Magnetic moments in the Fe–Co system

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Magnetic moment has been treated as an extensive thermodynamic variable because it scales with magnetic potential energy. With this assumption, values of partial magnetic moments on the individual atoms have been calculated using the tangent construction across the Fe–Co phase diagram. The values so obtained show excellent agreement with those inferred from neutron scattering at higher concentration of Co, there being serious disagreement at lower concentrations. The values of partial moments have also been calculated from those of partial volumes in this system, using an empirical relation between volume and moments. Excellent agreement between the moments calculated by tangent construction on two different variables gives us confidence in our approach.

Keywords: Atomic volume, Fe–Co system, magnetic moment, Slater–Pauling curves, tangent construction.

THE Fe–Co phase diagram[1] shows the presence of BCC phase up to a Co content of ~70% (atom concentrations/fractions are used here); a two-phase region follows BCC phase. Single phase FCC region is observed at ~85–96% Co. The disordered BCC phase undergoes an ordering reaction to B2 structure over a wide concentration range around 50% Co. The magnetic moments and atomic volume analysed in this communication purport to the disordered solid solutions with BCC and FCC structure.

The Fe–Co system forms the basis of some of the important commercial soft magnetic materials. It shows the highest value of the average (saturation) magnetic moment observed amongst the solid solutions. The system is one of the earliest to attract attention and its interpretation has played an important part towards understanding the well-known Slater–Pauling curves[2]. The variation of the average (integral) saturation magnetic moment μ with concentration shows interesting behaviour[2], indicating a broad maximum at around 35% Co. Polarized neutron diffraction experiments[3] at 30%, 50% and 70% Co indicate that whereas the magnetic moment on Co atoms μ_{Co} remains essentially unchanged at about 1.7 μ_B, the moment on Fe atoms μ_{Fe} increases from 2.6 to 3.0 μ_B. Neutron scattering experiments[4] at 1% Co indicate a value of 2.1 ± 0.5 μ_B for Co moment.

This communication justifies the treatment of magnetic moment as an extensive thermodynamic variable and proposes the use of tangent construction[5] to extract information about the partial moments from the experi-

mental values of average moments across a binary phase diagram. We show the application of this method to the Fe–Co system and discuss the values thus obtained vis-à-vis the reported values mentioned above. Additionally, we use experimental values of atomic volume to arrive at partial volume of the binary components in this system. We then explore the possibility of calculating partial magnetic moments from partial volumes in view of the strong correlation[6,7] observed between magnetic properties and lattice parameter (atomic volume) with concentration. The values of partial moments thus obtained are compared with those obtained by tangent construction of (average) magnetic moments. This approach also allows us to predict the experimental magnetic moments across the phase diagram using the lattice parameter data.

Knowledge of partial magnetic moments has played a critical role in developing theories and models of ferromagnetic behaviour. This information to date has been generated using the neutron scattering techniques. There are only a few facilities that exist the world over. The technique also requires specific isotopes with desirable scattering cross-section. These limitations have led to studies on a rather limited number of systems and that too for a few compositions. The method we propose here can generate values of partial moments at all compositions of a multi-component system provided average values are known at several compositions, especially in the regions where the average curves change slope.

The condensed matter scientists routinely use tangent construction with free energy/atom (chemical potential) as an extensive thermodynamic function in two-phase binary systems. Its use yields the equilibrium values of concentration between the two phases under consideration. However, its use in single-phase systems is not often resorted to, even though (as we show) it can give valuable information regarding the partial values of some properties of the single phase that change with composition.

In case one knows the variation of average values of an extensive thermodynamic function, X, with concentration across a binary system I–J, the partial values attributable to the constituent atoms X_I and X_J at any composition are the intercepts made by a tangent drawn at that composition[5]. Therefore, knowing the variation of function X with concentration, the commonly used tangent construction[5] can be used to extract partial values for the constituent atoms. Thus, if the average value X_c, and the slope, dX/dc at any concentration c are known, the values of X_I and X_J can be found using the following simple relations:

$$X_I = X_c - c\, dX/dc, \tag{1}$$

$$X_J = X_c + (1 - c)dX/dc, \tag{2}$$

Although the saturation magnetic moment is not a thermodynamic variable, it can be treated as an extensive

parameter in view of the simple arguments outlined below.

