|k, n_2, m\rangle$ of the continuous spectrum for $m = 0, n_2 = 0, 1, 2, 3, k \leq 10$, were considered. The resulting binding energies will be presented in section 3.8, together with the results obtained by different methods, including the naive two-level approximation.

3.7. The truncation method

The most recent and accurate calculations based on the system (3.19) consist in a truncation of the system and in a reduction to a Sturm–Liouville problem for a finite number of equations on a finite interval $(0, R_m)$ [Melezhik et al. (1981)]. The Hamiltonian is split into a two-center Coulomb Hamiltonian and a kinetic part according to the discussion of section 3.5,

$$H = T_a + h_a + 1/R,$$

and basis 2 is chosen (see section 3.5).

A discretization of the continuum spectrum is also introduced, so that the momentum k is limited within a range $I = (0, k_m)$, the interval I being divided for any value of $s = (n_2, m)$ into $N_k^{(s)}$ subintervals. Integrals are obviously substituted by sums.

For the discrete spectrum, were taken into account all the states with $n_1 + n_2 + m \leq 2$ and the states $|300\rangle, |210\rangle, |120\rangle$, whereas for the continuous spectrum were considered the states with $s = (0, 0), (1, 0), (2, 0), (0, 1), (1, 1), (3, 0)$. Taking into account the splitting of interval I , an amount of $N_s = 119$ states of the continuum spectrum were considered, which, when added to the $N_i = 13$ states of the discrete spectrum, yielded a system of 264 equations having the form:

$$\left\{ \begin{aligned} I \left\{ \frac{d^2}{dR^2} - \frac{J(J+1) - 2m_j^2}{R^2} - \frac{2M}{R} + 2M\epsilon_{Jv} \right\} \chi_i(R) &= \sum_{j=1}^{\infty} U_{ij}(R) \chi_j(R) + \sum_s \int_0^{\infty} d\kappa U_{is}(\kappa, R) \chi_s(\kappa, R) \\ I \left\{ \frac{d^2}{dR^2} - \frac{J(J+1) - 2m_j^2}{R^2} - \frac{2M}{R} + 2M\epsilon_{Jv} \right\} \chi_s(k, R) & \\ &= \sum_{i=1}^{\infty} U_{si}(k, R) \chi_i(R) + \sum_s \int_0^{\infty} dk' U_{ss'}(k, k', R) \chi_s(k', R), \end{aligned} \right. \quad (3.55)$$

with the boundary conditions

$$\begin{aligned} \chi_i(0) = \chi_i(R_m) = 0 \quad & 1 \leq i \leq N_i \\ \chi_s(k, 0) = \chi_s(k, R_m) = 0 \quad & 1 \leq s \leq N_s \quad 0 \leq k \leq k_m. \end{aligned}$$

One should notice that in the calculation the effective potential matrix was not included completely, its form being illustrated in fig. 4 (shaded area). This choice can be easily understood, on the grounds that, in the frame of perturbation theory, the omitted matrix elements give to the energy of the ground state and its vibrational and rotational excitations a contribution which is smaller than the contribution of the

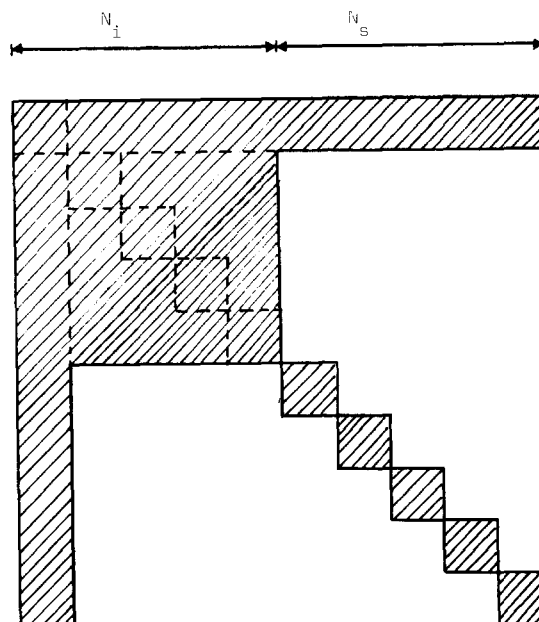


Fig. 4. Scheme of the effective potential matrix used in the truncation method (from Melezhik et al. 1981). N_d is the number of the discrete spectrum states taken into account, N_s the number of the continuum spectrum states.

included matrix elements by an order of magnitude in the parameter ($m_\mu/2M_0$). It is worth observing, however, that the approach exploiting the truncation method is not perturbative in principle, the approximation consisting in the number of the continuum and discrete spectrum states involved.

The included matrix elements were calculated with an accuracy $\sim 10^{-5}$. The relative accuracy in the solution of the Sturm–Liouville problem was estimated to be $\sim 10^{-4}$ – 10^{-5} . Once the wave functions $\chi_j(R)$ and $\chi_s(k, R)$ are evaluated, one can calculate the contribution of any state to the energy levels according to formulae (3.26).

The results concerning different mesomolecules are reported in table 2b. In Melezhik et al. (1981) one can find tables and figures describing the contribution to the energy of the discrete and continuum spectrum states. In this respect, one should notice that the evaluation of the partial contribution of the states included in system (3.55) is not identical with the perturbative calculation outlined in section 3.4, in that the wave functions χ differ from the functions introduced in perturbation theory already in the second order in ($m_\mu/2M_0$).

The error in the calculated energy levels is in the range $0.05 \div 0.1$ eV. For deep levels, the decisive contribution is due to the pair of states $|000g\rangle$ and $|000u\rangle$, whereas for weakly bound states it is necessary to take into account the contribution of the excited states of both discrete and continuum spectrum. In conclusion, the relative accuracy achieved in the calculation of the energy levels is of the order of 10^{-3} for deep levels, with an absolute error of 0.1 eV, and of 10^{-2} for weakly bound states, with an error of 0.05 eV.

3.8. Conclusions

We present here a collection of the results obtained for different mesomolecules by the techniques outlined above, together with some results obtained by other means, mainly by variational calculations [Halpern (1964) and Carter (1966, 1968)].

In table 2 the spectrum of all mesomolecules is presented, according to the most recent calculations. These are the results of the perturbative calculations and of the truncation method. The error is generally at the level of 0.1 eV, with the exception of the weakly bound levels. For these latter the truncation method yields an error of about 0.05 eV.

In table 3 we report the energy of the ground state of several mesomolecules, as obtained by the variational calculations (first line), two-level approximation (second line), two-level effective approximation (third line), perturbation theory (fourth line), truncation method (fifth line). Having regard to the errors ascribed to the various methods, one can check the consistency of the different calculations. The least accurate of them is the two-level calculation, whose error is of the order of a few eV. The variational calculations are affected by an error of about 1 eV. For the effective two-level approximation and the perturbative calculation the error is about 0.1 eV, while the truncation method is slightly more accurate.

Table 3
Binding energy (eV) of the $(J, v) = (0, 0)$ state of all the mesomolecules, according to different methods of calculation.

$(pp\mu)$	$(pd\mu)$	$(pt\mu)$	$(dd\mu)$	$(dt\mu)$	$(tt\mu)$	Method
253.09	221.28	213.00	324.27	318.07	361.4	Variational
247.31	215.68	207.28	322.69	317.04	361.56	Two-level
253.00	221.69	214.23	325.00	319.13	362.91	Eff. two-level
253.55	221.49	213.85	324.99	319.09	362.89	Perturbation
252.95	221.52	213.97	325.04	319.15	362.95	Truncation

To give an idea of the progress implied in the several approaches described above, we trace a kind of history of the weakly bound states of the $(dd\mu)$ and $(dt\mu)$ mesomolecules in table 4. It may be worthwhile observing that only the calculations performed in the frame of the adiabatic representation account for the existence of weakly bound states. One can see that, while hints of the existence of a weakly bound state for $(dd\mu)$ were found since the early seventies, the existence of a weakly bound state of $(dt\mu)$ mesomolecule was harder to establish. Moreover, the alleged values of the energies have changed a bit in the course of time.

Table 4
Evolution of the calculation of the binding energy (eV) of the weakly bound states of the $(dd\mu)$ and $(dt\mu)$ mesomolecules.

$(dd\mu)$	$(dt\mu)$	error	Method	Reference
7	-	few eV	two level	Belayev et al. (1960)
0.7	-	1	two level	Ponomarev et al. (1973)
0.64	-0.47	1	two level	Ponomarev et al. (1976)
1.96	0.85	0.1	perturbation	Ponomarev et al. (1979)
1.83	0.63	0.1	perturbation	Melezhik et al. (1981)
1.91	0.64	0.05	truncation	Melezhik et al. (1981)

In conclusion, from this short discussion one can realise that the investigation of the spectrum of mesomolecules was a rather delicate task. This is particularly true for the weakly bound states, whose existence can be proved on theoretical grounds only at the cost of solving a consistent number of integro-differential equations with a rather pushed accuracy. Although hints of their existence were found already in the early seventies, it took almost ten years to get a firm belief in the reliability of the calculations.

