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## Mössbauer Studies of $\text{BiFeO}_3\text{-PbTiO}_3$ Perovskite-Type Solid Solutions\*†

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The Mössbauer spectra of the ferroelectric solid solutions  $(\text{BiFeO}_3)_x(\text{PbTiO}_3)_{1-x}$  have been studied as a function of temperature, mainly between room temperature and  $600^\circ\text{C}$ , using  $^{57}\text{Fe}$  in the solutions as an absorber. The quadrupole splitting increased and the isomer shift decreased between  $x=0.05$  and  $x=0.70$ , a range in which the solutions are tetragonal and paramagnetic, and in which the unit cell volume increases with increasing  $x$ . The isomer shift, in particular, was primarily determined by the unit cell volume. Data are presented on the internal magnetic field of the antiferromagnetic solutions in the rhombohedral phase between  $x=0.7$  and  $x=1.0$ . The results demonstrate the similarity and continuity in internal structure between the ferroelectrics  $\text{PbTiO}_3$  and  $\text{BiFeO}_3$ .

### INTRODUCTION

$\text{PbTiO}_3$  and  $\text{BiFeO}_3$  form a continuous series of solid solutions,<sup>1</sup> all distorted perovskites at room temperature, which have been investigated using x-ray<sup>1,2</sup> and neutron diffraction,<sup>3,4</sup> as well as dielectric and magnetic susceptibility<sup>5,6</sup> measurements. The interest in the system results from the high ferroelectric Curie points and distortions of the solid solutions, as well as from the fact that compositions rich in  $\text{BiFeO}_3$  are antiferromagnetic and show weak ferromagnetism. The structural parameters and dielectric properties of the solid solutions were given most recently by Smith *et al.*<sup>2</sup> Briefly,  $\text{PbTiO}_3$  is a ferroelectric perovskite with tetragonal distortion, a  $c/a$  ratio of 1.063<sup>7,8</sup> at room temperature, and a Curie point of  $490^\circ\text{C}$ .<sup>9</sup>  $\text{BiFeO}_3$  is also a ferroelectric perovskite with rhombohedral distortion and a Curie point of about  $850^\circ\text{C}$ .<sup>1,2</sup> There had in the past been doubt as to whether  $\text{BiFeO}_3$  should be classified as ferroelectric, but a recent demonstration that single crystals of the material undergo dielectric hysteresis<sup>10</sup> has resolved this question. In addition,  $\text{BiFeO}_3$  is antiferromagnetic with a Néel temperature of  $370^\circ\text{C}$ .

The properties of solid solutions of the two materials are extraordinary. Usually, when solid solutions of two ferroelectric perovskites  $(\text{ABO}_3)_x(\text{A}'\text{B}'\text{O}_3)_{1-x}$  are made, the ferroelectric properties of the end members are weakened for the mixtures. That is, the Curie points, the spontaneous polarizations, and the spontaneous strains decrease for increasing  $x$  near  $x=0$ , and also decrease for decreasing  $x$  near  $x=1$ . There is often a central compositional region which can only be classified as pseudocubic, because there are random distortions which are not well defined. [A relatively continuous variation in ferroelectric properties may occur, however, when A and A' are identical as in  $(\text{PbTiO}_3)_x(\text{PbZrO}_3)_{1-x}$ , or when B and B' are identical as in  $(\text{BaTiO}_3)_x(\text{PbTiO}_3)_{1-x}$ .] The solid solutions  $(\text{BiFeO}_3)_x(\text{PbTiO}_3)_{1-x}$  show a relatively uniform and unanticipated increase in Curie point with  $x$  between  $x=0$  and  $x=1$ . Moreover the tetragonal distortion, measured at room temperature, increases strongly

from  $x=0$  to  $x=0.7$ , where the tetragonal  $c/a$  ratio is about 1.2. One need only compare this value with the  $c/a$  ratio in tetragonal  $\text{BaTiO}_3$ , which is 1.01, to realize the unusual strength of this effect. For  $x$  greater than 0.7, the solutions become rhombohedral in symmetry. Mössbauer spectrometry, using  $^{57}\text{Fe}$  in the solid solutions as the absorber, offers an important tool for the investigation of the environment of the iron atom in these solid solutions and for the correlation of such data with the dielectric and magnetic properties of the compounds.

