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Zaisho—a pallasite containing pyroxene and phosphoran olivine

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ABSTRACT. The Zaisho meteorite, one of only four known pallasite falls, contains pyroxene, which is rare in pallasites, and it is only the fourth meteorite known to contain phosphoran olivine. Except for a recent report by Goodrich (1984) such olivine appears to be restricted to pallasites, and to those containing the rare Mg phosphate, farringtonite. It is unique among olivines in having P in tetrahedral sites and having vacancies, for charge balance, in octahedral sites. Zaisho also contains stanfieldite and a phosphate having a composition between that of stanfieldite and farringtonite. Its metallographic cooling rate is similar to that of many other pallasites, but its average olivine composition, $Fa_{18.3}$, places it in a small group with more Fe-rich olivines than most pallasites.

THE Zaisho pallasite fell in 1898 (in Japan), but material was only recently made available for study. It is small, and the main mass is held privately. As a consequence, it was not included in several recent extended studies of the mineralogy and petrology of pallasites (Buseck and Goldstein, 1969; Buseck, 1977; Buseck and Holdsworth, 1977; Scott, 1977). The purpose of the current paper is (*a*) to update those studies by including data on Zaisho, and (*b*) to provide additional information on an intriguing mineral, phosphoran olivine. Shima *et al.* (1980) provided a brief description of Zaisho, but did not note some of its more unusual and interesting features such as the presence of pyroxene and phosphoran olivine.

We obtained a small (0.80 gm) chip of Zaisho, just large enough for a polished section. The mineralogy and texture of the meteorite are fairly typical of pallasites. It contains metal, olivine, chromite, phosphates, pyroxene, troilite, and schreibersite, with the last three in minor to trace amounts. Our specimen has a greater volume of chromite than olivine, which is atypical of pallasites; however, this is almost certainly a consequence of both the small and presumably unrepresentative sample and the heterogeneity of pallasites. Unusual features are the occurrence of phosphoran olivine (this is only the fourth meteorite from which this material has been described), pyroxene, and a phosphate intermediate in composition to farringtonite and stanfieldite.

Electron microprobe techniques are as described by Buseck and Holdsworth (1977). Analyses with summations between 98.5 and 101.5 wt. % are included; largely because of the small sizes and rounding of crystals near the edges of the section, a number of analyses fell outside this range and were excluded.

Olivine. The olivine crystals in our Zaisho sample are mostly small, ranging from 5 μm to 1000 μm in largest dimension, and located along the periphery of the chip. The crystals are somewhat angular, but with all their corners rounded, as would occur had the meteorite been annealed following olivine facturing. Two types of olivine–troilite intergrowths are present. Some of the trolite contains myriads of small (5 to 80 μm diameter) subhedral to euhedral olivine crystals. Elsewhere, along borders between troilite and the larger olivine crystals, there is a fine-grained symplectic intergrowth of the two minerals. Both features are unusual among pallasites. In a few places, small rounded fragments of olivine occur within troilite.

The compositions of the olivines are indicated in Table I. The average composition is similar to the analysis given by Shima *et al.* (1980). The analyses show more scatter than is typical of other pallasites. In Glorieta Mtn., Huckitta, and Marjalahti the small olivines show compositional scatter (Buseck and Goldstein, 1969) and, as many of the crystals in Zaisho are small, it is possible that a similar effect is present. Several analyses were performed to check for zoning within a single olivine crystal, but none was detected. The olivines enclosed in troilite are compositionally similar to those enclosed in metal and exhibit approximately the same range of compositional scatter.