Consider a system that is subjected to an external magnetic field H. The aligning of the magnetic moments associated with the system leads to a change in its internal energy (dU) that according to the basic laws of thermodynamics is given by,

$$dU = TdS + dw. \tag{3}$$

S and w refer to entropy and work done, positive when done on the system and negative if done by the system. If we consider only the magnetic work, dw equals the dot product of the magnetic moment M of the system and the magnetic field H,

$$dw = M \cdot H. \tag{4}$$

The reported values of average saturation magnetic moment refer to extrapolation to an infinite magnetic field at absolute zero temperature (even though the measurements are carried out at finite fields and higher temperatures). Infinite field implies alignment of all magnetic moments in the direction of the magnetic field. Therefore, if we consider a very large finite field, we find that at $T = 0$, eq. (3) yields the scalar relation,

$$dU = MH. \tag{5}$$

Since the field can be held constant, variations in dU result only from changes in M. The internal energy considered above is referred to as magnetic potential energy by Halliday *et al.*[8]. The reported values[2] of average saturation magnetic moment refer to total magnetic moment M of the system divided by the total number of atoms in the system. It can thus be treated as an extensive thermodynamic variable since it is proportional to internal energy of the system. The total magnetic moment of an alloy arises from the contribution of the constituent elements in proportion to their concentration. Thus it may be worthwhile to employ tangent construction to arrive at the contribution of individual elements to the magnetic moment of the alloy phase.

If X is considered to represent average magnetic moment, μ the values of partial moments μ_I and μ_J can be deduced using relations signified by eqs (1) and (2). The value of initial slope, $(d\mu/dc)_{c=0}$ has been extensively used in the literature since Stoner[9] to deduce the role of different solutes in influencing the magnetic behaviour. This parameter has played an important part in model building. According to these equations, the solvent moment μ_I is not altered when infinitesimal quantities of solvent J are added to it, whereas the solute moment is given by

$$\mu_{J(c=0)} = \mu_I + (d\mu/dc)_{c=0}. \tag{6}$$

We shall illustrate the use of this simple approach to calculate partial moments in the Fe–Co system.

Figure 1 shows the values of average atomic moment according to Bozarth[2], as also the calculated partial moments μ_{Fe} and μ_{Co} that the tangent construction yields. Experimental values obtained by neutron scattering are also shown in the figure. According to these, $\mu_{Fe} > \mu_{Co}$ at all the compositions studied. The values obtained by tangent construction showed excellent agreement with those of Collins and Forsyth[3] for Co atom-fractions 0.50 and 0.70. However, serious disagreement was observed at atom fraction 0.30 and at the infinite dilution limit. According to our approach, the Co moment has higher values at lower Co content, the crossover occurring at an atom fraction of ~ 0.35, the value at which the maximum is observed in the average magnetic moment. This is to be expected if we accept the treatment of magnetic moment as an extensive thermodynamic parameter, the maximum being the point at which the slope is zero and the partials have the same value. For small addition of Co to Fe (infinite dilution), neutron scattering data of Collins and Low[4] indicate a value of $2.1 \pm 0.5 \, \mu_B$ compared to $3.2 \, \mu_B$ indicated by tangent construction. The point arrived at by Mott and Stevens[10] (Figure 1) indicates a value of $3.0 \, \mu_B$. The above discussion pertains to the BCC phase.

To the best of our knowledge no neutron scattering data have been reported for the FCC phase. Our approach gives a value of $1.73 \, \mu_B$ for FCC Co, in reasonable agreement with reported values of $1.75 \, \mu_B$ (ref. 11) and $1.74 \, \mu_B$ (ref. 12). In the FCC phase μ_{Fe} has a value of $2.77 \, \mu_B$. The partial moments in the FCC phase are independent of the composition.

Atomic volume is an important thermodynamic variable, easily accessible to accurate measurements via density. After the advent of X-ray diffraction even more accurate data have been made available through precision
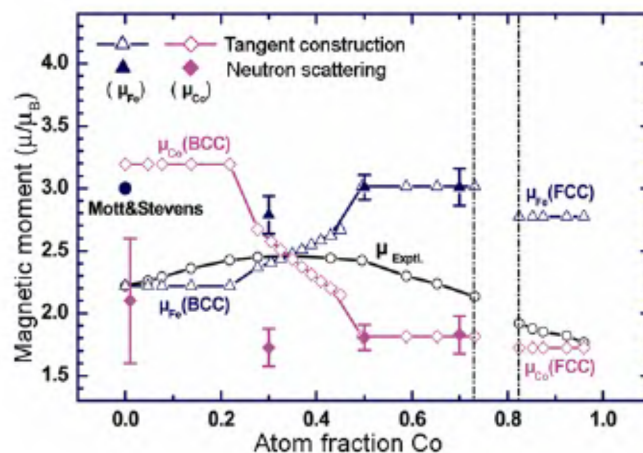

