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Exotic nuclei/Les noyaux exotiques

First observation of two-proton radioactivity from an atomic nucleus

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Presented by Guy Laval

Abstract

Nature uses radioactivity to gain stability for an unstable atomic nucleus. The discovery of radioactivity is considered the starting point of nuclear physics. The classical types of radioactivity, α, β, γ decay and nuclear fission, have allowed for many detailed studies of nuclear structure and have found a wide range of applications in other fields of science. According to theoretical predictions, other types of nuclear transformation should occur for very short lived atomic nuclei. In recent experiments at the GANIL and GSI laboratories, two-proton radioactivity was observed for the first time in the decay of proton-rich ^{45}Fe. ***To cite this article: B. Blank et al., C. R. Physique 4 (2003).***

Résumé

Première observation de la radioactivié deux-protons d'un noyau atomique. La Nature utilise la radioactivité pour stabiliser un noyau atomique instable. La découverte de la radioactivité constitue le point de départ de la physique nucléaire. Les radioactivités classiques, les desintégrations α, β, γ et la fission, ont permis une étude approfondie de la structure nucléaire et ont trouvé des applications très diverses dans d'autres domaines de science. Selon des prédictions théoriques, d'autres types de transformations nucléaires devraient exister pour des noyaux de très courte durée de vie. Lors des expériences récentes au GANIL et au GSI, la radioactivité deux-protons a été observée pour la première fois dans la décroissance de ^{45}Fe. ***Pour citer cet article : B. Blank et al., C. R. Physique 4 (2003).***

Keywords: Two-proton radioactivity; Nuclear transformation; ^{45}Fe

Mots-clés : Radioactivité deux-protons ; Transformation nucléaire ; ^{45}Fe

1. Introduction

The atomic nucleus is made up of positively-charged protons and neutral neutrons, the nucleons. To form a stable atomic species, a balance between the number of neutrons and of protons has to be respected. For light nuclei, this requires an equal number of both nucleons. Beyond mass or nucleon number $A = 40$, stable atomic nuclei accommodate more neutrons than protons to outbalance the repulsion of the charged protons. The number of neutrons with respect to the number of protons increases until reaching a neutron number of $N = 126$ and a proton number $Z = 82$ for lead.

Nuclei which do not respect this balance are unstable and decay by radioactive processes. For a minor disequilibrium, β decay occurs: an excess neutron is transformed into a proton and vice versa by emitting an electron/positron and an anti-

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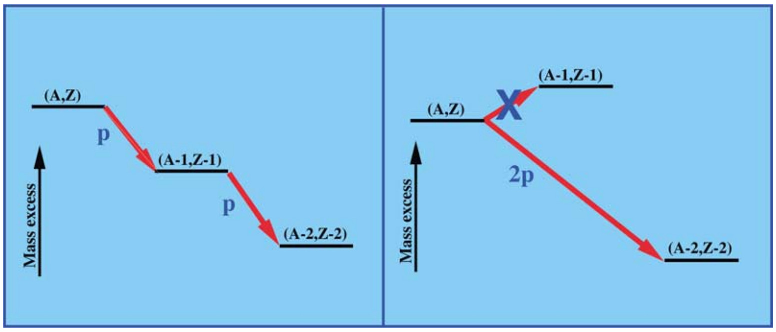

Fig. 1. Schematic representation of sequential (right) and simultaneous two-proton emission. If the one-proton daughter state is energetically not accessible, the emission has to be simultaneous.

neutrino/neutrino. If the imbalance between proton and neutron numbers is too large, the nuclear forces can no longer bind all nucleons and the atomic nucleus becomes particle-unstable. Thus the limits of nuclear stability are reached and the unstable nucleus will emit single excess nucleons or clusters of nucleons. The emission of clusters of two protons and two neutrons, a ^{4}He nucleus, was the first radioactivity observed and was called α radioactivity. It occurs mainly for very heavy nuclei in the lead to uranium region ($Z = 82$–92), but also for very proton-rich lighter nuclei. For the heaviest nuclear species, fission may occur, where the heavy nucleus splits into two medium-heavy pieces.

