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LETTER TO THE EDITOR

No magic numbers in neutral ${}^4\text{He}$ clusters[†]

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Abstract. By means of quantum Monte Carlo simulation we show that the binding energy of small neutral $({}^4\text{He})_N$ clusters is a smooth monotonic function of mass number N . Magic numbers observed in a recent experiment on charged He clusters are therefore *not* due to enhanced stability of neutral clusters at certain sizes.

In the recent literature there exists a number of investigations on clusters of rare gas atoms (Stephens and King 1983, van Deursen and Reuss 1975, Gspann 1981 (and references therein), Echt *et al* 1981, 1982, Farges 1975, Farges *et al* 1981, Pandharipande *et al* 1983). The most extensive experimental investigation on small medium-size quantum mechanical He clusters has been reported by Stephens and King (1983). As in the case of classical xenon clusters (Echt *et al* 1981, 1982) ‘magic numbers’ are those cluster sizes where the abundancy has a more or less pronounced maximum. In some cases this maximum is a rather dramatic peak. It has been argued (Echt *et al* 1981, 1982) that these ‘magic numbers’ were due to enhanced stability, i.e. stronger binding of the corresponding ‘magic’ neutral cluster before the ionisation process. In some cases the ‘magic numbers’ found experimentally agree with favourable geometrical packing of classical spheres. It is not clear, however, if the ‘magic numbers’ are a property of neutral clusters or if they are generated in the course of the ionisation process. It is the purpose of the present letter to demonstrate that no ‘magic numbers’ can be attributed to variations in relative stability of neutral ${}^4\text{He}_N$ clusters; they therefore must have been generated by the ionisation process.

In a previous Monte Carlo simulation (Pandharipande *et al* 1983) quantum mechanical ground states of the neutral ${}^4\text{He}$ clusters have been obtained by means of the Green’s function Monte Carlo (GFMC) method (Kalos *et al* 1981 and references therein), an approximation-free solution to the many-Boson Schrödinger equation. Unfortunately those cluster sizes where dramatic variations occur experimentally were not studied there.

Variations in the stability of clusters will appear most pronounced in the ground states. Taking thermodynamic averages will smear out any possibly existing structure. Moreover, the temperature of the condensed droplets is supposed to be extremely small so that the ground state will be the dominating component in the thermodynamic density matrix. It is therefore sufficient to concentrate on the ground state here.

In the same way as in Pandharipande *et al* (1983) we assume a system of N ${}^4\text{He}$ atoms interacting pairwise via the potential given by Aziz *et al* (1979). Executing the

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GFMC algorithm we conduct a random walk in $3N$ -dimensional configuration space guided by some suitable trial wavefunction. The only result considered in the present letter is the total ground state energy per atom as a function of number of atoms, $E(N)/N$. For every value of N to be considered an independent Monte Carlo simulation is performed. In table 1 we collect some previously unpublished results ($N = 5, 6, 7, 9, 10$) and several new calculations ($N = 11-15, 21-33$) together with two results from Pandharipande *et al* (1983) ($N = 8, 20$). The corresponding GFMC simulation for the infinite system (bulk ${}^4\text{He}$, in both the liquid and the solid phases) yields results in perfect agreement with experimental results at a variety of densities or pressures (Kalos *et al* 1981). The present model may therefore be assumed to be an accurate description of the physical system.

Table 1. Energy per atom as function of cluster size. The numbers in parentheses give the statistical error in the last digit.

N	$E/N(K)$	N	$E/N(K)$
5	-0.2514 (4)	20	-1.627 (3)
6	-0.3735 (5)	21	-1.690 (5)
7	-0.4965 (7)	22	-1.746 (5)
8	-0.6165 (6)	23	-1.811 (5)
9	-0.7274 (7)	24	-1.853 (4)
10	-0.830 (4)	29	-2.075 (6)
11	-0.933 (3)	30	-2.107 (7)
12	-1.029 (3)	31	-2.154 (7)
13	-1.123 (5)	32	-2.194 (13)
14	-1.208 (4)	33	-2.234 (10)
15	-1.293 (3)		

The data table 1 may accurately be fitted by the three-term liquid-drop formula (Pandharipande *et al* 1983)

$$E(N)/N = -7.36 + 21.4 N^{-1/3} - 15.7 N^{-2/3}(K). \quad (1)$$

The coefficients are slightly different from those given by Pandharipande *et al* (1983) since a different mass range limited to rather small clusters is fitted here.

In order to scrutinise the smoothness of $E(N)$ more closely we give in figure 1 the evaporation energy $E(N-1) - E(N)$ against N . This first derivative of $E(N)$ should show any irregularities significantly amplified. However, to within statistical error, the separation energy which gives a reliable indication of relative cluster stability does *not* exhibit any fluctuation. Even if the small, statistically insignificant deviations of the data points from the separation energy computed from equation (1) were taken seriously they appear much too small in order to explain the dramatic variations found in Stephens and King (1983). We conclude therefore that the production rate of neutral He clusters should be a smooth monotonic function of cluster size N .

The structure in the relevant abundancies of ${}^4\text{He}_N^+$ clusters found experimentally remains to be explained. Considering the difference in energy scale between neutral clusters where atoms interact by very weak Van-der-Waals forces (Kelvin scale) and electronic excitations as ionisation (eV scale, $1 \text{ eV} \approx 10^4 \text{ K}$) it is highly plausible that the electronic rearrangement taking place in forming the ion should be of much larger

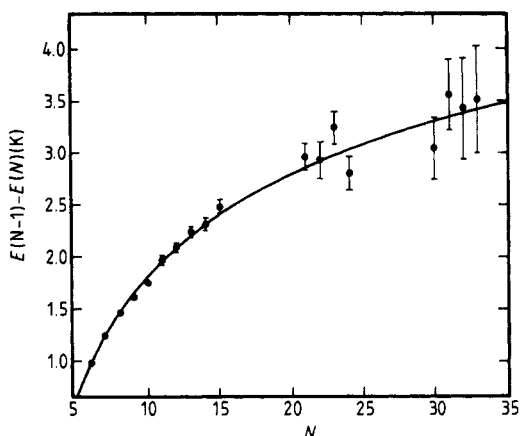


Figure 1. Evaporation energy $E(N-1) - E(N)$ as a function of cluster size. The error bars indicate one standard deviation statistical error. The full curve is computed from equation (1).

influence onto cluster (ion) stability than whatever were the properties of neutral clusters. In the case of argon atoms it has been suggested by Birkhofer *et al* (1984; Haberland, private communication) that the formation of a tightly bound dimer ion and subsequent fragmentation of the cluster is the mechanism responsible for observed mass spectra. In view of the present results and the strong similarities between rare gases it seems highly probable that the very same mechanism is also responsible for the observed ^4He mass spectra. Even in the case of fermion ^3He clusters where 'magic numbers' in neutral ground states may be expected due to exclusion-principle shell effects. These being on the Kelvin scale probably will be dominated by the electronic eV-scale rearrangement processes leading to qualitatively similar mass spectra for both species. Unfortunately Monte Carlo simulations for fermion clusters appear significantly more difficult than the simulations reported here. However, it can be demonstrated rather convincingly (Pieper 1983) that less than 20 ^3He atoms will not bind at all. The corresponding observed clusters in Stephens and King (1983) therefore are stabilised by the missing electron exclusively and do not even exist as a neutral cluster.

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