Fe–rich olivine from an andesite dike in Miocene Shitara volcanic rocks, central Japan: a revised relationship between Mg/Fe ratio and Raman spectrum in olivine

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Olivine is a major mineral in ultramafic and mafic rocks. Due to the higher Mg/Fe partition coefficient of olivine than the coexisting phases, the occurrences of Mg–poorer olivine grains, especially with Mg# [= Mg/(Mg + Fe2+)] = 0.2–0.6, are rarely reported, and thus, their spectroscopic data are limited. Fe–rich olivine phenocrysts showing compositional zoning with Mg# = ~0.5 (core) and ~0.3 (rim) and microphenocrysts with Mg# = ~0.4 (core) and ~0.2 (rim) occur in basaltic trachyandesite of Miocene Shitara volcanic rocks in central Japan. These olivine grains were investigated by Raman spectroscopy. Combining our data with the published values, we have revised the equation for Mg#–Raman spectrum relationship proposed by Mouri and Enami (2008) as follows: Mg# = 0.005446ω − 0.20259ω + 1.8442 (correlation coefficient r² = 0.984), where ω is the difference between the doublet peak positions (κ₂ − κ₁).

Keywords: Fe–rich olivine, Raman spectroscopy, Mg#, Solid solution, Shitara volcanic rocks

INTRODUCTION

Laser Raman spectroscopy is widely used in earth and planetary sciences because of its potential to analyze several kinds of information such as the temperature and pressure conditions of minerals at the time of crystallization, their chemical composition, and crystal structure (McMillan and Hofmeister, 1988; Dubessy et al., 2012; Bodnar and Frezzotti, 2020; Korsakov et al., 2020; Nasdala and Schmidt, 2020). Thus, Raman spectroscopy has been applied extensively to identify minute phases in terrestrial and extraterrestrial materials and to the non-destructive analysis of their chemical compositions.

Olivine grains occur widely in Earth’s crust and upper mantle, and in meteorites. They have a characteristic Mg# [= Mg/(Mg + Fe2+)], which is closely related to the host rocks and their modes of occurrence. Therefore, the composition of olivine grains retains important information on the petrogenesis of their host rocks. Guyot et al. (1986) demonstrated that the linear shift in the vibration peaks of SiO₄ tetrahedra, induced by Fe–Mg substitution, is useful for estimating the chemical composition of olivine. More specifically, the doublet near 820 and 850 cm⁻¹ has been used to identify the composition of olivine grains. Wang et al. (2004) and Kuebler et al. (2006) provided equations to describe the quantitative relationship between Mg# [= Mg/(Mg + Fe²⁺)] and doublet wavenumbers for the full range of forsterite (Mg₂SiO₄)–fayalite (Fe₂SiO₄) solid solution. Mouri and Enami (2008) and Ishibashi et al. (2012), respectively, proposed new parameters for the difference between the doublet wavenumbers (represented by ω) and a set of ΔMg# (= Mg#ref − Mg#) and Δν(= νref − ν) values with which the Mg# can be
estimated, where \( \nu \) is the wavenumber of the olivine spectrum for each Mg\# and \( \nu_{\text{ref}} \) is the wavenumber of an olivine grain with a reference value of Mg\#ref. Breitenfeld et al. (2018) analyzed 181 spectrum data using the partial least squares model and concluded that the peak near 850 cm\(^{-1} \) is best for estimating the Mg\# value.

Although the published spectroscopic data cover the forsterite–fayalite series, most of the data are reported for Mg\# > 0.6 or Fe-rich series with Mg\# < 0.2; therefore, data for olivine series with Mg\# = 0.25–0.5 are rare (Fig. 1). In this study, we conducted Raman spectroscopic analysis of Fe-rich olivine grains (Mg\# = 0.2–0.5) in Miocene volcanic rocks from Shitara province, central Japan. These data are useful for revising the compositional dependence of the Raman spectra on the forsterite–fayalite solid solution series, and based on our data, we propose a revised \( \omega - \text{Mg\#} \) diagram modified after Mouri and Enami (2008).

**SAMPLE DESCRIPTION**

The olivine–bearing sample studied was collected from the Toei-cho area where the Neogene Hokusetu Group and the Shitara igneous complex are widely distributed (e.g., Takada, 1978). Miocene andesitic dikes and sills, a member of the Shitara igneous complex, have intruded extensively into the sedimentary sequences of the Hokusetu Group (Geshi, 2003). The rock sample is dark greenish colored basaltic trachyandesite with a whole-rock composition of 54.4 wt% SiO\(_2\), 6.2 wt% Na\(_2\)O + K\(_2\)O, and FeO*/MgO ratio of 5.4 (Table 1), which is similar to the composition of other Shitara volcanic rocks such as alkali basalt–trachyandesite described by Geshi (2000, 2003).