4. Corrections to the Coulomb interaction

In order to get a sharp estimate of the energy levels of mesomolecules it is necessary to go beyond the approximation of eq. (3.5). Several sources of corrections arise. One has to take into account relativistic corrections to the particle motion, the corrections due to the finite size and polarization of the nuclei, spin effects, electron screening and so on. In particular, it is important to discuss the effect of vacuum polarization.

It is well known that electron-positron vacuum polarization dominates in muonic hydrogen atoms, where it gives rise to corrections to the energy levels of the order of 1 eV. The Coulomb potential $V_c = -e^2/r$ between two point charges is distorted due to the vacuum polarization:

$$V_c(r) \rightarrow V(r) = V_c(r) + \delta V(r). \quad (4.1)$$

The polarization correction $\delta V(r)$ can be represented as a series in the fine structure α . The principal term, i.e. the lowest order in α , is the so called Uehling potential [Lifshitz and Pitaevskii (1974)]:

$$\delta V(r) = -\frac{e^2}{r} \cdot \frac{2\alpha}{3\pi} \int_1^\infty \frac{dx}{x^2} \sqrt{x^2-1} \left(1 + \frac{1}{2x^2}\right) \exp(-2\gamma xr), \quad (4.2)$$

where

$$\gamma = m_e c / \hbar.$$

For mesonic atoms the shift of the energy levels is:

$$\Delta E_i^{v.p.} = \int |\psi_i|^2 \delta V d^3r, \quad (4.3)$$

where ψ_i is the wave function of a mesic atom in state i . In a similar way the level shift of a mesic molecule is:

$$\Delta E_{Jv}^{v.p.} = \int \psi_{Jv}(r, R) \delta V(r, R) d^3r d^3R - \Delta E_a^{v.p.}, \quad (4.4)$$

where $\Delta E_a^{v.p.}$ is the polarization shift of the ground state of the ($a\mu$) atom, defined by eq. (4.3), and ψ_{Jv} is the wave function of the mesomolecule in the (J, v) state. The Uehling potential δV_{mol} for a mesomolecule has to be taken as a sum of potentials representing the interaction between all the particles:

$$\delta V_{\text{mol}} = \delta V(r_{\mu a}) + \delta V(r_{\mu b}) + \delta V(R). \quad (4.5)$$

Calculations of $\Delta E_{Jv}^{v.p.}$ are performed [Melezhik and Ponomarev (1978)] using for ψ_{Jv} the unperturbed solutions of the Coulomb Schroedinger problem, eq. (3.5). The values reported in table 5, fifth column, are accurate at the level of 5 meV.

The electromagnetic structure of the particles and relativistic effects of the order α^2 , which do not depend on the orientation of the particle spins, have been discussed in Bakalov (1981). The calculated values, reported in table 5, are again accurate to the level of 5 meV.

Table 5
Relativistic corrections and corrections for the electromagnetic structure of the nuclei to the energy levels of the mesomolecules (from Bakalov 1981). The energies are in eV.

Mesic molecule	$J\nu$	Electromagnetic structure	Contact interaction	Squared Coulomb	Recoil	Vacuum polarization	Total shift $\Delta\epsilon_{\text{tot}}$
$pp\mu$	{00	+0.0032	+0.0199	-0.0684	+0.0383	-0.285	-0.292
	10	+0.0002	+0.0013	-0.0268	+0.0225	-0.064	-0.067
$pd\mu$	{00	-0.0660	+0.0282	-0.0740	+0.0699	-0.290	-0.342
	10	-0.0737	+0.0110	-0.0371	+0.0440	-0.096	-0.152
$pt\mu$	{00	-0.0390	+0.0289	-0.0758	+0.0688	-0.325	-0.342
	10	-0.0438	+0.0122	-0.0409	+0.0532	-0.124	-0.143
$dd\mu$	{00	+0.0386	+0.0251	-0.0828	+0.0681	-0.397	-0.348
	10	+0.0185	+0.0120	-0.0494	+0.0543	-0.227	-0.196
	20	-0.0037	-0.0024	-0.0189	+0.0395	-0.016	-0.002
	01	+0.0034	+0.0022	-0.0176	+0.0463	-0.030	+0.004
	11	-0.0094	-0.0010	-0.0049	+0.0440	+0.008	+0.037
$dt\mu$	{00	+0.0624	+0.0267	-0.0864	+0.0777	-0.428	-0.348
	10	+0.0476	+0.0147	-0.0608	+0.0648	-0.267	-0.201
	20	+0.0293	+0.0002	-0.0273	+0.0496	-0.058	-0.006
	01	+0.0281	+0.0033	-0.0221	+0.0550	-0.056	+0.008
	11	+0.0022	-0.0006	-0.0069	+0.0524	-0.003	+0.044
$tt\mu$	{00	+0.0289	+0.0290	-0.0902	+0.0815	-0.479	-0.430
	10	+0.0185	+0.0186	-0.0683	+0.0701	-0.329	-0.290
	20	+0.0047	+0.0048	-0.0376	+0.0553	-0.130	-0.103
	30	-0.0063	-0.0063	-0.0095	+0.0440	+0.044	+0.066
	01	+0.0058	+0.0058	-0.0295	+0.0581	-0.097	-0.057
	11	+0.0010	+0.0011	-0.0177	+0.0533	-0.034	+0.004

So far, spin effects have been ignored. The hyperfine structure (H.S.) of the energy levels of mesomolecules has been calculated with an accuracy of 1 meV in the lowest order in α [Bakalov and Vinitzky (1980), Bakalov et al. (1980)]. Again, the solution of the nonrelativistic problem with Coulomb interactions in the adiabatic representation is used as the zero approximation. In Bakalov et al. (1980), the population of the stationary states of the H.S. of mesic molecules produced in collisions between ($p\mu$), ($d\mu$) or ($t\mu$) mesic atoms in the para or ortho states with p, d or t nuclei have been calculated (see table 6 for details).

Beyond the electromagnetic interaction, one has also to consider the strong interaction between the nuclei of the mesomolecule. Recently, this problem has been carefully reconsidered in Bogdanova et al. (1981), for the particular case of the ($dt\mu$) mesomolecule. The energy shifts and widths caused by the nuclear interaction have been derived by solving the eigenvalue problem for the Hamiltonian of the ($dt\mu$) system. For the calculation of the molecular wave function the adiabatic representation of the three-body problem has been used. The nuclear interaction was chosen in a form which can fit the experimental data available on the fusion cross section for the reaction $d + t \rightarrow n + {}^4\text{He}$, and the data on the elastic $d - t$ cross section. The values for the level shifts and widths are reported in table 7.

Finally, the effect of electron screening is being calculated by the Dubna group. Preliminary results, which include only the monopole and dipole contributions, give a shift of 22 meV for the ($dt\mu$) mesomolecule, and 16 meV for the ($dd\mu$) mesomolecule (Bakalov and Melezhik, private communication).

The above presentation shows that the calculation of the energy levels of the mesomolecules has been the subject of intense study in the last few years. Clearly, due to the errors of order 0.1 eV in the calculation of the Coulomb energies, most of the terms just discussed cannot be of practical use in the comparison between the theoretical and experimental values. On the other hand, the substructure

Table 6

Hyperfine structure of the $(dd\mu)$ mesomolecule (a) and of the $(dt\mu)$ mesomolecule (b), from Bakalov et al. 1980. F is the total spin, the index N labels the different states with the same F , \mathcal{E}_{Fv}^{FN} is the hyperfine energy shift, in eV, w is the relative probability of forming the (J, v, F, N) state of the mesomolecule starting from a specific spin state of the mesoatom.

(a)						(b)						
Jv	F	N	\mathcal{E}_{Fv}^{FN} (eV)	$w_{Fv}^{FN}(\uparrow)$	$w_{Fv}^{FN}(\uparrow\uparrow)$	Jv	F	N	\mathcal{E}_{Fv}^{FN} (eV)	$w_{Fv}^{FN}(\uparrow)$	$w_{Fv}^{FN}(\uparrow\uparrow)$	
00	1	1	0	0.1667	0.1667	00	1	0	0.0173	0	0.1111	
		1	-0.0286	0.8333	0.0833			1	0.0282	0.0096	0.3301	
		1	0.0191	0	0.7500			2	-0.1107	0.9904	0.0032	
01	1	1	0	0.1667	0.1667	01	1	2	0.0463	0	0.5556	
		1	-0.0246	0.8333	0.0833			0	0.0239	0	0.1111	
		1	0.0164	0	0.7500			1	0.0312	0.0043	0.3319	
10	2	1	-0.0169	0.2213	0.0560	2	1	1	-0.1123	0.9957	0.0014	
		2	0.0070	0.0565	0.1384			2	0.0439	0	0.5556	
	2	1	-0.0180	0.4436	0.1115	0	2	1	0.0277	0.0007	0.0368	
		2	0.0084	0.1119	0.2774			2	-0.1039	0.1104	0.0002	
	11	2	1	0.0097	0.1667	0.4167	1	2	1	0.0162	0.0000	0.1111
			1	-0.0159	0.2222	0.0555			2	0.0249	0.0031	0.1101
2		1	0.0077	0.0555	0.1389	3	2	3	-0.1035	0.3303	0.0010	
		1	-0.0161	0.4444	0.1111			4	0.0406	0.0000	0.1111	
11	2	1	0.0079	0.1112	0.2778	1	3	1	0.0273	0.0056	0.1833	
		1	0.0082	0.1667	0.4167			2	-0.1041	0.5499	0.0019	
	3	1	0.0433	0	0.2593	2	3	2	0.0447	0.0000	0.1852	
		1	0.0445	0.0001	0.0370			3	0.0433	0	0.2593	
11	3	1	-0.1424	0.1110	0.0000	0	3	1	0.0407	0.0000	0.1111	
		2	0.0439	0.0002	0.1110			2	-0.1424	0.1110	0.0000	
	3	1	-0.1422	0.3331	0.0001	1	3	1	0.0407	0.0000	0.1111	
		2	0.0501	0.0000	0.1111			2	0.0439	0.0002	0.1110	
	3	3	1	0.0443	0.0004	0.1851	1	3	3	-0.1422	0.3331	0.0001
			2	-0.1424	0.5552	0.0001			4	0.0501	0.0000	0.1111
		3	2	0.0511	0.0000	0.1852	2	3	1	0.0443	0.0004	0.1851
			3	0.0508	0	0.2593			2	-0.1424	0.5552	0.0001

