Canadian Mineralogist
Vol. 22, pp. 383-390 (1984)

OPTICAL DISPERSION AND ZONING IN MAGMATIC PLAGIOCLASE: LASER-INTERFERENCE OBSERVATIONS

THOMAS H. PEARCE
Department of Geological Sciences, Queen's University, Kingston, Ontario K7L 3N6

Abstract

Using the data of Tsuboi and others, the dispersion of the indices of refraction has been calculated for the plagioclase series at wavelengths of the common lasers (HeNe and Argon-ion). Sellmeier's equation fits the experimental data quite well. Errors are typically between 2 × 10^{-4} to 1 × 10^{-5}, usually well within the experimental error. Using these data and a new laser-interference microscope, the shape of typical oscillatory zoning in magmatic plagioclase from some andesites has been determined from the profile of the index of refraction. The zoning is complicated, yet a simplified pattern may be discerned. Repeated periods of resorption are extremely common in andesitic feldspar.

Keywords: laser, index of refraction, dispersion, plagioclase, zoning, interference microscope.

Sommaire

On présente, à partir des données de Tsuboi et d'autres auteurs, les coefficients de dispersion des indices de réfraction de la série des plagioclases pour les longueurs d'onde des lasers courants (HeNe et Ar ionisés). L'équation de Sellmeier prédit bien les données expérimentales. L'écart-type dans l'indice de réfraction, entre 2 × 10^{-4} et 1 × 10^{-5}, est généralement inférieur à l'erreur expérimentale. L'application de ces données aux observations par microscopie à interférence de lasers permet de délimiter, par le profil des indices de réfraction, la forme des zones oscillatoires du plagioclase magmatique de certaines andésites. La zonation est complexe en détail, mais on peut discerner une zonation générale assez simple. Les périodes de résorption répétées sont très fréquemment observées dans le plagioclase andésitique.

(Traduit par la Rédaction)

Mots-clés: laser, indice de réfraction, dispersion, plagioclase, zonation, microscopie à interférence.

Introduction

Lasers are becoming fairly common light sources in many laboratories. Common lasers, such as the HeNe and argon-ion types, produce spectrally pure light of wavelengths different than were used in laboratories a generation ago. Because of dispersion, the index of refraction of minerals varies with wavelength and is quite different in laser light and sodium light, for example. In this work, I present a recalculation of the optical dispersion for the magmatic plagioclase series using the original data of Tsuboi and others, reported in Tsuboi's 1923 paper. I use these essential data to determine the shape of the oscillatory zoning in intermediate plagioclase phenocrysts from some andesites from South America and the Cascades of Washington and Oregon. The latter data have been obtained using a multiple frequency laser-interference microscope of my design, located in my laboratory at Queen's University. New data on dispersion of indices of refraction of magmatic plagioclase are now being collected by the writer and P.L. Roeder, and will be published at a later date.

In this present work, I will assume that the reader is familiar with the problems of zoning in magmatic plagioclase, as outlined in Smith (1974) and discussed by Bottinga *et al.* (1966), Ewart (1963), Homma (1932) and Leedal (1952), for example. Since I am concerned with the empirical shape of the zones, I will make no assumptions about their origin, although it may be possible to reach some preliminary conclusions.

Calculation of Dispersion

Calculation of the variation in the index of refraction with wavelength for transparent substances may be found using Cauchy's (1) or Sellmeier's (2) equations (Bloss 1961):

$$N = B + C/\lambda^2 \qquad (1)$$

$$N^2 = 1 + \frac{A\lambda^2}{\lambda^2 + \lambda_0^2} \qquad (2)$$

where N is the index of refraction for a given wavelength λ, and A, B, C and λ_0 are constants that vary for each composition of plagioclase.

