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PRECISE IDENTIFICATION OF EXTRACTED ION BEAM SPECTRUM INITIALLY OBTAINED IN SYNTHESISING IRON-ENDOHEDRAL FULLERENES ON ECRIS

I. Owada†[1], T. Omori[1], W. Kubo[1], S. Harisaki[1], K. Sato[1], K. Tsuda[1],
M. Muramatsu[2], A.Kitagawa[2],Y. Yoshida[3], and Y. Kato[1],
[1]Graduate School of Engineering, Osaka University, Suita, Osaka, Japan
[2]National Institutes of Quantum and Radiological Science and Technology, Chiba, Chiba, Japan
[3]Faculty of Science and Engineering, Toyo University, Kawagoe, Saitama, Japan

Abstract

We have been trying to produce iron endohedral fullerenes in the electron cyclotron resonance ion source (ECRIS). They are expected to be used as quantum computing and contrast agents with high sensibility for MRI. We have conducted initial experiments on producing them only in the second stage of the ECRIS. We have been using iron vapor source by induction heating (IH) from the mirror end along to the geometrical axis, and C60 crucible from the side wall, respectively. We succeeded in realizing ECR plasma that fullerene and iron ions coexist on the single stage ECRIS, even by 1kV extraction voltage. We have performed a detailed identification of the ions of endohedral fullerene by the typical charge state distribution (CSD) using different criteria. By these experimental series, it is suggested that there is possibility of slight formation of iron endohedral fullerenes. We have confirmed the reproducibility of the spectrum of the ions corresponding to Fe@C60+ in several data sets. We will increase the amount of iron vapor and conduct additional experiment.

INTRODUCTION

We have produced the electron cyclotron resonance ion source (ECRIS) plasma to generate a synthetic ion beam in Osaka university [1,2]. The ECRIS has been applied in various fields such as heavy ion cancer therapy and space propulsion [3]. In the field of bionanomaterials, various kinds of atoms have been studied to be encapsulated in fullerenes. Fullerenes have a cage shape, and their scientific and physical properties are dependent on the atoms they contain [4]. Iron endohedral fullerenes are expected to be used as quantum computing or magnetic resonance imaging (MRI) contrast agents. In previous work, various synthesis technologies for producing endohedral fullerene have been developed, e.g. laser-vaporization, arc-discharge, and ion irradiation technique [5-8]. We have been trying to produce endohedral fullerene by collision reaction in the vapor phase in the ECR plasma. We have already succeeded in generating a plasma in which iron and fullerene ions coexist [9]. We have also improved the iron evaporation source for iron ion production. In this study, we have been developing an evaporation source using an induction heating method that allows for non-contact, high-purity material vapor to be extracted. The coexistence of iron and fullerene ions in the ECR plasma was observed in the experimental system. From the analysis of the mass/charge (m/q) value distribution and the result of detailed identification of the ions, the spectrum of the endohedral fullerene ion is confirmed around C_{60}^{+}. We have confirmed the reproducibility of the spectrum in several data sets. We will increase the amount of iron vapor and conduct additional experiment. In this paper, we report the schematic diagram of the constructed system and the results of the detailed identification of charge state distribution (CSD). We also show the results of preliminary experiments conducted without using the iron evaporator is also presented.

EXPERIMENTAL APPARATUS

A. ECR Ion Source

The top view of ECRIS in Osaka Univ. is shown in Fig. 1. The magnetic configuration is formed by the two mirror coils (Coil A, Coil B), additional coil (Coil C), and permanent octupole magnets. The current in coils A, B, and C are I_A, I_B, and I_C, respectively. The I_C is used for controlling the resonance region. We take the center of the vacuum vessel as the origin of the cartesian coordinates system. The IH iron evaporation source is located at z=-280mm and the fullerene crucible is at z=175mm.

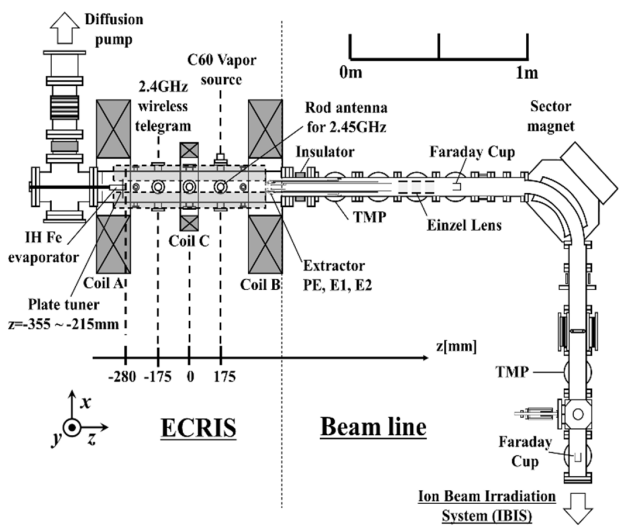

Figure 1: The top view of the ECRIS (Osaka Univ).

