# GENERALIZED SYMMETRY AND NEUMANN'S PRINCIPLE

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ABSTRACT

Neumann's principle is examined in the context of the recent generalizations relating to symmetry and physical properties of crystals. It is shown that the difficulties encountered in applying the principle to transport properties are only apparent and that it is not necessary to impose restrictions on the principle. A proper interpretation of the symmetry of crystals resolves the difficulties and brings out clearly the axiomatic nature of this principle and its universal applicability.

## 1. GENERALIZED POINT GROUPS

RECENT studies of magnetic properties revealed that the concepts of symmetry in respect of crystals need generalization. It is realized that in addition to the symmetry of spatial disposition of atoms in the crystal, the symmetry of the spatial orientation of the atomic magnetic moments is relevant. This necessitated introduction of new symmetry operations. The reversal of atomic magnetic moments is a possible symmetry operation and it is denoted by $\mathscr{R}$. The magnetic moment reversal operation is physically equivalent to the reversal of the electric current that gives rise to each magnetic moment and is therefore called the current reversal operation. It is also sometimes referred to as the time-reversal operation. The combinations of $\mathscr{R}$ with the conventional operations are possible symmetry operations and they are called complementary or anti-symmetry operations. The introduction of these new operations increases the crystallographic point groups from 32 to 122.

Among these 122 generalized groups, there are 32 which are obtained by augmenting each of the 32 classical point groups by $\mathscr{R}$ and its products with the elements of the classical group. These are known as grey groups. Of the remaining 90 groups, 32 are identical with the classical groups in the sense that they do not contain either the operation $\mathscr{R}$ or any anti-operation. They are called single-colour groups. The remaining 58 groups do not contain $\mathscr{R}$ but contain classical as well as anti-symmetry operations. They are called double-colour groups. In the double-colour groups, the classical operations in themselves form a subgroup of index 2.

Ferromagnetic and antiferromagnetic crystals are characterized by a spatial orientation of magnetic moments. Their symmetries have to be classified under the 90 single and double-colour groups, since $\mathcal{R}$ cannot be a symmetry operation in such crystals. The para and diamagnetic crystals can be considered to possess the operation $\mathcal{R}$ as a symmetry operation and they are classified under the 32 grey groups.

## 2. NEUMANN'S PRINCIPLE

Neumann's principle states that every physical property exhibited by a crystal must possess at least the symmetry of the crystal. In a restricted sense, symmetry of the crystal as used in this statement was conventionally understood as its symmetry belonging to one of the 32 classical crystallographic point groups.

The generalization of symmetry concepts and classification of crystals as enumerated above has to be taken into account in utilizing Neumann's principle for linking the symmetry of crystals with their physical properties (Bhagavantam and Pantulu, 1964). Any physical property of a crystal defines a relation between two physical quantities. The relation may be represented by a tensor equation of the form:

$$B_{ijk\cdots} = a_{ijk\cdots lmn\cdots} \; A_{lmn\cdots} \tag{1}$$

In (1), $A_{lmn\cdots}$ and $B_{ijk\cdots}$ are physical quantity tensors and $a_{ijk\cdots lmn\cdots}$ is the physical property tensor. Neumann's principle requires that the tensor $a_{ijk\cdots lmn\cdots}$ must be invariant under the point group symmetry of a crystal in which it is observed. In the context of the generalization outlined in Section 1, by 'point group symmetry of the crystal' is meant 'the generalized point group symmetry of the crystal'. It is necessary to know how the physical tensors transform under the new operations introduced. Since $\mathcal{R}$ is a magnetic moment reversal operation, we can take that the symmetry group of the magnetic field H, the magnetic moment I and the magnetic induction B cannot contain $\mathcal{R}$ explicitly. The symmetry group of the electric current vector J cannot contain $\mathcal{R}$ explicitly for similar reasons. The electric field vector E is equivalent to a static charge distribution by which it is generated and we may therefore conclude that its symmetry group contains the operation $\mathcal{R}$ explicitly. The symmetry of more complicated tensors is determined by interpreting them as the outer products of simple tensors like the above whose symmetry is known. For this purpose, we note that (i) the product of two tensors which are both symmetric or asymmetric under the operation $\mathscr{R}$ is symmetric under $\mathscr{R}$ and (ii) the product of two tensors one of which is symmetric and the other asymmetric under $\mathscr{R}$, is asymmetric under $\mathscr{R}$.