Sellmeier's equation is somewhat sounder in its theoretical basis (Born & Wolf 1980); experience has shown that it predicts known indices of refraction with less error than Cauchy's equation. Accordingly, I have taken the original data used by Tsuboi (1923) and calculated the dispersion of the indices of plagioclase for the wavelengths of the most common lasers. The errors generated by fitting the data to Sellmeier's model range from 2 × 10^{-4} to 1 × 10^{-5} in the index of refraction over the complete

TABLE 1. DISPERSION IN PLAGIOCLASE: α AS A FUNCTION OF λ

MOLE %	REFRACTIVE INDEX FOR A GIVEN COLOR (ληm)					SELLMEIER'S CONSTANTS	
An%	632.8	589.3	514.5	488.0	457.9	A	λ_o
0.4	1.52721	1.5289	1.53279	1.53466	1.53720	1.30027	98.23065
2.5	1.52763	1.5292	1.53300	1.53479	1.53723	1.30272	96.37543
3.3	1.52793	1.5293	1.53290	1.53456	1.53682	1.30586	92.83870
3.6	1.52824	1.5300	1.53407	1.53602	1.53868	1.30200	100.25722
24.0	1.53861	1.5403	1.54443	1.54638	1.54903	1.33364	99.33513
37.0	1.54315	1.5450	1.54926	1.55131	1.55410	1.34586	101.37803
62.3	1.56018	1.5623	1.56691	1.56917	1.57225	1.39477	104.87075
92.6	1.57191	1.5736	1.57811	1.58018	1.58300	1.43425	99.89054
95.4	1.57173	1.5738	1.57847	1.58073	1.58381	1.43059	104.03186

TABLE 2. DISPERSION IN PLAGIOCLASE: γ AS A FUNCTION OF λ

MOLE %	REFRACTIVE INDEX FOR A GIVEN COLOR (ληm)					SELLMEIER'S CONSTANTS	
An%	632.8	589.3	514.5	488.0	457.9	A	λ_o
0.4	1.53744	1.5392	1.54321	1.54514	1.54778	1.33031	99.05979
2.5	1.53781	1.5393	1.54307	1.54483	1.54722	1.33430	94.68712
3.3	1.53728	1.5390	1.54309	1.54503	1.54768	1.32964	99.33872
3.6	1.53855	1.5401	1.54400	1.54582	1.54829	1.33558	96.18247
24.0	1.54639	1.5481	1.55212	1.55403	1.55664	1.35797	97.98808
37.0	1.55106	1.5529	1.55710	1.55912	1.56187	1.37055	100.18883
62.3	1.56918	1.5713	1.57593	1.57817	1.58125	1.42269	104.18228
92.6	1.58524	1.5872	1.59204	1.59431	1.59741	1.47260	103.41113
95.4	1.58500	1.5872	1.59224	1.59466	1.59797	1.46935	106.57982

visible spectrum. These errors are usually well within the experimental error associated with the determination of the indices. Errors tend to be somewhat larger toward the blue end of the spectrum.

The results of these calculations are presented in Tables 1 and 2. Unfortunately, Tsuboi's data do not include β, which thus has to be estimated. A rough estimate of β is obtained by averaging α and γ.

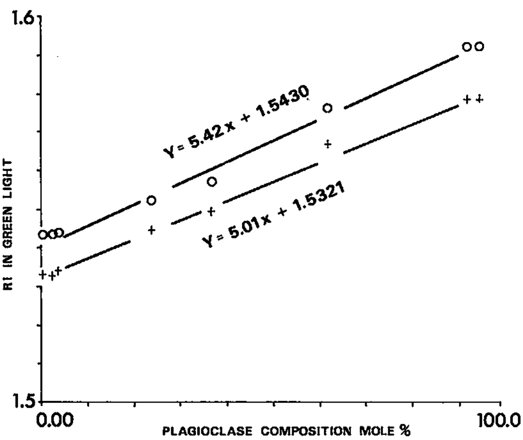

FIG. 1. Dispersion of indices of refraction α (lower line) and γ (upper line) with composition in the plagioclase series. Data from Tsuboi (1923) recalculated for green light of wavelength 514.5 nm (argon-ion laser line) according to Sellmeier's equation (see Tables 1 and 2).

TABLE 3. LINEAR FIT OF REFRACTIVE INDEX AND COMPOSITION FOR A GIVEN COLOR

			WAVELENGTH IN NANOMETERS				
			632.8	(589.3)	514.5	488.0	457.9
	α	SLOPE	4.88447	4.92293	5.01015	5.05259	5.11068
		INTERCEPT	1.52663	1.52825	1.53211	1.53395	1.53644
(EST.)	β	SLOPE	5.04114	5.08623	5.18625	5.23504	5.30210
		INTERCEPT	1.53115	1.53278	1.53664	1.53848	1.54098
	γ	SLOPE	5.19781	5.24971	5.36234	5.41749	5.49353
		INTERCEPT	1.53567	1.53731	1.54117	1.54301	1.54552

For any given composition, the dispersion curve as given by Sellmeier's equation is definitely not linear (Sellmeier's constants are given in Tables 1 and 2). However, if one holds the wavelength constant, then one obtains a linear relationship between index of refraction and composition. The fit to a straight line is quite good, as may be seen from Figure 1, a graph of α and γ as a function of composition. The slope and intercept of the linear fit for the common laser lines, as well as for the sodium line (589.3 nm), are given in Table 3.