Some olivines close to or enclosed by chromite have unusually low Fe contents, ranging from 10.0 to 16.4 mole % fayalite. Similarly, the host chromite exhibits Mg depletion in the regions immediately surrounding the olivine inclusions. This clearly

Table I Microprobe analyses of P-free olivines in Zaisho

	Oxide weight %							Atomic Proportions (0=4)					
	FeO	MnO	MgO	SiO_2	Sum	%Fa		Fe	Mn	Mg	Si	Total Cations	Fe+Mn+Mg
	17.4	0.38	43.3	39.1	100.2	18.4		0.37	0.01	1.64	0.99	3.01	2.02
	17.2	0.38	44.2	39.0	100.8	17.9		0.36	0.01	1.66	0.98	3.01	2.03
	17.4	0.43	43.1	38.8	99.7	18.4		0.37	0.01	1.64	0.99	3.01	2.02
	17.2	0.40	43.6	39.3	100.5	18.1		0.36	0.01	1.64	0.99	3.00	2.01
	17.3	0.43	43.6	38.4	99.7	18.2		0.37	0.01	1.66	0.98	3.02	2.04
	16.1	0.47	43.4	39.0	99.0	17.2		0.34	0.01	1.65	1.00	3.00	2.00
	17.8	0.36	42.5	39.3	100.0	19.0		0.38	0.01	1.61	1.00	3.00	2.00
	17.4	0.37	42.8	39.4	100.0	18.6		0.37	0.01	1.62	1.00	3.00	2.00
	17.6	0.41	42.9	39.4	100.3	18.7		0.37	0.01	1.62	1.00	3.00	2.00
	a) 16.5	0.37	43.6	39.4	99.9	17.5	a)	0.36	0.01	1.65	1.00	3.02	2.02
	b) 17.1	0.45	43.0	39.2	99.8	18.3	b)	0.36	0.01	1.63	1.00	3.00	2.00
	c) 17.7	0.43	42.6	39.2	99.9	18.9	c)	0.38	0.01	1.62	1.00	3.01	2.01
$\overline{X}$	17.2	0.41	43.2	39.1	99.9	18.3	$\overline{X}$	0.37	0.01	1.64	0.99	3.01	2.01

a)Lacy troilite region A, average of 2 crystals

b)Lacy troilite region B, average of 5 crystals

c)Lacy troilite region C, average of 5 crystals

indicates the interchange of Mg and Fe between the two coexisting minerals. Owing to the anomalous nature of these Zaisho olivines, they are not included in Table I.

Phosphoran olivine. A unique feature of certain pallasites is that some olivine crystals contain appreciable phosphorus (Buseck, 1977). Koritnig (1965) discussed the substitution of P for Si in rock-forming silicates, and his olivine analyses show a maximum of several hundred p.p.m. P. Bradley *et. al.* (1966) examined Si-P substitution in a variety of silicates, with emphasis on olivines. They reported a lack of evidence for significant solid solution of phosphate in olivine, both as

Table II Microprobe analyses of phosphoran olivines

Meteorite	Crystal No.	FeO	MnO	MgO	SiO_2	P_2O_5	Sum	% Fa
		Oxide Weight %						
Zaisho	1+	15.9	0.40	42.9	36.5	4.36	100.1	17.2
(Zaisho	2	16.7	0.38	44.2	38.6	0.03	99.9	17.5)
Zaisho	3	16.2	0.42	42.8	37.4	3.00	99.8	17.5
Zaisho	4	12.3	0.40	46.1	36.8	3.76	99.4	13.0
Brahin*		13.7	0.30	43.2	36.5	3.82	97.5	15.1
Rawlinna*		14.3	0.26	44.8	35.9	4.91	100.2	15.2
Springwater*		16.2	0.45	43.0	35.5	4.14	99.3	17.5

		Atomic Proportions (0=4)						
		Fe	Mn	Mg	Si	P	Total Cations	Fe+Mn+Mg
Zaisho	1	0.33	0.01	1.60	0.91	0.09	2.94	1.94
(Zaisho	2	0.36	0.01	1.67	0.98	<0.01	3.02	2.04)
Zaisho	3	0.34	0.01	1.61	0.94	0.06	2.96	1.96
Zaisho	4	0.26	0.01	1.71	0.92	0.08	2.98	1.98
Brahin*		0.29	0.01	1.63	0.93	0.08	2.94	1.93
Rawlinna*		0.30	0.01	1.66	0.89	0.10	2.96	1.97
Springwater*		0.34	0.01	1.62	0.90	0.09	2.96	1.97

+Crystals #1 and 2 are adjacent to one another. *Buseck (1977) reported in the literature and in their experiments on forsterite and $LiMgPO_4$, which is isostructural with olivine. Goodrich (1984) has found olivine and pyroxene in basalts from Disko Island that contain up to *c.* 3 wt. % P as P_2O_5. This is the only known terrestrial occurrence of these phosphoran species.