The application of Neumann's principle adopting these generalizations in respect of symmetry of crystals and physical tensors cannot give new results for non-magnetic properties of para or diamagnetic crystals. The generalizations, as already pointed out, are effected to take into account the fact that the magnetic properties of crystals are influenced by the symmetry of magnetic moment orientation in the crystal. Non-recognition of this fact gave rise to an incorrect conclusion that magnetic properties like piezomagnetism were altogether impossible. A recognition of the 122 generalized classes and proper application of Neumann's principle to them reveals that such properties are possible in some of the single-colour and double-colour classes (90 magnetic classes). In particular, piezomagnetism is possible in 66 of these classes. It also shows that magnetic properties and in fact all properties that are represented by tensors asymmetric under $\mathscr{R}$ are forbidden in the 32 grey classes.

## 3. Transport Properties

A common transport property, namely electrical resistivity, is represented by a second order symmetric tensor $\rho_{ik}$ defined by the relation:

$$E_i = \rho_{ik}\, J_{ik}. \tag{2}$$

Since $E_i$ is a polar vector symmetric under $\mathscr{R}$ and $J_k$ is a polar vector asymmetric under $\mathscr{R}$, $\rho_{ik}$ is a second order polar tensor asymmetric under $\mathscr{R}$. An uncautious application of Neumann's principle would lead to the erroneous result that electrical resistivity and electrical conduction are forbidden in para and diamagnetic crystals. This situation has to be clarified. When a steady transport process is taking place in a crystal, the system is not under thermodynamic equilibrium. The crystal is in a steady state and entropy is being produced at a constant rate at every point in the crystal. The application of time-reversal operation $\mathscr{R}$ would result in a negative rate of entropy production which is physically impossible. Thus when transport phenomena are occurring, a definite direction in time has to be recognized. Neumann's principle in its conventional form amounts to stating that free space is isotropic. In the context of the generalizations of symmetry mentioned above, it amounts to stating that free space is isotropic and there is no preference in the direction of time. When a transport process is taking place, the steady rate of production of entropy imposes a preference to one direction in time. For this reason, Birss (1964) concludes that Neumann's principle cannot be applied to transport properties. He states that to find the symmetry of a transport property observed in a crystal, its tensor should be reduced applying only the classical operations contained in its generalized symmetry group. Thus, (i) if the crystal belongs to one of the grey classes, the tensor should be subjected to the operations of the classical point group that is contained in it as a subgroup of index 2; (ii) if the crystal belongs to a single-colour group, all the operations of the group should be used; (iii) if the crystal belongs to a double colour group, the classical operations which form a subgroup of index 2 should be used.

We wish to point out here that these conclusions can be reached in a different manner and conforming with the general principles such that it fits into the scheme of generalizations invoked. It is also felt that any new principles introduced in respect of transport properties should not appear to be imposing *ad hoc* restrictions on the applicability of Neumann's principle in view of its axiomatic nature.

We note that when Neumann's principle is being applied, it is implied that the appropriate symmetry of the crystal is considered. That is to say, any change in symmetry of the crystal due to a change in its state has to be taken account of. Such changes in symmetry cannot be predicted by Neumann's principle. For example, a change in temperature of a crystal would cause a strain (thermal expansion) in the crystal. The form of the thermal expansion tensor in any particular crystal can be determined by applying Neumann's principle. A change in temperature might lead to a changeover into a magnetic state with a different symmetry. This cannot be predicted by Neumann's principle. Given the new (changed) symmetry, it can find the form of the same tensor appropriate to it. In a similar manner, we note that when a steady transport process is taking place in a crystal which results in a constant positive rate of entropy production at every point in the crystal, the symmetry of the crystal is no longer the same as what it was in its equilibrium state. The constant rate of entropy production $ds/dt$ is a scalar quantity which is asymmetric under the operation $\mathcal{R}$. Its symmetry is given by the full orthogonal group. The symmetry of the crystal in which a steady irreversible process is taking place is the symmetry common to that of the scalar $ds/dt$ and that of the crystal in its previous equilibrium state. This is according to Curie's principle. The above conclusions (i), (ii) and (iii) naturally follow because the intersection of any of the 122 generalized groups with the full orthogonal group selects out the subgroup of classical operations contained in it.