Table 7

Energy shifts (ΔE) and widths (Γ) of the levels of the $(dt\mu)$ mesomolecule induced by the nuclear interaction, in eV (from Bogdanova et al. 1981).

Jv	00	01	10	11	20
ΔE	9.6×10^{-4}	8.0×10^{-4}	9.8×10^{-8}	3.0×10^{-8}	8.0×10^{-11}
Γ	8.2×10^{-4}	6.8×10^{-4}	6.6×10^{-8}	2.5×10^{-8}	6.7×10^{-11}

induced by hyperfine interactions, which does not depend on the value of the Coulomb energy, can be nicely evidenced, as will be discussed in section 5.2.

5. Formation of mesomolecules

The formation of mesomolecules occurs through collisions:



where $X, X' = p, d$ or t .

The formation reactions are classified according to the way the binding energy is released. This energy can be transferred to an Auger electron (see fig. 5a), or it can be transferred to the excitation of the vibrational and rotational degrees of freedom of ordinary molecules (fig. 5b).

We shall see that the former mechanism, referred to as the nonresonant process, can explain the experimental data for the $(pp\mu)$ and $(pd\mu)$ mesomolecules. On the other hand, the latter process is needed in order to account for the formation of $(dd\mu)$ and $(dt\mu)$ mesomolecules.

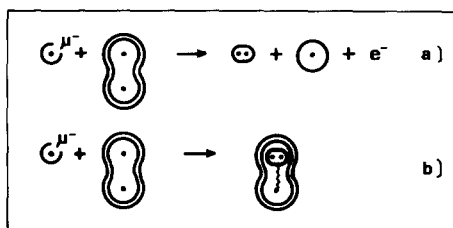


Fig. 5. Mechanisms of mesomolecule formation: (a) Auger (non-resonant) process. (b) The resonant process.

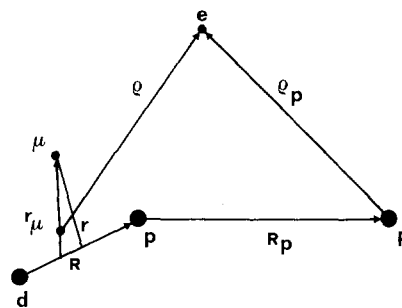


Fig. 6. The coordinates used for the description of reaction (5.2).

5.1. Nonresonant formation

To the purpose of giving a description of the calculational scheme, we discuss the formation of the $(pd\mu)$ system in some detail, following Ponomarev and Faifman (1976). The reaction is:



The formula for the differential cross section corresponding to the electron being emitted in the momentum range q to $q + dq$ is*:

$$V d\sigma = \frac{2\pi}{\hbar} \delta(E_f - E_i) \cdot d^3q \sum_{m_L} \left| \int d^3R d^3r \Psi^{(f)}(r, R) \psi^{(f)}(\rho) H_{int} \psi^{(i)}(\rho) \Psi^{(i)}(r, R) \right|^2. \quad (5.3)$$

V is the relative velocity of the colliding system, $\psi^{(i,f)}$ is the wave function for the emerging electron in the initial and final state. $\Psi^{(f)}$ is respectively the wave function for the system $(d\mu) + p$ and for the $(pd\mu)$ mesomolecule. The δ function expresses the energy conservation and the sum is over the allowed values of the angular momentum of the mesomolecule which is formed.

The perturbing Hamiltonian which is responsible for the mesomolecule formation is taken as:

$$H_{int} = -e^2/R_{1e} - e^2/R_{2e} + e^2/R_{\mu e} + e^2/\rho \quad (5.4)$$

R_{ie} ($i = 1, 2, \mu$) is the distance from the i -th nucleus and from the muon to the electron.

Taking into account the inequality:

$$R_i \ll \rho \quad (5.5)$$

* See fig. 6 for the description of the coordinates.

where R_i are the distances of the nuclei and of the muon from the center of mass of the $(pd\mu)$ mesomolecule, we have, in the dipole approximation:

$$H_{\text{int}} = -ed \cdot \rho/\rho^3. \quad (5.6)$$

d is the electric dipole moment of the mesomolecule with respect to the center of mass.

The Heitler–London expression is used for the wave function of the electron in the initial state. For the final state one uses a Coulomb wave function with an effective charge Z' .

The wave functions representing the motion of the muon and of the nuclei are calculated within the two-level approximation of the adiabatic representation (see section 3).

The formation rate is:

$$\lambda_{pd\mu} = \sigma \cdot V \cdot D. \quad (5.7)$$

D is the density of the target.

$\lambda_{pd\mu}$ is shown in fig. 7 as a function of the collision energy, ϵ . One sees that in the low energy region, $\epsilon < 1$ eV, the rate $\lambda_{pd\mu}$ is almost constant, as a consequence of the Bethe law (see section 2).

The principal source of error in the calculation is associated with an insufficient knowledge of the wave function for the emitted electron. In Ponomarev and Faifman (1976) the $((pd\mu)pe)$ complex was replaced by a Coulomb center with an effective charge $Z' = 1$. By varying Z' it was possible to conclude that the error arising from the insufficient knowledge of $\psi^{(e)}(\rho)$ amounts to some 5–10% of the quoted value.

The experimental results on the $(pp\mu)$ and $(pd\mu)$ formation rate are reported in table 8. It is worth observing that the results obtained at L.H.T. and R.T. are equal within the errors. In table 8 we report

Table 8
Formation rates of the $(pp\mu)$ and $(pd\mu)$ mesomolecules. The experimental values (Exp.) were obtained at room temperature (R.T.) and liquid hydrogen temperature (L.H.T.). The theoretical values (Th.) are also given. All the rates are normalized to liquid hydrogen density.

Source		$\lambda_{pp\mu}$ 10^6 s^{-1}	$\lambda_{pd\mu}$ 10^6 s^{-1}
Dzhelepov et al. (1962)	Exp., R.T.	1.5 ± 0.6	
Bleser et al. (1963)	Exp., L.H.T.	1.89 ± 0.20	5.8 ± 0.3
Conforto et al. (1964)	Exp., L.H.T.	2.55 ± 0.18	6.82 ± 0.25
Bodyashov et al. (1968)	Exp., R.T.	2.74 ± 0.25	
Bystritsky et al. (1975)	Exp., R.T.	2.34 ± 0.17	5.53 ± 0.16
Cohen et al.	Th.	3.9	3.0
Zel'dovich and Gershtein (1960)	Th.	2.6	1.3
Ponomarev and Faifman (1976)	Th.	2.20	5.91

also the theoretical values calculated for a muonic atom with room thermal energy. In fact, it is clear from fig. 7 that the formation rate is weakly sensitive to the muonic atom energy as long as this is smaller than 1 eV.

One concludes that the calculations just presented account for the experimental data on $(pp\mu)$ and $(pd\mu)$ mesomolecules.

On the other hand, as it is clear from fig. 8, the Auger process cannot account for the shape and the values of the $(dd\mu)$ formation rate. An additional mechanism is needed in order to explain the formation of $(dd\mu)$.

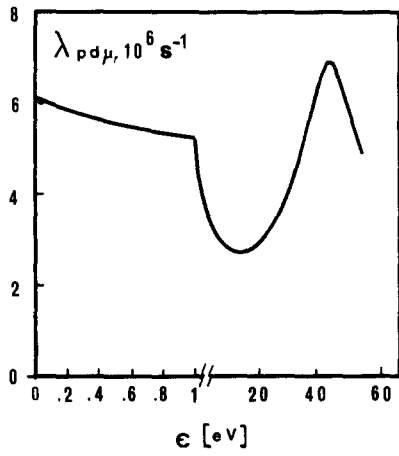


Fig. 7. The formation rate $\lambda_{pd\mu}$ as a function of the collision energy (from Ponomarev and Faifman 1976).

