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Novel process of isotope separation of silicon by use of IR FEL

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Abstract

We found that silicon isotopes can be effectively enriched when mono-phenyl derivative of SiF_4 was used as the working substance. The chemical processes following the multi-photon dissociation of this molecule yield gaseous SiF_4 and solid powders of the by-product substances. By tuning the wavenumber of FEL beam at 961 cm^{-1} , the minority isotopes ^{29}Si and ^{30}Si have been successfully enriched in the residual gas of the working substance. Further, ^{28}Si could be enriched to above 98% when irradiated with FEL beam of 934 cm^{-1} .

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1. Use of Si_2F_6

Recently, a strong interest has appeared in the silicon isotopes in relation with the development of new electronic devices [1,2]. The isotope separation technology is considered to be one of the most interesting topics of future application of high-power infrared FEL. Thus it is of great interest to investigate strategically an effective process of isotope separation by strong infrared beam. In fact, there have been already several such studies [3–5]. Hitherto, Si_2F_6 has been used as the working substance in most studies of laser isotope separation of silicon. It was demonstrated that the enrichment

of ^{28}Si could be achieved by using TEA- CO_2 laser [6] and FEL [7] as the stimulating light source. However, the minority isotopes ^{29}Si and ^{30}Si could not be enriched above 50% by using Si_2F_6 as the working substance, because Si_2F_6 contains a couple of silicon atoms in a symmetrical position.

We started the investigation of new working substances suitable for the isotope separation of silicon at IR FEL Research Centre of the Tokyo University of Science (FEL-SUT). In this paper, we will report the experimental results obtained by the use of phenyltrifluorosilane (Ph-SiF_3) as working substance.

2. Experiments

Experiments were carried out by use of MIR-FEL of the IR FEL Research Center. MIR-FEL

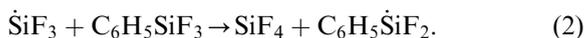
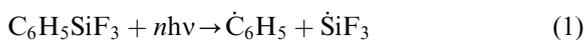
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covers the wavelength range 4–16 μm . In this experiment, we used FEL light of the wavelength range 9–11 μm , where the maximum macro-pulse power was 17 mJ when measured in front of the reaction cell. The micropulse width was 2 ps and the repetition rate was 2856 MHz. The macropulse width was 2 μs and the repetition was 5 Hz. The vapour of the working substance Ph-SiF₃ was filled in a cylindrical stainless-steel cell with ZnSe windows at the both ends. The length of the cell was 130 mm and the diameter was 40 mm. The collimated FEL light with the diameter of about 2 mm was focused at the centre of the cell by use of a ZnSe lens with the focal length of 25 cm. The initial pressure of Ph-SiF₃ was set at 2 Torr. A FT-IR spectrometer and a mass spectrometer were used for the analysis of the working gases. Ph-SiF₃ is a stable compound and no change could be identified by the FT-IR spectrometer after keeping its vapour in the cell for 12 h.

3. Results and discussions

The FT-IR spectra observed before and after the irradiation with FEL beam at $\nu_{\text{FEL}} = 971 \text{ cm}^{-1}$ are shown in Fig. 1, where the spectrum of Ph-SiF₃ before the irradiation is shown with solid. Ph-SiF₃ gives three vibration bands at 863, 971, and 1146 cm^{-1} , respectively. The Ph-SiF₃ band at 971 cm^{-1} is very strong and expected to have a large isotope shift, the isotope shift being predicted to be 8 cm^{-1} for Ph-²⁹SiF₃ and 16 cm^{-1} for Ph-³⁰SiF₃, respectively, from Ph-²⁸SiF₃ by ab initio molecular orbital calculations. When irradiated with 971 cm^{-1} FEL beam, the intensities of the bands due to Ph-SiF₃ decreased with time and a new band corresponding to SiF₄ appeared at 1031 cm^{-1} . These results seem to indicate the following main reaction pathway:



We could also see the formation of yellow solid powders in the cell. These powders are considered to be produced by the secondary reactions of the produced radicals.

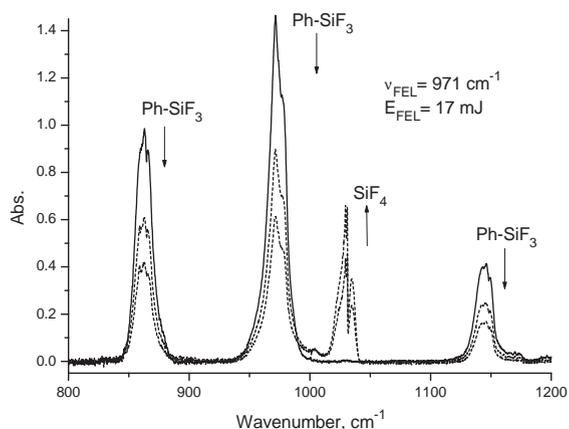


Fig. 1. The change of the FT-IR Spectra during the irradiation with FEL light of $\nu_{\text{FEL}} = 971 \text{ cm}^{-1}$; the spectrum being observed at every 1-h.