Figure 1. A plot of average saturation magnetic moment[2] $\mu_{Exptl.}$, and of partial moments μ_{Fe} and μ_{Co} (tangent construction) in the Fe–Co system. Neutron scattering values of partial moments[3,4] are shown as also the value according to Mott and Stevens[10].

lattice parameter measurements. It also occupies centre-stage as far as the electron theories are concerned. Despite the extensive use of average atomic volume in electron theories, the partial volume that is easily accessible via the tangent construction has rarely been used. King[13] pointed out that many physical properties could be correlated to volume size factor. He tabulated the data for volume size factor in several binary solid solutions. He based his table on extensive lattice parameter data. King defined volume size factor, $\Omega_{sf} = (V_{solvent} - V^*_{solute})/V_{solvent}$, $V_{solvent}$ being the solvent volume and V^*_{solute} the partial atomic volume of the solute. He only considered the region wherein linear variation of atomic volume with solute concentration was observed, i.e. the solid solution regime in which the solvent volume remains unaltered from its elemental state and V^*_{solute} stays constant. He did not consider the more interesting behaviour; the departure from linearity in the atomic volume plots that implies simultaneous variation of the solvent and solute volume.

Bhatia et al.[14,15] have used values of Ω_{sf} given by King to predict lattice parameters in binary and ternary $L1_2$ and $B2$ structures. They have also tried to unravel the pattern underlying volume changes that accompany the formation of several structures[16–18], and disordering[19,20]. In our view this approach can help delineate the role of the constituent atoms on different physical properties in multicomponent systems as shown here.

Figure 2 shows the variation of atomic volume in the Fe–Co system according to precise lattice parameter measurements of Rodgers and Maddocks, Ellis and Greiner, and Sutton and Hume–Rothery as compiled by Pearson[6]. The measurement accuracy is such that one can repose confidence in the values plotted. The line joining the elemental volumes of Fe and Co would give the variation of volume across the phase diagram according to Vegard's law. Positive deviation from Vegard's law is shown at all compositions.
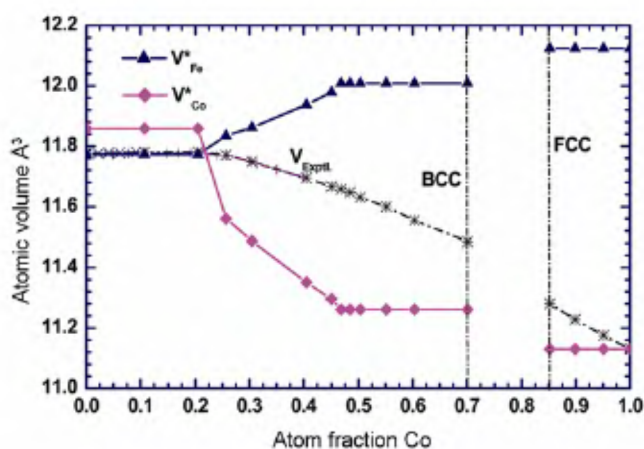

Figure 2. Variation of atomic volume with Co content according to data reported by Pearson[6]. Partial volumes V^*_{Fe} and V^*_{Co} obtained by the use of tangent construction are also shown.

The experimental data on atomic volume have been converted to yield values of partial volumes V^*_{Fe} and V^*_{Co} across the Fe–Co system by the use of tangent construction. These are also shown in Figure 2. The more accurate data of Sutton and Hume–Rothery have been used to arrive at the slope for the initial linear portion that extends to 0.21 Co fraction. The values calculated by us for this concentration range are in good agreement with King's data.

A cursory look at Figures 1 and 2 shows the existence of strong correlation between magnetic moments and atomic volume. Both show linear variation up to Co fraction of ~ 0.21, when the departure from linearity sets in. Linear behaviour in both the functions is observed at Co fractions above ~ 0.5, the slope being different in the BCC and FCC phase. However, although the moments increase beyond 0.21, achieving a maximum at Co fraction ~ 0.35, the volume decrease sets in at 0.21. The partial volumes show interesting behaviour with increasing Co content. Whereas V^*_{Fe} increase from ~ 11.77 (in pure BCC Fe) to ~ 12.12, as Co content increases, V^*_{Co} decreases from ~ 11.81–11.13 Å^3 (in pure FCC Co).

