Irreducible fourth-rank Cartesian tensors

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The complete reduction of a fourth-rank Cartesian tensor into parts which are irreducible under the three-dimensional rotation group is presented. Results are given for the general case of a tensor without index permutational symmetry, and also for tensors with specific index symmetry properties. Applications in the realm of crystal physics are indicated with particular reference to four-photon absorption, and a table of irreducible tensor representations is provided for the seven crystal classes.

I. INTRODUCTION

In many areas of physics involving the use of tensor calculus, the adoption of irreducible tensors leads to a simplification of the theory and a clarification of the physical principles. In recent years, irreducible tensor methods have increasingly been used in studies on nonlinear optical and electro-'optical processes, ^{1,2} light scattering in fluids multiphoton spectroscopy,^{7–10} and the physica multiphoton spectroscopy,^{\prime -10} and the physical properties of condensed matter.^{11–13} Some of the important advantages which can be gained from the use of such methods can be summarized as follows.

(1) In a given process, a distinct physical meaning can usually be attributed to each irreducible tensor characterized by a certain weight.

(2) Irreducible tensors of different weight never mix under rotational frame transformation, so that relations between microscopic and macroscopic properties assume a particularly transparent form.

(3) Each irreducible tensor has a unique group theoretical representation which facilitates the derivation of spectroscopic selection rules from symmetry arguments.

Irreducible tensors have traditionally been treated in the spherical tensor formalism, and the relation between Cartesian and spherical tensors has 'been examined by several authors.^{4,13-15} However much less work has been done on the explicit reduction of Cartesian tensors, despite the fact that problems involving vector algebra naturally give rise to tensors in a Cartesian form. Also, Cartesian tensors clearly exhibit directional properties in three-dimensional space which are obscured in spherical tensors. Whilst the reduction of a

second-rank Cartesian tensor is well known, the general reduction of the third rank has only been available in recent years, following the pioneering studies of Coope et al ^{8,11,16} Little work appears to have been done on the reduction of fourth-rank Cartesian tensors; Harris³ and Jerphagnon¹³ and their co-workers have derived certain results, but only for the irreducible tensors in their natural form. (An irreducible tensor of natural form is a tensor with equal rank and weight; such tensors are fully index symmetric and traceless with respect to every pair of indices.) These results are not amenable to immediate use since they are not embedded in the appropriate tensor space of rank four. Explicit results for the fourth-rank reduction have only recently been presented for the special case where the tensor has complete index symmetry.¹⁰

As far as we are aware, the complete and general reduction of a fourth-rank Cartesian tensor has not hitherto been accomplished, and it is our purpose to present the results in this paper. In the following section we outline Coope's reduction procedure and in Sec. III we give the explicit results for the general case of a tensor with no index permutational symmetry. We also show how these results are modified for tensors with specific index symmetry properties. Finally, in Sec. IV, we briefly discuss applications in the realm of crystal physics, and we provide a table of irreducible tensor representations in the seven crystal classes.

II. REDUCTION PROCEDURE

An irreducible tensor of rank n is characterized by a weight $j \leq n$, and it has $(2j + 1)$ independent

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components. The reduction of a Cartesian tensor $T_{(n)}$ generally results in a sum of irreducible tensors, with some weights represented more than once. Hence we can write

 $\overline{}$

$$
T_{(n)} = \sum_{j=0}^{n} \sum_{q=1}^{N_{n}^{(j)}} T_{(n)}^{(j;q)} , \qquad (2.1)
$$

where q is called the seniority index of the irreducible tensor $T^{(j;q)}_{(n)}$ and $N^{(j)}_{n}$ is the multiplicity of weight j in this reduction. This number can be determined using the rules of angular-momenta coupling,¹⁷ and is given by

$$
N_n^{(j)} = \sum_{k} (-1)^k {n \choose k} \left[2n - 3k - j - 2 \atop n - 2 \right], \quad (2.2)
$$

where $0 \le k \le [(n - j)/3]$. Each irreducible tensor has $(2j+1)$ independent components, so that the total number of components in the reduction is

$$
\sum_{j=0}^{n} (2j+1)N_n^{(j)} = 3^n
$$
 (2.3)

as required.