OSCILLATORY ZONING

The general method of interference microscopy and its application in geology have been explained by Chao (1976), and the laser-interference technique used in this study has been reported by Pearce (1982b, 1983a). The interested reader is referred to these works. Preliminary work on plagioclase zoning has been reported (Pearce 1982a,c,d).

Using a normal thin section and by suitably adjusting the interference microscope to produce the wide-fringe mode of observation, one may obtain a picture of the zonation of plagioclase contoured with respect to index of refraction and, hence, composition. Figure 2 shows a plagioclase grain from a Chilean andesite (PT–5, Queen's petrology collection), collected by M. Zentilli and A.H. Clark. In studying the pattern of zoning of crystals such as this, I letter the discontinuities from the rim (A) in toward the core. The zones are identified by the discontinuity on the outermost side of the zone. The AB zone, therefore, is the outer rim of the crystal. The BC zone is the zone immediately inside it, and separated from the A zone by the B discontinuity and from the next innermost zone BC by the C discontinuity. In this system of nomenclature, I number any finer features within the zones thus: AB–1, AB–2. This form of nomenclature from youngest to oldest, the reverse of the usual type of nomenclature, is rendered necessary by the random nature of sections through the phenocrysts. The rim is always present in any section that cuts a crystal, but the exact centre of the crystal may only be present in very few sections (if at all). Equations for predicting the probability of a random section being close to a desired section have been published by Pearce (1983b). Calculations

FIG. 2. Wide-fringe laser interferogram of a plagioclase phenocryst from a Chilean andesite, double-beam interference in green light (514.5 mm). The distance from the centre R to the rim A is 1 mm.

show that depending on the number of crystals present, the centre of a crystal may not be present in any given thin section. Therefore, it is not possible to begin labeling from the centre outward.

In Figure 2, the apparent centre of the crystal is R, and the distance R–A is 1000 micrometres. Note that the dark and light patterns seem to appear in pairs between discontinuities, thus giving an apparently double nature to the zones from A to D and from I to O. Zones between E and I are very narrow, and it is difficult to subdivide such fine and complex zones. Nevertheless, a system of nomenclature based on the discontinuities seems to be practical. When these observations are combined with data obtained using the narrow-fringe method (see below), a general pattern of zonation appears to be common.

As is apparent, the wide-fringe technique is well suited to a display of the details of the zonation of such crystals (each shade is a different composition of plagioclase). However, this method is not well suited to quantitative measurements, since estimates of the fringe shift (change in index of refraction) are difficult to obtain.

Once suitable adjustments are made to the interference microscope, narrow fringes are obtained (the narrow-fringe mode of observation). This method yields quite a different sort of interferogram, as illustrated in Figure 3. It is suited to quantitative analysis but requires dispersion data of the sort listed in Tables 1 and 2. Using these data and the fundamental equation (3) below, it is possible to

