SUPPLEMENTARY MATERIALS

Slodzian *et al.* (2004) reported the Quasi-Simultaneous Arrival (QSA) effect on mass fractionation of sulfur and silicon isotopes using a NanoSIMS. When the average number K of secondary ions ejected per primary ion is large, the probability of more than one secondary ion per primary impact is not negligible. Those ions might arrive simultaneously on the EM detector. The detector might then recognize them as a single ion, even though a few ions actually enter the detector. Consequently, the QSA effect might reduce the ion counting efficiency for the major isotope. A similar QSA effect was observed by Nishizawa *et al.* (2010) when they measured sulfur isotopes of pyrite. In our case, observed Mg isotopes might vary with K according to the following equation.

Table S1. Chemical compositions of partly altered plagioclase grains

Oxide	Na_2O wt (%)	Al_2O_3 wt (%)	SiO_2 wt (%)	CaO wt (%)	FeO wt (%)
Grain 1					
#1	2.85	27.5	53.0	15.3	1.34
#2	3.22	29.7	52.5	13.4	1.12
#3	3.26	29.0	49.8	13.1	4.82
#4	1.75	29.9	47.2	19.8	1.33
#5	2.86	23.5	58.4	13.7	1.54
average	2.79	27.9	52.2	15.1	2.03
Grain 2					
#1	3.61	30.9	48.8	14.3	2.37
#2	3.96	31.2	49.7	13.6	1.54
#3	2.70	25.9	43.0	10.2	18.3
#4	3.13	32.7	48.0	15.2	0.96
#5	4.88	30.5	50.6	13.6	0.49
#6	4.36	30.8	49.4	12.9	2.62
#7	3.81	29.2	50.5	14.0	2.39
#8	3.48	26.2	45.8	10.9	13.7
average	3.74	29.7	48.2	13.1	5.29
Grain 3					
#1	3.79	29.6	50.4	13.6	2.63
#2	3.14	26.7	46.8	13.3	10.0
#3	2.90	24.4	41.1	9.5	22.1
#4	2.72	26.4	40.9	11.1	18.8
#5	4.45	32.6	48.5	13.9	0.61
#6	3.78	31.4	50.5	14.2	0.13
#7	2.94	29.0	51.7	15.6	0.82
average	3.39	28.6	47.1	13.0	7.89
Grain 4					
#1	2.81	31.0	51.6	12.9	1.70
#2	3.20	30.2	51.6	13.4	1.57
#3	3.30	30.6	51.0	13.5	1.57
#4	2.92	28.8	50.4	12.0	5.89
average	3.06	30.2	51.2	13.0	2.68

$$(^{25}Mg/^{24}Mg)_{obs} = (^{25}Mg/^{24}Mg)_{cor} \times (1+1/2K).$$

In that equation, $(^{25}Mg/^{24}Mg)_{obs}$ and $(^{25}Mg/^{24}Mg)_{cor}$ respectively denote the observed and corrected $^{25}Mg/^{24}Mg$ ratios. A similar equation is applicable to the $^{26}Mg/^{24}Mg$ ratios. Slodzian *et al.* (2004) recommended that corrected K (K_{cor}) should be considered when the actual number of ions reaching the EM detector is not known.

$$K_{cor} = K_{exp}/(1 - K_{exp} \times 0.69).$$

Therein, K_{exp} is the experimental ratio of secondary intensity over primary intensity. In this work, the primary beam is 1 pA and the secondary beam is 5×10^4 cps. Then the K_{cor} value is calculated as 8.7×10^{-3}, which engenders the QSA correction of 5.5‰ decrease of both δ^{25}Mg and δ^{26}Mg values. The corrected $\Delta^{26}Mg_{ex}$ values vary from –1.8‰ to 2.0‰ and are consistent with zero excess within the experimental error margin (see Fig. S2 where most corrected data are located on the slope 1/2 line within experimental error margin).

Two methods exist for minor adjustment of the analyzed mass by a NanoSIMS: to change the magnetic