Schlosser[21] proposed an empirical relation between atomic volume and the magnetic moment associated with it for the 3d transition elements. He based this relation on a phenomenological approach that involved observations of volume and magnetic moments in several binary alloys. According to this relation,

$$V = V_0 \, (\mu = 0) + 0.108\mu^2. \tag{7}$$

Using the above relation Schlosser[22–24] tried to rationalize a large amount of experimental data on atomic volume in several binary alloys of Fe, Co, Mn and Ni. He gave values of $V_0 \, (\mu = 0)$ for different elements, being 11.2484 and 10.7782 Å^3 for Fe and Co respectively.

With the help of the above relation, and values of $V_0(\mu = 0)$, μ_{Fe} and μ_{Co} have been calculated across the Fe–Co phase diagram, using the values of partial volumes plotted in Figure 2. The partial moments so calculated are shown in Figure 3. For comparison the partial moments derived from the tangent construction on moments (Figure 1) are also plotted in this figure. Surprisingly, good agreement was observed for partial moments arrived at by two very different routes up to Co fraction of 0.45, the agreement being poor at higher Co contents in the BCC phase.

Figure 4 shows the plot of (average) magnetic moment calculated using the partial values derived from partial volume (Figure 3) and its comparison with the reported experimental values as given in Figure 1. The agreement can be considered to be satisfactory, especially at low Co content. The general shape of the experimental curve is well reproduced. Thus in principle we can have a fairly good prediction of the magnetic moments if accurate values of lattice parameter across the phase diagram are available.
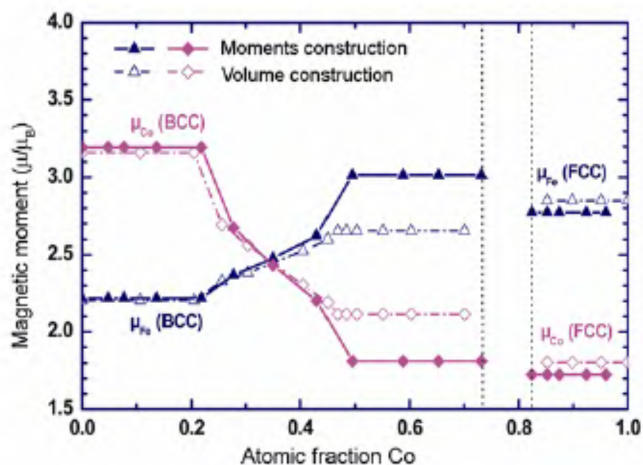
Figure 3. A plot of partial magnetic moments obtained from partial volumes according to Schlosser[21] compared with values obtained from tangent construction of moments curve as shown in Figure 1.
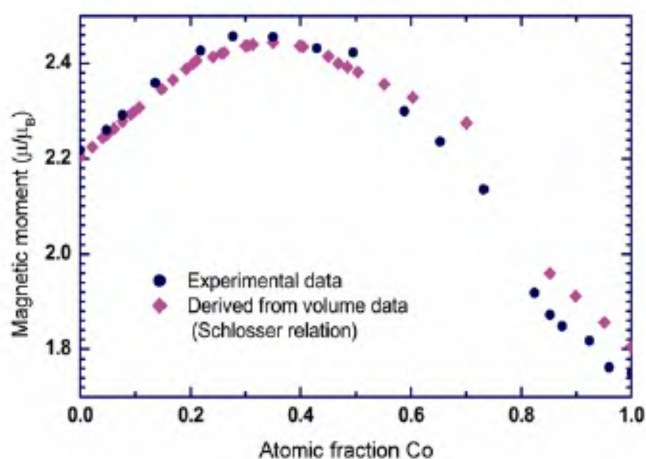

Figure 4. Values of (average) magnetic moments derived from partial volume data compared with experimental data[2] as shown in Figure 1.

At the outset we would like to state the well-known cliché that thermodynamics does not give us any clues about the mechanisms underlying the conclusions inferred from its application. No model for the magnetic moments in the Fe–Co system is being proposed. However, in our view as applied to the moment problem on hand, it gives us quantitative information that is likely to be correct.

Table 1 summarizes the data on partial moments obtained using the tangent construction on the magnetic moment curve; the reported values obtained by neutron scattering are also indicated. As mentioned earlier, perfect agreement is observed at Co fraction of 0.50 and 0.70. However, serious disagreement is observed at 0.3 Co and at infinite dilution. A cursory look at the curve for the experimental data shows that it has zero slope at ~0.35 Co, implying, $\mu_{Co} = \mu_{Fe} = 2.45$ μ_B. Further, the slope indicates that $\mu_{Co} > \mu_{Fe}$ at lower Co content, and $\mu_{Co} < \mu_{Fe}$ at higher concentration.