### EXPERIMENTAL

The Mössbauer spectra were obtained with an Austin Science Associates conventional spectrometer operated with an RIDL-400 multichannel analyzer. A Mössbauer source in a copper matrix, nominally 10 mCi, was used during the course of the study. For every spectrum, approximately 400 000 counts were accumulated in each channel, so that the statistical error never exceeded 0.15%. The apparatus was mounted vertically to enable the use of a powder sample with a minimum absorption loss due to the sample holder. The absorbers were prepared by spreading the powder on a thin beryllium foil using acetone as a suspension medium. A vibromet polishing wheel was used to spread the powder uniformly on the beryllium foil. During the experiment the absorber was heated in a short tube furnace. The sample temperature was monitored with a Chromel-Alumel thermocouple to  $\pm 3^\circ\text{C}$ .

The solid solutions were prepared by reacting stoichiometric amounts of the constituent oxides, wrapped in Pt foil, in a morganite crucible. The  $\text{Fe}_2\text{O}_3$  was enriched to 80%-90%  $^{57}\text{Fe}$ . The samples were fired between  $700^\circ$  and  $1050^\circ\text{C}$  for a period of two hours. Samples with a higher concentration of  $\text{BiFeO}_3$  were sintered at lower temperatures. The solid solutions were air quenched and characterized by x-ray diffraction analysis.

The Mössbauer spectra were analyzed, using a least-squares fit to a linear approximation of the Lorentzian

TABLE I. Summary of the experimental results from the Mössbauer spectra for the solid solutions with magnetic phases. ( $^{57}\text{Co}/\text{Cu}$  source at room temperature).

Percent $\text{BiFeO}_3$	Absorber temp. ( $^{\circ}\text{K}$ )	Site	Isomer shift relative to metallic iron (mm/sec)	Quadrupole interaction (mm/sec) ( $\Delta_{12}-\Delta_{36}$ )	Effective magnetic field (kOe)	Néel temp. ( $^{\circ}\text{K}$ )
74	298	Paramagnetic	$0.30 \pm 0.02$	$0.47 \pm 0.02$	0	$458 \pm 5$
	298	Magnetic	$0.39 \pm 0.04$	$0.20 \pm 0.04$	$415 \pm 10$	
	526	Paramagnetic	$0.22 \pm 0.02$	$0.49 \pm 0.02$	0	
80	298	Magnetic	$0.38 \pm 0.04$	$0.22 \pm 0.04$	$447 \pm 10$	$495 \pm 5$
	328	Magnetic	$0.35 \pm 0.04$	$0.16 \pm 0.04$	$412 \pm 10$	
	370	Magnetic	$0.33 \pm 0.04$	$0.14 \pm 0.04$	$376 \pm 10$	
	432	Magnetic	$0.31 \pm 0.04$	$0.08 \pm 0.04$	$305 \pm 10$	
	473	Magnetic	$0.27 \pm 0.04$	$0.08 \pm 0.04$	$209 \pm 10$	
	513	Paramagnetic	$0.22 \pm 0.04$	$0.52 \pm 0.02$	0	
	571	Paramagnetic	$0.21 \pm 0.02$	$0.50 \pm 0.02$	0	
90	298	Magnetic	$0.40 \pm 0.04$	$0.24 \pm 0.04$	$473 \pm 10$	$595 \pm 5$
	609	Paramagnetic	$0.18 \pm 0.04$	$0.49 \pm 0.02$	0	
100	298	Magnetic	$0.38 \pm 0.04$	$0.26 \pm 0.04$	$502 \pm 10$	

lines,<sup>11</sup> to yield the quadrupole splitting, isomer shift, and internal magnetic field of the solid solutions.

### RESULTS AND DISCUSSION

Figures 1 and 2 show typical room-temperature Mössbauer spectra for nonmagnetic ( $x=0.40$ ) and antiferromagnetic ( $x=0.90$ ) solid solutions, respectively. It is seen that the Lorentzian curves generated by the computer program yield a very close fit to the data. In agreement with previous measurements of the magnetic susceptibility of the solid solutions, those in the tetragonal phase were found to be paramagnetic at room temperature, while rhombohedral materials had relatively elevated Néel temperatures.