Three of the olivines in our chip of Zaisho are phosphoran; their compositions are given in Table II, together with those of the other known phosphoran olivines. There is some compositional spread among the Zaisho olivines. Except for the slightly higher content of SiO_2, the Zaisho phosphoran olivines resemble those from the other pallasites, and two of the Zaisho crystals have similar fayalite contents to that from Springwater.

Crystal no. 1 in Table II is the largest phosphoran olivine encountered in Zaisho (250 μm in largest dimension) and is separated by a crack from non-phosphoran olivine, which is either a distinct crystal or another portion of the phosphoran crystal. This is the only phosphoran olivine that is clearly zoned. Three microprobe traverses separated by approximately 70 μm were run across this crystal, with P analyses performed each 5 μm. The P distributions are roughly parallel. Fig. 1 shows the averaged P intensities across the phosphoran olivine crystal. The P contents are high near the chromite and lowest next to the P-free olivine.

The average of analyses at five spots on adjacent, essentially P-free olivine is given as crystal no. 2 in Table II. The non-phosphoran crystal contains greater amounts of Fe, Mg, and Si and has a slightly higher fayalite content. This closely parallels Springwater, whose phosphoran olivine also has a slightly lower fayalite content than coexisting P-free olivine. By contrast, in Brahin the phosphoran olivine is appreciably more fayalitic than coexisting P-free olivine.

Crystal no. 3 in Table II is the only phosphoran olivine in our polished section that is visibly adjacent to phosphate (fig. 2). A separate P-free olivine is also adjacent to the phosphate. Crystal no. 4 in Table II, the smallest phosphoran olivine encountered in Zaisho (40 μm in largest dimension), is somewhat anomalous in its low fayalite content. This crystal, along with several P-free olivines, is

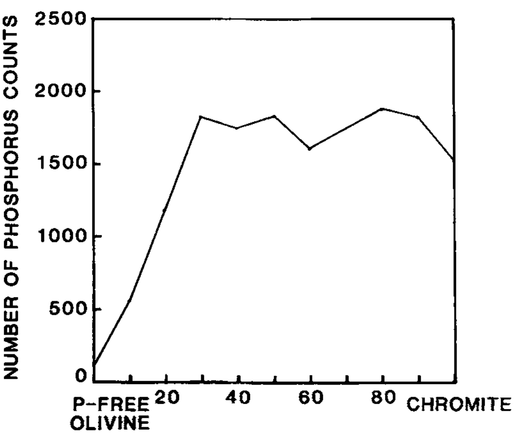

FIG. 1. P intensities averaged for three parallel traverses (of lengths 45, 65, and 30 μm) across a phosphoran olivine. This plot was obtained by expanding the two shorter traverses to that of the longest one and then by averaging the values at 10% intervals of distance from P-free olivine to chromite. Although there is appreciable scatter, in the high-P region the maximum spread from the average is less than 500 counts.