† issei0211@me.com

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The 2.45 GHz and 2.4 GHz micro wave are usually launched by magnetron (5~100W, z=175mm) and wireless telegram (0.1~0.9W, z=-175mm), respectively. A plate tuner installed on the z-axis. It can be moved at z=-355~-215mm and the introduction of microwaves can be optimized by moving it. It is effective when we manipulate low power micro wave. The extracted ion beam is separated by sector magnet for mass-charge ratio (m/q), detected by a faraday cup, and the CSD of the extracted ion current was obtained.

B. Induction Heating Iron Evaporator and Fullerene Vapor Source

The diagram of inducting heating iron evaporator [10] in Osaka Univ. is shown in Fig. 2. A radio frequency (RF) power supply is used as the power source, and the input power is usually about 600~800W. It is located at z=-280mm.

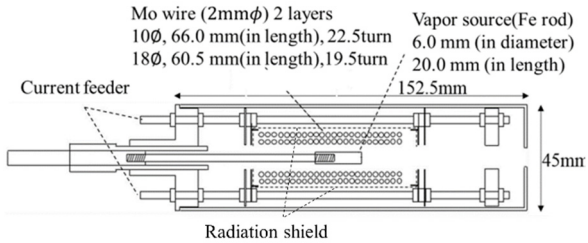

Figure 2: IH Fe evaporator.

The diagram of Fullerene crucible in Osaka University, is shown in Fig. 3. A direct current (DC) power supply is used as the power source, and the input power is usually about 10 W. It is located at z=175mm.

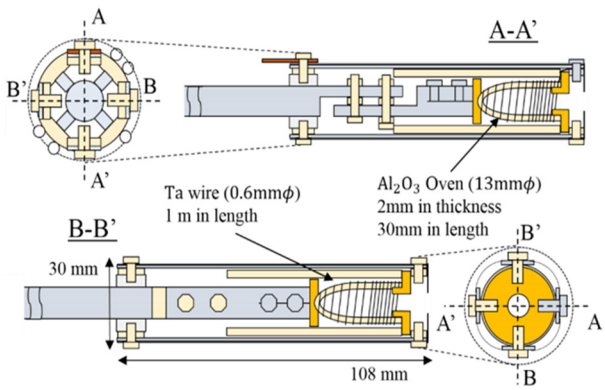

Figure 3: C_{60}^{+} crucible.

C. Experimental Procedure

We mainly use argon or xenon as the support gas. It is suitable for generating iron ions in high power micro wave and high extraction voltage. However, in generating fullerene ions, low power microwave and low extracted voltage is preferred. It is difficult to coexist both iron ion and fullerene ion. At first we preheat Fe evaporator and C_{60} crucible and set the extraction voltage at 10 kV to generate iron ions. The microwave about 10~20 W are launched from the magnetron. After confirming the generation of iron ions on the CSD, we decrease the extraction voltage to 2kV gradually. Likewise, the introduction of microwaves is switched to the wireless telegram and the power is reduced to less than 1W. It is necessary to generate fullerene ions while maintaining the existence of iron ions. Finally, we optimize the magnetic field conditions for production of endohedral fullerenes ions.

EXPERIMENTAL RESULTS AND DISCUSSIONS

A. Typical CSD of Fe^{+}, C_{60}^{q+}, and Their Compound Ions

Figure 4 (a) shows the typical CSD of fullerene ions. The horizontal axis represents the magnetic field and the vertical axis represents the amount of beam current. We can confirm the coexistence of C60+ ~ C603+ ions, Fe ions and residual Xe ions. Fig. 4(b), shows the right-hand side of the spectrum of C60+. From the identification of them, the spectrum to the right-hand side of the spectrum of C60+ are considered as Ar@C60+, Fe@C60+, Xe@C56+ and C70+. We used magnetron microwaves with 10W power, IH iron evaporation source was performed at 700 W input power. The extraction voltage is 2 kV. The support gas is argon with xenon remaining in it.