Figure 1. (a) Cumulative diagram and (b) histogram of Mg\# of natural and synthetic olivine grains with the Raman spectroscopic data reported in the literature. The data are from Breitenfeld et al. (2018), Chopelas (1991), Guyot et al. (1986), Ishibashi et al. (2012), Kuebler et al. (2006), Mouri and Enami (2008), Weber et al. (2014), Yasuzuka et al. (2009), and RRUFF (https://rruff.info: 02 Nov., 2020).

Table 1. Whole-rock composition of an olivine-bearing andesite from the Miocene Shitara volcanic rocks in central Japan

|          | (wt\%) | (ppm) |
|----------|--------|-------|
| SiO\(_2\) | 54.35  | V     |
| TiO\(_2\) | 1.90   | Cr    |
| Al\(_2\)O\(_3\) | 14.62 | Co    |
| FeO*     | 13.10  | Ni    |
| MnO      | 0.23   | Cu    |
| MgO      | 2.17   | Zn    |
| CaO      | 5.89   | Pb    |
| Na\(_2\)O | 4.47  | Sr    |
| K\(_2\)O | 1.73   | Y     |
| P\(_2\)O\(_5\) | 0.87  | Zr    |
| Total    | 99.33  | Nb    |
|          |        | Ba    |
|          |        | Pb    |
|          |        | Th    |

The analytical procedures used in this study were identical to those in Wakasugi et al. (2020).
* Total iron as FeO.

The studied olivine grains occur as euhedral/subhedral phenocrysts (1–1.5 mm in size) and subhedral microphenocrysts (10–100 \( \mu \)m in size) in groundmass. The phenocrysts show clear compositional zoning with Mg\# = ~ 0.5 in the core and Mg\# = ~ 0.3 in the rim; the microphenocrysts are slightly Mg–poorer (Mg\# = 0.4–0.2) than the phenocrysts. The abundance of MnO, NiO, and CaO are 0.7–1.3, <0.1, and 0.2–0.4 wt%, respectively. In addition to olivine, the studied sample contains euhedral/subhedral phenocrysts of augite (Mg\# = 0.64–0.40), plagioclase (An\(_{31-39}\)), and a holocrystalline groundmass consisting of fine-grained plagioclase (An\(_{12-28}\)), augite (Mg\# = 0.60–0.50), ilmenite, and magnetite. The olivine phenocrysts are partly replaced by sheet silicate, probably stilpnomelane. Augite and plagioclase phenocrysts occasionally form aggregates with interfinger texture. These petrographical characteristics are similar to the P1-type Shitara volcanic rocks studied by Geshi (2000).
RESULTS AND DISCUSSION

Quantitative analysis of olivine was performed using an electron probe microanalyzer [EPMA, JEOL JXA–8900R (WDS)] at the Petrology Laboratory of Nagoya University. The accelerating voltage, specimen current, and beam diameter were maintained at 15 kV, 12 nA, and 2–3 µm, respectively. Matrix corrections were performed using the α-factor table of Kato (2005).

Raman spectra were obtained using a laser Raman micro–spectrophotometer (Nicolet Almega XR, Thermo Scientific) with a grating of 2400 lines/mm at the Petrology Laboratory of Nagoya University. The instrument was equipped with a 532 nm Nd–YAG laser, Peltier-cooled charge-coupled device (CCD) detector with 256 × 1024 pixels (Andro Technology), and an automated confocal microscope (Olympus BX51). The 100× objective lens from Olympus Mplan–BD has a numerical aperture of 0.9. The wavenumber resolution is about 1 cm$^{-1}$. The analytical reproducibility determined using the 520 cm$^{-1}$ band of Si wafer is ±0.3 cm$^{-1}$ (1σ) (Enami et al., 2007).

The Raman spectrum was separated using PeakFit v. 4.12 (SeaSolve) via a Voigt area function by subtracting background employing a linear baseline in the spectral range of 600–1200 cm$^{-1}$. Although Mouri and Enami (2008) denoted the doublets near 820 and 850 cm$^{-1}$ as κ2 and κ1, respectively, considering the converse notations in the literature, we referred these peaks as κ1 and κ2, respectively. In accordance with this change, the parameter ω (= κ2 – κ1), proposed by Mouri and Enami (2008), is now defined as ω = κ1 – κ2.