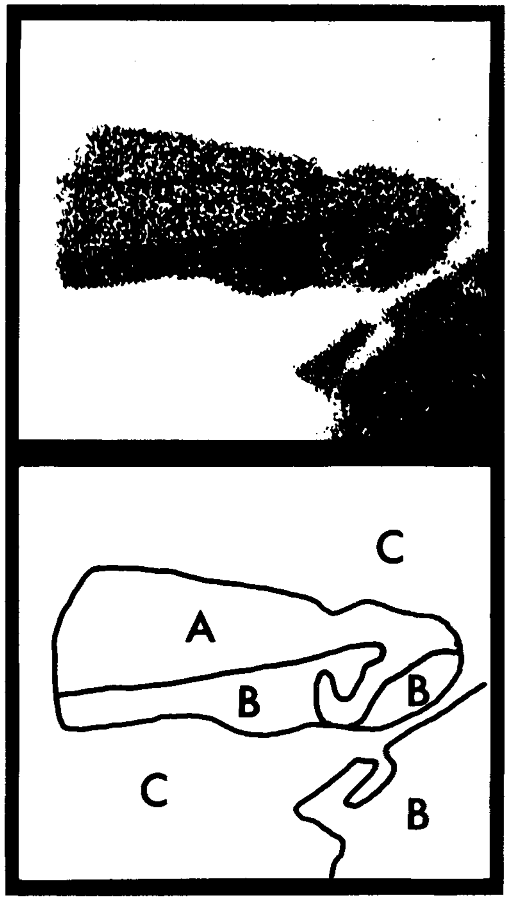

FIG. 2. X-ray scanning photograph (*top*) and sketch (*bottom*) of P in phosphoran olivine. The white regions in the top photograph indicate the presence of P. A, phosphoran olivine; B, non-phosphoran olivine; C, farringtonite. The crystal is 90 μm in its longest dimension.

present as an inclusion in a large (500 μm in largest dimension) chromite crystal. The average mole % fayalite (11.9) of the P-free olivines is correspondingly low. Interchange of Mg and Fe between the chromite and olivine is indicated by the depletion of Mg in the chromite regions immediately surrounding the olivine inclusions.

The phosphoran olivine of Zaisho appears to be similar to those in the other pallasites. However, occurrence and crystallographic location of the P in phosphoran olivine remains a problem. Li orthophosphate (triphylite) is isostructural with olivine, and Li could conceivably provide the required charge balance for P in olivine. We searched for Li in Springwater phosphoran olivine using an ion microprobe. This work was done with J. V. P. Long of Cambridge University. Although strong Li signals were obtained from standards, no Li was detected in the olivine.

In a related attempt to characterize the mineral, a portion of the phosphoran olivine from the Springwater meteorite was extracted for X-ray analysis by R. Von Dreele of Arizona State University. A full anisotropic least-squares refinement including occupancy factors was performed (46 parameters and 713 reflections). The residuals are $R = 0.0342$ and $R_w = 0.0236$, and the resulting lattice parameters are $a = 4.7689$ (6), $b = 10.2647$ (16), and $c = 5.9997$ (8) Å. The refinement, although of high quality, did not establish the presence or location of P or of compensating vacancies in the structure.

Although the question of charge balance for the P in the olivine has not been determined, related phosphate minerals do provide insight. Heterosite [$\square(Fe^{3+},Mn)PO_4$] and sarcopside [$\square FeFe_2(PO_4)_2$] have similar structures to olivine; they achieve charge balance by having vacant octahedral sites (Quensel, 1937; Eventoff *et al.*, 1972; Moore, 1972). The compositions of the phosphoran olivine are not conclusive, but they are least suggestive of a similar situation. Table I shows that the atomic proportions of the octahedral cations in the P-free olivines total 2.02 ± 0.02; Table II shows proportions for the phosphoran olivine of 1.96 ± 0.03. The atomic proportions of P in these olivines range from 0.06 to 0.10 and so, for charge balance by vacancies, we would predict a cation deficit of 0.03 to 0.05 in the octahedral sites. These predicted values are remarkably close to the observed value of 0.04 ± 0.03. It thus appears as if phosphoran olivine may achieve charge balance by octahedral site vacancies, in a fashion similar to heterosite and sarcopside. It should be noted, however, that such vacancies have not been reported from olivine.

Measurements of electron channelling along (010) planes indicates that the P occurs on either the octahedral *M* (2) sites or the tetrahedral sites; crystal-chemical considerations suggest that the P occupies the tetrahedral sites (Buseck *et al.*, 1983; Self and Buseck, 1983).