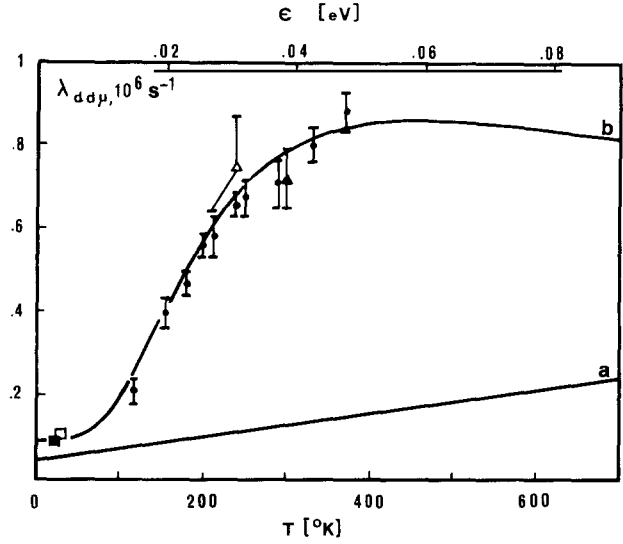


Fig. 8. Formation rate of $(dd\mu)$. Points = experimental results (from Bystritsky et al. 1979). (a) = the non resonant contribution (from Vinitsky et al. 1978). (b) = resonant + non resonant contribution (from Bystritsky et al. 1979).

5.2. Resonant formation

As it was discussed in section 3, the $(dd\mu)$ mesomolecule has a level $(J, v) = (1, 1)$ with a binding energy $E_b = 1.91$ eV. Since this value is smaller than the dissociation energy of the D_2 molecule,

$$E_{\text{dis}} = 4.5 \text{ eV}, \quad (5.8)$$

it is possible to form the mesomolecule by transferring energy to the vibrations of the molecule $((dd\mu)d2e)$ produced in the reaction depicted in fig. 5b:



This reaction proceeds from the initial state of the continuous spectrum of the system $(d\mu) + d$ with $J = 0$ and from the ground state $(K, v) = (0, 0)$ of the D_2 molecule into a bound state $(J, v) = (1, 1)$ of the mesic molecule $(dd\mu)$ and an excited vibrational state of the $((dd\mu)d2e)$ molecule with orbital angular momentum $K = 1$ and with vibrational quantum number $v = 7$, see fig. 9.*

Due to the quantized structure of the vibrational level this process is possible only if the collision energy (kinetic energy in the c.m. system), ϵ , equals a resonant value ϵ_R (see fig. 9):

$$\epsilon_R = \Delta E - E_b. \quad (5.10)$$

Therefore the resonant formation rate has the shape:

* The analysis of Vinitsky et al. (1978) was grounded on slightly unprecise value of the binding energy, $E_b = 1.96$ eV. Consequently, in their calculation they assumed that the $v = 8$ level was excited. We report here the results of a more recent investigation performed by the Dubna group (private communication by L.I. Ponomarev).

$$\Lambda_{\text{res}}(\epsilon) = W\delta(\epsilon - \epsilon_{\text{R}}). \quad (5.11)$$

One assumes that the kinetic energy distribution inside the target at temperature T is described by the Maxwell distribution, with $\epsilon_{\text{T}} = 3/2 kT$:

$$\gamma(\epsilon, \epsilon_{\text{T}}) = (27\epsilon/2\pi\epsilon_{\text{T}})^{1/2} \cdot (1/\epsilon_{\text{T}}) \exp(-3\epsilon/2\epsilon_{\text{T}}). \quad (5.12)$$

Hence, by averaging over such a distribution, one gets the contribution of the resonant process to the formation rate:

$$\lambda_{\text{res}} = \int d\epsilon \Lambda_{\text{res}}(\epsilon) \gamma(\epsilon, \epsilon_{\text{T}}) = W\gamma(\epsilon_{\text{R}}, \epsilon_{\text{T}}). \quad (5.13)$$

In conclusion, the molecular formation rate is the sum of the two contributions,

$$\lambda_{\text{dd}\mu} = \lambda_{\text{nres}} + \lambda_{\text{res}}. \quad (5.14)$$

These represent, respectively, the nonresonant term, which is weakly dependent on the $(d\mu)$ kinetic energy, i.e. on the temperature, and the resonant term, which varies with the temperature following the Maxwell distribution.

The constant W can be calculated in terms of the electric dipole matrix element between the initial and final state of the system. It is important to remark that this quantity is extremely sensitive to the vibrational number of the molecular level one is exciting.

The resonant energy ϵ_{R} has to be smaller than the energy difference between two consecutive vibrational levels of the D_2 molecule. This splitting is of the order of 0.1 eV. As already mentioned, the accuracy of the calculations of the binding energy of mesomolecules is again of the order of 0.1 eV. Hence the value of the resonant energy cannot be predicted from the theory and it has to be left as a free parameter in the fit of the experimental data.

In this way one finds [Bystritsky et al. (1979)]:

$$\epsilon_{\text{R}} = (0.050 \pm 0.003) \text{ eV}. \quad (5.15)$$

For the excitation of the $v = 7$ level one has [Vinitsky et al. (1978)] $\Delta E = 2.002$. Accordingly, from eq. (5.10) one easily calculates the energy of the $(J, v) = (1, 1)$ level:

$$E_{\text{b}} = (1.952 \pm 0.003) \text{ eV}. \quad (5.16)$$

It is clear from the above analysis that the study of the resonant formation can provide rather accurate determinations of the mesomolecular energy levels. For a comparison, we remember that the vacuum polarization shift of the $J = 1, v = 1$ level of the $(dd\mu)$ mesomolecule is 8 meV. On the other hand it is worth observing that the above analysis is somehow oversimplified.

For example, one should take into account that in the target the D_2 molecules are distributed among the rotational levels following the Boltzmann distribution. Hence the mesomolecule can be formed as a result of several $K \rightarrow K \pm 1$ transitions. The resonant energies for these transitions differ by amounts of the order of $K\hbar^2/(M_0 a_{\mu}^2)$. The resonant contribution to the formation rate is then a superposition of

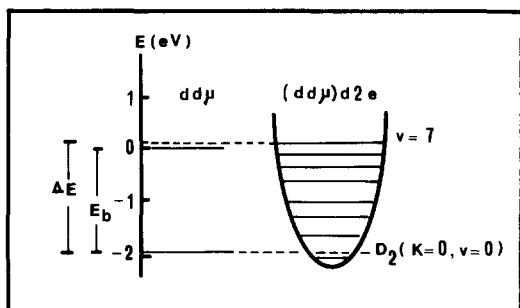


Fig. 9. Scheme of the resonant formation for $(dd\mu)$.

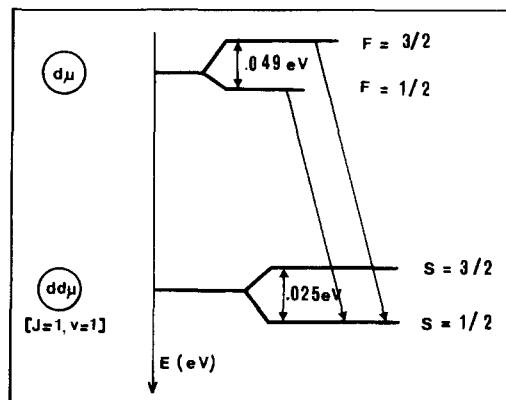


Fig. 10. Hyperfine structure of the $(d\mu)$ atom and of the $(dd\mu)$ $J=1$, $v=1$ state. $F = S_d + S_\mu$, and $S = S_{d1} + S_{d2} + S_\mu$. Additional splittings arising from the spin-orbit interactions are neglected.

Maxwell functions, at different values of ϵ_R , and not just a single term like in eq. (5.13). If this effect is taken into account when fitting the experimental data, the resulting value of E can be modified by a few meV.

More important and interesting effects arise when taking into account the Hyperfine Structure (H.S.) of mesic atoms and mesomolecules. The transition observed by the Russian group can be interpreted as being the transition $F=1/2 \rightarrow S=1/2$ (see fig. 10). As we just discussed, it corresponds to a resonant energy $\epsilon_R = 0.050$ eV, and the resonant formation rate is maximum at a temperature of about 400 K. The $F=3/2 \rightarrow S=1/2$ will correspond (see again fig. 10) to a resonant energy $\epsilon_R = (0.001 \pm 0.003)$ eV. Accordingly, the formation rate reaches a maximum at a temperature of a few kelvin.

In fact, evidence for such a resonance has been found by a Vienna-SIN group in an experiment which was performed at SIN in 1979. The group continued working on this subject and the analysis of their experimental work is still in progress. Ultimately, they will be able to completely clarify the H.S. of the $(dd\mu)$ mesomolecule with an accuracy of the order of 1 meV. These measurements are particularly meaningful, since their interpretation does not rely on a detailed knowledge of the Coulomb energy levels, which, as already discussed, is affected by an error of about 50 meV. We refer to their forthcoming publications for a more detailed analysis of hyperfine effects. However, it results already clear that the tiny H.S. can induce large effects on the formation rates of mesomolecules.

It is also clear that for the $(dd\mu)$ system we are now at the second generation of experiments, and we are close to a full understanding of the mesomolecule formation. On the other hand, the study of the $(dt\mu)$ mesomolecule, which we are going to discuss, is still in a rather primitive stage.