The reduction procedure is based on extraction of the natural forms of tensors with weights $j = 0, 1, \ldots, n$ from $T_{(n)}$, followed by their embedding in the tensor space χ^n of rank n. This is accomplished by means of mappings between tensor subspaces of the same symmetry but different rank. We shall denote the tensor subspace of rank *n* and weight j by \mathcal{H}_j^n , which in turn is expressible as a sum of $N_n^{(j)}$ elementary irreducible subspaces $\mathcal{H}_{i,q}^n$. We first need to introduce the natural projection $E^{(j)}_{(j|j)}$, defined as the projection of χ^j onto the natural tensor subspace \mathcal{H}_j^j . The natural projection is an invariant double tensor of order 2j, which is separately symmetric and traceless in both sets of indices; explicit expressions for $E^{(j)}_{(j|j)}$ with $j \leq 4$ may be found in the work of Coope and Snider.¹⁵ We also need to define isotropic tensors $f_{(m)}^{(0)r}$ of rank m , which are products of $m/2$ Kronecker deltas if m is even, and products of $(m-3)/2$ Kronecker deltas and one Levi-Civita antisymmetric tensor, if m is odd.^{18,19} The index r in $f_{(m)}^{(0)r}$ is used to differentiate the various index permutations of f , each of which contracts with a tensor of rank n to give one of rank j .

The appropriate order m is determined by the contraction properties of the Kronecker delta and Levi-Civita tensors; double contraction by δ , e.g.,

$$
\delta_{s_1s_2}T_{s_1s_2}\cdots_{s_n}\equiv \delta\odot^2 T_{(n)}
$$

lowers the rank by 2 whilst leaving the weight unchanged; similarly double contraction by ϵ , e.g.,

$$
\epsilon_{s_{\tau}s_1s_2}T_{s_1s_2}\cdots s_n\equiv \epsilon\odot^2 T_{(n)}
$$

lowers the rank by ¹ and leaves the weight unchanged. Naturally the result of any such contraction which lowers the rank to less than the weight is a null tensor.

Thus, a tensor of rank $j=(n-2p)$ can be obtained from $T_{(n)}$ by double contraction with p delta tensors; hence the appropriate order for the tensor f here is $m = 2p = (n - j)$. A tensor of rank $j = (n - 2p - 1)$ can be obtained from $T_{(n)}$ by a further double contraction with one epsilon tensor; hence the order of f is

$$
m = (2p + 3) = (n - j + 2)
$$

in this case.

The principal element in the reduction procedure is the mapping $G_{(n|j)}^{(0;q)}$ of the minimal rank tensor subspace $\mathcal{H}_{j,q}^j$ onto $\mathcal{H}_{j,q}^n$. This mapping is constructed by taking the tensor product of the natural projection $E^{(j)}_{(j|j)}$ with one of the isotropic tensors $f_{(m)}^{(0)r}$. In the case of weight 0, the natural projection is unity, and the mappings $G_{n}^{(0)}_{(n)}$ are ident
ical with the isotropic tensors $f_{(n)}^{(0)q}$. For other weights $j \leq n$, we have the following expressions weights $j \le$
for $G_{(n\,|\,j)}^{(0;q)}$:

$$
G_{(n|j)}^{(0;q)} = E_{(j|j)}^{(j)} \otimes {}^{n-j}f_{(n-j)}^{(0)q}
$$
\n
$$
(n-j \text{ even}) \qquad (2.4)
$$
\n
$$
G_{(n|j)}^{(0;q)} = E_{(j|j)}^{(j)} \odot \otimes {}^{n-j+1}f_{(n-j+2)}^{(0)q}
$$
\n
$$
(n-j \text{ odd}) \qquad (2.5)
$$