Buseck (1977) found phosphoran olivine in only three of the thirty-eight pallasites examined. It is curious that two of these, Rawlinna and Springwater, are also the only pallasites in which Buseck and Holdsworth (1977) found farringtonite. (Brahin, the other pallasite that has phosphoran olivine, contains no phosphate in the available specimens.) The fact that Zaisho also contains the unusual phosphate farringtonite and phosphoran olivine is strongly suggestive of a related origin between these two rare phases.

Pyroxene and symplectic intergrowths. Pallasites were long thought to contain only one type of silicate, olivine, and it occurs in abundance. However, Buseck (1977) described minor amounts of pyroxene from seven pallasites. Zaisho now joins that select group. As in the other pallasites, the Zaisho pyroxenes occur in symplectic intergrowths around some of the olivines; lacy trolite is a prominent feature of these intergrowths.

The origins of the symplectites and thus the pyroxenes in pallasites are not known. However, Fleet and MacRae (1982) propose that olivine reaction with an Fe–Ni monosulfide and graphite could produce symplectic intergrowths. Although there are differences between their experimental results and our observations (we see neither niningerite nor Fe–Ni–Si alloy, but these may be quench products in their experiments), their results may provide the most reasonable model for these curious intergrowths. If correct, the symplectites indicate the former presence of graphite in these pallasites as well as a post-formation event of sulphide metasomatism.

Microprobe analyses of pyroxenes from Zaisho were obtained from three distinct regions of the polished section, and between five and ten analyses, taken at points separated by 100 μm or more, were obtained from each region. Within experimental error the analyses are identical The average pyroxene composition in oxide wt. % is: MgO 31.4(0.3), FeO 11.3(0.3), MnO 0.43(0.08), CaO 0.05(0.05), SiO_2 56.4(0.3), total 99.6 %. The numbers in parentheses represent the standard deviations, based on analyses of twenty crystals. Corresponding atomic proportions, based on six oxygen atoms, are: Mg 1.66, Fe 0.33, Mn 0.01, Ca 0.01, and Si 2.00. The ratio of octahedral cations to Si is 1.00, and the % ferrosilite content is 16.8. The pyroxene is stoichiometric, has unusually low Ca content, and is identical in composition to the pyroxene in Springwater (Buseck, 1977).

Zaisho belongs to the small group of pallasites

(Springwater and Phillips Co.) with both olivine and pyroxene having Fe/(Fe+Mg) ratios overlapping those of the H-group chondrites (Dodd, 1969; Buseck, 1977). As in the case of pyroxene from the other pallasites, Zaisho pyroxene contains less Ca than the chondritic pyroxenes.

Chromite. Our Zaisho sample contains several grains of chromite; one is the largest single crystal in the specimen, comprising approximately 12% of the surface area of the polished section. The chromite characteristics are similar to those described by Buseck (1977), with the crystals in Zaisho being subhedral and having somewhat rounded corners. Some crystals contain small inclusions of olivine.

Eight chromite crystals were analysed with the electron microprobe. All have similar compositions. Average oxide wt. % is: Cr_2O_3 67.9(0.5), Al_2O_3 0.69(0.23), V_2O_3 0.60(0.04), TiO_2 0.09(0.03), SiO_2 0.09(0.03), FeO 27.4(0.5), MgO 3.33(0.19), MnO 0.71(0.04), ZnO 0.11(0.07), total 100.9%. The numbers in parentheses represent the standard deviations, based on analyses of eight crystals. Atomic proportions based on thirty-two oxygens are Cr 15.40, Al 0.23, V 0.11, Ti 0.02, Si 0.03, Fe 6.57, Mg 1.42, Mn 0.17, Zn 0.02, and total cations equal 24.0. The Zaisho chromite contains less Al_2O_3 and MgO and slightly more FeO than most of the chromites from pallasites studied by Bunch and Keil (1971).

Phosphates. Phosphate minerals are widespread minor constituents of pallasites. Zaisho contains a large number of phosphate grains, ranging in diameter from 50 μm to one grain that is 3500 μm long. Two phosphate grains are apparently surrounded by chromite and one by troilite. In one spot the phosphate contains small fragments of olivine, one of which is phosphoran.