5.3. Formation of the $(dt\mu)$ mesomolecule

The theoretical calculations predict the existence of a weakly bound state also in the $(dt\mu)$ mesomolecule (see section 3 and table 4). It is again the $(J, v) = (1, 1)$ state, the energy being $E_b = (0.64 \pm 0.05)$ eV. Consequently, the resonant formation mechanism can work also for this system.

Since the binding energy is smaller than in the $(dd\mu)$ case it is now necessary to excite a molecular

level with a smaller vibrational quantum number v . Actually one has $v = 3$ [Ponomarev (1981)]*.

This results in a considerable enhancement of the factor W in eq. (5.11), by about two orders of magnitude. Consequently, one expects a corresponding enhancement of the resonant formation rate. The formation rate of $(dd\mu)$ mesomolecule being of order 10^6 s^{-1} , one can predict for the $(dt\mu)$ system rates of the order 10^8 s^{-1} [Gershtein and Ponomarev (1977)].

One has to observe, however, that it is hard to make a definite prediction on the value of the formation rate since it is crucially dependent on the resonant energy, ϵ_R .

Experimentally, only a lower limit for the formation rate has been obtained so far, which indeed confirms the prediction of Gershtein and Ponomarev (1977):

$$\lambda_{dt\mu} \geq 2 \times 10^8 \text{ s}^{-1}. \quad (5.17)$$

The result was obtained [Bystritsky et al. (1980a,b,c)] by measuring the yield of fusion neutrons Y_n in a deuterium target with tritium concentration up to 3% and a pressure up to 66 atm.

In these situations the $(t\mu)$ atoms are mostly originated through the transfer reaction:



Neglecting the time for the $(d\mu)$ formation as well as the fusion time compared to the muon lifetime, the expression for the neutron yield is:

$$Y_n = \lambda_{Tr} C_D \lambda_{dt\mu} C_T / (\lambda_\mu + \lambda_{Tr} C_D) (\lambda_\mu + \lambda_{dt\mu} C_T), \quad (5.19)$$

where C_D and C_T are the deuterium and tritium densities (normalized to L.H.D.) and λ_{Tr} and $\lambda_{dt\mu}$ are the transfer and formation rates (at L.H.D.).

Clearly, for

$$\lambda_{dt\mu} C_T \gg \lambda_\mu \quad (5.20)$$

the neutron yield is insensitive to the precise value of the mesomolecular formation rate. This is in essence the source of the experimental limit, eq. (5.17).

The experiment was performed at several temperatures, in the range 93–613 K, and showed no variation of the neutron yield, whereas one could expect a significant temperature dependence, on the grounds of the discussion for the $(dd\mu)$ ion.

There are two possible explanations of this behaviour [Bystritsky et al. (1980c)]. According to the first one, the rate $\lambda_{dt\mu}$ changes with temperature in accordance with the resonance dependence, still remaining so high that at any investigated temperature eq. (5.20) is fulfilled. Another possibility, pointed out by Ermolov (quoted in Bystritsky et al. (1980c)), is that the $(t\mu)$ atoms, which are formed through reaction (5.18) with a kinetic energy of 19 eV, do not reach thermal energy before the mesomolecule is formed. Therefore the variations of the temperature of the target do not correspond to variations of the $(t\mu)$ kinetic energy.

It should be noted that in both cases the value of the maximum of the formation rate can exceed substantially the limit given by eq. (5.17).

* Earlier calculations gave $E_b = 0.85 \text{ eV}$, which corresponds to excitation of the $v = 4$ level. We report here the result of more recent and more accurate calculations.

It is clear that, in order to have a determination and not just a limit of the formation rate, as well as in order to understand the kinetics of the muon catalysis in d – t mixtures, new experiments performed at intense muon beams and at higher tritium concentrations are needed. We refer to Gershtein et al. (1980) for a detailed analysis about the significance of a series of future experiments.

6. Muon sticking and reactivation

One has to remark that having large values of the mesomolecule formation rate is not a guarantee that there are many fusions. Beyond the mesomolecule formation there are other processes which can limit the number of fusions.

In particular it is important to discuss the possibility that the muon should stick to a charged product of the fusion reaction. For example, in the $(dt\mu)$ system, in which the fusion can occur via the process



the muon can stick to the α particle, thus forming a muonic helium ion $(\alpha\mu)$. If this occurs, and if the muon stays bound to the α particle, it is lost to the chain of fusions.

The sticking probability w can be calculated using the sudden approximation [Migdal (1939)]. For the $(dt\mu)$ case one finds [Bracci and Fiorentini (1981a)]:

$$w = 1.2\% , \quad (6.2)$$

so that on the average the muon sticks to the α particle after catalysing some eighty fusions, no matter how high is the mesomolecule formation rate.

In order to evaluate the actual number of fusions which a muon can catalyse, it is necessary to discuss if the muon stays bound to the α particle after it sticks to it.

As soon as it is formed, the $(\alpha\mu)$ ion has a kinetic energy

$$E_{\alpha\mu} = 3.5 \text{ MeV} , \quad (6.3)$$

and then it slows down like a heavy proton through a series of ionising collisions with the surrounding molecules. On the other hand, before the kinetic energy has become smaller than the appropriate thresholds, as a consequence of several reactions the $(\alpha\mu)$ ion can be stripped and the muon can be made again available for the cycle of nuclear reactions (muon reactivation).

The probability w_{ef} that at the end of the slowing down process the muon is still bound to the α particle will be reduced with respect to the initial value:

$$w_{\text{ef}} = (1 - R)w , \quad (6.4)$$

where the coefficient R is termed the reactivation efficiency. Clearly it is w_{ef} that ultimately determines the number of possible fusions.

It is easy to see that the slowing down and the stripping processes are really competitive. Indeed, if $\sigma_{\text{st}}^{(\mu)}$ is the $(\alpha\mu)$ stripping cross section and S is the stopping power – energy loss of the $(\alpha\mu)$ ion per unit length and per unit density – the reactivation efficiency is approximately

$$R = 1 - \exp(-I), \quad (6.5)$$

$$I = \int_0^{E_{\alpha\mu}} dE \sigma_{st}^{(\mu)}/S. \quad (6.6)$$

The stopping power S is given by an electron stripping cross section $\sigma_{st}^{(e)}$ for the hydrogen molecule by impact of the $(\alpha\mu)$ ion, times the average energy loss per collision, $\bar{E} \approx 30$ eV. By scaling arguments one expects:

$$\sigma_{st}^{(\mu)} \approx (a_\mu/2a_0)^2 \sigma_{st}^{(e)}. \quad (6.7)$$

In this way one gets:

$$I \approx (E_{\alpha\mu}/4\bar{E}) \cdot (m_e/m_\mu)^2 \approx 0.7. \quad (6.8)$$

Since the reactivation efficiency depends exponentially on I , the calculation of R requires a careful evaluation of the relevant cross sections.

Until recently the reactivation process had been discussed only by Jackson (1957). In that paper the treatment was at the level of estimating the order of magnitude of the reactivation probability. In order to calculate the actual number of fusions the process has been recently reanalysed by two groups [Bracci and Fiorentini (1981a), Gershtein et al. (1981)].

Following Bracci and Fiorentini (1981a) we discuss in some detail the case of $(dt\mu)$, which is the most interesting as a consequence of its high mesomolecule formation rate. The full chain of competing processes after the fusion is depicted in fig. 11, where it is taken into account that muon sticking can occur in the 1s state as well as in excited states $(\alpha\mu)^*$. These latter are most likely the n-s states, with a relative distribution $\sim 1/n^3$.

The muon reactivation can occur through several channels. Besides direct ionization, lines e and h in fig. 11, one has also to consider transfer reactions, lines f and i:



and multistep processes, i.e. collisional excitations of the $(\alpha\mu)$, line g, followed by a stripping reaction of the $(\alpha\mu)^*$.

In competition with these processes one has the slowing down of the $(\alpha\mu)$, lines c and d, and the electromagnetic cascade of the $(\alpha\mu)^*$, lines k and j.

The stopping power for the $(\alpha\mu)$ was derived from the existing data for protons in hydrogen, under the assumption that the stopping power is a function of the velocity of the particle, and not of its mass.

The cross sections for the reactions corresponding to lines e-i of fig. 11 have been calculated using the Born approximation and several versions of the semiclassical approximation. The comparison of the calculated values with the experimental data available for stripping of $(e\alpha)$ by proton impact shows that the validity of the approximation is at the level of 10–20%.

On these grounds one calculates the reactivation efficiency R :

$$R = 0.24, \quad (6.10)$$

with an estimated relative error of order 20%.