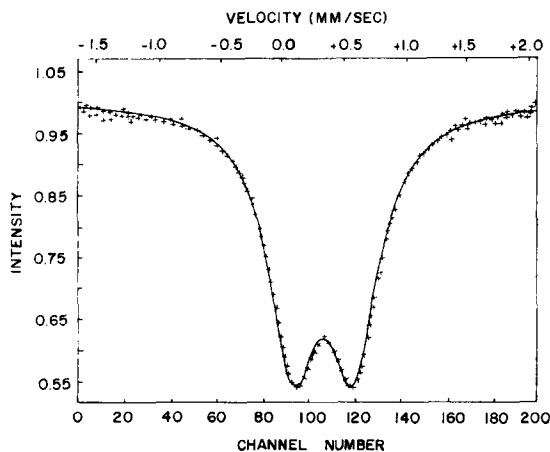


FIG. 1. Mössbauer absorption spectrum of  $(\text{BiFeO}_3)_{0.40}$   $(\text{PbTiO}_3)_{0.60}$  at room temperature. The source was  $^{57}\text{Co}$  in copper, and the zero of the velocity scale is the center for the spectrum of iron at room temperature.

There is a considerable decrease in unit cell volume in passing from the tetragonal to the rhombohedral phase,<sup>2</sup> and the lower volume of the latter apparently favors the magnetic ordering. The properties measured for magnetic solid solutions are summarized in Table I. There is a wide mixed-phase region (about 70–76 mol%  $\text{BiFeO}_3$ ) in which the data were analyzed by the superposition of a magnetic and a nonmagnetic spectrum.

In the paramagnetic state all of the solid solutions showed a quadrupole splitting indicative of the presence of an electric field gradient. The magnitude of this quadrupole splitting at room temperature is shown in Fig. 3 for the paramagnetic materials. As might be expected, the quadrupole splitting increased for solid solutions of higher  $c/a$  ratio. As had been previously

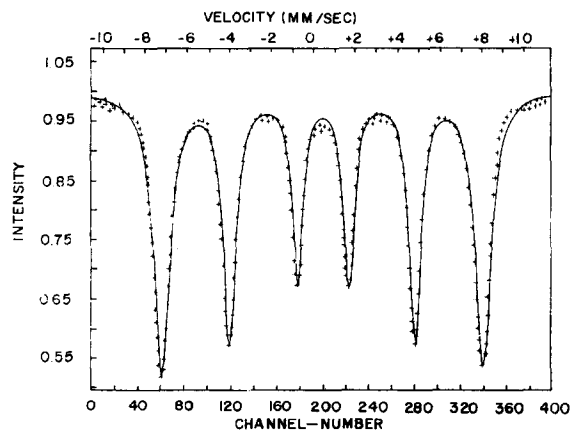


FIG. 2. Mössbauer absorption spectrum of  $(\text{BiFeO}_3)_{0.90}$   $(\text{PbTiO}_3)_{0.10}$  at room temperature.

reported for (BiFeO<sub>3</sub>)<sub>0.05</sub>(PbTiO<sub>3</sub>)<sub>0.95</sub>,<sup>12</sup> the quadrupole splitting did not disappear in the cubic phase at high temperature. The apparently asymmetric environment of the iron atom substituted for a titanium atom within a perovskite oxygen octahedron had been previously reported by Bhide<sup>13</sup> in studies on BaTiO<sub>3</sub>. In that case the asymmetry was attributed to oxygen vacancies required for valence balancing. Bell,<sup>14</sup> in a study of the ferroelectric perovskite PbNb<sub>0.5</sub>Fe<sub>0.5</sub>O<sub>3</sub>, analyzed his data in terms of the quadrupole splitting at the iron nucleus caused by a random distribution of iron and niobium atoms. He was able to account for the observed line splitting, which was similar in magnitude to that observed in the present work, as a superposition of lines caused by iron atoms with differing niobium-neighbor configurations. There was a relatively small change in the line splitting in passing through the ferroelectric Curie point.

The iron environment in the solid solutions of (BiFeO<sub>3</sub>)<sub>x</sub>(PbTiO<sub>3</sub>)<sub>1-x</sub> is more complicated than that in PbNb<sub>0.5</sub>Fe<sub>0.5</sub>O<sub>3</sub>. The reason is that, in the latter case, all of the A cations of the ABO<sub>3</sub> perovskite are the same (Pb), the B cations being randomly distributed, while, in the former case, both the A and B ions may vary. The A cation may be either Pb or Bi, while the B cation may be Fe or Ti. There is not complete randomness, since local valency balance effects would strongly favor a Bi atom as a neighbor for each Fe atom, even in the more dilute solutions. This situation is difficult to analyze quantitatively, but there is a qualitative conclusion which can be drawn. The measured quadrupole splitting in BaTiO<sub>3</sub> or PbNb<sub>0.5</sub>Fe<sub>0.5</sub>O<sub>3</sub> is about 0.4 mm/sec, and for BiFeO<sub>3</sub>, about 0.5 mm/sec. All of these are perovskitic ferroelectrics. A calculation of the value for PbTiO<sub>3</sub> yields a similar result. In addition, measurements of the perturbed directional correlation of the <sup>44</sup>Sc gamma-gamma cascade (resulting from the electron capture decay of <sup>44</sup>Ti), show that the electric field gradient is about 50% higher in PbTiO<sub>3</sub> than in BaTiO<sub>3</sub>.<sup>15,16</sup> In contrast, our measurements imply that the field