Because the phosphates are located mainly along the edges of our chip and were severely rounded during the polishing process, they were the most difficult phases in Zaisho to analyse. Of twenty-six distinct regions where analyses were performed, only eight provided summations within the allowable limits; these are given in Table III. All of these phosphates contain Mg and P as their major cations and are thus compositionally equivalent to farringtonite; in their survey of phosphates in thirty-one pallasites, Buseck and Holdsworth (1977) found farringtonite in only two meteorites, indicating it to be an unusual mineral among the pallasitic phosphates, in spite of its relative abundance in Springwater. The alkali elements are greatly depleted in pallasites. Those that do occur reside in the phosphate minerals. All of the farringtonite grains contain detectable K and one contains detectable Na.

All of the phosphate analyses (including those that were excluded from Table III because of poor summations) were normalized to give Ca+Mg+Fe = 100 and are indicated in fig. 3. The result shows that most crystals plot in the farringtonite region, but one plots as stanfieldite [$Ca_4(Mg,Fe)_5(PO_4)_6$], and another has a composition between that of stanfieldite and farringtonite. The normalized 'stanfieldite' contains 8.0 wt. %

Table III Microprobe analyses of farringtonites in Zaisho

Oxide Weight %

	FeO	CaO	MgO	MnO	Na_2O	K_2O	P_2O_5	Al_2O_3	SiO_2	Sum
	2.32	0.06	43.1	0.13	n.d.*	0.02	53.7	0.10	0.04	99.5
	3.97	0.06	42.8	0.23	n.d.	0.01	53.4	0.03	0.06	100.6
	7.70	0.01	40.7	0.21	n.d.	0.48	50.0	0.03	0.10	99.2
	4.29	0.04	42.4	0.19	n.d.	0.01	53.2	0.06	0.07	100.3
	3.68	0.04	42.2	0.22	n.d.	0.01	52.5	0.06	0.07	98.8
	3.83	0.05	41.2	0.19	n.d.	0.01	53.6	0.05	0.07	99.0
	4.24	0.03	40.9	0.04	0.69	0.04	52.7	0.12	0.25	99.0
	4.13	0.05	41.2	0.32	n.d.	0.02	53.2	0.05	0.08	99.1
$\bar{X}$	4.27	0.04	41.8	0.19	0.09	0.08	52.8	0.06	0.09	99.4

Atomic Proportions (O=24)

	Fe	Ca	Mg	Mn	Na	K	P	Al	Si	Total Cations	Metal: Phosphorus
	0.26	0.01	8.55	0.02	<0.01	<0.01	6.05	0.02	0.01	14.92	1.46
	0.44	0.01	8.48	0.03	<0.01	<0.01	6.01	<0.01	0.01	14.98	1.49
	0.89	<0.01	8.38	0.02	<0.01	0.08	5.85	<0.01	0.01	15.24	1.61
	0.48	0.01	8.44	0.02	<0.01	<0.01	6.01	0.01	0.01	14.98	1.49
	0.42	0.01	8.50	0.03	<0.01	<0.01	6.01	0.01	0.01	14.99	1.49
	0.43	0.01	8.25	0.02	<0.01	<0.01	6.10	0.01	0.01	14.83	1.43
	0.48	<0.01	8.25	0.01	0.18	0.01	6.03	0.02	0.03	15.00	1.49
	0.47	0.01	8.28	0.04	<0.01	<0.01	6.07	0.01	0.01	14.89	1.45
$\bar{X}$	0.48	0.01	8.39	0.02	-	0.01	6.02	0.01	0.01	14.98	1.49

*Not detected (detection limit is 0.13 percent by weight)