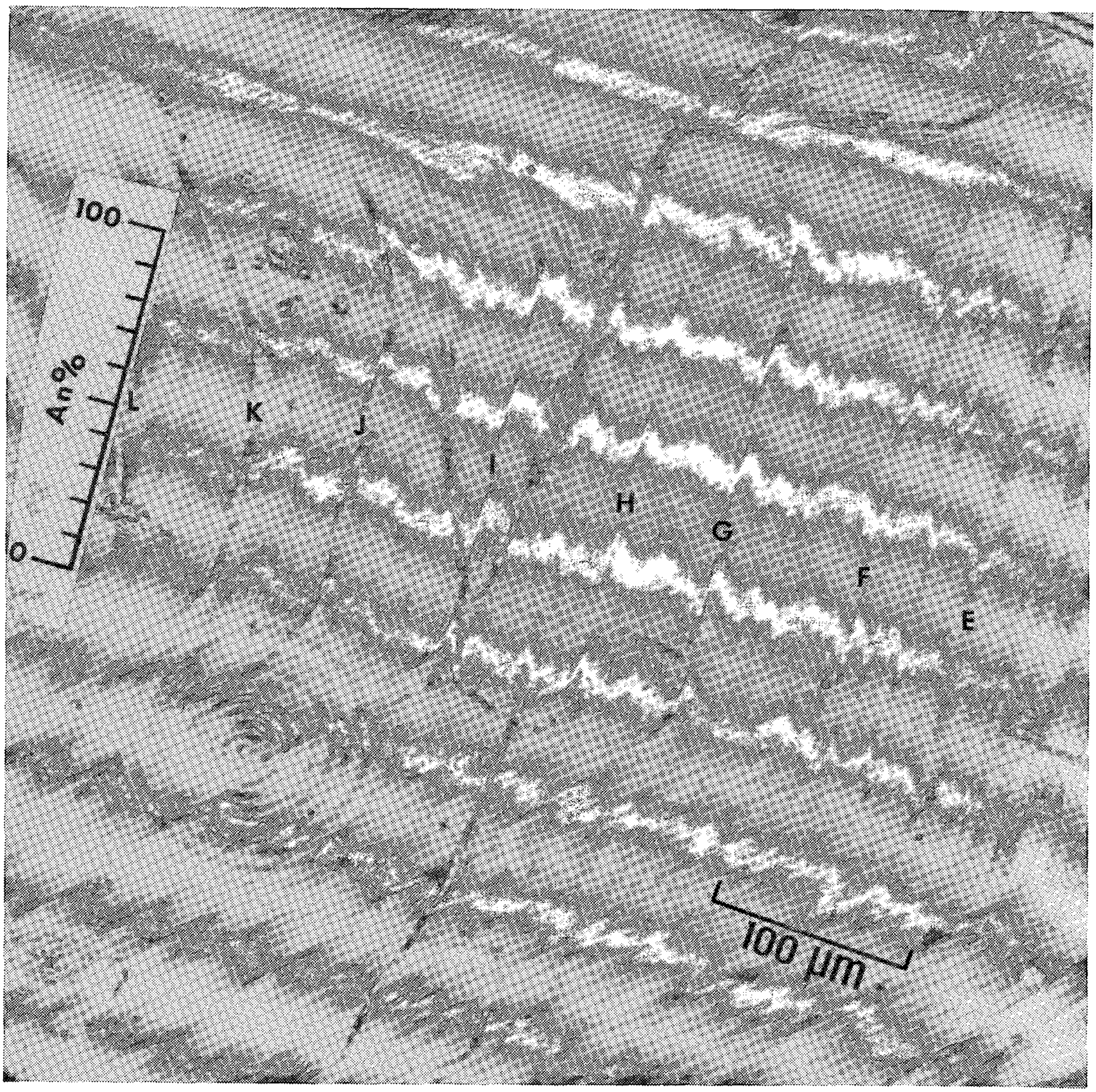

FIG. 3. Narrow-fringe laser interferogram (514.5 mm green line) of the same crystal as in Figure 2. The IJ zone is 65 μm wide. The approximate scale of %An for the lettered fringe is as indicated. The jump in %An across the lettered discontinuities is about 7%. The trace of the fringe is a graph of the %An across the crystal.

TABLE 4. NUMBER OF WAVELENGTHS SHIFT FOR 100% An DIFFERENCE AT DIFFERENT WAVELENGTHS FOR AN OPTICAL PATH OF 30 μm

RI	632.8	(589.3)	514.5	488.0	457.9
α	2.3154	2.5061	2.9213	3.1061	3.3483
γ	2.4642	2.6725	3.1267	3.3304	3.5991

calculate the An% of a plagioclase zone from the fractional shift as the fringes cross the fringe boundary, as in Figure 3:

$$N1 - N2 = S \times \lambda/T \quad (3)$$

where $N1$ and $N2$ are the indices of refraction of two phases, S is the shift in apparent wavelength, T is thickness and λ is the wavelength of the light used.