In Eq. (2.5), the contraction symbols denote an outer product over $(n - j + 1)$ indices together with contraction of one index pair. In both cases the $N_n^{(j)}$ mappings, distinguished by index q, are obtained by taking the tensor products with different index permutations; a suitable linearly independent set of mappings is chosen such that a regular symmetric matrix g_{pg} may be defined through the relation

$$
g_{pq} E^{(j)}_{(j \,|\, j)} = G^{(0;p)}_{(n \,|\, j)} \odot {^n}G^{(0;q)}_{(n \,|\, j)} \ . \tag{2.6}
$$

We now introduce a set of mappings $\widetilde{G}^{(0;p)}_{(j|n)}$ dual to $G^{(0;q)}_{(n|j)}$, and defined by

$$
\widetilde{G}_{(j\,|\,n)}^{(0;p)} = \sum_{q} g^{pq} G_{(n\,|\,j)}^{(0;q)} \,, \tag{2.7}
$$

where g^{pq} and g_{pq} are elements of inverse matrices, i.c.,

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The dual mappings extract the natural forms $t_{(i)}^{(j;q)}$ from the tensor $T_{(n)}$ as follows:

$$
t_{(j)}^{(j;q)} = \widetilde{G}_{(j\,|\,n)}^{(0;q)} \odot^{n} T_{(n)} . \tag{2.9}
$$

Consequently, these tensors may be embedded in the tensor space of order n through the mapping

$$
T_{(n)}^{(j;q)} = G_{(n|j)}^{(0;q)} \odot {}^{j}t_{(j)}^{(j;q)} . \qquad (2.10)
$$

In summary, the irreducible tensors $T_{(n)}^{(j;q)}$ are derived from the relation

$$
T_{(n)}^{(j;q)} = \Pi_{(n}^{(j;q)} \odot {^n}T_{(n)} , \qquad (2.11)
$$

where $\Pi_{(n)}^{(j;q)}$ is an operator given by

$$
\Pi_{(n|n)}^{(j;q)} = G_{(n|j)}^{(0;q)} \odot {}^{j}\widetilde{G}_{(j|n)}^{(0;q)}, \qquad (2.12)
$$

which projects out the qth irreducible subspaces $\mathcal{H}_{j,q}^{n}$ of symmetry j from χ^{n} . The results for tensors of fourth rank given in the following section are entirely based on the application of Eqs. (2.11) and (2.12). Table I shows the key results in the reduction program, and a worked example in the Appendix illustrates the details of the calculations.

III. RESULTS

In this section we give the explicit results for the various weights of a general fourth-rank Cartesian

tensor $T_{s_1s_2s_3s_4}$. Here the reducible representation of the tensor under $0(3)$ is obtained from the prod-
uct of four representations of polar vectors uct of four representations of polar vectors $\mathscr{D}^{1-} \otimes \mathscr{D}^{1-} \otimes \mathscr{D}^{1-} \otimes \mathscr{D}^{1-}$, giving the result $3\mathscr{D}^{0+}\oplus 6\mathscr{D}^{1+}\oplus 6\mathscr{D}^{2+}\oplus 3\mathscr{D}^{3+}\oplus \mathscr{D}^{4+}$. The superscript on each representation labels the weight j and the coefficient corresponds to the multiplicity $N_4^{(j)}$. With no index symmetry the result is an 81dimensional representation.