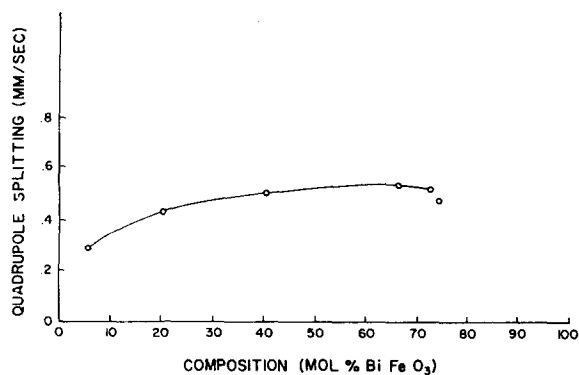


FIG. 3. The quadrupole splitting at room temperature as a function of composition for the BiFeO<sub>3</sub>-PbTiO<sub>3</sub> solid solutions.

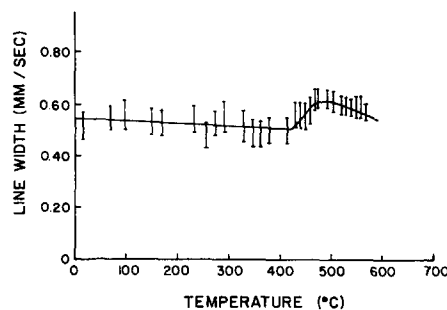


FIG. 4. Linewidth of the Lorentzian resonance for (BiFeO<sub>3</sub>)<sub>0.05</sub>(PbTiO<sub>3</sub>)<sub>0.95</sub> as a function of temperature.

gradient in most of the solid solutions of (BiFeO<sub>3</sub>)<sub>x</sub>(PbTiO<sub>3</sub>)<sub>1-x</sub> is about the same or lower than that in BaTiO<sub>3</sub>. It follows that the present experiment does not represent a measurement which is applicable to pure PbTiO<sub>3</sub>, but is uniquely applicable to the solid solutions of BiFeO<sub>3</sub> in PbTiO<sub>3</sub>.

The relatively small changes in the quadrupole splitting observed when the solutions pass through the Curie point and the generally low variation of the splitting with temperature (see Fig. 3, Ref. 12) indicate that the primary source of the electric field gradient is not the distortion due to ferroelectricity, but rather the local charge and position unbalance caused by Bi<sup>3+</sup> and Fe<sup>3+</sup> substituting for Pb<sup>2+</sup> and Ti<sup>4+</sup>, respectively. It is interesting, however, that when the linewidth of the resolved Lorentzian peaks for (BiFeO<sub>3</sub>)<sub>0.05</sub>(PbTiO<sub>3</sub>)<sub>0.95</sub> is plotted against temperature (see Fig. 4, and refer also to Fig. 3, Ref. 12), there is a distinct maximum at the Curie point. The electrical environment of the Fe atom is thus most varied at the Curie point, which is the temperature of maximum lattice softening and nonlinearity.

In the antiferromagnetic solutions of higher BiFeO<sub>3</sub> content, the solutions decomposed above the ferroelectric Curie point, and the spectra could not be measured at those temperatures. Measurements were made in the paramagnetic state between the Néel temperature and the Curie temperature, and a well-resolved quadrupole splitting was found. Below the Néel temperature, the quadrupole splitting was small. It will be recalled that the quadrupole interaction due to an axially symmetrical electric field gradient at an angle  $\theta$  with respect to a magnetic axis varies as  $(3 \cos^2\theta - 1)$ , so that, for appropriate values of  $\theta$ , the quadrupole splitting measured in magnetic substances can become very small. In the case of the solutions investigated here, most of the iron nuclei do not possess a fully axially symmetrical environment, but the above consideration is undoubtedly responsible for the observed decrease in quadrupole splitting below the Néel point. A comparison of the observed line positions with the Kundig<sup>17</sup> calculations for transition energies and line intensities, given for