Theoretical investigations [1,2] in the 1960s predicted that for medium-mass ($A = 50$–100) nuclei, other new radioactive decays might occur. For very proton-rich odd-Z nuclei, one-proton radioactivity, i.e., the emission of one single excess proton, should be observable, whereas for even-Z nuclei with a large proton excess, two-proton radioactivity, the emission of a pair of protons, should occur. Although these one-proton or two-proton emitting nuclei are particle-unbound, they may exhibit a measurable half-life due to the Coulomb barrier the protons have to tunnel through.

It was soon realized that two-proton emission could probably proceed via different decay mechanisms. Depending on the position of the intermediate one-proton daughter state, two-proton emission might be a sequential process, where one-proton emission populates a well defined quantal state in the one-proton daughter, before a second one-proton emission occurs to a two-proton daughter state. However, the nuclear pairing force, which couples two nucleons of the same type to gain energy, is responsible for the fact that the masses of even-Z nuclei (in our case those of the two-proton emitter and the two-proton daughter) are smaller that the one of the one-proton daughter, thus rendering one-proton emission impossible (see Fig. 1). In such a case, the two protons have to be emitted simultaneously for energy conservation.

The simultaneous two-proton decay can be described by two extreme pictures: (i) the two protons have no angular or energy correlation and the decay mechanism is dictated only by phase space. This decay is commonly referred to as democratic three-body decay; (ii) the decay proceeds via the ^{2}He resonance and, as in α decay, a preformed cluster is emitted, decaying into two protons as the two-proton system is not bound. This decay mode is called ^{2}He emission. A sound theoretical description has certainly to accommodate both pictures as extremes of a more realistic modeling of two-proton radioactivity.

2. Two-proton emission from ground and excited states

One-proton radioactivity was observed as early as 1981 [3,4] and constitues today a powerful tool to study nuclear structure far from stability [5,6].

2.1. Two-proton emission from light unbound nuclei

Two-proton emission from a nuclear ground state was first observed indirectly in the decay of ^{6}Be by Geesaman et al. [7]. More detailed studies were performed by Bochkarev and co-workers [8]. ^{6}Be decays by the emission of two protons to the ^{4}He ground state with a half-life of the order of 10^{-21} s. It turned out than the decay can be described by a phase-space picture and it was thus termed ‘democratic’ decay [8]. However, due to the large width of the ^{6}Be ground state and especially of the ground state of the one-proton daughter ^{5}Li, the decay might as well be, at least in part, sequential.

A similar case is the one of ^{12}O the decay of which was recently studied at Michigan State University [9,10]. In contrast to the decay of ^{6}Be, where the intermediate state, although very large, lies higher in energy than the two-proton daughter state, the sequential branch is widely open in ^{12}O decay and the experimental data can thus be described by a sequential as well as a three-body picture. The half-life is about 10^{-21} s.

2.2. Two-proton emission from excited states

Two-proton emission from an excited state was first observed in experiments in Berkeley when the decay of ^{22}Al was studied. The β decay of this isotope populates strongly the isobaric analog state in the β-decay daughter ^{22}Mg. This highly excited state decays than by emission of two protons to the ground and first excited states of ^{20}Ne. In this decay, many intermediate states in the one-proton daughter ^{21}Na are energetically accessible and therefore a sequential decay pattern could be nicely observed.

Since these early studies of β-delayed two-proton (β2p) emission more examples of β2p emitters have been found (see [11] for a recent example). All cases studied in more detail revealed a sequential decay pattern.

The decay of other excited states populated by, e.g., inelastic scattering were also studied. It was for example argued [12] that an excited 2^+ state in ^{14}O might decay directly to the ^{12}C ground state. However, a sequential picture was observed where the decay proceeds via the first excited state in ^{13}N. Another case was the decay of the first excited state in ^{17}Ne which could decay by simultaneous two-proton emission to the ground state of ^{15}O. However, this nuclear level was found to disintegrate by γ decay [13]. Finally a recent experiment at Oak Ridge National laboratory has observed two-proton events from an excited state in ^{18}Ne without being able to saying anything about the decay mechanism [14].

3. Two-proton radioactivity of ^{45}Fe

The main drawback of experiments with light nuclei as described in the previous sections is that either levels in the intermediate one-proton daughter are accessible and the decay is then likely to be sequential or levels in the intermediate nucleus, although with their central value not accessible, are so broad that tails of these states reach the allowed region, thus opening the one-proton channel. In all cases, the half-lives are very short ($\approx 10^{-21}$ s).