Table 1 also summarizes the values obtained for partial volume, as also partial moments corresponding to these volumes according to the Schlosser relation. Reasonably good agreement between partial moments so derived with values derived from tangent construction on moment curve suggests the correctness of our approach. We feel that the neutron scattering data and their interpretation according to which $\mu_{Co} < \mu_{Fe}$ in BCC phase at 0.30 Co and at infinite dilution, need to be revisited. If we accept the treatment of magnetic moment as an extensive thermodynamic parameter, eq. (6) follows. According to Vogt[12] $(d\mu/dc)_{c=0} = 1$, for additions of Co to Fe. Thus, for infinitesimal additions of Co, eq. (6) would give a value of 3.22 μ_B for μ_{Co}, in agreement with our approach, as we would expect. Mott and Stevens[10], after a careful consideration of several parameters (that included resistivity, bonding and absolute X-ray scattering factors), arrived at a value of 3.0 μ_B for μ_{Co} (Figure 1). Also, the paper by Mott and Stevens antedates neutron scattering information.

King's[13] comment on the atomic volume plots is illuminating. According to him, 'The tendency of a metallic solid solution to deviate from Vegard's law may be taken as a measure of the modification of the electronic environment of the solute atom. By the same reasoning, the breakaway of the volume plots from linear trends indicates that sufficient solute element has now been added to modify the electronic environment of the solvent atoms.'

Let us consider the above statement that seems reasonable for small additions of Co to Fe. With reference to Figure 2, the implications are that there is no change in the electronic environment of the Fe atoms when small additions of Co are made to Fe; thus they should have the same electronic environment and therefore magnetic moment as pure Fe. This situation ought to continue till a Co fraction of 0.21. If so, the increase in magnetic moment shown by the experimental data (Figure 1) has to come from the Co atoms and they have to possess a moment greater than that on the Fe atoms. The significantly altered moment on Co atoms is a result of the very different environment (BCC lattice compared to HCP and Fe neighbours) in which they find themselves, and is reflected in significantly higher value of partial volume compared to that in the elemental state.

A sophisticated, first-principle electron theory has been used by Schwartz et al.[25] to calculate the partial and integral magnetic moments in the Fe–Co system. At the composition Fe_3Co, assuming the Fe_3Al structure, moments of 2.40 and 2.57 μ_B for the two kinds of Fe atoms in this structure, and a moment of 1.65 μ_B for Co atoms have been calculated. This is in agreement with the accepted data of Collins and Forsyth at 0.30 Co, that in our view needs to be carefully re-evaluated. However, the underlying basic assumption by the authors that Fe and Co atoms have the same volume ought to make us vary of their prediction; the strong correlation between magnetic moment

Table 1. Values of partial magnetic moments obtained using tangent construction of moment curve[2], the reported values obtained by neutron scattering[3,4], partial volume derived from reported lattice parameter data[6] by tangent construction, and partial magnetic moment values obtained using the Schlosser relation[21]. Values are in μ_B for moments and the structure is BCC up to $_{0.70}$Co

| Co atom fraction | μ_{Fe} | μ_{Co} | μ_{Fe} | μ_{Co} | V^*_{Fe} | V^*_{Co} | μ_{Fe} | μ_{Co} |
	(Tangent construction)		(Neutron scattering)		(Å^3)		(Schlosser relation)	
0.00	2.22	3.19	–	2.1 ± 0.5	11.77	11.86	2.20	3.16
0.30	2.41	2.58	2.78 ± 0.15	1.72 ± 0.15	11.86	11.49	2.38	2.57
0.50	3.02	1.81	3.01 ± 0.10	1.81 ± 0.10	12.01	11.26	2.65	2.11
0.70	3.02	1.81	3.01 ± 0.15	1.83 ± 0.15	12.01	11.26	2.65	2.11
0.85–1.00 (FCC)	2.78	1.73	–	–	12.12	11.13	2.85	1.81

and volume having been demonstrated by us. The integral moment calculated by them for Fe₃Co is 2.30 μ_B, the experimental value being 2.44 μ_B. Our rather simplistic approach (Figure 4) using the lattice parameters and Schlosser relation yields a value of 2.43 μ_B.

