Production of multiply charged fullerene and carbon cluster beams by a 14.5 GHz ECR ion source

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Summary

I. Introduction (the goal of the research)

II. Experimental setup
   • The ECR ion source
   • The fullerene oven
   • Tuning of the ECRIS

III. Production of $C_n$ ion beams
   • Multiply charged fullerene beams
   • Carbon cluster beams

IV. Formation of $NC_n$ and $N@C_{60}$
   • $NC_n$ beams
   • $N@C_{60}$ beams
   • $N@C_{60}$ solids

V. Conclusion, plans
The goal of the research

- Endohedral fullerenes ($X@C_{60}$) generally
- if $N@C_{60}$: $N$ always in the center
- $N$ weakly interacting with cage
- electron-spin-resonance (ESR) lines are narrow

The $N$ atom is the most attractive probe to **measure/monitor**

- molecular distortions
- molecular motions (incl. in living tissues)
- internal fields
- exohedral chemical addition reactions
Traditional methods to produce N@C$_{60}$

By ion bombardment of C$_{60}$ on a **surface**

or

In electrical arc discharge tube (surface & **volume**)

- low concentration of N@C$_{60}$
- this should be increased
- production mechanism?
Production of $X@C_{60}$ ions in the ECRIS plasma

In the ECRIS trap:
- high confinement times
- ionization & excitation
- evaporation solved
- cooled surfaces exist
- extraction?
- beam?
The ATOMKI-ECRIS

- 14.5 GHz/2 kW
- Axial field: 1.2/0.4/1.0 T
- Radial field: 0.95 T
- Sextupole: Halbach-24
- Chamber: φ58x200
- Extraction: upto 30 KV (typical 10 KV)
- Gases: He, N, O, Ar, Kr, Xe (medium charges & currents)
- Solids (MIVOC & oven): C, C_n, C_{60}, Fe, Ni, Zn, Pb
The beamline system

14.5 GHz ATOMKI ECRIS
Modifications on the ECRIS

1. Low RF power (~1 W)
2. Iron plug removed
3. Biased disc removed
4. Al tubes (cooled collectors)
5. Extraction voltage max. 700 V (typical 10 KV)
The Fullerene Oven

1. Thermocouple
2, 3. Electrical connections & mechanical holders
4. Kanthal heater
5. Shielding
6. Crucible (SS)
7. BN insulator

- **Temp:** < 1000 C
- **Movement:** 50 mm axial
The tuning of the ECRIS

- “Unusual”
- 100…200 mg of 99.5% C\textsubscript{60} powder
- degassing, dehydration...
- Temperature: 400…600 C; typical: 450 C
- 1 mbar local fullerene pressure
- microwave power < 1W (optimum exists)
- oven position is critical
- gas flow effects on X@C\textsubscript{60} rate
Multiply charged fullerene beams

- Mostly $\text{C}_{60}^+$ and $\text{C}_{60}^{++}$
- $\text{N}_2^+$ and $\text{H}_2\text{O}^+$

Highest intensities:
- $\text{C}_{60}^+$ : 410 nA (700 V)
- $\text{C}_{60}^{++}$ : 245 nA (1300 V)
- $\text{C}_{60}^{+++}$: 105 nA (1900 V)
- $\text{C}_{60}^{4+}$ and $\text{C}_{60}^{5+}$ observed
The fine structure of the fullerene spectrum

- 3 main parts
- $C_{58}^+$, $C_{56}^+$, …, $C_{32}^+$
- $n=30...60$: only even mass numbers
- $C_{60}^+$: by electron impact ionization, then successive loss of $C_2$ molecules?
- $C^+$ and $C_2^+$ surprisingly low
Optimization for 1+, 2+, 3+
Carbon cluster beams

- Tuned for $\text{C}_{11}^+$
- $n=2\ldots15$: odd & even
- $U=1900$ V

- 10…100 nA cluster beams
- single and double charged
- the 4n+3 “magic” rule
Formation of NC\textsubscript{n} beams

- As the N flow increases, *new peaks* appear
- by mass: NC\textsubscript{n} (N=46…58, even)
- they are *most probably* N@C\textsubscript{n} beams

- Upper curve: \( p=9.5\times10^{-7} \) mbar
- Middle curve: \( p=2.7\times10^{-6} \) mbar
- Lower curve: \( p=5.2\times10^{-6} \) mbar

Slit: 2 mm
N@C_{60} beams

- Pure mass resolution of the bending magnet
- “ugly” spectrums
- but still clearly shows the existence of the 734=60*12+14 mass molecule
- N@C_{60} found in the beam!

C_{60} (720), NC_{59} (722), N@C_{60} (734), C_{62} (744), NC_{61} (746)
N@C$_{60}$ solids

- About 10 evaporated Al tubes were analysed
- time of evaporation: from 2-3 hours till 2 days
- ~ 80% of the evaporated material gained back
- thick black layer of C, C$_{60}$, clusters, N@C$_{60}$
- chemical treatment
- checking by electron-spin-resonance (ESR)
- **best sample**: 2 times higher N@C$_{60}$ concentration than with other methods
Conclusion

- High intensity single and multicharged fullerene (C_{60}) beams were produced by a 14.5 GHz ECRIS
- Large carbon cluster ion beams were also obtained
- The operation mode for the production of N-capsulated fullerene ions was found.
- The deposited macroscopic concentration was 2 times higher than with other methods.
Future plans

• higher concentration of N@C$_{60}$
• direct connections between the plasma (beam) parameters and the ESR results.
• information on the encapsulation process.
• other types of endohedral fullerene beams