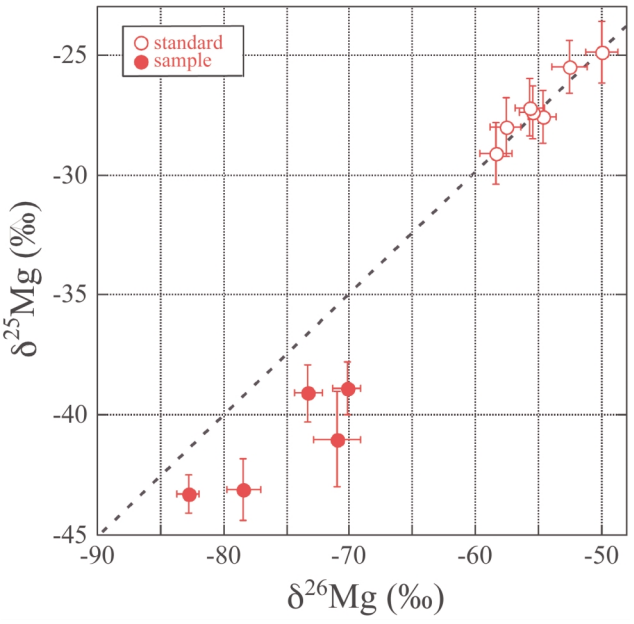

Fig. S1. Correlation diagram between the δ^{25}Mg and δ^{26}Mg values of standard plagioclase (Miyake-jima) and plagioclase grain #1 in the Efremovka chondrule. Errors are two sigma values. A dotted line shows a mass fractionation line with a slope of 1/2. Although the difference between δ^{25}Mg of the sample and standard is great, excess ^{26}Mg observed in the sample might be attributable to extinct ^{26}Al because a positive correlation exists between the Al/Mg ratio and the excess ^{26}Mg (see Fig. 2).

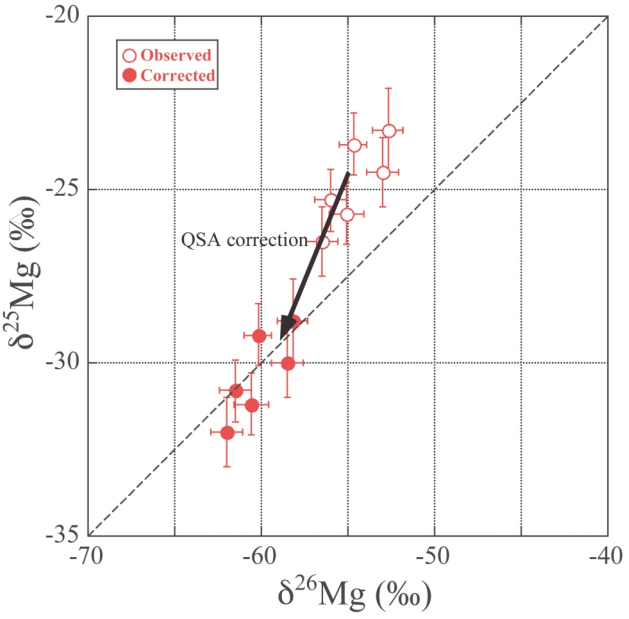

Fig. S2. Correlation diagram between the $\delta^{25}Mg$ and $\delta^{26}Mg$ values of standard olivine (San Carlos). Errors are two sigma values. A dotted line shows a mass fractionation line with a slope 1/2. Open and solid circles represent observed values and corrected values for the QSA effect.

field and/or the deflector plate voltage located in front of an EM detector. The former changes all mass quantities of multi-collector at once, whereas the latter adjusts the mass number of the individual detector. Because of the variable environmental conditions such as the room temperature and power supply fluctuation, the magnetic field might change slightly during the day. We can adjust the magnetic field as well as the deflector voltage slightly for the centering of a flat topped peak at the requested mass number. Then, the secondary ion trajectory might change and ions probably hit different positions on the first Cu–Be plate of the EM detector. This might cause a slight difference in the amplification factor. In the multi-collector system, the voltage adjustment of each detector is the same direction to compensate the magnetic field variation. Then the target position of the first plate might change the same way, which might produce systematic amplification alteration. That is not a random method because the EM detector might have similar characteristic of the first plate with a position-related efficiency. A large instrumental mass discrimination observed in Miyake-jima plagioclase (δ^{25}Mg = –24.9‰ – –29.1‰) might be attributable to the trajectory effect and amplification. Although the discrimination is large, data are distributed on the mass fractionation line of slope 1/2 in three isotope plots (Fig. S1). The relation between the trajectory effect and the mass dependent fractionation cannot be determined quantitatively. Moreover it is difficult to explain the discrimination with a slope 1/2 when the fractionation is putatively a random process.

References

Nishizawa, M., Maruyama, S., Urabe, T., Takahata, N. and Sano, Y. (2010) Micro-scale (1.5mm) sulphur isotope analysis of contemporary and early Archean pyrite. *Rapid Commun. Mass Spectrom.* **24**, 1397–1404.

Slodzian, G., Hillion, F., Stadermann, F. J. and Zinner, E. (2004) QSA influences on isotopic ratio measurements. *Appl. Surface Sci.* **231–232**, 874–877.