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LUZONITE, FAMATINITE AND SOME RELATED MINERALS¹

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Abstract

A complete solid solution series extends between the tetragonal minerals luzonite, Cu₃AsS₄, and famatinite, Cu₃SbS₄. Luzonite is dimorphous with the orthorhombic mineral enargite; the solid solubility of Sb in enargite is restricted, and the orthorhombic dimorph of famatinite is not known in nature. Chemical, morphological, physical and x-ray data are given for luzonite and famatinite. The crystal structure of luzonite-famatinite is closely related to that of chalcopyrite and basically is of the sphalerite-type. Space group I42m; $a_0 5.290$ Å, $c_0 10.465$ (synthetic luzonite); $a_0 5.38$ Å, $c_0 10.76$ (synthetic famatinite); cell contents 2 [Cu₃(As, Sb)S₄].

HISTORICAL SURVEY

Since first described over eighty years ago, luzonite and famatinite have been the subject of continuing controversy. The current work establishes them as a valid mineral species, forming a complete isomorphous series, with both intermediate and end compositions occurring in nature.

Zerenner (1869) made the first reference to luzonite or famatinite in noting the presence of an unknown massive brown mineral associated with enargite and chalcopyrite in a suite of ore specimens from Mankayan, Luzon, Philippines. F. W. Fritzsche (1869) analyzed the same material and reported it similar in composition to dufrenoysite, as described by Damour in 1845. Damour's "dufrenoysite" was later shown to be tennantite, and the name dufrenoysite was applied to a lead mineral by Des Cloizeaux in 1855.

Next Stelzner (1873), while studying a group of ore minerals from the San Pedro and other mines in the Sierra de Famatina, La Rioja, Argentina, noted a massive mineral of reddish black color, lacking cleavage. Chemically it was a copper antimony arsenic sulfide, with Sb:As=4:1. Stelzner named it famatinite, from the locality, and stated, correctly, that it was not isomorphous with enargite. The erroneous assumption that they were isomorphous later became deeply entrenched in the literature.

Noting the reference by Zerrenner (1869), Weisbach redescribed specimens of a "brown" material from Mankayan, Luzon, which he had put aside after a superficial examination in 1866. He stated that the new mineral, which Fritzsche had mistakenly called dufrenoysite, resembled niccolite or bornite. The chemist Winkler's analysis showed copper, arsenic,

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antimony, and sulfur, with a trace of iron, in proportions identical with enargite. Weisbach proposed the name luzonite for it, also after the locality, stating that it was different in appearance from enargite and was isomorphous with famatinite. Once again the original description was correct, particularly in reference to its relationship to enargite and famatinite. However, later incorrect assumptions so confused the literature that even today many do not consider luzonite a valid species.

Further references were not long in appearing. Frenzel (1874) described an intermediate member from Cerro de Pasco with As:Sb=1:1, assuming it to be triclinic. Klockmann (1891) reached the same conclusions as Weisbach, using material from Sierra de Famatina, Argentina. Moses (1905) examined luzonite from Mankayan, Luzon, and, on the basis of goniometric studies, declared it crystallographically identical with enargite. He concluded that luzonite was merely a color variety of enargite. The writer, using Moses' original material, determined that he was mistaken in assuming the small enargite crystals upon which he worked to be identical with their luzonite matrix.

The sixth edition of Dana's System of Mineralogy states that enargite and luzonite are identical and that enargite and famatinite are isomorphous.

On the basis of nearly identical powder photographs of enargite from Silverton, Colorado and famatinite from Argentina, De Jong (1928) concluded that they had identical structures. His results were accepted by Schneiderhöhn and Ramdohr (1931) and confirmed through x-ray work by Waldo (1935). De Jong's "famatinite," however, was unquestionably a mixture of fine-grained enargite and some famatinite. Pure famatinite from the same mine gives a powder pattern markedly distinct from enargite.

Frebold (1927), studying ores from Mankayan, Luzon, held that luzonite was not a valid species name. An extended study by Harcourt (1937) further corroborated the work of those previously mentioned. His conclusions suffered from the fact that, although his samples of luzonite and famatinite came from five localities, the group was not large enough to be truly representative.