The situation changes when one deals with medium-mass (A $\simeq$ 40–50) nuclei. In these cases, the Coulomb barrier is already sufficiently high to quasi-bind the unbound protons and to yield therefore half-lives in the microsecond to millisecond range. Several theoretical predictions [15–17] found that ^{45}Fe, ^{48}Ni, and ^{54}Zn are promising candidates for this radioactivity. Their two-proton decay energies should be of the order of 1.1–1.5 MeV. All commonly used mass models predict that the decay of these nuclei has to be simultaneous because the one-proton daughter state lies higher in energy than the parent nucleus. From an experimental point of view ^{45}Fe was expected to be the best candidate, as its production rate was predicted to be highest.

3.1. Experimental observation

In experiments at the LISE3 separator of GANIL [18] and at the FRS of GSI [19], the decay of ^{45}Fe was recently studied. Both experiments produced ^{45}Fe by projectile fragmentation of a stable ^{58}Ni beam impinging on a natural nickel and beryllium target, respectively. Behind the target, a powerful fragment separator was used in both cases to separate ^{45}Fe and other exotic nuclei from the bulk of less exotic species. Fig. 2 shows a schematic drawing of the GSI fragment separator and the detection setup.

At the exit of the separators, a detection setup consisting of detectors for tracking, time-of-flight (TOF), and energy-loss (ΔE) measurements was installed to identify the projectile fragments transmitted to the exit of the separators and to study, after implantation in a silicon telescope, their decays. Fig. 3 shows the ΔE–TOF identification plot from the GANIL data demonstrating the nice separation and identification of the different isotopes.

The implantation device consisted in silicon detectors, in the GSI experiment of a telescope of seven detectors and in the GANIL experiment of a silicon strip detector with 16×16 strips yielding a total of 256 pixels. The decay of the radioactive species was observed in the same detectors. Therefore, a correlation in time and space could be performed. In such a way, ^{45}Fe implants could be unambiguously correlated with their decays.

Fig. 4 shows the decay energy spectrum from both experiments. In the GSI experiment, six ^{45}Fe implantations were observed after which five decays could be observed. In one case, a temporary malfunctioning of the experiment electronics prevented the observation of the decay. Four of these five decay events are characterized by an energy release of 1.1(1) MeV. In the GANIL experiment, 22 ^{45}Fe implantations were observed. Twelve of them are followed by a decay with a decay energy of 1.14(5) MeV. The decay half-life determined in both experiments yields a value of $3.8^{+2.0}_{-0.8}$ ms.

In both experiments, additional detectors were mounted to search for β particles from the concurrent decay branch, β-delayed particle emission. At GSI, a NaI barrel was used to observe the 511 keV photons from the positron annihilation. At GANIL, a 6 mm thick silicon detector was able to detect directly the positrons. None of the events in the 1.14 MeV peak was found in coincidence with positrons.

Additional evidence for two-proton radioactivity as the origin of the 1.14 MeV peak comes from the fact that one-proton or two-proton peaks from β-delayed decays usually show a broadening of the peak due to β pile-up. No such broadening could be observed in the GANIL data, where the somewhat higher statistics allows for such a study. The GANIL data allowed also to