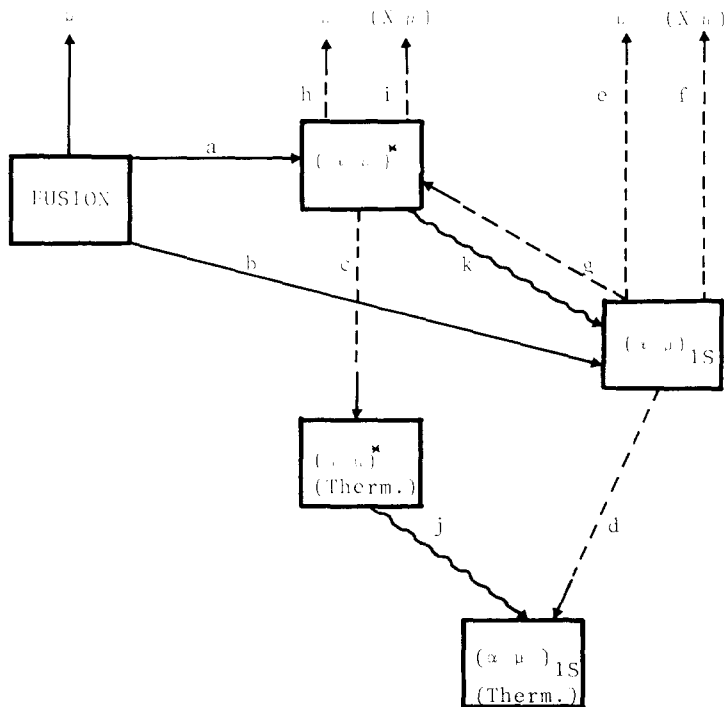


Fig. 11. Scheme of the competing processes in the reactivation phase (from Bracci and Fiorentini 1981a).

It is interesting to consider the contribution of the various processes to the reactivation (table 9). One sees that the direct ionization from the 1s state gives the main contribution, $R = 0.17$. This should be compared with the results of Jackson (1957), $R = 0.22$. The discrepancy is due to the different values of the 1s ionization cross section and stopping power. One also sees that the transfer channel and the combined effect of $1 \rightarrow n$ excitations together with stripping from the excited levels provide appreciable contributions to the reactivation efficiency.

The final result is in substantial agreement with Jackson's estimate. However, this agreement is somehow accidental, arising from the concurrence of several factors [Bracci and Fiorentini (1981a)].

The result of the Russian group [Gershtein et al. (1981)] for the $(dt\mu)$ problem is:

$$R = 0.23. \quad (6.11)$$

This is in agreement with the value previously reported, eq. (6.10), within the limits of the approximations used.

Table 9
Contributions of the various channels to the reactivation efficiency R .
The various channels are labelled as in fig. 11. (From Bracci and
Fiorentini 1981a.)

Channels included	(e)	(e) + (f)	(e) + (f) + (h) + (i)	(e) + (f) + (g) + (h) + (i)
R	0.17	0.20	0.22	0.24

By using eqs. (6.2), (6.4) and (6.10) one finds:

$$w_{ef} = 0.91\% . \quad (6.12)$$

This means that, on the average, a muon can catalyse at most 110 fusions before being trapped around a He-nucleus at thermal energy.

In the Russian paper one also finds calculations of w and w_{ef} for all the other mesomolecules (see table 10).

Table 10
Sticking probability (w), reactivation efficiency (R) and effective sticking probability (w_{ef}) for different fusion reactions according to (a) Bracci and Fiorentini (1981a) (b) Gershtein et al. (1981).

Reaction	$w, 10^{-2}$	R	w_{ef}
$(dt\mu) \rightarrow ({}^4\text{He}\mu)^+ + n$	(a) 1.20	0.24	0.91×10^{-2}
	(b) 1.12	0.23	0.86×10^{-2}
$(tt\mu) \rightarrow ({}^4\text{He}\mu)^+ + 2n$	(b) -	-	$0.05 \div 0.18$
$(dd\mu) \rightarrow ({}^3\text{He}\mu)^+ + n$	15.5	0.05	0.15
$(dd\mu) \rightarrow (t\mu) + p$	2.11	0	0

A warning is needed: all the information we have so far on the sticking effect are just theoretical. It is nice that two groups found similar results independently. Nevertheless some important point might have been missed, since the chain of competing processes is rather involved. It is therefore extremely desirable to have experimental results on this important point*.

An obvious question is whether one can avoid the limitations imposed by the sticking effect. A possible way out could be to provide energy to the $(\alpha\mu)$ in order that it stays a longer time at energies higher than the thresholds for the reactivation reactions. In principle this can be done by applying an electric field E . One expects this can balance the frictional force $F = SD$ arising from the collisions with the hydrogen molecules in a target of density D , provided that:

$$eE \approx S \cdot D . \quad (6.13)$$

The equation of motion of the $(\alpha\mu)$ in the presence of electric fields has been discussed by Bracci and Fiorentini (1981b) for several geometrical arrangements. It comes out that at a density of 10^{19} atoms/cm³ R doubles with respect to the zero field case if the electric field is of the order of 20 kV/cm.

Clearly, this results in a limited gain for the number of fusions and, more important, it is extremely hard to avoid breakdown before reaching so high an electric field.

In conclusion, the sticking problem looks as a severe limitation to the number of fusions a muon can catalyse. Presumably the problem is not hopeless, as one can argue for example from the naive scheme just outlined.

* The group of Prof. Vorobyov is presently analysing the result of an experiment performed at Gatchina on muon sticking following fusion in $(dd\mu)$ mesomolecules (private communication by Prof. Vorobyov).

7. Energy production from muon catalysed fusion?

The large value of the $(dt\mu)$ mesomolecule formation rate, $\lambda_{dt\mu} \geq 2 \times 10^8 \text{ s}^{-1}$, has revived the idea of using m.c.f. to the purpose of energy production.

In this spirit, Petrov has proposed an ingenious scheme [Petrov (1978, 1980)], the so called Mesocatalytic Reactor, (M.R.), or Hybreeder, where m.c.f. is used together with the electronuclear breeding (E.N.) in order to produce a positive energy outline. The logical scheme of the M.R. is shown in fig. 12.

Light nuclei, such as d or t, are accelerated up to an energy of about 1 GeV/nucleon and hit a target where fast nucleons and π mesons are produced. As π^- are most likely produced in n-n collisions, it is convenient to have both beam and target rich of neutrons.

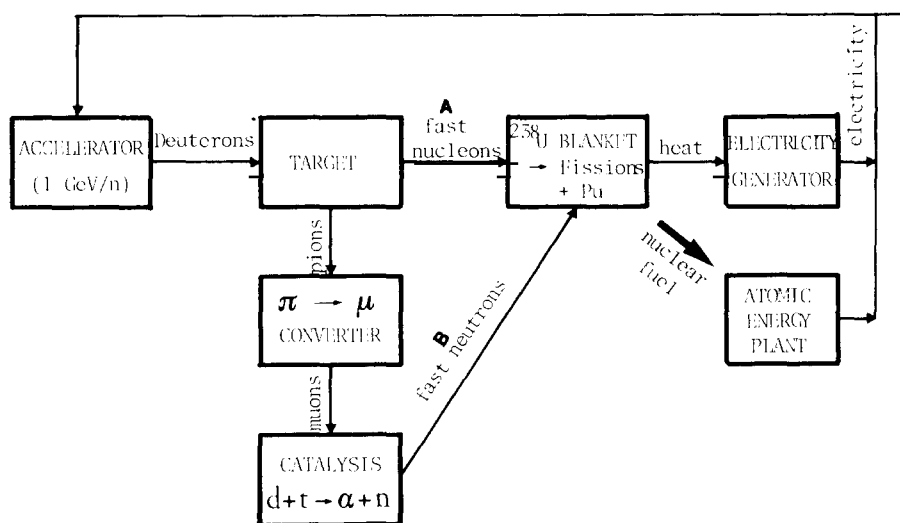


Fig. 12. Scheme of the Mesocatalytic Reactor (Hybreeder), (from Petrov 1980).

The fast neutrons impinge onto a U-238 blanket* where they cause the fission of uranium and produce fissile isotopes (Pu). In this way one gets out heat, which can be converted into electric energy in the Electric Generator, as well as nuclear fuel, which can be used in an atomic energy plant.

Essentially, this is the scheme of electronuclear energy production.

The pions, which have been trapped in a magnetic device around the target, will decay into muons and neutrinos in the converter. In principle, this system provides a source of muons which is much more economical than the standard way of producing a muon beam. Petrov and Shabelski (1981) estimate that the beam energy necessary for the production of one negative pion in a cylindrical berillium target is about 4.5 GeV.

In a suitable deuterium tritium mixture inside the so-called Synthesizer the muon will catalyse nuclear fusions. Petrov observes that, by using for electronuclear breeding the 14 MeV neutron originated from the dt fusion it is possible to recover much more energy than the fusion energy itself†.

* Note that most (99.28%) of available uranium consists of U-238, which is not fissile by impact of thermal neutrons. Energy production in atomic energy plants occurs through burning of the rare U-235 isotope.

† This idea is also used in the so-called Tohamak + blanket systems.

The theoretical estimates, and also experimental data [Weale et al. (1961)], show that in a thick blanket made of U-238 any 14 MeV neutron gives rise to one fission and four additional neutrons. These will produce about 2.4 Pu nuclei available for further burning, and one tritium nucleus for replacing the burned nucleus. Since the consumption of one Pu nucleus in a thermal reactor gives rise to 1.6 fissions, taking into account Pu breeding, one reaches the conclusion that from any fusion neutron it is possible to recover the energy of some 5 fission reactions, i.e. about 1 GeV.