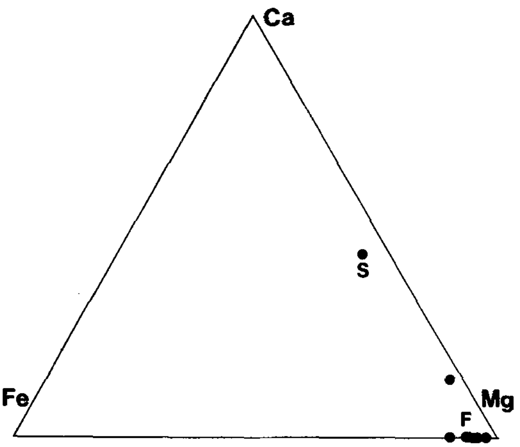

FIG. 3. Ternary composition diagram of phosphate analyses, normalized to give Ca+Mg+Fe = 100 (in atomic %). Note the well-defined cluster at the composition of farringtonite (F) and the sample with the composition of stanfieldite (S).

FeO, 42.0% MgO, and 50.0% CaO. It is compositionally similar to the averaged, normalized values of 6.9% FeO, 42.0% MgO, and 51.1% CaO for the stanfieldites in other pallasites [Table II of Buseck and Holdsworth (1977)].

Buseck and Holdsworth (1977) described a phosphate (from two crystals in the Springwater pallasite) that has a composition between stanfieldite and farringtonite. Its major oxide components, normalized to 100% are 10.8 wt. % FeO, 22.0% CaO, and 67.2% MgO, with a Ca:Mg atomic ratio of 0.27 and metal:phosphorus ratio of 1.54. In contrast, the values for the Zaisho phosphate that is compositionally between stanfieldite and farringtonite is 4.2% FeO, 17.6% CaO, and 78.2% MgO, with a Ca:Mg ratio of 0.16 and metal:phosphorus ratio of 1.50. Both of the intermediate phosphates contain unusually large amounts of Na (3.7±0.2 wt. % Na_2O). Although the Zaisho intermediate phosphate is clearly different from the Springwater ones, the two phosphates are more similar to each other than they are to other known meteoritic phosphates.

Cooling rate. The Zaisho cooling rate was determined by the same maximum Ni content ('Cγ max.') method that was used by Buseck and Goldstein (1969). Using an unoriented sample of Zaisho, we obtained a maximum Ni content of 49.0±0.2 wt. %, which corresponds to a cooling rate of 0.5 °C per million years, similar to that of many other pallasites. However, Narayan and Goldstein (1984) report that the published iron meteorite cooling rates are too low by two orders of magnitude. Presumably the pallasite cooling rates are similarly too low, in accord with the results of Pellas *et al.* (1981) which suggest that the published pallasite cooling rates are too slow. In any event, based on its cooling rate, Zaisho will remain grouped with the bulk of pallasites.

Olivine–chromite equilibration temperature. Bunch and Keil (1971) determined the temperatures of equilibration of olivine and chromite in ten pallasites using relationships developed by Jackson (1969). Their values ranged between 1143 and 1359 °C. Shima *et al.* (1980) obtained 1220 °C for Zaisho. This compares to our values of 1170 and 1178 °C for Zaisho; the former is for a pair of olivine and chromite crystals that are adjacent to one another, and the latter is for average compositions. Our values differ slightly from those of Shima *et al.* because of slightly different Mg/Fe ratios in the chromite analyses.

Conclusion. Zaisho is an unusual pallasite; it is one of only four pallasites known to contain phosphoran olivine and one of only three in which farringtonite is confirmed. It contains symplectic intergrowths and pyroxene, a mineral unusual in pallasites. It is also one of the few pallasites with olivine close to Fa_{20}; it apparently belongs to the small number of samples to which Scott (1977) referred as anomalous and that may have experienced a more oxidizing environment than the main group of pallasites.

Springwater, like Zaisho, exhibits all of the above features. The two meteorites landed on opposite sides of Earth and so it is unlikely that they are parts of the same fall (Springwater was found in Saskatchewan, Canada, in 1931, thirty-three years after Zaisho fell in Japan). Moreover, the appearance of our small Zaisho sample differs considerably from that of Springwater, primarily in sizes of the olivines and the distribution of farringtonite. None the less, the unusual features shared by the two pallasites suggest that they have a related origin and source.

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