## 4. The Axiomatic Nature of Neumann's Principle

We shall now examine the merits of keeping the sanctity of Neumann's principle. The symmetry of a crystal is in principle the highest common symmetry of the symmetries of various physical properties it is capable of exhibiting. The problem of determining the possible point groups of real three dimensional orthogonal transformations is a mathematical problem which can be studied independently of their application to crystal physics. The enumeration of 32 (classical) point groups as possible symmetry groups of crystallographic significance, however involves the lattice hypothesis which is based on observations that are physical. The construction and enumeration of 122 point groups of symmetry is a generalization effected on the 32 groups so determined, taking into account the spatial orientations of magnetic moments. The practical determination of the point group of a crystal is carried out by studying its response to various physical influences. For instance, simple X-ray diffraction studies give us the symmetry of the charge distribution in a crystal and by such studies, the crystal can be only assigned to one of the 11 X-ray classes, *i.e.*, those groups which contain the operation inexplicitly. A further and more detailed analysis has to be done by using other tools and by observing other properties of the crystal if we want to distinguish between the subgroups within one of the X-ray classes. This means that if the property chosen depends on the charge distribution only (symmetrical under the operation $\mathscr{R}$), we arrive at the classical point group of charge distribution symmetry of the crystal. We will not yet know its space-time or magnetic symmetry. If we choose suitable magnetic properties for our study, the magnetic symmetry group can be determined. Thus, we note that the symmetry of a crystal in the generalized sense as belonging to one of the 122 classes is determined by its electrical and magnetic properties.

This general classification is established to be necessary and will be considered adequate until any new properties, expected or observed, require a further generalization. Neumann's principle should be applied in this generalized sense until a contradiction or anomaly is noticed. Whereas it is possible for such a contradiction to lead to a new generalization of the notions of symmetry, it cannot result in a revision of or a restriction on the Neumann's principle. This is because Neumann's principle is in effect equivalent to a tautological statement. It demands that any property of a crystal must possess at least the symmetry of the crystal which is determined as the greatest common symmetry of all such properties. Any apparently anomalous result would only mean that either the symmetry of the crystal is not properly ascertained or a new generalization in the notion of symmetry is called for. If a new generalization is effected, the principle is also automatically generalized because then "the symmetry of the crystal" would mean the new generalized symmetry. Neumann's principle is thus inescapable. We have seen that the transport properties can be fitted into the existing scheme of ideas by noting that the symmetry of a crystal in the steady irreversible state is different from what it was in its previous equilibrium state. Neumann's principle is in this case applied, using the changed symmetry of the crystal.

## 5. Transition from a Non-Magnetic State to a Magnetic State

In this section, we shall consider another kind of an apparent non-conformity between the observed results and those expected from the application of Neumann's principle. In the transition of a crystal from a non-magnetic state belonging to a grey class to a magnetic state, say by cooling below a transition temperature, two alternatives are possible. The first is one in which the symmetry group in the magnetic state may be the classical group contained as a subgroup of index 2 in the grey group or a magnetic variant thereof. By a magnetic variant of a classical group is meant the group obtained by choosing half the elements in the classical group as anti-symmetry operations. In this case, one can see that there is no deterioration in symmetry for conventional purposes. The second is one in which the symmetry group in the magnetic state may be a proper subgroup of the classical group or a magnetic variant thereof. In this case, one can see that there is a deterioration in symmetry due to transition. In the first alternative, observations conducted on properties symmetric with respect to $\mathscr{R}$ will not show a change in symmetry at the transition point. In the second alternative, such properties should in principle be expected to show a deterioration in symmetry. In practice, if such a deterioration is observed, it conclusively establishes that the generalizations effected in symmetry are physically significant. If no such deterioration in symmetry is observed, that fact itself cannot serve as evidence contradicting Neumann's principle. The change in symmetry at the transition point is always such that the lower symmetry in the magnetic state forms a subgroup of the higher symmetry in the non-magnetic state. From this it follows that the scheme of coefficients of a non-magnetic property tensor in the higher symmetry can be obtained from the scheme appropriate to the lower symmetry by applying the extra symmetry operations to the tensor coefficients and demanding invariance. Thus, we can be sure that a coefficient that is ruled out by considerations of symmetry in the lower symmetry state of the crystal cannot appear in the higher symmetry state. It follows that if a deterioration in symmetry due to transition, as shown by magnetic properties, is not shown by non-magnetic properties, the higher symmetry exhibited by the property can obviously be explained by recognizing that the influence of spin orientations is not enough to resolve the degeneracy. In other words, the physical property exhibits the symmetry of the crystal and in fact a little more. Neumann's principle continues to hold.

## REFERENCES

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