The wide variance of opinion held by the several investigators as to the existence and nature of luzonite and famatinite is due in a large measure to the fact that the same material has never been used for chemical analysis, polished section investigation, morphological work, and x-ray powder photography. Additional uncertainties arose because, as far as could be ascertained, none of the men who investigated the problem personally collected the specimens on which they worked, or visited the mines in which they occurred. Competent mineralogists visiting these localities probably would have found more luzonite-famatinite crystals. Luzonite-famatinite, moreover, are not always easily distinguishable from enargite because they usually occur more or less intergrown and because in polished section enargite often has a deceptively pink tinge.

The most recent work done on this problem has been by three Japanese: H. Imai (1943), who studied the Mankayan, Philippine Islands locality; M. Watanabé (1943), who studied luzonite from three mines in Japan and one in Formosa; and H. Sawada (1944), who made x-ray studies on luzonite crystals from Japan. Imai concluded that luzonite was a valid mineral species and that, although arsenic-rich, it did contain a little more antimony than enargite, yet considerably less than famatinite. Hence, in the series Cu₃AsS₄-Cu₃SbS₄, the mineral enargite corresponds to Cu₃AsS₄, while luzonite is an intermediate compound with a different structure (based on x-ray powder photographs). Watanabé reached a similar conclusion, even though his analysis of luzonite (intermixed with gangue) showed no antimony. Imai (1949, p. 59) refers to the fact that Sawada had determined the crystal structure of luzonite. Although Sawada's original paper, in Japanese, is not available in this country, a summary of his work was obtained by the writer via personal communication.

Using minute, imperfect, twinned crystals from the Hokuetsu mine, Japan, Sawada determined that luzonite is tetragonal, pseudo-cubic, with cell dimensions $a_1 = a_2 = 5.28$ Å; c = 5.22 Å. He admitted the possibility that the true cell might be a multiple of the one he determined, but had no positive evidence for this. Imai's reference to Sawada contains the misprint " $a_1 = a_2 = 4.28$ Å." His figure "c = 10.44 Å" is double Sawada's, with no explanation given. Imai's reference also has a discrepancy in the determination of the space group, which Sawada correctly determined to be $I\overline{4}2m$. Although Sawada's structure determination and other data are not consistent with that obtained in the present work, his is certainly the best study of the crystallography and structure of luzonite that has appeared to date. Any deficiency in his results can be attributed to the poor quality of the material with which he worked.

CRYSTALLOGRAPHY

Small crystals of luzonite were found by the writer on specimens from Mankayan, Luzon, Philippines, and Goldfield, Nevada. Interfacial angles, determined on the two-circle reflecting goniometer, indicated that the crystals are tetragonal, scalenohedral. The largest crystal observed was roughly equant and measured 2 millimeters across, but the largest dimension of those suitable for measurement was about 0.5 milli-

Forms	ϕ Meas.	ϕ Calc.	ρ Meas.	ρ Calc.	A Calc.	MCalc
001			0–00	0-00	90-00	90-00
110	45-00	45-00	90-00	90-00	45-00	90-00
012	0-50	0-00	46-00	45-00	90-00	60-00
011	0-20	0-00	63-00	63-26	90-00	50-46
112	45-10	45-00	54-00	54-44	54-44	90-00
111	45-00	45-00	71-00	70-32	48-11	90-00
132	19-00	18-26	72-00	72-27	72-27	64-46

TABLE 1. ANGLE TABI	LE OF FAMATINITE AND OF A	Measured
LUZONITE-FAMATINIT	e Crystal from Goldfield), NEVADA

 $p_0:r_0=2:1$

c/a = 2.000

Doubtful forms— $q\{1 \cdot 1 \cdot 10\}, w\{1 \cdot 3 \cdot 12\}, v\{125\}.$

For luzonite, the axial ratio c/a=1.978, and p:r=1.978:1. Since the difference between this and the axial ratio for famatinite is small, the luzonite angle table differs only slightly from this one.

meters. All crystals measured were twins, with irregular and unequally developed faces. Fairly sharp reflections were obtained with a crystal from Goldfield, Nevada. The forms listed in Table 1 were measured on this crystal. Its analysis is shown in Table 4. It corresponds to a mineral about midway between the two end members of the series. Angles were calculated on the basis of the cell derived from precision measurement of the powder photograph of pure synthetic famatinite, shown in Fig. 5 and Table 2. Figure 1 shows a measured crystal of luzonite-famatinite from Goldfield. Portions of another crystal attached to this one as an interpenetration twin were eliminated for the sake of clarity.