Table II summarizes the reduction of fourthrank tensors with index symmetry. We adopt the standard representation²⁰ of complete index symmetry in any group of indices by enclosing them in parentheses, e.g., $T_{(s_1s_2)s_3s_4}$ is symmetric with respect to interchange of the indices s_1 and s_2 alone. The multiplicity of each weight in the reduction scheme is determined using Jerphagnon's procedure,¹³ giving the number of independent components shown. The explicit results for each case of index symmetry can be calculated from the correlations shown in Table III; for example, $T^{(0,1)}_{(s_1s_2s_3s_4)}$ is obtained by summing the results of the permutational symmetry in the first three indices. Further results for tensors with index antisymmetry are readily deduced by subtraction of the expressions for the corresponding indexsymmetric tensor from the general results which follow:

$$
T_{s_1s_2s_3s_4}^{(0,1)} = \frac{1}{30} (4 \delta_{s_1s_2} \delta_{s_3s_4} - \delta_{s_1s_3} \delta_{s_2s_4} - \delta_{s_1s_4} \delta_{s_2s_3}) T_{s_\rho s_\rho s_\sigma s_\sigma} \,,\tag{3.1}
$$

$$
T_{s_1s_2s_3s_4}^{(0,2)} = \frac{1}{30}(-\delta_{s_1s_2}\delta_{s_3s_4} + 4\delta_{s_1s_3}\delta_{s_2s_4} - \delta_{s_1s_4}\delta_{s_2s_3})T_{s_\rho s_\sigma s_\rho s_\sigma},\tag{3.2}
$$

$$
T_{s_1s_2s_3s_4}^{(0,3)} = \frac{1}{30}(-\delta_{s_1s_2}\delta_{s_3s_4} - \delta_{s_1s_3}\delta_{s_2s_4} + 4\delta_{s_1s_4}\delta_{s_2s_3})T_{s_\rho s_\sigma s_\sigma s_\rho},\tag{3.3}
$$

$$
T_{s_1s_2s_3s_4}^{(1,1)} = \frac{1}{10} (3\epsilon_{s_{\pi}s_1s_2} \delta_{s_3s_4} - \epsilon_{s_{\pi}s_1s_3} \delta_{s_2s_4} - \epsilon_{s_{\pi}s_1s_4} \delta_{s_2s_3} + \epsilon_{s_{\pi}s_2s_3} \delta_{s_1s_4} + \epsilon_{s_{\pi}s_2s_4} \delta_{s_1s_3} \epsilon_{s_{\pi}s_{\rho}s_{\sigma}} T_{s_{\rho}s_{\sigma}s_{\tau}s_{\tau}},
$$
(3.4)

$$
T_{s_1s_2s_3s_4}^{(1,2)} = \frac{1}{10}(-\epsilon_{s_{\pi}s_1s_2}\delta_{s_3s_4} + 3\epsilon_{s_{\pi}s_1s_3}\delta_{s_2s_4} - \epsilon_{s_{\pi}s_1s_4}\delta_{s_2s_3} - \epsilon_{s_{\pi}s_2s_3}\delta_{s_1s_4} + \epsilon_{s_{\pi}s_3s_4}\delta_{s_1s_2}\epsilon_{s_{\pi}s_\rho s_\sigma}T_{s_\rho s_\tau s_\rho s_\tau},
$$
(3.5)

$$
T_{s_1s_2s_3s_4}^{(1,3)} = \frac{1}{10}(-\epsilon_{s_{\pi}s_1s_2}\delta_{s_3s_4} - \epsilon_{s_{\pi}s_1s_3}\delta_{s_2s_4} + 3\epsilon_{s_{\pi}s_1s_4}\delta_{s_2s_3} - \epsilon_{s_{\pi}s_2s_4}\delta_{s_1s_3} - \epsilon_{s_{\pi}s_3s_4}\delta_{s_1s_2})\epsilon_{s_{\pi}s_{\rho}s_{\sigma}}T_{s_{\rho}s_{\tau}s_{\rho}s_{\sigma}},
$$
(3.6)