Indeed, if oriented correctly, the fringes form a graph of %An. Following the trace of a fringe across the phenocryst in Figure 3 outlines the change in composition across the zones. The sensitivity of this method for plagioclase in a section of normal thickness (30 μm) is given in Table 4, which indicates the maximum shift possible (An_{100} to An_0). As a general rule, fractional shifts of 1/10 to 1/20 of a wave may be readily measured, and in special cases 1/1000 of a wave has been reported in the literature (Steel 1967). This probably represents the ultimate sensitivity for the technique. Table 5 lists the actual shift factor F, which allows calculation of the An% difference in the zone. Absolute values may be obtained if an internal standard (such as quartz or a known mounting medium) is available, preferably in contact with the feldspar.

The compositional changes across the discontinuities from F to K are examined in Figure 3. The shift is about the same in all cases, and the high value of An content is on the outermost side of the discontinuity. For example, for the plagioclase on either side of discontinuity H, the shift is ¼ of a wave. Using the data from Table 5 for observations in green light, this corresponds to a difference of 8.25% An. Electron-probe data for these zones give 40% An for zone HI and 47% An for zone GH, a difference of 7% An. This is actually quite good agreement because the electron beam samples a somewhat larger volume of material than is resolved optically.

In examining the shape of the zones in Figure 3, note that the lines labeled G, H, *etc.*, mark major discontinuities in the crystal and represent events at which the crystal stopped growing one composition and began to grow plagioclase of another (usually more calcic) composition. The time represented by the change from one composition to another is not known. Locally, however, angular unconformities and cut-and-fill structures leave no doubt that the surfaces are due to solution (in stratigraphic terms, a hiatus). Thus the time interval represented by the surface may be substantial. In some cases (not illustrated in this present work), judging from the depth of the embayments, as much as 10% or more of the crystal may have dissolved. Clearly, this is not an insignificant feature. In other cases, little of the crystal appears have been dissolved. This observation requires more study.

After staring at more of these fringes than the author cares to remember, it appears that a general pattern exists. This idealized general pattern is illustrated in Figure 4a in its most complete form, and schematically in Figure 4b. Note that the growth of the crystal is from left to right in this figure. The scale of the zoning is of the order of 7 to 12 %An, and the width of the zones usually varies with an order of magnitude from 10 to 100 micrometres (zone IJ is 65 μm wide), although there may be finer fluctuations within the larger zones. Broadly speaking, an idealized form of zoning may be described as follows. After an event that led to a discontinuity (indicated by D in Fig. 4), the crystal abuptly formed a more calcic layer (part I of Fig. 4b). The %An steadily decreases outward, and may reach a plateau (part II). The %An then increases slightly (part III) prior to a "final" decrease (part IV). Another discontinuity D and an abrupt increase in %An (rising to about the same composition as in the previous case) begin the next cycle.

It is clear from the shape of the fringes in Figure 3 that this postulated generalized pattern is often cut short by successive discontinuities, such as illustrated in Figure 4c, in which the complexities of the naturally occurring zones mask somewhat the generalized pattern described above. The idealized pattern may also be distorted slightly. For example, in rare cases the third part of the sequence (part III) may increase to a higher An content than part I (see zones GH and JK in Fig. 3). Locally, part II may form a relatively large plateau, as in zone IJ in Figure 3. Such a zone of constant composition might represent the "steady state" postulated by Bottinga *et al.* (1966).

In some cases, parts I and II form a distinct subzone different from parts III and IV, but without any obvious discontinuity between them (the line indicated by B in Fig. 4b; see zones G to L already referred to). Such "double zones" and even some multiple zones are fairly common in some rocks, but their significance is not known. These patterns may

TABLE 5. FRACTIONAL SHIFT FACTOR "F" (An % = F x SHIFT)

RI	632.8	(589.3)	514.5	488.0	457.9
α	43.19	39.90	34.23	32.19	29.86
β (EST).	41.88	38.66	33.10	31.11	28.80
γ	40.58	37.41	31.98	30.02	27.78