We believe that the values of partial moments calculated using tangent construction of the experimental moment curve (Figure 1) are more reliable. In our view the divergence between the values of partial and integral moments (Figures 3 and 4), calculated using volume and those obtained from tangent construction on the moment curve points towards the need for developing better theoretical models for the relation between magnetic moment and volume. These relations are likely to depend on the element concerned and its crystal structure, unlike the single relation putforth by Schlosser. Availability of such relations would lead to better agreement with experimental moments (Figure 4) and partial moments derived from partial volume (Figure 3).

The procedure (tangent construction) used here for the Fe–Co system should yield useful information on partial magnetic moments in other binary systems as well, e.g. it should be applicable to the different Slater–Pauling curves[2]. The procedure would also be applicable to ternary and quaternary systems[26–28] that can be treated as pseudo-binary. For example, the data on magnetic moments and volume reported in the $Mn_{(1-x)}Ni_xAl$ system by Hara et al.[26] can be analysed by tangent construction and the role of Mn and Ni atoms delineated. In explaining their results, Hara et al.[26] make the assumption that Ni atoms carry no magnetic moment which is unlikely to be correct.

We expect that linear portions of magnetic moment curve clearly give values of partial moments, and the average atomic volume should also show a linear variation over the same concentration range, as observed by us for the system under consideration. Similarly, the concentration region in which a nonlinear behaviour is observed for the average moments, the average atomic volume should show similar variation. Our analysis (not presented here) shows a nonlinear behaviour in the moment curve in the Ni–Mn system, but the volume curve shows a straight line in the same concentration range, indicating that the procedure outlined by us is not applicable. Thus

to obtain partial moments in the regions in which the moment curve shows nonlinear behaviour, one needs to confirm similar behaviour in the atomic volume/concentration curve.

Finally, the tangent construction can yield useful information on other physical properties that scale with extensive thermodynamic parameters, especially when used in conjunction with variation in volume, as shown in this paper. We are now looking at the possibility of obtaining useful information from the reported specific heats[29,30] in the Fe–Co system, as also trying to tie up the experimental information on density-of-states using concepts outlined here.

We have demonstrated that the tangent construction is a powerful tool to calculate partial magnetic moments from the experimental data on (integral) magnetic moments in the Fe–Co system. Till date partial moments have been arrived at by neutron scattering experiments. There is good agreement with reported experimental data at higher Co content, serious disagreement being observed at lower concentrations. Keeping in view the fact that there is a strong correlation between magnetic moment and volume, and the observation that tangent construction on volume curve independently confirms the values derived above, especially at lower Co content, give us confidence in our approach. Thus, in our view the interpretation of experimental data at lower concentrations needs to be carefully re-assessed.

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On acoustic theory of conch shell

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We recently reported a study on the generation and resonance of sound in a shell assuming the straightened conch-cavity as a truncated horn. However, the tomographic picture reveals that the cavity grows as an Archimedes spiral carried forward in length. To account for the realistic case, in this study, the equation of propagation of sound waves in the shell is set up as Webster's horn equation in three dimensions in circular cylindrical coordinates. Solution of the angular and radial components of the equation on application of appropriate boundary conditions leads to an expression for frequency which is the same as the one reported earlier. However, solution of the *z*-component of the wave equation subjected to available boundary conditions poses the possibility of existence of very high frequency components, almost in the ultrasonic range. The intensity of radiation saturates in the high frequency range.

THE conch shell is one of the earliest wind instruments found in nature. In view of its wide use as a musical instrument and trumpet by various cultures the world over through the ages and in view of the fact that little study has gone into its acoustics, we have taken up a close study of the system. In this process we reported the result of our study on morphology and tomographic pictures of the shell samples to show that the structure of the shell follows the Fibonacci pattern[1].

Further in a recent report we have presented our understanding of the mechanism of generation of musical sound in the conch shell[2]. We have presented the sound spectra graphs from a number of shell samples produced on a computer using Spectra Plus software. The spectral characteristics have been observed and outlined. It has been pointed out that the shell sounds in 'lock-in' mode accompanied by frequencies of all integral multiples of the peak (i.e. lock-in) frequency.

In order to correlate the resonance frequency with the shell parameters, we have assumed the straightened shell cavity spiral to approximate a truncated horn. A well-known theory of conical horn has been applied, which leads to a frequency expression that agrees fairly well with the observed spectra.

However, as seen in the tomographic pictures, the conch cavity grows as an Archimedes' spiral carried