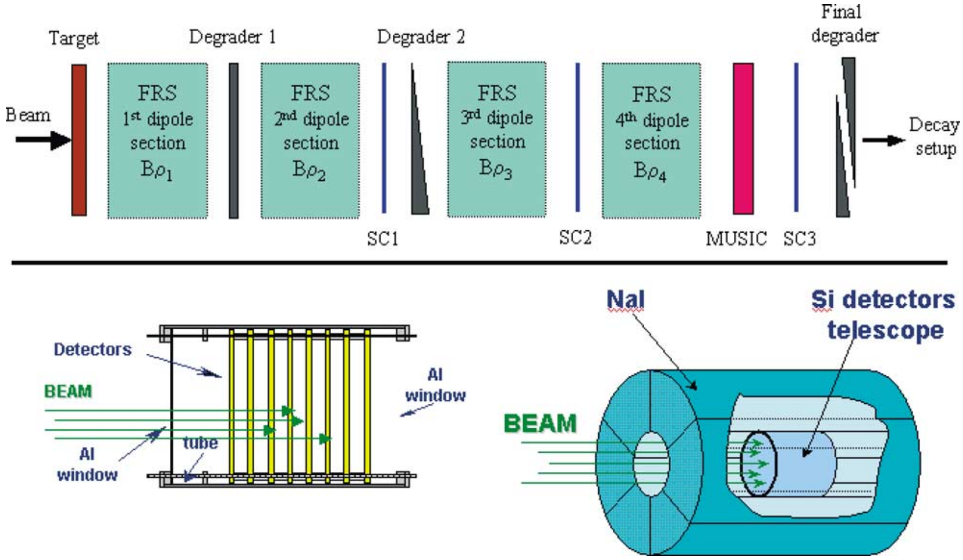

Fig. 2. Schematic representation of the experimental setup used at GSI. The top part of the figure shows the fragment separator FRS with the target, the four dipole sections, the energy-loss degraders between them as well as the tracking detector of the standard FRS detection system. The bottom part shows the setup mounted at the exit of the FRS consisting of a silicon detector telescope surrounded by a high-efficiency NaI crystal.

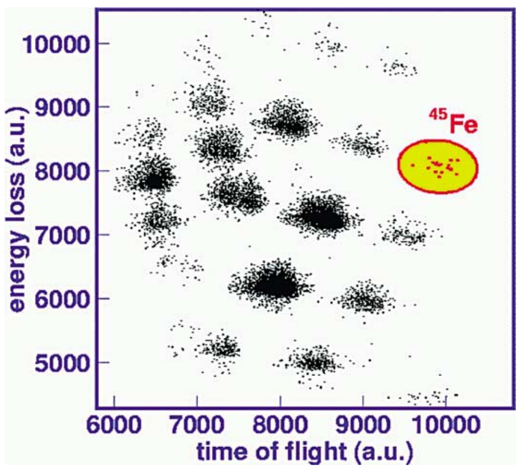

Fig. 3. Two-dimensional identification plot of energy loss versus time of flight for the GANIL data. The different isotopes transmitted to the end of the LISE3 separator are nicely identified and separated.

search for the decay of the 2p daughter nucleus, ^{43}Cr. The decay time determined for the daughter decay is consistent with the known half-life of ^{43}Cr [20]. In the same way, the daughter decay spectrum, despite its low statistics, is in agreement with the spectrum measured for the decay of ^{43}Cr previously [20].

The GSI data show an additional decay event at about 6 MeV. Three events with a comparable energy are also observed in the GANIL data. These events are explained by a small decay branch of ^{45}Fe via β-delayed one- or two-proton emission. According to both data sets, ^{45}Fe decays with a branching ratio of about 80% by two-proton radioactivity, whereas 20% of the decays show a βp or β2p picture. The missing seven decay events of the GANIL experiment are explained by data acquisition dead-time losses or losses due to implantation between two silicon strips where the charge collection is known to be bad.

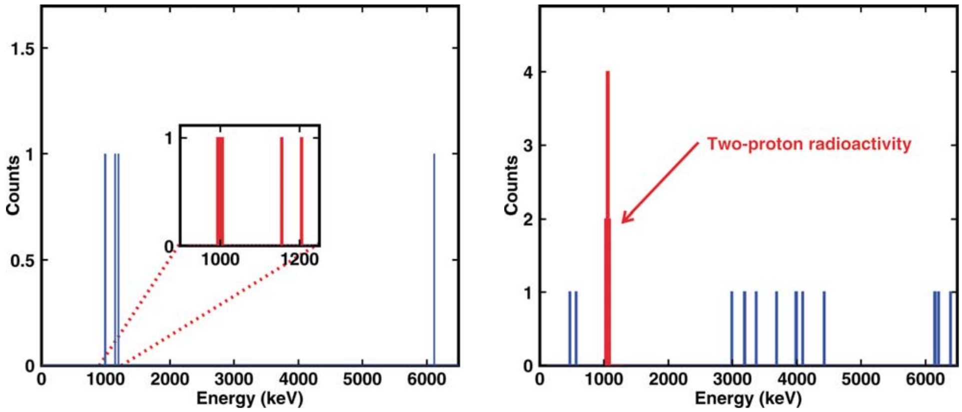

Fig. 4. Left part: Decay energy spectrum from the GSI experiment. The events shown are correlated with a ^{45}Fe implantation. Right part: Decay energy spectrum from the GANIL data. The decay events are correlated to ^{45}Fe implants in the same detector pixel for 100 ms.