We remember that the fusion energy itself is only 17 MeV, so that the idea of using the fusion neutrons for electronuclear breeding gives a substantial gain in the energy production.

The total multiplication factor of the initial beam energy

$$f = E_{\text{out}}^{(\text{th})}/E_{\text{beam}} \quad (7.1)$$

is thus the sum of two terms, arising from the electronuclear energy production in the two channels, A and B, of fig. 12.

Petrov estimates:

$$f = 15 + 0.25N\Phi. \quad (7.2)$$

The first figure arises from the electronuclear breeding in channel A. Standard estimates for the multiplication factor of E.N. systems is $f_{\text{en}} = 18$. Petrov calculates that some 20% of the primary beam energy is spent for pion production. Hence the contribution of channel A is reduced by this factor.

The other term in eq. (7.2) represents the contribution due to the m.c.f.; Φ is the probability of having a muon available for the catalysis per produced pion, and N is the number of fusions which a muon can catalyse.

Petrov assumes $N = 100$ and $\Phi = 0.8$, so that the total amplification factor is $f = 35$, almost twice the value for pure E.N. systems.

The efficiency of the system, i.e. the ratio between the electric energy which the system can release and the thermal energy produced is:

$$q = (E_{\text{out}}^{(\text{el})} - E_{\text{beam}}^{(\text{el})})/E_{\text{out}}^{\text{Th}} = \eta_e - 1/f\eta_a, \quad (7.3)$$

where η_e is the efficiency of the nuclear plant, $\eta_e = 0.35$, and η_a is the efficiency of the accelerator. By using $\eta_a = 0.6$ one gets out with an efficiency of 30%, which looks quite acceptable.

The relationship between the beam power P_{beam} and the electric power one can deliver $P_{\text{del}}^{(\text{el})}$ is of course:

$$P_{\text{del}}^{(\text{el})} = fqP_{\text{beam}}. \quad (7.4)$$

If one requires, as usual for nuclear plants, a value $P_{\text{del}}^{(\text{el})} \approx 1 \text{ GW}$, it is then necessary to have $P_{\text{beam}} \approx 80 \text{ MW}$.

It is worth recalling that at modern meson factories such as LAMPF one reaches a beam power $P_{\text{beam}} \approx 0.4 \text{ MW}$ and accelerator efficiency $\eta_a \approx 0.1$ for 800 MeV protons. Therefore, the values quoted above for P_{beam} and η_a are quite large and require the solution of severe technical problems. However, accelerators with $P_{\text{beam}} \approx 300 \text{ MW}$ and $\eta_a \approx 0.6$ are now being designed for electronuclear breeding systems [Schriber et al. (1977)].

Besides these problems, which are common to any electronuclear energy production scheme, it is necessary to have a detailed discussion of the mechanism of confinement of pions and nucleons until fusion takes place.

The assumption $\Phi = 0.8$ looks optimistic, as Petrov himself remarks. In fact, if the produced pions are trapped in a region containing the d-t mixture necessary for fusion, most of these pions will be stopped and absorbed by the nuclei before they decay into muons. On the other hand, if the pions are trapped in a very low density region, where they can decay before being stopped, then one has to solve the problem of transporting the muons to a region where they can induce fusions. Indeed, as it is clear from eq. (7.2), if it comes out that practical values of Φ are an order of magnitude smaller, then it makes no sense to add the muon catalysis to the electronuclear system. However, it is the product $N\Phi$ which determines the energy output of the muon catalysis. Clearly it is necessary to determine the actual value of N , the number of fusions. Taking $N = 100$ looks now as a safe estimate. If N can be made appreciably larger, then the hope for energy production from m.c.f. becomes more realistic.

In conclusion, Petrov's scheme brings two interesting ideas into the subject of m.c.f.:

(i) it presents an "economical way" of getting a source of muons, in that all the energy of the primary beam is eventually used for energy production – through channels A and B of fig. 12 – and the particle losses of the muon beam are appreciably reduced.

(ii) it shows that, by a suitable use of the fusion products, it is possible to recover much more energy than from the fusion itself.

On the other hand, several problems have to be discussed in detail before one can reach conclusions about the feasibility of the system.

We would also like to mention a different scheme proposed by Tan (1976). The idea is to use the m.c.f. in order to provide the energy needed for the ignition of ordinary d-t fusion reactions in an inertially confined deuterium tritium pellet. The pellet has to be first compressed to ~ 1000 times L.H.D. and preheated to 1 keV by a laser or by an electron beam acting as a prepulsor. Simultaneously, or a short time beforehand, a pulse of muons is injected into the pellet. The energy released in the m.c.f. should provide further heating of the plasma, so that, when the catalytic reactions are complete, the ignition temperature is reached and the main burn takes place by means of ordinary fusion reactions.

This method avoids the sticking problem. Indeed, once the $(\alpha\mu)$ is formed it travels through a medium which is almost completely ionized, so that the usual mechanism of energy loss – ionization of ordinary atoms – is no longer effective. It is then clear that stripping reactions can be well efficient here, resulting in a prompt breaking of the $(\alpha\mu)$ ion soon after it is formed.

On the other hand, since the pellet is almost completely ionised, the formation of $(dt\mu)$ mesomolecules is quite unlikely. The fusion reaction has therefore to occur in flight, i.e. directly in the collision between a $(t\mu)$ and a d nucleus. The rate for this reaction is estimated to be 10^9 s^{-1} (following Jackson 1957)* and the confinement time of the plasma is of the order of 1 ns, so that, on the average, a muon can catalyse just one fusion during the confinement time. As pointed out by Hincks et al. (1977), this spoils the effectiveness of Tan's scheme. See also the same paper for further criticism.

8. Conclusions

From the foregoing discussion it appears that the theoretical and experimental work performed in the last few years has opened the door to a new, refined spectroscopy. This is interesting for several

* Nuclear reactions in flight and in mesomolecules have been recently re-discussed by Bogdanova et al. (1980).

reasons:

(i) it is possible to perform refined tests of the methods of solution of the Coulomb three-body problem.

(ii) the study of mesomolecules is also relevant for the investigation of the energy levels of ordinary molecules. This occurs since nonadiabatic effects are extremely enhanced ($m_\mu/m_p \approx 0.1$ whereas $m_e/m_p \approx 0.0005$).

(iii) in mesomolecules it is possible to study the nucleon–nucleon interaction at distances of the order of some hundreds fm. This is interesting from a general point of view. For example, one can get information about the long distance behaviour of hadronic forces.

Theory and experiment are in substantial agreement for the interpretation of the formation process of ($pp\mu$), ($pd\mu$) and ($dd\mu$) mesomolecules. The physics of the last system is particularly rich as a consequence of the sensitivity of the resonant formation process to the H.F. structure. Evidence of such effects has just been found by a Vienna-SIN group [Breunlich (1981) and Breunlich et al. (1981)]. Now we have to understand the H.F. structure in detail. In conclusion, it appears that in the study of the ($dd\mu$) system we are now at the second generation of experiments and we are close to a full understanding of the mesomolecular formation process. On the other hand, the study of the ($dt\mu$) system is still in a primitive stage and deserves substantially new experimental effort.

It is also remarkable that, besides the determination of the mesomolecular energy levels, it is now possible to detect transitions between mesomolecular states. The recent result of Bardin et al. (1981), who measured the ortho-para transition rate in the ($pp\mu$) mesomolecule, has an intrinsic interest, apart from its relevance for the study of weak interactions.

The theoretical predictions (later confirmed by experimental results) of a particularly high value of the ($dt\mu$) formation rate have revived the idea of using the m.c.f. for energy production. In this respect, several points have to be further investigated. First, one has to clarify the formation mechanism of the ($dt\mu$) mesomolecule and provide definite estimates on the number of possible fusions. For this purpose, new experiments especially devoted to the study of the kinetics of the m.c.f. are needed. It is also interesting to analyse more deeply the feasibility of the various components of Petrov's Mesocatalytic Reactor. Actually, we feel that Petrov's scheme has to be considered just as an interesting example of an approach to the exploitation of m.c.f. Maybe this is not the best possible approach and it could be useful to investigate other schemes of practical applications of m.c.f. The primary application of m.c.f. may even turn out to be something else from energy production.

The field is really open and no firm statement can be made presently about the possibility of practical applications of m.c.f. As a conclusion, an Italian proverb seems appropriate*:

“Se son rose fioriranno”.

Acknowledgements

We benefited from conversations on this subject with several people. Particularly, we would like to express our gratitude to Drs. Bakalov, Breunlich, Hofer, Leon, Melezhik, Petitjean, Petrov, Polikanov, Ponomarev, Rafelski, Rosati, Vorobyov and Zavattini.

The critical reading by Drs. Ericson and Ponomarev is warmly acknowledged.

Finally, we thank Dr. Hildebrand for kindly allowing us to produce a photograph from his experiment.

* Approximately: if they are roses, they will bloom.

Appendix. The continuous analog of Newton's method

The system of integro-differential equations, one obtains in the frame of the adiabatic representation, was solved numerically by the Russian group by means of a method we will briefly outline below. For details see Ponomarev et al. (1973). The mathematical aspects were mainly worked by I.V. Puzynin.