Figure 2 shows an idealized crystal from Mankayan, Luzon, Philippines. Crystals from Mankayan gave poor reflections. No crystals of famatinite were observed on material from Famatina, Argentina.

STRUCTURE

The space group of luzonite-famatinite is $I\overline{4}2m$. The writer determined approximate cell dimensions by means of single-crystal x-ray rotation photographs of crystals from Mankayan, Luzon and Goldfield, Nevada. Later, precise cell dimensions were obtained from powder photographs of synthetic luzonite and famatinite made by hydrothermal processes. These photographs were made with filtered Cu radiation with $\lambda = 1.5418$ Å. The unit cells contain two formula weights and have dimensions as follows:

Luzonite:	a₀ 5.290 Å,	c ₀ 10.465,	c/a = 1.978
Famatinite:	a ₀ 5.38	c ₀ 10.76	c/a = 2.000





Frg. 1. Luzonite-famatinite crystal from Goldfield, Nevada.

FIG. 2. Idealized crystal from Mankayan, Luzon, Philippines.

m

The 0-layer photograph brought out the tetragonal nature of luzonitefamatinite beyond question. Excellent zero, first, and second layer Precession photographs, shown in Figs. 3 and 4, were obtained with a fragment of famatinite from massive, coarsely crystalline material from Sierra de Famatina, Argentina. These showed that the cell is body-centered tetragonal, with no glide planes or screw axes. Eight different space groups would fulfill the criteria shown by these photographs, viz: $I\bar{4}$, $I\bar{4}m2$, $I\bar{4}2m$, I4, I4/m, I4mm, I42, I4mmm. $I\bar{4}2m$ was chosen on the basis of the structure as it was finally determined.

Powder photographs were taken of luzonite and famatinite from all the principal localities from which specimen material was available, and of artificial material made by hydrothermal processes. The synthetic material included the arsenian and antimonian end-members and several intermediate members of the series. Powder photographs of the natural and artificial material were in complete agreement. Careful comparison of many photographs established which lines were unquestionably due to luzonite-famatinite and which should be rejected as representing impurities.

These powder photographs were then measured and indexed, using the



FIG. 3. Famatinite, Argentina. Precession photograph, a-axis, 0-level.



FIG. 4. Famatinite, Argentina. Precession photographs, *a*-axis, 1-level (left), 2-level (right).

Straumanis technique of measurement to compensate for film shrinkage and errors in camera diameter. The photographs of the two end-members and of natural luzonite and famatinite from the two type localities and from Goldfield, Nevada are shown in Fig. 5. The indexed lines of the two photographs are tabulated in Table 2. It will be noted that considerable differences in intensity exist for some of the equivalent lines of luzonite and famatinite. It was possible to obtain sharper and clearer photographs of famatinite than of luzonite. However, the most important cause of this