$$
T_{s_1s_2s_3s_4}^{(1,4)} = \frac{1}{10} (\epsilon_{s_{\pi}s_1s_2} \delta_{s_3s_4} - \epsilon_{s_{\pi}s_1s_3} \delta_{s_2s_4} + 3\epsilon_{s_{\pi}s_2s_3} \delta_{s_1s_4} - \epsilon_{s_{\pi}s_2s_4} \delta_{s_1s_3} + \epsilon_{s_{\pi}s_3s_4} \delta_{s_1s_2}) \epsilon_{s_{\pi}s_{\rho}s_{\sigma}} T_{s_{\tau}s_{\rho}s_{\sigma}s_{\tau}},
$$
(3.7)

$$
T_{s_1 s_2 s_3 s_4}^{(1,5)} = \frac{1}{10} (\epsilon_{s_{\pi} s_1 s_2} \delta_{s_3 s_4} - \epsilon_{s_{\pi} s_1 s_4} \delta_{s_2 s_3} - \epsilon_{s_{\pi} s_2 s_3} \delta_{s_1 s_4} + 3 \epsilon_{s_{\pi} s_2 s_4} \delta_{s_1 s_3} - \epsilon_{s_{\pi} s_3 s_4} \delta_{s_1 s_2}) \epsilon_{s_{\pi} s_{\rho} s_{\sigma}} T_{s_{\tau} s_{\rho} s_{\tau} s_{\sigma}},
$$
(3.8)

$$
T_{s_1s_2s_3s_4}^{(1,6)} = \frac{1}{10} (\epsilon_{s_4s_1s_3} \delta_{s_2s_4} - \epsilon_{s_4s_1s_4} \delta_{s_2s_3} + \epsilon_{s_4s_2s_3} \delta_{s_1s_4} - \epsilon_{s_4s_2s_4} \delta_{s_1s_3} + 3\epsilon_{s_4s_3s_4} \delta_{s_1s_2} + \epsilon_{s_4s_4s_5} T_{s_4s_4s_5} \delta_{s_1s_2} \tag{3.9}
$$

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T^{2,1}_{1,2,1}
\n
$$
T_{1,2,1,1}^{(1)} = \frac{1}{u} \delta_{1,1,1} (T_{1,p,p,p,q} + T_{1,p,p,p,q} - \frac{2}{3} \delta_{2,1,1} T_{1,p,p,q,p} - \frac{1}{16} \delta_{1,1,1} T_{1,p,p,q,p} - \frac{1}{16} \delta_{1,2,1} T_{1,p,p,q,p} - \frac{2}{3} \delta_{2,2,1} T_{1,p,q,p,q} - \frac{2}{3} \delta
$$

$$
+\frac{1}{4}\epsilon_{i_1i_2i_3}\epsilon_{i_1i_3i_4}\epsilon_{i_1i_4i_5}\epsilon_{i_1i_4i_5}\epsilon_{i_1i_4i_5}\epsilon_{i_1i_4i_5}\epsilon_{i_1i_4i_5i_5i_5}\epsilon_{i_1i_4i_5i_5i_5}\epsilon_{i_1i_4i_5i_5i_5}\epsilon_{i_1i_4i_5
$$

$$
+ S_{s_1s_4} T_{s_3s_7s_6} + S_{s_1s_4} T_{s_2s_7s_7s_8} + S_{s_2s_1} T_{s_2s_7s_8s_9})
$$

\n
$$
+ \frac{3}{4} \epsilon_{s_1s_3s_4} \epsilon_{s_2s_7s_9} \left[\frac{1}{6} (S_{s_2s_7} T_{s_1s_2s_7s_7} + S_{s_2s_1} T_{s_2s_1s_7s_7} + S_{s_2s_1} T_{s_2s_1s_7s_9} + S_{s_2s_1} T_{s_1s_1s_7s_9} + S_{s_2s_1} T_{s_1s_1s_7s_9} + S_{s_2s_1} T_{s_1s_1s_7s_9} + S_{s_2s_1} T_{s_1s_1s_7s_9} + S_{s_2s_1} T_{s_1s_2s_7s_9} + S_{s_2s_1} T_{s_1s_2s_7s_9} + S_{s_2s_1} T_{s_2s_7s_7s_9} + S_{s_2
$$