First of all, one truncates the complete system of equations, so to have a finite number of unknown functions. The system can then be written in the form:

$$y_i'' + \sum_{j=1}^n (q_{ij}(x) - \lambda \delta_{ij}) y_j = 0 \quad i = 1, \dots, n, \quad (\text{A.1})$$

with the boundary conditions:

$$y_i(0) = 0, \quad y_i(\infty) = 0. \quad (\text{A.2})$$

For the sake of simplicity we consider a Sturm–Liouville problem consisting of a single equation, in order to enucleate the peculiarities of the method without any extrinsic complication, following Ponomarev et al. (1973).

Even with the simpler problem

$$y'' + (q(x) - \lambda)y = 0, \quad y(0) = 0, \quad y(\infty) = 0 \quad (\text{A.3})$$

and the auxiliary normalization condition:

$$\int_0^{\infty} dx y^2 = 1, \quad (\text{A.4})$$

one has to meet the difficulty that the function $q(x)$ (the effective potential in physical applications) is known only numerically. The method proposed has the advantage of a simultaneous determination of the eigenfunction and the eigenvalue, whereas one generally starts with the determination of λ , and subsequently y is evaluated.

In order to further simplify the problem, one first considers a limited interval $(0, a)$. The couple $(\lambda, y(x))$ can be viewed as an element of the space $Z = \mathbf{R} \times C^2[(0, a)]$, $C^2(0, a)$ being the space of the functions doubly differentiable on the interval $(0, a)$. The Sturm–Liouville problem and the normalization condition can be viewed as a nonlinear operator φ acting in Z and transforming $z \in Z$ into the null element of the space $W = \mathbf{R}^3 \times C(0, a)$:

$$\varphi(z) = 0, \quad (\text{A.5})$$

where, in component form, one has:

$$\varphi^{(1)}(\lambda, y) = y'' + [q - \lambda]y, \quad (\text{A.6a})$$

$$\varphi^{(2)}(\lambda, y) = y'(0) + f(\lambda, 0)y(0), \quad (\text{A.6b})$$

$$\varphi^{(3)}(\lambda, y) = y'(a) + g(\lambda, a)y(a), \quad (\text{A.6c})$$

$$\varphi^{(4)}(\lambda, y) = \int_0^a y^2 dx - 1. \quad (\text{A.6d})$$

For the physical problem of interest $f = g = 0$, but the more general boundary conditions described above do not introduce any serious difficulty.

The crucial point consists in finding a continuous parameter t such that:

$$\frac{d}{dt} [\varphi(z(t))] = -\varphi(z(t)); \quad z(0) = z_0, \quad (\text{A.7})$$

z_0 being any initial choice of z . Since one has:

$$\varphi(z(t)) = \varphi(z_0) \exp(-t), \quad (\text{A.8})$$

one expects that $\lim_{t \rightarrow \infty} z(t)$ is a solution of the problem. This actually happens, under fairly large conditions, as proved in Zhidkov and Puzynin (1967 and 1968). The authors name the method a continuous analog of Newton's method since, if eq. (A.7) is written in the form

$$z' = -(\varphi'(z))^{-1} \cdot \varphi(z), \quad z(0) = z_0, \quad (\text{A.9})$$

it reminds the well known method of successive approximations for the solution of the equation $f(x) = 0$:

$$x_{k+1} - x_k = -f'(x_k)^{-1} \cdot f(x). \quad (\text{A.10})$$

In order to actually find the parameter t and the limit of $z(t)$ for $t \rightarrow \infty$, one builds a step-by-step procedure in the following way. Defining:

$$\mu(t) = \lambda(t) + d\lambda(t)/dt, \quad (\text{A.11a})$$

$$v(x, t) = \partial y(x, t)/\partial t, \quad (\text{A.11b})$$

and using eq. (A.3) the four equations (A.6) can be written as

$$\partial^2 v(x, t)/\partial x^2 + [q - \lambda(t)]v(x, t) = -\varphi^{(1)}(t) - [\lambda(t) - \mu(t)]y, \quad (\text{A.12a})$$

$$\partial v(0, t)/\partial x + f(\lambda(t), 0)v(0, t) = -\varphi^{(2)}(t) + [\lambda(t) - \mu(t)] \partial f(\lambda(t), 0)/\partial \lambda, \quad (\text{A.12b})$$

$$\partial v(a, t)/\partial x + g(\lambda(t), a)v(a, t) = -\varphi^{(3)}(t) + [\lambda(t) - \mu(t)] \partial g(\lambda(t), a)/\partial \lambda, \quad (\text{A.12c})$$

$$2 \int_0^a y(x, t)v(x, t) dx = -\varphi^{(4)}(t), \quad (\text{A.12d})$$

where $\varphi^{(i)}(t) = \varphi^{(i)}(z(t))$ and the conditions for $t = 0$ are:

$$z_0 = [\lambda_0, y_0(x)]; \quad \lambda(0) = \lambda_0; \quad y(x, 0) = y_0(x). \quad (\text{A.13})$$

Dividing the real axis into intervals (t_k, t_{k+1}) of length τ_k , introducing the notations

$$\begin{aligned} \lambda_k &= \lambda(t_k), \mu_k = \mu(t_k), y_k(x) = y(x, t_k), \\ v_k(x) &= v(x, t_k), f_k = f(\lambda(t_k), 0), g_k = g(\lambda(t_k), a) \end{aligned} \quad (\text{A.14})$$

and replacing derivatives with respect to t by finite differences, the system (A.12) is transformed into a boundary value problem for the functions $v_k(x)$, whose solution can be written in the form:

$$v_k(x) = v_{1k}(x) + \mu_k v_{2k}(x). \quad (\text{A.15})$$

The functions v_{ik} solve μ -independent boundary value problems:

$$v_{ik}(x) + (q(x) - \lambda_k)v_{ik}(x) = P_{ik}(x), \quad v'_{ik}(0) + f_k v_{ik}(0) = \psi_{ik}, \quad v'_{ik}(a) + g_k v_{ik}(a) = \theta_{ik}, \quad (\text{A.16})$$

where

$$\begin{aligned} P_{1k} &= -(y_k''(x) + q(x)y_k), & P_{2k} &= y_k(x) \\ \psi_{1k} &= -[y_k'(0) + f_k y_k(0)] + \lambda_k \partial f_k / \partial \lambda, & \psi_{2k} &= -\partial f_k / \partial \lambda \\ \theta_{1k} &= -[y_k'(a) + g_k y_k(a)] + \lambda_k \partial g_k / \partial \lambda, & \theta_{2k} &= -\partial g_k / \partial \lambda. \end{aligned} \quad (\text{A.17})$$

Knowing $z_k = (\lambda_k, y_k)$, one can determine the v_{ik} 's, which in turn give μ_k via the normalization condition:

$$\mu_k = \left[\frac{1}{2} - \frac{1}{2} \int_0^a y_k^2 dx - \int_0^a y_k v_{1k} dx \right] / \int_0^a y_k v_{2k} dx. \quad (\text{A.18})$$

At this point, one can determine y_{k+1} via the finite difference relation:

$$(y_{k+1} - y_k) / \tau_k = v_k \quad (\text{A.19})$$

and pass to next step.

In order to take into account the fact that the original equation is defined over the interval $(0, \infty)$, one can exploit the knowledge of the asymptotic form of the effective potential $q(x)$, and consequently of the asymptotic behaviour of $y(x)$. The asymptotic solution $\tilde{y}(x)$ has the form

$$\tilde{y}(x) = C \cdot \exp[-\sqrt{\lambda}x] \sum_{n=0}^N b_n x^{-n}, \quad (\text{A.20})$$

where the b_n 's are determined by $q(x)$. The constant C is so chosen as to connect smoothly \tilde{y} with $y(x)$ at the point a . The boundary conditions then are modified as follows:

$$\varphi^{(2)}(\lambda, y) = y(0) = 0, \quad \varphi^{(3)}(\lambda, y) = y'(a) + y(a) \left[\sqrt{\lambda} \sum_{n=0}^N n b_n a^{-n-1} \right] / \sum_{n=0}^N b_n a^{-n}$$

$$\varphi^{(4)}(\lambda, y) = \int_0^a y^2 dx - 1 + \int_a^\infty \bar{y}^2 dx = 0. \quad (\text{A.21})$$

One should notice that it is convenient to choose as z_0 a couple $(z_0, y_0(x))$ which represents an analytical solution of the equation obtained from eq. (A.3) by replacing $q(x)$ with a potential $V(x)$ which allows analytical solution and, to some extent, approximates the effective potential $q(x)$. For the case of mesomolecules a suitable choice is offered by the Morse potential:

$$V(x) = D(\exp(-2c(x - x_0)) - 2 \exp(-c(x - x_0))). \quad (\text{A.22})$$

The case of a system of coupled equations does not present any substantially new feature.

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Note added in proof

The muon sticking coefficient in the muon catalysed d-d fusion has been recently measured at the Leningrad synrocyclotron [Balin et al. (1981)]. The value obtained, $w = 0.14 \pm 0.01$, is in agreement with the theoretical prediction.

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