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Luzonite				Famatinite					
Intensity	hkl	<i>d</i> measured	d calculated 1	Intensity	hkl	d measured	d calculated		
1	002	5.19 Å	5.240 Å	2	002	5.261 Å	5.380 Å		
3	101	4.73	4.724	3	101	4.731	4.811		
10	112	3.046	3.046	1	110	3.752	3.805		
4	103	2.928	2.914	10	112	3.071	3.106		
4	200	2.649	2.645	2	103	2.952	2.984		
3	004	2.605	2.615	5	$\begin{cases} 004 \\ 200 \end{cases}$	2.664	2.690		
1	213	1.954	1.959	2	202	2.390	2.405		
9	220	1.855	1.870	3	211	2.336	2.348		
7	312	1.592	1.592	2	114	2.181	2.197		
6	116	1.578	1.581	4	213	1.985	1.997		
2 2 5 4 4	224	1.520	1.522	8	220	1.895	1.903		
2	321	1.448	1.452	2	006	1.787	1.793		
5	400	1.321	1.323	2	301	1.763	1.768		
4	008	1.302	1.308	1	310	1.693	1.701		
4	332	1.212	1.213	7	312	1.614	1.622		
6	316	1.204	1.207	2	303	1.597	1.603		
2	420	1.179	1.183	3	224	1.547	1.553		
6	424	1.078	1.078	1	206	1.485	1.492		
6 2 6 5 4	228	1.071	1.072	2	321	1.469	1.477		
4 4	512 336	$\begin{array}{c}1.018\\1.014\end{array}$	$\begin{array}{c}1.017\\1.014\end{array}$	2 2	314	1.434	1.437		
3	1.1.10	1.004	1.014	5	323 ∫008∖	1.373 1.342	1.377 1.345		
2	440	.9349	.9350	1	$1400 \\ 402$	1.304	1.343		
4	408	.9293	.9305	2	411	1.294	1.295		
4	532	.8935	.8936	ĩ	330	1.266	1.268		
4	516	.8912	.8915	Ĝ	332	1.232	1.234		
4	$3 \cdot 1 \cdot 10$.8854	.8875	1	413	1.225	1.226		
4	620	.8363	.8367	4	420	1.200	1.203		
4	622	.8263	.8263	1	422	1.174	1.174		
4	536	.8049	.8047	1	307	1.164	1.166		
3	$3 \cdot 3 \cdot 10$.8007	.8016	1	334	1.145	1.147		
				1	415	1.114	1.115		
				6	424	1.099	1.098		
				1	431	1.072	1.070		
				1	510	1.055	1.055		
				5	512	1.037	1.035		
				1	521	.9960	.9948		
				2	523	.9628	.9638		
				4	440	.9491	.9516		
				5	516	.9105	.9097		
				1	$600 \\ 0 \cdot 0 \cdot 12 $.8974	.8967		
				2	611	.8815	.8813		
				2	534	.8736	.8730		
				1	613	.8588	.8584		
				5	620	.8510	.8506		
				1	$ \begin{cases} 446 \\ 622 \end{cases} $.8405	.8402		
				1	518	.8304	.8300		
				4	536	.8208	.8208		
				1	624	.8120	.8112		
				2	606	.8018	.8020		
				1	3.1.12	.7929	.7933		
				2	633	.7836	.7825		
				4	448	.7768	.7767		

 TABLE 2. INDEXED POWDER PHOTOGRAPHS OF SYNTHETIC LUZONITE

 AND FAMATINITE. Cu RADIATION, Ni Filter



FIG. 5. X-ray powder photographs of luzonite-famatinite. Cu radiation, Ni filter. 1. Synthetic famatinite. 2. Synthetic luzonite. 3. Luzonite, Mankayan, Luzon, Philippines. 4. Luzonite-famatinite, Goldfileld, Nevada. 5. Famatinite, Famatina, Argentina.

effect is undoubtedly the large difference in scattering power exhibited by arsenic atoms as compared to antimony atoms.

Luzonite and famatinite have a sphalerite-type structure, since their powder photographs closely resemble those of sphalerite. Chalcopyrite and stannite, both of which are tetragonal, also have a sphalerite-type structure. Both, moreover, have other properties similar to luzonitefamatinite: they are almost never untwinned and in polished section show a polysynthetic type of twinning, they have similar hardness and specific gravity, and they possess closely similar cell dimensions. All four minerals have indistinct cleavages and show very limited variation in composition. A tabulation of their comparative properties follows in Table 3. Assuming, then, that luzonite-famatinite had a structure closely similar to chalcopyrite or stannite, a determination of this structure involved the distribution of the atoms in luzonite-famatinite so as to comply with a body centered tetragonal cell, and the comparison of the theoretical intensity of reflections from such a cell with the measured intensity of natural material.