Verification of the above results can be accomplished as follows. Each result is expressible in the form

$$
T_{s_1s_2s_3s_4}^{(j;q)} = \sum_{r=1}^{105} a_r^{(j;q)} T_{s_5s_6s_7s_8} f_{s_1s_2s_3s_4s_5s_6s_7s_8}^{(0)r} ,
$$
\n(3.20)

i.e., as a linear combination of contractions of the original tensor with the 105 index permutations of the eighth-rank isotropic tensor $\delta_{s_1s_2}\delta_{s_3s_4}\delta_{s_5s_6}\delta_{s_7s_8}$. Addition of the complete set of $T_{s_1s_2s_$

$$
T_{s_1s_2s_3s_4} = \sum_{r=1}^{105} A_r T_{s_5s_6s_7s_8} f^{(0)r}_{s_1s_2s_3s_4s_5s_6s_7s_8} \t\t(3.21)
$$

where

 \sim

TABLE I. Key results in the reduction of a fourth-rank Cartesian tensor.

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Index symmetry	Number of independent	Weight				
	components	0				
$s_1s_2s_3s_4$	81		6	6		
$(s_1s_2)s_3s_4$	54					
$(s_1s_2)(s_3s_4)$	36					
$(s_1s_2s_3)s_4$	30					
$[(s_1s_2)(s_3s_4)]$	21		0			
$(s_1s_2s_3s_4)$	15		0			

TABLE II. Number of independent components and the multiplicity of each weight in the reduction of a fourth-rank tensor.

TABLE III. Correlation of irreducible tensors with different index symmetries.

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Weight 4

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Class

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 $1 + \frac{1}{2}$

 $R_{\rm g}+E_{\rm g}+T$

 $T+\mathbf{1}$

 $\ddot{+}$

 $2g + B_1$ $\frac{1}{1}$

 $3A_{g} + 2B_{1}$ $A_{g}+4B_{g}$

 $_{g}+2A_{2g}+2E$

 $1g + 2E$

$$
A_r = \sum_{j=0}^{4} \sum_{q=1}^{N_q^{(j)}} a_r^{(j;q)} . \tag{3.22}
$$

However, the 105 eighth-rank isotropic tensors are not all linearly independent, and use has to be made of the relationships between them which we have derived elsewhere.²¹ Equation (3.21) can then be written in such a form that the coefficient sum A_r is zero for all r , except that which corresponds *A*, is zero for all *r*, except that which corresponds
to the isotropic tensor $\delta_{s_1s_5}\delta_{s_2s_6}\delta_{s_3s_7}\delta_{s_4s_8}$, where its value is unity; hence the equality is proven.

IV. APPLICATIONS

The results given in this paper can be applied to many areas of physics, as outlined in the Introduction. One important area for consideration is that of solid-state physics, and in particular, thc applications to optical, electromagnetic, and mechanical properties of crystals. For example, a tensor with properties of crystals. For example, a tensor with
the structure $T_{(s_1s_2s_3s_4)}$ arises in the theory of laser four-photon absorption, the tensor $T_{(s_1s_2)(s_3s_4)}$ is required for the Kerr effect, and the tensor $T_{[(s_1,s_2)(s_3,s_4)]}$ can be applied in the theory of crystal elasticity.

To facilitate such applications to crystals of a particular symmetry class, we present in Table IV the representations spanned by irreducible polar tensor components in the holosymmetric point groups of each class; the corresponding representations for groups of lower symmetry are readily obtained by the use of correlation tables. (For axial fourth-rank tensors, the corresponding ungerade representations apply.) For processes such as the Kerr effect, where the tensor must possess thc same symmetry properties as the crystal, only those tensor components which transform under the totally symmetric representations are nonzero; thus, for example, in holosymmetric cubic crystals only weight-0 and weight-4 contributions arise.