Table 2 lists the sets of Raman spectra and chemical compositions for the newly analyzed Fe-rich olivine grains. The Raman spectra data plotted in Figures 2–4 of Mouri and Enami (2008) are listed in Supplementary Table S1 (available online from https://doi.org/10.2465/jmps.201204). The abbreviations for minerals and end-members follow Whitney and Evans (2010).

### Table 2. Doublet peak data and chemical compositions of olivine

| Anal. No. | Texture | $\kappa_1$ (cm$^{-1}$) | $\kappa_2$ (cm$^{-1}$) | $\omega$ (cm$^{-1}$) | Chemical composition (apfu for total divalent cations = 2) |
|-----------|---------|------------------------|------------------------|---------------------|---------------------------------------------------------|
| 18        | P (core)| 817.1 (0.0)            | 845.5 (0.1)            | 28.4 (0.1)          | Fe$^2+$ = 0.021, Mn = 0.947, Mg = 0.009, Ca = 0.001, Ni = 0.481 |
| 32        | P (rim) | 815.7 (0.2)            | 842.0 (0.4)            | 26.3 (0.1)          | Fe$^2+$ = 0.033, Mn = 0.592, Mg = 0.011, Ca = 0.002, Ni = 0.303 |
| 30        | G       | 815.9 (0.2)            | 844.2 (0.1)            | 28.3 (0.2)          | Fe$^2+$ = 0.027, Mn = 0.764, Mg = 0.012, Ca = 0.001, Ni = 0.390 |
| 34        | G       | 815.4 (0.2)            | 841.9 (0.1)            | 26.5 (0.2)          | Fe$^2+$ = 0.035, Mn = 0.462, Mg = 0.009, Ca = 0.002, Ni = 0.236 |
| 22        | G       | 815.2 (0.0)            | 841.2 (0.1)            | 26.0 (0.1)          | Fe$^2+$ = 0.033, Mn = 0.429, Mg = 0.009, Ca = 0.002, Ni = 0.219 |

Note: $\omega$ = $\kappa_2$ – $\kappa_1$; apfu, atom per formula unit; Mg# = Mg/(Mg + Fe$^{2+}$); P, phenocryst; G, microphenocryst in groundmass.

* Total iron as Fe$^{2+}$.

The number in parentheses indicates the standard deviation (1σ).

Figure 2. Relationship between the positions of the strong doublet peaks ($\kappa_1$ and $\kappa_2$) and the Mg# of forsterite–fayalite series. * Linear 1 indicate the regression line for 22 data points [E (LT)]; and Linear 2 indicate the regression line for a set of two sample groups with high Mg# (>0.25, E (LH): 17 data points] and low Mg# (≤0.25, E (LL): 5 data points], respectively. ** Quadratic indicates regression curve for 22 data points [E (Q)]. In Figure 3 of Mouri and Enami (2008), the $\kappa_1$ and $\kappa_2$ data for 99–01, 99–02, and 7232915_G02 were plotted in the incorrected positions. These errors have been revised in this figure.

Mg# dependency of Raman spectra

Figure 2 shows the relationship between $\kappa_1$–Mg# and $\kappa_2$–Mg# obtained using the dataset provided in Mouri and Enami (2008) and in Table 2 (this study). The natural Fe–rich olivine contains varying amounts of tephroite (Mn$_3$SiO$_4$) component, which has lower $\kappa_1$ and higher $\kappa_2$ values ($\kappa_1$ = 807.5 ± 0.1 cm$^{-1}$ and $\kappa_2$ = 839.3 ± 0.1 cm$^{-1}$) than fayalite ($\kappa_1$ = 814.7 ± 0.3 cm$^{-1}$ and $\kappa_2$ = 836.8 ± 0.5 cm$^{-1}$).
cm$^{-1}$). Thus, with the incorporation of the tephroite component, the $\kappa_1$ value of the Fe-rich olivine decreases whereas the $\kappa_2$ value increases (Mouri and Enami, 2008). Regression analyses of the relationship between $\kappa_1$–Mg$#$ and $\kappa_2$–Mg$#$ in the forsterite–fayalite series (Fig. 2) were applied to the Mn–poor Mg–Fe olivine (Mn < 0.05 apfu) following Mouri and Enami (2008).