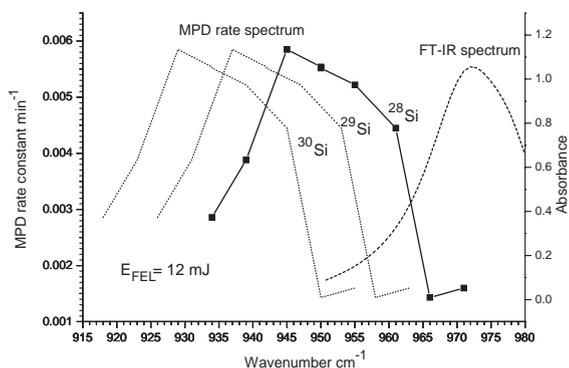


Fig. 2. The wavelength dependence of the MPD rate constant (MPD spectrum). The solid squares are observed data. Dotted lines are the expected MPD spectra for ²⁹Si and ³⁰Si components (refer text). The broken line is spectrum the concerned IR band of Ph-SiF₃.

Fig. 2 shows the wavelength dependence of the multi-photon dissociation (MPD) rate constant. The broken line shows the FT-IR spectrum corresponding to the anti-symmetric stretching mode of Ph-SiF₃. The solid squares are the observed value of MPD rate constant. We can consider that the solid line connecting those observed points gives the wavelength dependence of the MPD rate constant (MPD spectrum) of Ph-²⁸SiF₃ because the natural abundance of silicon

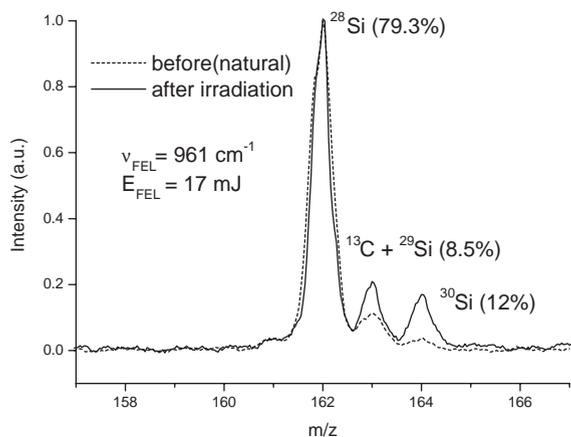


Fig. 3. The mass spectra of Ph-SiF₃ before (dotted line) and after (solid line) the irradiation with FEL beam of $\nu_{\text{FEL}} = 961 \text{ cm}^{-1}$; the irradiation time being 80 min. The spectra have been normalized by the peak height of Ph-²⁸SiF₃.

isotopes is ²⁸Si:²⁹Si:³⁰Si = 92.23:4.67:3.10. In Fig. 2, we have drawn the MPD spectrum expected for Ph-²⁹SiF₃ and Ph-³⁰SiF₃ (dotted lines) by shifting the MPD spectrum of Ph-²⁸SiF₃ by 8 and 16 cm⁻¹ toward lower frequency, respectively.

The mass spectra before and after 380 min irradiation with the FEL light of $\nu_{\text{FEL}} = 961 \text{ cm}^{-1}$ are shown in Fig. 3. The mass numbers $m/z = 162, 163, 164$ correspond to the Ph-²⁸SiF₃, Ph-²⁹SiF₃, and Ph-³⁰SiF₃, respectively. It should be noted that the relative intensity of the peak at $m/z = 163$ in the mass spectrum before irradiation is about 6% higher than the one expected from the natural abundance because the carbon atoms in the phenyl ring contain about 1% of ¹³C. Taking this into account, it can be seen that the minority isotopes ²⁹Si and ³⁰Si were enriched 8.5% and 12%, respectively, in the residual gaseous components. Although the data are not shown here, ²⁸Si was enriched to more than 98% in the residual gas when the wavenumber of the irradiating FEL beam was $\nu_{\text{FEL}} = 934 \text{ cm}^{-1}$.

Fig. 4 shows the irradiating energy (E_{FEL}) dependence of the MPD rate constant at $\nu_{\text{FEL}} = 961 \text{ cm}^{-1}$. From this figure, the MPD rate constant was found to be proportional to $E_{\text{FEL}}^{3.3}$. The irradiating FEL energy dependence of the isotope selectivity is being investigated at present.

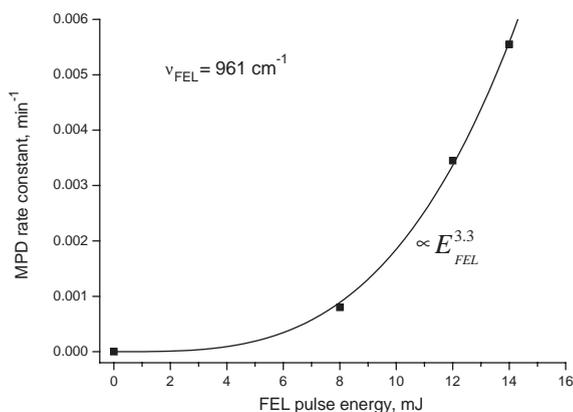


Fig. 4. The irradiating energy (E_{FEL}) dependence of the MPD rate constant at $\nu_{\text{FEL}} = 961 \text{ cm}^{-1}$.

4. Conclusion

The isotope separation of silicon by the use of IR FEL has been successfully done when the Ph-SiF₃ was used as the working substance. By tuning the FEL wavenumber at $\nu_{\text{FEL}} = 961 \text{ cm}^{-1}$, the minority isotopes ²⁹Si and ³⁰Si were enriched to 8.5% and 12%, respectively, in the residual gas. On the other hand, ²⁸Si was enriched to above 98% when irradiated with FEL beam of $\nu_{\text{FEL}} = 934 \text{ cm}^{-1}$. Although further experiments are necessary, the present finding will open the possibility to establish a new and effective process of isotope separation of silicon.

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