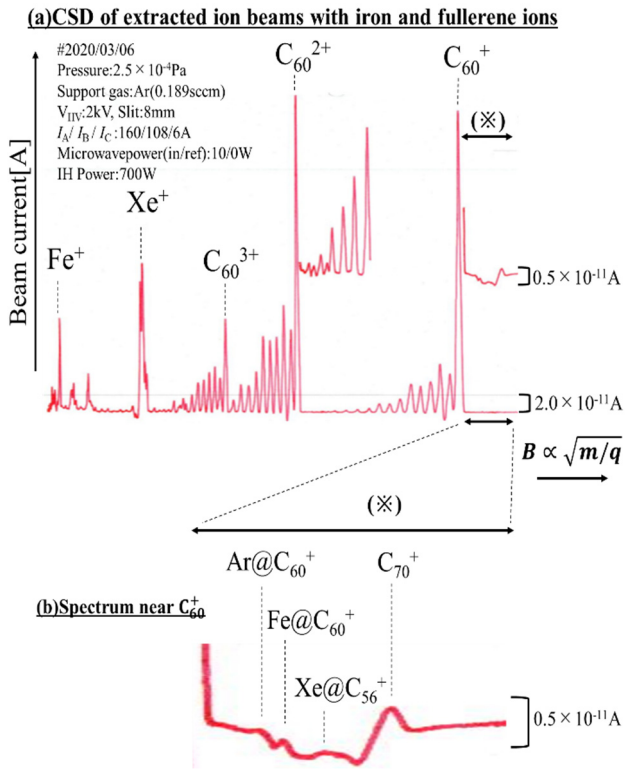

Figure 4 (a): CSD of extracted ion beams with iron and fullerene ions v.s.$\sqrt{m/q}$ (b): Spectrum near C_{60}^{+}.

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B. Error Evaluation

We performed relative error evaluation with different combinations of identification criteria. The relative error [%] is defined by the following Eq. (1).

$$\text{relative error}[\%] = \left|\frac{\alpha_1 - \alpha_2}{\alpha_1}\right| \times 100 \qquad (1)$$

α_1 : theoretical mass/charge (m/q) value
α_2 : mass/charge (m/q) value measured by CSD

The results of the evaluation are shown in Table 1. We conduct using a combination of various ions, including fullerene. We were able to identify the spectrum corresponding to the endohedral fullerene with an average relative error of 0.36%~1.35%. The spectrum corresponding to the iron endohedral fullerene were identified with an average relative error of 0.68%.

C. Typical CSD without Iron Ions

The results of the fullerene ion beam generation without the iron ions are shown in Fig. 5.

Table 1: Relative Error Evaluation in Each of the Identification Criteria. [%]

Identification creteria	ArC_{60}^+	FeC_{60}^+	XeC_{56}^+	C_{70}^+
C_{60}^+ & C_{60}^{2+}	0.10	0.50	0.34	0.24
C_{60}^+ & C_{60}^{3+}	0.05	0.44	2.08	0.19
C_{60}^+ & Fe^+	0.60	0.44	1.07	0.47
C_{60}^{2+} & Fe^+	1.47	1.32	1.92	0.52
Ave. error	0.56	0.68	1.35	0.36

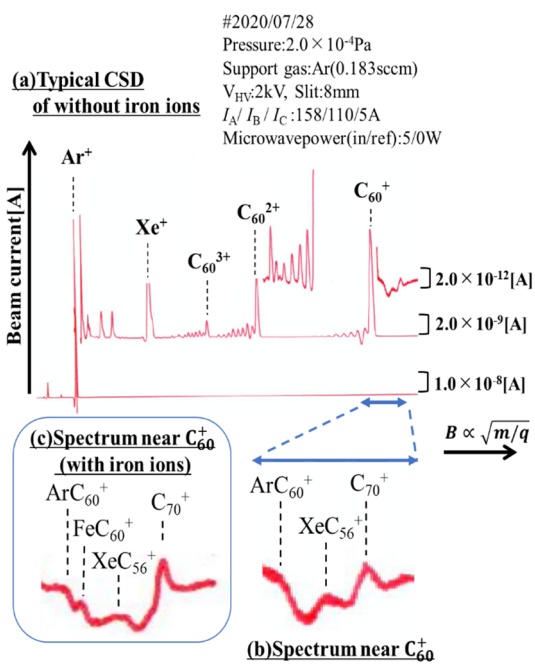

Figure 5: CSD of no iron ion introduction.

It is showed that Fig. 5(a) is the typical CSD of without iron ions and Fig. 5(b) focus on the right side of C60+. Fig. 5(c) is in the case of introducing the iron ions and focusing on the right side of C60+. The results of the similar identification with different criteria, we find the spectrum corresponding to ArC60+, XeC56+, and C70+, but no spectrum corresponding to FeC60+. This shows the reliability of the spectrum corresponding to Fe@C60+ when iron ions are introduced.

D. Conclusions and Future Planning

We have conducted experiments on the formation of iron endohedral fullerene using the inductively heated iron evaporation source. The spectrum corresponding to the iron endohedral fullerene were identified with an average error of 0.68% by using different combinations of the reference ions. We have confirmed the reproducibility of the spectrum of the ions corresponding to Fe@C60+ in several data sets. Additional experiments performed without the introduction of iron ions confirmed that the spectrum corresponding to $Fe@C_{60}^+$ were not seen in the CSD.

We will conduct time-of-flight and chemical analyses of actual products. We are also planning to improve the iron evaporater to produce high-purity iron ions and conduct additional experiment in a situation of increased iron vapor.

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