Mouri and Enami (2008) used the $\omega$ value for discussing the Mg$#$ dependency of the Raman spectra to avoid systematic errors in individual measurements of $\kappa_i$ values. By considering the five new data points of olivine with Mg$# = 0.22$–$0.48$ (Table 2), we have revised the $\omega$–Mg$#$ diagram from Mouri and Enami (2008) as shown in Figure 3.

**Regression analysis**

For regression analyses of the Mg$#$ dependencies on Raman spectra, the former studies have either used quadratic formulas to fit their Mg$#$ and $\kappa_i$ (or $\omega$) datasets (Wang et al., 2004; Kuebler et al., 2006; Mouri and Enami, 2008; Foster et al., 2013; Breitenfeld et al., 2018) or used linear formulas (Guyot et al., 1986; Yasuzuka et al., 2009; Ishibashi et al., 2012). In particular, Ishibashi et al. (2012) reported a clear dataset where the Raman spectra displayed linear variation with Mg$#$ for olivine grains with Mg$# > 0.63$.

Kolesov and Tanskaya (1996) analyzed the dependency of totally symmetric vibrations of olivine on compositions and reported that there were significant changes (steps) of the Raman wavenumber at around Mg$# = 0.6$ and 0.3; the step was most evident on the peak around 820 cm$^{-1}$. Gaisler and Kolesov (2007) also reported linear Mg$#$ dependencies on the peak positions of olivine groups with Mg$# > ~ 0.3$ and Mg$# \leq 0.3$, suggesting a one-step change of the wavenumber at around Mg$# = 0.2$–0.3. They attributed this dynamic effect to the interaction of spin and vibration that generates a phonon–magnon excitation in the disordered magnetic medium. Therefore, the relationships between $\kappa_1$–Mg$#$ and $\kappa_2$–Mg$#$ (Fig. 2) and $\omega$–Mg$#$ (Fig. 3) are probably explained also by the combination of the two regression lines for the olivine groups, with Mg$#$ bordering ~ 0.2–0.3. Table 3 lists the intercepts and coefficients of the equations that depict the relationship between $\kappa_1$–Mg$#$, $\kappa_2$–Mg$#$, and $\omega$–Mg$#$ after calibration using quadratic [E (Q)] and linear [E (LT), E (LH), and E (LL)] formulae, where LT, LH, and LL indicate the regression lines for Mg$#$ = 0–1, Mg$# > 0.25$, and Mg$# \leq 0.25$, respectively.

The controversies regarding the formulation of the Mg$#$ dependencies on the Raman spectra of olivine are in terms of the following conditions, i.e., if the use of (1) linear or polynomial formula, (2) single group data for the total solid solution or two groups bordering at ~ 0.2–0.3 Mg$#$, or (3) $\kappa_i$ or $\omega$ as the Raman parameter is more appropriate for regression analysis of a forsterite–fayalite series. Although there is no logical basis for adopting a quadratic or higher-order formula (Breitenfeld et al.,
we employed a quadratic formula to fit the 17 Mn-poor data points in our previous paper (Mouri and Enami, 2008) because of the following reasons: (1) significant differences in the trends between olivine groups with Mg# > 0.6 and < 0.2, (2) lack of Raman data for olivine with Mg# = 0.2–0.6, and (3) slightly higher correlation coefficient ($r^2$) in the quadratic fitting ($r^2 = 0.956, 0.988$, and 0.991 for $\kappa_1$, $\kappa_2$, and $\omega$ values, respectively) compared to the linear fitting ($r^2 = 0.921, 0.980$, and 0.988 for $\kappa_1$, $\kappa_2$, and $\omega$ values, respectively).

Furthermore, while calibrating the 22 Mn-poor data that included the five values where Mg# ranged from 0.2 to 0.5, the quadratic fitting $E[Q]$ provided a higher correlation coefficient ($r^2$) in the quadratic fitting ($r^2 = 0.956, 0.988$, and 0.991 for $\kappa_1$, $\kappa_2$, and $\omega$ values, respectively) compared to the linear fitting ($r^2 = 0.921, 0.980$, and 0.988 for $\kappa_1$, $\kappa_2$, and $\omega$ values, respectively).