In the case of four-photon absorption, the excited-state symmetry dictates the representation under which the tensor components must transform. We have recently shown how the application of this rule to four-photon absorption in gaseous or liquid media, where the constituent molecules are randomly oriented, enables the symmetry properties of the excited states to be determined from a combination of experiments with different laser-beam polarizations.¹⁰ In order to further illustrate the details of such an application, we now briefly discuss the case of four-photon absorption in oriented crystals.

Orthorhombic
Rhombohedral $\frac{1}{2}$ $\frac{1}{2}$ $\frac{1}{2}$ FS
OU
E

 mmm \mathbf{a}

> Tetragona
Hexagona
Cubic etragonal

> $4/mmm$ $6/mmm$ $m\bar{3}m$

Using the principles of quantum electrodynamics, it is readily shown that the rate of four-photon absorption from a single laser beam of intensity I is given by¹⁰

$$
\Gamma = \frac{3\pi^5 I^4 \rho_f}{\hbar c^4} |T_{(s_1 s_2 s_3 s_4)} S_{(s_1 s_2 s_3 s_4)}|^2 , \qquad (4.1)
$$

where ρ_f is the density of final states, $T_{(s_1 s_2 s_3 s_4)}$ is a polar nonlinear susceptibility tensor, and $S_{(s_1 s_2 s_3 s_4)}$ is a radiation tensor consisting of a product of components of the polarization vector \vec{e} ;

$$
S_{(s_1s_2s_3s_4)} = e_{s_1}e_{s_2}e_{s_3}e_{s_4} \t\t(4.2)
$$

Since this tensor is fully index symmetric, it may be expressed as a sum of weight-o, -2, and -4 terms, and the results of Sec. III lead to the following expressions:

$$
S_{s_1 s_2 s_3 s_4}^{(0)} = \frac{1}{15} (\delta_{s_1 s_2} \delta_{s_3 s_4} + \delta_{s_1 s_3} \delta_{s_2 s_4} + \delta_{s_1 s_4} \delta_{s_2 s_3}) (\vec{e} \cdot \vec{e})^2 ,
$$
\n(4.3)

$$
S_{s_1s_2s_3s_4}^{(2)} = \frac{1}{7} (\delta_{s_1s_2} e_{s_3} e_{s_4} + \delta_{s_1s_3} e_{s_2} e_{s_4} + \delta_{s_1s_4} e_{s_2} e_{s_3} + \delta_{s_2s_3} e_{s_1} e_{s_4} + \delta_{s_2s_4} e_{s_1} e_{s_3} + \delta_{s_3s_4} e_{s_1} e_{s_2}) (\vec{e} \cdot \vec{e}) - \frac{10}{7} S_{s_1s_2s_3s_4}^{(0)},
$$
(4.4)

$$
S_{s_1s_2s_3s_4}^{(4)} = S_{s_1s_2s_3s_4} - S_{s_1s_2s_3s_4}^{(0)} - S_{s_1s_2s_3s_4}^{(2)} \t\t(4.5)
$$

Note that $(\vec{e} \cdot \vec{e})$ is unity for plane polarized light, but zero for circularly polarized light, where the polarization vector is complex.

From Eq. (4.1) we now have

 ϵ - ϵ

$$
\Gamma = \frac{3\pi^5 I^4 \rho_f}{\hbar c^4} \left| T^{(0)}_{(s_1 s_2 s_3 s_4)} S^{(0)}_{(s_1 s_2 s_3 s_4)} + T^{(2)}_{(s_1 s_2 s_3 s_4)} S^{(2)}_{(s_1 s_2 s_3 s_4)} + T^{(4)}_{(s_1 s_2 s_3 s_4)} S^{(4)}_{(s_1 s_2 s_3 s_4)} \right|^2. \tag{4.6}
$$