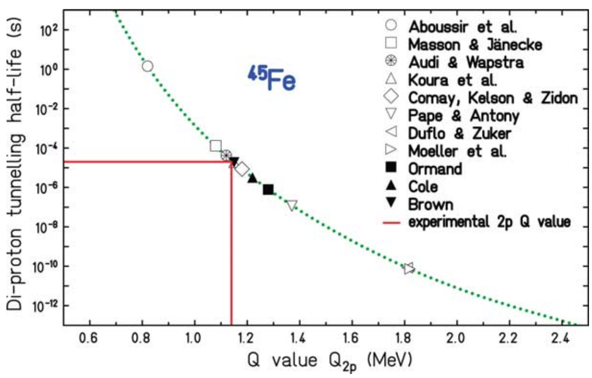

Fig. 5. Barrier-penetration half-lives are calculated as a function of the two-proton decay energy Q_{2p} within the di-proton model. The decay energy predictions of different mass models are also indicated. Without any spectroscopic factor, the barrier penetration calculations yield a half-life of 0.02 ms (see text). The experimental decay energy and the associated barrier penetration half-life is indicated by the red line.

3.2. Theoretical interpretation

Different model calculations are available to describe two-proton emission. The simplest of them is the di-proton model. This model deals with a structureless, but stable 'di-proton' particle which traverses the Coulomb barrier with a given energy by tunneling. A calculation with such a model, taking into account spectroscopic factors as calculated by Brown [21], yields half-lives about an order of magnitude too short compared to experiment when the experimental two-proton energy is used. In Fig. 5, we use such a model to calculate the barrier-penetration half-life for ^{45}Fe with different 2p decay energies.

More realistic values can be obtained with the R-matrix model of Barker [22,23] which takes the ^{2}He resonance potentially formed by the two protons explicitly into account. Depending on the optical model parameters used, half-lives of 8–24 ms are obtained for the experimental decay energy [24].

Probably the most realistic model available today is the three-body model of Grigorenko et al. [25] which explicitly treats the proton–proton and the proton–nucleus interactions. Different model assumptions allow us to calculate decay half-lives for the protons in different quantum orbitals. As shown in Fig. 6, nice agreement with experiment is obtained with the assumption that the protons be emitted from a pure p state. However, this is in contradiction with expectation based on mirror symmetry which rather predict the protons to be in a f state.

4. Future studies

The experiments recently performed on ^{45}Fe clearly show that it decays most probably by two-proton radioactivity. All commonly used mass models predict the decay to be simultaneous. However, beyond that nothing is known experimentally

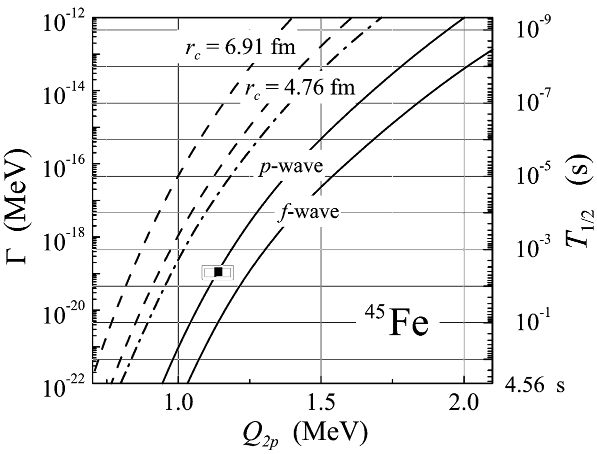

Fig. 6. Three-body calculations (full lines) are compared to di-proton calculations (dashed lines) with different asumptions. All di-proton calculations predict too large widths and thus too short half-lives. Assuming a pure p state of the protons, the three-body calculations are in agreement with the experimental data point (see text for a more detail discussion).

about the decay mechanism. To study the decay mechanism, measurements are needed which observe directly the two protons and measure their relative emission angle as well as their individual energies. Such measurements can not be performed with setups used up to now. The energy of the decay protons is too low and they stay captured in the silicon detector. To detect the protons as well as to measure their trajectories, a development of a time-projection chamber (TPC) was started which should allow us to visualize in 3D the tracks of the protons and therefore to get access to their energies and angles.