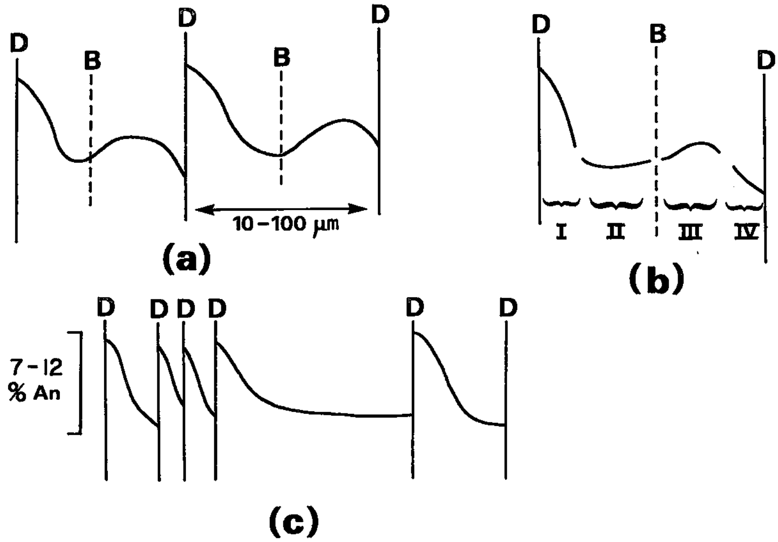

FIG. 4. An idealized model of the generalized pattern of distribution of An content in the oscillatory zones studied in this work. Figure 4a shows the generalized pattern of a complex double zone, which is schematically illustrated in Figure 4b. Figure 4c shows some of the complexities that have been noticed . Discontinuities are indicated by D, whereas changes in composition not accompanied by a discontinuity are indicated by B.

all be variations on a general theme, as indicated in Figure 4b.

DISCUSSION

In this preliminary report, I have shown that laser-interference microscopy is a useful tool in studies of plagioclase zoning. It is advantageous in studies of this sort to be able to see the composition of the zones represented either as contours or as a curve in profile. Some previous workers, using conventional optical techniques, have determined the correct general appearance of the zoning profile (Homma 1932, Leedal 1952). However, the use of optical-extinction angles to obtain detailed profiles of the zonation may lead to inconsistent results (see Ewart 1963, p. 418, for example). These difficulties may be overcome using the narrow-fringe method of laser interferometry. Complex double zones appear not to have been noted previously, although they are fairly common in many of the andesites studied by the writer.

Recently, several numerical simulations based on diffusion models have been published as an extension and modification of the earlier work of such investigators as Harloff (1927), Bottinga *et al.* (1966) and Sibley *et al.* (1976): see, for example, Haase *et al.* (1980, Fig. 2), Lasaga (1982, Figs. 8, 10) and Loomis (1982, Fig. 13). Whereas results of this type of work are encouraging, these numerical simulations seem to have produced results that are quite different from the natural pattern of zonation with regard to the exact form and, in some cases, even the direction of compositional changes. Clearly, more effort must be devoted to determining the exact shape and variety of naturally occurring zonation, both to firmly establish its nature and to serve as a guide in studies of numerical modeling.

Major discontinuities in the zonation, some of which are unquestionably associated with important events of solution, should be assigned a more significant role in developing models of plagioclase zonation. The irregular periodicity of these discontinuities (*i.e.*, of solution events ?) appears to indicate that some control external to the local environment of the growing crystal is important. Indeed, it is tempting to infer that the surfaces of major solution are a result of volcanic eruptions (and consequent intrusion), as earlier suggested by Ewart (1963) and Wiebe (1968). The fine-scale zoning (< 10 μm across) between major discontinuities may be due to a complex interplay of local diffusion gradients set up by the growing crystal and, possibly, fluctuations in water pressure, as suggested by numerous authors (see review in Smith 1974). With regard to the significance of solution surfaces, one might expect that the essentially regular nature of diffusion-related zoning would be complicated by a release of total pressure as the magma moves upward in the magma column prior to an eruption. The irregularly periodic nature of the discontinuities that I have drawn in Figure 4 are consistent with such an interpretation. Plagioclase with the most complicated zoning tends to come from andesitic and dacitic magmas, whose propensity for violent and frequent eruption is notorious. This empirical field observation should not be overlooked in theoretical simulations.

It is hoped that the results of this present note will help clarify the complex nature of oscillatory zoning in plagioclase and lead to further work. A numerical analysis of the form of the zoning profiles would seem to be a logical result of this type of study. Much more research needs to be done on this fascinating problem.