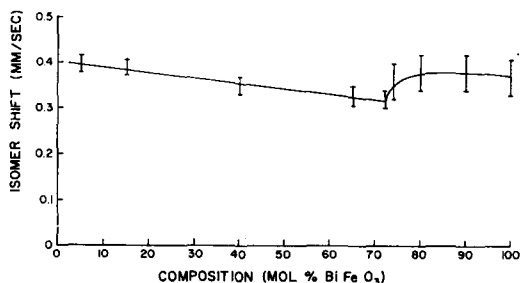


FIG. 5. Isomer shift at room temperature as a function of composition for the  $\text{BiFeO}_3$ - $\text{PbTiO}_3$  solid solutions.

an axially symmetrical field gradient and for different angles  $\theta$  showed a best fit for  $\theta$  in the neighborhood of  $90^\circ$ , in agreement with the value obtained from neutron diffraction measurements on  $\text{BiFeO}_3$ . In view of the assumption of axial symmetry, however, the above value cannot be taken as precise. The sign of the quadrupole interaction was found to be negative for all of the compositions.

The isomer shift, referenced to metallic iron at  $25^\circ\text{C}$ , for  $(\text{BiFeO}_3)_x(\text{PbTiO}_3)_{1-x}$  is given as a function of composition in Fig. 5. The observed values are characteristic of trivalent iron. The decrease in isomer shift with increasing  $x$  in the tetragonal phase may be due to several causes. The first of these is the increased volume of the more tetragonal solutions, whose unit cell increases about 3% from  $x=0$  to  $x=0.7$ .<sup>2</sup> This would be expected to expand the  $d$ -electron shell of the  $\text{Fe}^{3+}$  which would, in turn, through the decrease of screening effects, increase the  $s$ -electron density at the nuclear site and decrease the isomer shift. This is not the complete explanation, since the rhombohedral phase is about 5% lower in volume than the tetragonal phase, while the increase in isomer shift in going from tetragonal to rhombohedral solutions is not as great as would be expected from this figure. There is probably also a lowering of the isomer shift with increased Bi content because of a greater degree of covalency in the material, which is to be expected in substituting bismuth for lead in the A cation position. This effect would not be expected to be very large, however, because the strongest covalent bonding of the iron and titanium is primarily with the oxygen atoms in the surrounding octahedron, and the nature of the bonding will not be greatly changed by the type of A cation.

The relative temperature coefficient of the isomer shift is about  $-2.3 \times 10^{-15}/^\circ\text{K}$  for tetragonal solutions and  $-1.9 \times 10^{-15}/^\circ\text{K}$  for rhombohedral solutions. The high-temperature limit (second-order Doppler shift) for this parameter is  $-2.41 \times 10^{-15}/^\circ\text{K}$ , in reasonable agreement with the value for tetragonal solutions. The disagreement between the isomer shift temperature coefficient for the rhombohedral solutions and the high-temperature limit is not understood, but may be connected with the high electrical Curie points of the latter.

The room-temperature spectra of  $(\text{BiFeO}_3)_{0.90}(\text{PbTiO}_3)_{0.10}$  and  $(\text{BiFeO}_3)_{0.80}(\text{PbTiO}_3)_{0.20}$  showed six peaks, characteristic of a strong magnetic hyperfine interaction associated with the known antiferromagnetism of these solutions, the area ratios of lines being 3:2:1. In Fig. 6, which shows the variation of the magnetic field with temperature, the effective magnetic field for  $(\text{BiFeO}_3)_{0.90}(\text{PbTiO}_3)_{0.10}$  at  $0^\circ\text{K}$  is extrapolated to  $525 \pm 10$  kOe, not greatly different from the value of 550 kOe reported for pure  $\text{BiFeO}_3$ .<sup>18</sup> The indicated temperature for the disappearance of the magnetic field was  $595^\circ \pm 5^\circ\text{K}$ .

