The structure of C4 as studied by the Coulomb explosion method

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COMMUNICATIONS

The structure of C_4 as studied by the Coulomb explosion method

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The structure of carbon clusters in general and C_4 in particular has been investigated recently both theoretically[1] and experimentally.[2,3] Although a number of high level calculations indicated that even clusters that contain only four atoms have a stable cyclic (rhombic) form, experimentally it was found that only when the number of atoms exceeds nine can a cyclic form be detected. In the present work the Coulomb explosion imaging (CEI) method was used to probe the structure of C_4. In the results presented here one observes for the first time a rhombic C_4.

Yang *et al.*[3] applied the laser sputtering source to study the photoelectron spectroscopy of negatively charged carbon clusters. They found that clusters with less than eleven carbon atoms exhibit an even–odd alternation in their electron affinity value, the odd having lower electron affinity. For clusters having more than ten carbon atoms it was observed that a new series can be drawn in which the reverse odd–even dependence of the electron affinity exists. In conjunction with theory, the two series were attributed to chain and ring structures, respectively. By varying the cluster formation conditions, the authors could observe, in the case of C_{11}^-, both the linear and the ring form.

Bernholdt *et al.*[1] in a theoretical investigation predicted, however, that a cyclic rhombic form exists and is isoenergetic with the linear form. It was further claimed that while the electron affinity of the linear C_4 is about 3.3 eV, the electron affinity of the rhombic form is about 2 eV.

Recently we developed a system which combines a laser photodetachment setup with the Coulomb explosion imaging method.[4] At the Pelletron accelerator at the Weizmann Institute, a chopped negatively charged carbon cluster beam was produced by sputtering with Cs. After mass selection the clusters were accelerated by a field of 12 MV. When reaching a field free region at the middle of the accelerator, the chopped ion beam interacted with a frequency doubled Nd:Yag laser (Quantell-535 nm, 25 mJ/cm^2, 20 ns long pulse). Electrons were photodetached and the neutral clusters drifted 30 m till they impinged upon a thin Formvar foil (about 30 Å thick). Following the foil, the charges of the four "Coulomb exploding" carbon ions were separated by a magnetic field. Their position and time of arrival were measured by a detector that combines a microchannel plate with hybridized anode and digitized video.[5] It is evident from the two dimensional projection of the "exploded" structure, that the neutral C_4 enters the foil as a ring. Hence we present here the analysis of two-dimensional projections in which four C^{3+} ions were selected. This information is sufficient for distinguishing between the linear and cyclic forms.

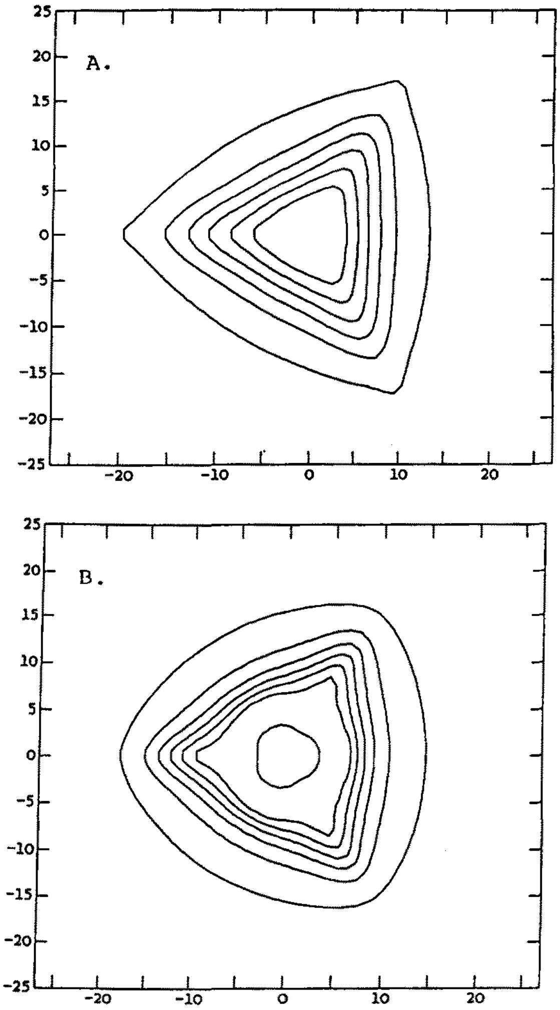
For the analysis we used special body coordinates[6] for the two-dimensional projection. These are

$$E_b = \frac{1}{\sqrt{12}}\{2(r_{12}+r_{34}) - (r_{13}+r_{14}+r_{23}+r_{24})\},$$
$$E_t = \frac{1}{2}(r_{13}-r_{14}+r_{24}-r_{23}),$$

where r_{ij} $(i,j = 1,...,4)$ are the carbon–carbon distances.

In Fig. 1(A) the data is presented using these two coordinates. For comparison in Figs. 1(B) and 1(C) simulations of the results for a rhombic and linear forms, respectively, are shown. Since the molecules are expected to contain internal energy,[7] some internal motion was invoked in the simulation. The data prove unambiguously that many of the neutrals measured have the rhombic form. The experiment was repeated with a laser energy of about 3.6 eV (third harmonic of a Nd:Yag laser), and identical results were obtained.

The apparent inconsistency between the results present-



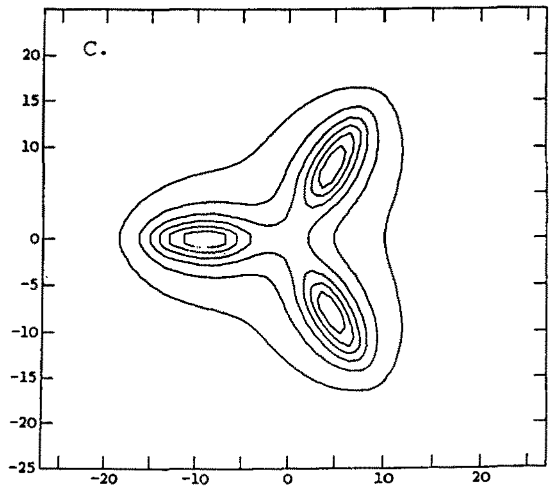


FIG. 1. The data presented in the body coordinates as discussed in the text (A), and simulation of the data assuming rhombus configuration (B) and linear shape (C).

ed here and those shown in Ref. 3 can be resolved as follows. Yang *et al.* applied the second harmonic of a Nd:Yag laser to sputter carbon, and an excimer laser for producing photoelectrons in the nozzle's throat. In both processes species with electron affinity below 2.34 (corresponding to 535 nm of the doubled Nd:Yag), can be easily neutralized. In our source on the other hand no discrimination against the low electron affinity species exists, and hence it is more probable to observe them. Thus, it is probable that cyclic C_n with $n < 10$ exist, although so far only the cyclic structure of C_4 has been established experimentally. Further work is on the way to obtain the structure of larger carbon clusters, and the full three-dimensional structure as a function of the laser frequency.

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