The structure of famatinite can be derived from two sphalerite unit cells, one on top of the other. As there are only two antimony atoms in a

TABLE 3. COMPARATIVE PROPERTIES OF LUZONITE, FAMATINITE, STANNITE,								
AND CHALCOPYRITE								

	Composition	Hardness	Specific Gravity	Cell dimensions		Space	Twinning
				a.	Go	group	- orthorne
Luzonite	Cu ₃ AsS ₄	3.5	4.35-4.50	5.29	10.465	142m	Polysynthetic, etc
Famatinite	Cu ₃ SbS ₄	3.5	4.50-4.65	5.38	10.76	$I\overline{4}2m$	Polysynthetic, etc
Stannite	Cu2FeSnS4	4.0	4.3 -4.5	5.46	10,725	$I\overline{4}2m$	Polysynthetic, etc
Chalcopyrite	Cu2Fe2S4	3.5-4.0	4.1 -4.3	5.24	10.30	$I\overline{4}2d$	Polysynthetic, etc

unit cell of famatinite, the problem of distributing these so as to give a body-centered tetragonal cell can be simply resolved by replacing the zinc atoms of the doubled sphalerite cell with antimony atoms at all the corners and at the center. The eight corner atoms contribute $\frac{1}{8}$ atom each, and the center one a full atom, for a total of two atoms. Copper atoms are then put in place of the remainder of the zinc atoms and the sulfur atoms remain undisturbed. This gives a cell containing $2(Cu_3SbS_4)$, as illustrated in Fig. 6. The derivation of luzonite and sphalerite is exactly analogous. The structure may also be derived from that of stannite



FIG. 6. Arrangement of atoms in the unit cell of luzonite-famatinite.

by substituting arsenic or antimony for the iron atoms and copper for the tin atoms.

Confirming this structure involved making an estimation of the intensity of the spots corresponding to each hkl value on the precession or Weissenberg photographs, and then calculating the intensities of reflections which would theoretically be derived from the assumed structure. Estimation of intensities was made entirely by visual comparison, and the results are correspondingly approximate. The estimated intensities for luzonite-famatinite were in good agreement with the intensities calculated from the assumed structure.

HABIT

Luzonite-famatinite is usually massive and fine-grained. More rarely, it is fairly coarse-grained, and some famatinite from Argentina occurs in massive granular intergrowths with single grains attaining 5 mm. in their largest dimension. Distinct equant crystals, found in vugs and druses and not exceeding 2 mm. in size (usually less than 1 mm.) are rare. Usually crystals are rough, with some or all faces curved and etched. It is more common to find tufts, rosettes, and botryoidal crusts of indistinct crystals lining vugs and druses. Crystals suitable for goniometric or x-ray single crystal work are invariably inconspicuous and have not been noted except in association with massive luzonite. It is understandable that earlier workers, with the exception of Sawada, were unable to find material suitable for describing the crystal structure and habit of this mineral series.

TWINNING

As previously noted, crystals of luzonite-famatinite usually show evidence of twinning and in polished section it exhibits polysynthetic twinning, similar in appearance to that found in albite. Since twinning is invariably very fine, it is difficult to secure mineral grains for x-ray single crystal work known to consist of a single individual. One twin law was determined by x-ray work on a twinned crystal of the interpenetration type, and subsequent confirmation of this law was obtained by morphological measurements made on the same crystal. The composition plane and the twinning plane is (112). It is not known if this twin law will account for the common polysynthetic twinning of luzonite-famatinite seen in polished section.

PHYSICAL PROPERTIES

Cleavage: (101), good; (100), distinct. Cleavage is not usually observed because of the fine grain of natural material. Fracture: uneven, conchoidal. Tenacity: brittle. Hardness: about 3.5. Specific gravity: the value measured on luzonite from Mankayan, Luzon, with the Berman balance is 4.380; the value calculated from the x-ray data is 4.438. The value measured on famatinite from Famatina, Argentina, by pyknometer is 4.635; the value calculated is 4.660.

OPTICAL PROPERTIES

Color: deep pinkish brown. Similar to bornite, although darker and with more of a greyish tinge. Streak: black. Luster: usually dull, metallic. Opaque. In polished section pale brownish pink in color. Shows weak pleochroism and strong anisotropism. Polarization colors: greenish yellow and purplish red. Different members of the luzonite-famatinite series did not show any differences in color, luster, streak, or behavior under polarized light.

CHEMICAL PROPERTIES

Luzonite is a copper sulf-arsenide corresponding to the formula Cu_3AsS_4 . Famatinite is a copper sulf-antimonide, Cu_3SbS_4 . A complete solid solution series exists between the two end-members. Luzonite and enargite have identical formulas, and hence are dimorphs. There is no mineral with enargite type structure having the composition of famatinite, although a maximum of about 6% Sb will enter into the enargite structure, as determined from scores of analyses of natural crystals.