Table 3. Results of least-squares regressions for the relationship between the Mg# and the Raman parameters

| Parameter | Equation | Formula | Composition | $Mg# = M0 + M1 \times 10^{-3} \times (κ_1^*, \omega)$ | $M0$ | $M1$ | $M2$ | $r^2$ |
|-----------|----------|---------|-------------|---------------------------------|------|------|------|------|
| $κ_1^*$   | E (Q)    | Quadratic | $0 \leq Mg# \leq 1.0$ | $-2.9556$ | $2.9263$ | $-5.438$ | 0.966 |
|           | E (LT)   | Linear   | $0 \leq Mg# \leq 1.0$ | $-0.9621$ | $0.8020$ | $0.937$  | 0.935 |
|           | E (LH)   | Linear   | $0.25 < Mg#$ | $-0.6404$ | $0.6607$ | $0.124$  | 0.107 |
|           | E (LL)   | Linear   | $Mg# \leq 0.25$ | $-0.5830$ | $0.4985$ | $-0.124$ | 0.107 |
| $κ_2^*$   | E (Q)    | Quadratic | $0 \leq Mg# \leq 1.0$ | $-2.7746$ | $0.9485$ | $-0.510$ | 0.985 |
|           | E (LT)   | Linear   | $0 \leq Mg# \leq 1.0$ | $-1.6435$ | $0.4630$ | $0.982$  | 0.966 |
|           | E (LH)   | Linear   | $0.25 < Mg#$ | $-1.4892$ | $0.4344$ | $0.966$  | 0.964 |
|           | E (LL)   | Linear   | $Mg# \leq 0.25$ | $-1.2855$ | $0.3667$ | $0.764$  | 0.764 |
| $ω$       | E (Q)    | Quadratic | $0 \leq Mg# \leq 1.0$ | $1.8442$ | $-2.0259$ | $5.446$  | 0.984 |
|           | E (LT)   | Linear   | $0 \leq Mg# \leq 1.0$ | $-2.3284$ | $1.0113$ | $0.963$  | 0.964 |
|           | E (LH)   | Linear   | $0.25 < Mg#$ | $-2.8849$ | $1.1922$ | $0.964$  | 0.964 |
|           | E (LL)   | Linear   | $Mg# \leq 0.25$ | $-1.0392$ | $0.4869$ | $0.894$  | 0.894 |

Note that the parameter $κ_1^*$ ($= κ_1 - 800$) was used instead of $κ_1$ to avoid a large absolute value of the intercept (M0) of the regression equation.

Figure 5. Comparisons of Mg# values calibrated using the new quadratic formula [E (Q),] with those calibrated using linear formulae E (LL) and E (LH) of $κ_1$, $κ_2$, and $ω$ values, and quadratic formula (ME08) proposed by Mouri and Enami (2008). Solid line indicates the Mg#(Q):Mg#(LL/LH) or Mg#(Q):Mg#(ME08) = 1:1.

Figure 5 compares the Mg#(Q) value calibrated using the quadratic function $E[Q]$ with those calibrated using the linear formulae $E[LL]$ and $E[LH]$ of $κ_1$, $κ_2$, and $ω$ values, and quadratic formula (ME08) proposed by Mouri and Enami (2008). The results were compared with those of linear fittings for two compositional groups of Mg# ≤ 0.25 [E (LL)] and Mg# > 0.25 [E (LH)].

The Mg# values have lower $D_{LL/LH–Q}$ than a relatively large difference in the Mg# between the two calibrations $D_{LL/LH–Q} = Mg#(LL/LH) - Mg#(Q)$: −0.05–0.08 and −0.02–0.13 for the olivine groups of Mg# ≤ 0.25 and Mg# > 0.25, respectively. The $D_{LL/LH–Q}$ values of the $κ_2$ peak also have relatively large differences of −0.04–0.05 and −0.01–0.04 for Mg# ≤ 0.25 and Mg# > 0.25, respectively. The $ω$ values have lower $D_{LL/LH–Q}$ than...
those of the $\kappa_1$ and $\kappa_2$ peaks; these values are $-0.03$–$0.02$ and $-0.04$–$0.03$ for $\text{Mg}# \leq 0.25$ and $\text{Mg}# > 0.25$, respectively. Consequently, the $\omega$ value produces similar $\text{Mg}#$ values for all forsterite–fayalite solid solutions with a difference of less than 0.04, regardless of which of the following regression formulae of the quadratic and linear fits is adopted:

$$\text{Mg}#(Q)_\omega = 0.005446\omega^2 - 0.20259\omega + 1.8442 \quad (r^2 = 0.984) \quad E(Q)_\omega,$$

$$\text{Mg}#(LH)_\omega = 0.04869\omega - 1.0392 \quad (r^2 = 0.894) \quad E(LH)_\omega,$$

and

$$\text{Mg}#(LH)_\omega = 0.11922\omega - 2.8849 \quad (r^2 = 0.964) \quad E(LH)_\omega.$$

The regression equation in Mouri and Enami (2008) overestimates the $\text{Mg}#$ values up to 0.06 compared to the revised $E(Q)_\omega$ equation, over a wide $\text{Mg}#$ range of about 0.1–0.8.