For a better understanding of two-proton radioactivity, other candidates have to be studied as well. ^{48}Ni and ^{54}Zn are other prime candidates to exhibit this type of decay. The first one was recently observed for the first time and production rates of one count per day can be achieved [26]. ^{54}Zn was not yet observed experimentally. However, extrapolations from the production rates of ^{55,56}Zn recently observed in a GANIL experiment [27] give estimated rates of about 10–15 counts per day, if the isotope is sufficiently stable to be observed ($T_{1/2} > 1$ μs). These two nuclei are expected to exhibit similar decay characteristics as ^{45}Fe.

For the lighter nuclei, ^{19}Mg seems to be a promising candidate. Although relatively broad, the intermediate one-proton daughter state lies high in energy, so that only the extreme tail of this state is in the energetically allowed region. This fact could sufficiently hinder the sequential decay and favor the simultaneous emission. However, as the half-life is expected to be rather short ($T_{1/2} < 10^{-15}$ s), complete kinematics measurements are necessary to study this decay (see, e.g., [10]).

5. Conclusion

For the first time, two-proton radioactivity has been observed in a rather clear way. For the moment this new radioactivity appears as a curiosity of nuclear structure. However, continuous developments in experiment and theory may help to develop it into a powerful nuclear structure tool which will allow us to determined nuclear masses far beyond the limits of stability, to study nuclear pairing, to determine the sequence of single-particle levels, to study deformation and the tunneling process for deformed nuclei and many more things. It is a common believe that a deeper understanding of nuclear structure can be achieved by studying nuclei far from stability. Two-proton radioactivity is certainly a promising route in this endeavor.

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References

[1] V.I. Goldansky, Nucl. Phys. 19 (1960) 482.

[2] J. Jänecke, Nucl. Phys. 27 (1965) 648.
[3] S. Hofmann, et al., Z. Phys. A 305 (1982) 111.
[4] O. Klepper, et al., Z. Phys. A 305 (1982) 125.
[5] P. Woods, C. Davids, Annu. Rev. Nucl. Part. Sci. 47 (1997) 541.
[6] K. Rykaczewski, Proc. ENAM conference, Hämeenlinna, Finland, 2001, Eur. Phys. A 15 (2002) 81.
[7] D.F. Geesaman, et al., Phys. Rev. C 15 (1977) 1835.
[8] O.V. Bochkarev, et al., Sov. J. Nucl. Phys. 55 (1992) 955.
[9] A. Azhari, R. Kryger, M. Thoennessen, Phys. Rev. C 58 (1998) 2568.
[10] R.A. Kryger, et al., Phys. Rev. Lett. 74 (1995) 860.
[11] H. Fynbo, et al., Nucl. Phys. A 677 (2000) 38.
[12] C. Bain, et al., Phys. Lett. 373B (1996) 35.
[13] M. Chromik, et al., Phys. Rev. C 66 (2002) 024313.
[14] J. Gomez del Campo, et al., Phys. Rev. Lett. 86 (2001) 43.
[15] B.A. Brown, Phys. Rev. C 43 (1991) R1513.
[16] W.E. Ormand, Phys. Rev. C 53 (1996) 214.
[17] B.J. Cole, Phys. Rev. C 54 (1996) 1240.
[18] J. Giovinazzo, et al., Phys. Rev. Lett. 89 (2002) 102501.
[19] M. Pfützner, et al., Eur. Phys. J. A 14 (2002) 279.
[20] J. Giovinazzo, et al., Eur. Phys. J. A 10 (2001) 73.
[21] B.A. Brown, Phys. Rev. C 44 (1991) 924.
[22] F. Barker, Phys. Rev. C 59 (1999) 535.
[23] F. Barker, Phys. Rev. C 63 (2001) 047303.
[24] B.A. Brown, Private communication.
[25] L. Grigorenko, et al., Phys. Rev. Lett. 85 (2000) 22.
[26] B. Blank, et al., Phys. Rev. Lett. 84 (2000) 1116.
[27] J. Giovinazzo, et al., Eur. Phys. J. A 11 (2001) 247.