Neither luzonite nor famatinite have been found in absolutely pure masses. Even the purest obtainable material reveals, in polished section under high magnification, minor amounts of accessory materials; most commonly pyrite, tetrahedrite-tennantite, chalcopyrite, and covellite. Although the total of these impurities may be 1% or less, they serve to obscure the nature of elements which may substitute in the luzonite-famatinite structure, for they cannot be separated from material being prepared for analysis. Many of the old analyses, made on material even more difficult to purify than that used by the writer, were recalculated after subtracting anywhere from 10% to over 50% for insoluble material or pyrite.

The samples analyzed were all hand-picked under a microscope from -40 mesh material. All grains showing any foreign mineral attached or included were rejected. A representative sample of the purified material was then reserved for making a polished section, and the remainder was analyzed. Any minor recalculation of the analysis was based on the mineral impurities shown to have been present in the polished section. Analyses are listed in Table 4.

Spectrographic analysis of the purest material available, from Famatina, Argentina, showed, in addition to copper, arsenic, antimony, and iron, the following elements:

.1%-.01%-Al, Ca, Ti, Si

.01%-.001%-Mg, Na, K, Bi, Mn, Zr, Pb, Cr, Mo, V, Cb, Ta, Ag, Au, Ba

The most important point brought out by this analysis is the very small amount of such elements as Bi, Pb, and Ag present, and the spectroscopic absence of such heavy elements as Zn, Cd, and Sn, which might normally be expected to be present in traces in most base metal deposits. Actually, the mine from which this specimen came is primarily a silver-gold mine. Galena and sphalerite are minor accessories in the ore. The indication would seem to be, therefore, that the amount and

	1	2	3	4	5	6	7	8	
Cu	48.42	47.55	48.32	43.94	45.72	45.70	42.98	43.27	
Fe		.26		.48			.26		
Bi				1.79					
Sb		1.46	1.48	13.19	13.72	14.59	24.36	27.63	
As	19.02	17.07	17.35	9.08	9.45	8.98	3.31		
S	32.56	32.63	32.85	30.86	31.11	30.73	28.97	29.10	
Rem.		.97		.17					
Total	100.00	99.94	100.00	99.51	100.00	100.00	99.88	100.00	
-	ALL NOT THE								

TABLE 4. ANALYSES

1) Cu_3AsS_4 .

2) Mankayan, Luzon, Philippines. Gonyer analysis.

3) Analysis #2, recalculated after deduction of .97% insol. and .56% pyrite.

4) Goldfield, Nevada. Gonyer analysis.

5) Analysis #4, recalculated after deduction of .17% insol., 1.03% pyrite, and 2.20% bismuthinite.

6) $Cu_3As_{1/2}Sb_{1/2}S_4$.

7) Sierra Famatina, Argentina. Gonyer analysis.

8) Cu₃SbS₄.

kind of substitution of other elements for Cu, As, or Sb in the luzonitefamatinite structure is very limited. Probably even the iron usually reported is chiefly due to small amounts of admixed pyrite, chalcopyrite, or tetrahedrite. The chemical behavior of this group therefore contrasts markedly with that of the tetrahedrite-tennantite group, in which extensive substitution of other elements for Cu, As, and Sb is the rule.

Occurrence

Members of this series are found in low to medium intensity copper deposits, particularly in the former, or in lower-intensity phases of higher intensity deposits. They are generally absent from copper deposits of the hypothermal, pyrometasomatic, or mesothermal "porphyry" type. The most common associated sulfide minerals are: enargite, tetrahedritetennantite, pyrite, chalcopyrite, covellite; and more rarely sphalerite, bismuthinite, ruby silvers, native silver, gold, and marcasite. Among gangue minerals barite and drusy quartz are so common as to be almost characteristic. Alunite is also found in some places.

The most important localities are Mankayan, Luzon; Famatina, Argentina; Goldfield, Nevada; Hokuetsu, and other localities in Japan; Kinkwaseki, Formosa; Cerro de Pasco, Peru; Morococha, Peru; and minor quantities are found at Butte, Montana and Cananea, Mexico. Measurable crystals have been observed only from Mankayan, Luzon; Goldfield, Nevada; and Hokuetsu, Japan.

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