**Comparison with previous studies**

Figure 6 compares the results of regression analyses in this study and data with $\text{Mn} < 0.05$ apfu in the literature. The data from the terrestrial olivine grains with $\text{Mg}# > 0.6$ are most consistent with the regression curve $E(Q)_\omega$ (Fig. 6a). However, the data for terrestrial olivine grains with $\text{Mg}# < 0.6$ were more scattered than that for $\text{Mg}# > 0.6$. As the $\text{Mg}#$ of olivine decreases, the $\kappa_1$ and $\kappa_2$ doublet peaks gradually approach each other, and thus, their degree of overlapping increases (e.g., Kuebler et al., 2006). Therefore, with the decrease in $\text{Mg}#$, it becomes increasingly difficult to separate the two peaks and determine their exact positions. In addition, the results of the peak separation can vary depending on the function and model employed for peak fitting (Breitenfeld et al., 2018). The combined consequence is that the data of olivine grains with a lower $\text{Mg}#$ tend to have higher uncertainties and may be more scattered than the data of high $\text{Mg}#$ olivine grains.

The synthetic grains display relatively more scattered data than the terrestrial grains, with some of the former data being plotted far from the regression curve and the lines proposed in this study (Fig. 6b). Considerable cation re-ordering occurs at the octahedral sites of the Fe–$\text{Mg}$–$\text{Mn}$ olivine during the quenching process (e.g., Akamatsu and Kumazawa, 1993; Henderson et al., 1996), and it was shown that the degree of order under room temperature reflects the sample cooling rate (Redfern et al., 1996). The cooling rates undergone by the synthetic olivine grains might not have fully recovered to those expected at room temperature conditions and from their $\text{Mg}#$ values. Con-
Consequently, each synthetic olivine grain might have retained various recovery rates for the ordering degree depending on its production process. Such a difference in the re-ordering degree might be one of the main factors for the variability in the Raman spectra of the synthetic olivine grains. However, this interpretation cannot explain why the Fe$_2$SiO$_4$ olivine data reported by Breitenfeld et al. (2018) deviates from the regression curve proposed here, and other factors need to be considered.

An olivine grain in a meteorite with Mg\# = 0.34 studied by Kuebler et al. (2006) produces a low \( \omega \) value (24.5 cm$^{-1}$), which is far from the expected \( \omega \) value. Foster et al. (2013) reported that an experimentally impacted olivine displays a different \( \omega - \text{Mg\#} \) trend, which systematically shifts to the lower Mg\# and/or higher \( \omega \) side compared to the un-impacted olivine. Although the olivine data reported by Kuebler et al. (2006) show a shift toward the higher Mg\# and/or lower \( \omega \) side, which is contrary to the effect reported by Foster et al. (2013), we suggest that any impact or other astronomical events experienced by the host meteorite might have some influence on the Raman spectra of the olivine.

**CONCLUSIONS**

EPMA and Raman spectroscopic analyses were performed on Fe-rich olivine grains (Mg\# = 0.2–0.5) in the andesitic rocks from the Miocene Shitara volcanic complex. Considering these data, we modified the equation proposed by Mouri and Enami (2008) to determine Mg\# using the wavenumber difference of a high-intensity doublet between 800 and 860 cm\(^{-1}\) (\( \omega = \kappa_2 - \kappa_1 \)). Some published data on the synthetic forsterite–fayalite series and olivine in meteorites are plotted far from the \( \omega - \text{Mg\#} \) regression equation proposed here. In some of the synthesized crystals, the degree of Mg–Fe ordering between the M(1) and M(2) sites may not reach the values expected from the room temperature and chemical composition due to different cooling rates during experiments. The difference in the degree of ordering might have affected the wavenumber of the Raman spectra. In addition, impact events experienced by meteorites might have affected the Raman spectra of olivine. We should take these points into account while applying the proposed regression equation to synthetic crystals or olivine in samples that may have been impacted.

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**SUPPLEMENTARY MATERIAL**

Supplementary Table S1 is available online from https://doi.org/10.2465/jmps.201204.

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