Since one of the maximum resonance peaks in the paramagnetic state occurs very near the zero velocity with a  $^{57}\text{Co}$  in the copper matrix source, the antiferromagnetic-paramagnetic transition temperature was also determined by measuring the count rate transmitted through a stationary absorber. An abrupt change in the count rate was found with the onset of hyperfine structure at a temperature of  $590^\circ \pm 5^\circ\text{K}$ , within the experimental error of the previous method. This method was employed for other magnetic solid solutions, and the temperatures below which the magnetic field appeared are given in Table I. These temperatures, determined by Mössbauer spectroscopy, are  $50^\circ$ - $100^\circ\text{C}$  higher, however, than the Néel temperatures determined from magnetic susceptibility anomalies. This is probably because the latter method measures the disappearance of permanent long-range order, while the Mössbauer spectrum is also affected by the magnetic field due to rapidly fluctuating short-range order, which persists to higher temperatures.

These measurements were intended to study a solid solution system in which there was an unusual continuity of ferroelectric properties and also which offered the possibility of magnetic-electric interaction. The measurements showed that the compositions are divided into two categories: tetragonal paramagnetic and rhombohedral antiferromagnetic. Within each category the changes in the environment of the iron atom are small. This is probably because of the

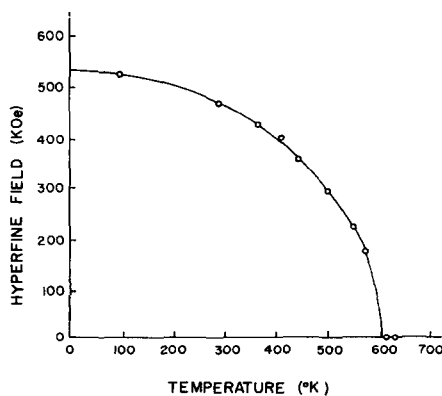


FIG. 6. The variation of the magnetic field at the iron nuclei in  $(\text{BiFeO}_3)_{0.90}(\text{PbTiO}_3)_{0.10}$  as a function of temperature.

closeness of the mass and ionic radius of Bi<sup>3+</sup> and Pb<sup>2+</sup> and of Fe<sup>3+</sup> and Ti<sup>4+</sup>, and also because of the high polarizability of all of the atoms involved. It appears that an oxygen octahedron with Fe<sup>3+</sup> as the central atom is capable of dielectric effects similar to those found with Ti<sup>4+</sup>, in spite of the fact that the latter ion has a noble gas configuration and Fe<sup>3+</sup> does not. With respect to magnetic electric interaction, the sharp change of Néel temperature and unit cell volume with composition in the vicinity of the rhombohedral-tetragonal phase boundary suggests that for these compositions the interaction of electrical, magnetic, and crystallographic parameters will be maximized.

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## Light Scattering by a Rotating Disk\*

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The theoretical expression of the power spectrum of laser light scattered by a ground-glass rotating disk is investigated. In particular, it is shown that the coherence time is not always determined by the transit time of the disk through the illuminated area. The results of several measurements of the conditional probability of scattered light, which exhibit a good agreement with the theory, are presented.

### I. INTRODUCTION

The light scattered by a rotating ground glass screen, containing a random distribution of scattering centers, exhibits properties similar to those of a thermal light source. This device was introduced by Martienssen and Spiller<sup>1</sup> in order to obtain radiation fields with extremely long coherence time, suitably varying the angular velocity of the screen. Since then, the ground-glass rotating disk has been used as the most reliable scattering medium for obtaining light of Gaussian statistical properties from a laser beam. While the Gaussian nature of such a radiation has been experimentally tested by means of photon-counting experiments,<sup>2,3</sup> to the best of our knowledge no detailed analysis has been attempted of the form of the power spectrum and of its dependence from the various parameters involved.

We have theoretically investigated the expression of the correlation function  $\langle \mathbf{E}(t) \cdot \mathbf{E}^*(t+\tau) \rangle$  of the scat-

tered electric field, under the hypothesis of a completely random distribution of dishomogeneities in the glass disk. The resulting expression exhibits direct explicit dependence from the scattering angle, the linear dimensions of the scattering region and the angular velocity of the disk. In particular, two characteristic times exist. The first one is the transit time of the disk through the illuminated area, while the second one is associated with the linear dimensions of the scattering zone and with the scattering angle. The smallest one determines in practice the coherence time.

While the time Fourier transform of  $\langle \mathbf{E}(t) \cdot \mathbf{E}^*(t+\tau) \rangle$ , that is, the power spectrum of the scattered radiation, is a rather involved expression, this is not true for the correlation itself. Therefore a conditional probability experiment seems more apt for testing the theoretical predictions. As a matter of fact, assuming the radiation field to possess Gaussian statistical properties, the time-dependent part of the conditional probability is simply