Acknowledgements

The research in this study was supported by Natural Sciences and Engineering Research Council of Canada (operating grant A8709). It is a result of theoretical work done in connection with studies of zoned crystals using a new laser-interference microscope (Canadian patent #394,168, Laser Interferometer). The laser-equipped microscope laboratory at Queen's University was funded by an NSERC New Research Idea grant (A5226) and an NSERC equipment grant (E6622). I thank Professor P.L. Roeder for analyzing the plagioclase in some specimens. Personnel costs and some equipment costs for the initial part of this research were supported by grants from the Graduate School and Advisory Research Committee, Queen's University. Photography and drafting were done by Chris Peck.

References

Bloss, F.D. (1961): *An Introduction to the Methods of Optical Crystallography*. Holt, Rinehart & Winston, New York.

Born, M. & Wolf, E. (1980): *Principles of Optics* (6th Edition). Pergammon Press, New York.

Bottinga, Y., Kudo, A. & Weill, D. (1966): Some observations on oscillatory zoning and crystallization of magmatic plagioclase. *Amer. Mineral.* **51**, 792-806.

Chao, E.C.T. (1976): The application of quantitative interference microscopy to mineralogic and petrologic investigations. *Amer. Mineral.* **61**, 212-228.

Ewart, A. (1963): Petrology and petrogenesis of the Quaternary pumice ash in the Taupo area, New Zealand. *J. Petrology* **4**, 392-431.

Haase, C.S., Chadam, J., Feinn, D. & Ortoleva, P. (1980): Oscillatory zoning in plagioclase feldspar. *Science* **209**, 272-274.

Harloff, E.A. (1927): Zonal structure in plagioclases. *Leidsche Geol. Med.* **2**, 99-113.

Homma, F. (1932): Über das Ergebnis von Messungen an zonaren Plagioklasen aus Andesiten mit Hilfe des Universaldrehtisches. *Schweiz. Mineral. Petrog. Mitt.* **12**, 345-352.

Lasaga, A.C. (1982): Toward a master equation in crystal growth. *Amer. J. Sci.* **282**, 1264-1288.

Leedal, G.P. (1952): The Cluanie igneous intrusion, Inverness-shire and Ross-shire. *J. Geol. Soc. London* **108**, 35-63.

Loomis, T.P. (1979): An empirical model for plagioclase equilibrium in hydrous melts. *Geochim. Cosmochim. Acta* **43**, 1753-1759.

______ (1982): Numerical simulations of crystallization processes of plagioclase in complex melts: the origin of major and oscillatory zoning in plagioclase. *Contr. Mineral. Petrology* **81**, 219-229.

Pearce, T.H. (1982a): Laser interferometric observations of plagioclase from Mt. St. Helens. *Geol. Assoc. Can. - Mineral. Assoc. Can. Program Abstr.* **7**, 72.

______ (1982b): Multiple frequency laser interference microscopy: a new technique for mineralogy. *Geol. Assoc. Can. - Mineral. Assoc. Can. Program Abstr.* **7**, 72.

______ (1982c): Observations of plagioclase phenocrysts from andesites using multiple frequency laser interference microscopy. *Geol. Soc. Amer. Abstr. Program* **14**, 585.

______ (1982d): Direct observations of feldspar zoning using laser interference microscopy. *Amer. Geophys. Union Trans.* **63**, 1136 (abstr.).

______ (1983a): Multiple frequency laser interference microscopy: a report. *The Microscope* (in press).

______ (1983b): The analysis of zoning in magmatic crystals, with emphasis on olivine. *Contr. Mineral. Petrology* **86**, 149-154.

Sibley, D.F., Vogel, T.A., Walker, B.M. & Byerly, G. (1976): The origin of oscillatory zoning in plagioclase: a diffusion and growth controlled model. *Amer. J. Sci.* **276**, 275-284.

SMITH, J.V. (1974): *Feldspar Minerals. 2. Chemical and Textural Properties*. Springer-Verlag, New York.

STEEL, W.H. (1967): *Interferometry*. Cambridge University Press, London, England.

TSUBOI, S. (1923): A dispersion method of determining plagioclase in cleavage flakes. *Mineral. Mag.* **20**, 108-122.

WIEBE, R.A. (1968): Plagioclase stratigraphy: a record of magmatic conditions and events in a granite stock. *Amer. J. Sci.* **266**, 690-703.

Received August 23, 1983, revised manuscript accepted December 21, 1983.