Substituting from Eqs. (4.3) to (4.5) and making use of the index symmetry in the susceptibility tensor then leads to the result

$$
\Gamma = \frac{3\pi^5 I^4 \rho_f}{\hbar c^4} + \frac{1}{5} T^{(0)}_{(s_1 s_1 s_2 s_2)} (\vec{e} \cdot \vec{e})^2 + \frac{6}{7} T^{(2)}_{(s_1 s_1 s_2 s_3)} (\vec{e} \cdot \vec{e}) e_{s_2} e_{s_3} + T^{(4)}_{(s_1 s_2 s_3 s_4)} e_{s_1} e_{s_2} e_{s_3} e_{s_4} |^2. \tag{4.7}
$$

This result is generally applicable to four-photon absorption in crystals of any symmetry class, but only certain terms will arise for a given transition; for example, reference to Table IV shows that a four-photon transition to an E_g state in a holosymmetric cubic crystal would involve only the weight-2 and -4 terms. Several other interesting features emerge, however. Suppose, for example, that four-photon absorption is observed in such a crystal at an optical frequency ω , and the transition proves to be forbidden for two-photon absorption at frequency 2ω ; in other words the transition is forbidden on symmetry grounds rather than on the basis of energy matching. In this case the only nonzero irreducible components of the susceptibility tensor are those of weight 4, and hence it is immediately clear from Table IV that the excited-state symmetry is $T_{1_{g}}$.

From the brief analysis above, it is evident that irreducible Cartesian tensor methods have a great deal to offer in several areas of crystal physics. For a fuller demonstration of the power of applying these methods to physical problems, however, we would again refer the reader to our recent work on multiphoton absorption in fluids.¹⁰

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APPENDIX: ILLUSTRATION OF THE REDUCTION PROGRAM

We illustrate the scheme for the reduction of a fourth-rank Cartesian tensor by reference to the case of weight $j=1$. Using the results of Sec. II, we employ the six isotropic tensors of order $n -j +2=5$ shown in Table I. These generate a set of six linearly independent mappings, the first of which is

$$
G_{(4|1)}^{(0;1)} = \sum_{s_{\tau}} E_{(r_1|s_{\tau})}^{(1)} \epsilon_{s_{\tau}s_1s_2} \delta_{s_3s_4} ,
$$

from Eq. (2.5) ; the natural projection operator in this expression is given by¹⁵

$$
E^{(1)}_{(r_1|s_{\tau})} = \delta_{r_1s}
$$

and hence $G^{(0;1)}_{(4|1)} = \epsilon_{r_1s_1s_2} \delta_{s_3s_4}$. The matrix g_{pq} is generated using Eq. (2.6); for example, the first element is obtained from the result

$$
G_{(4|1)}^{(0;1)} \odot {}^4G_{(4|1)}^{(0;1)} \! = \! \epsilon_{r_1s_1s_2} \delta_{s_3s_4} \epsilon_{r_1's_1s_2} \delta_{s_3s_4} \! = \! 6 \delta_{r_1r_1'} \! = \! 6E_{(r_1|r_1')}^{(1)}
$$

so that $g_{11} = 6$. Calculation of the matrix g_{pq} and its inverse g^{pq} leads to the expressions for the dual mappings using (2.7); for example,

$$
\widetilde{G}_{(1|4)}^{(0;1)} = \frac{1}{10} (3G_{(4|1)}^{(0;1)} - G_{(4|1)}^{(0;2)} - G_{(4|1)}^{(0;3)} + G_{(4|1)}^{(0;4)} + G_{(4|1)}^{(0;5)} + 0G_{(4|1)}^{(0;6)})
$$

Finally, the projection operator $\Pi_{(4|4)}^{(0;1)}$ which projects out the first irreducible subspace $\mathcal{H}_{1,1}^4$ of weight 1 from χ^4 follows from Eq. (2.12):

$$
\Pi_{(4|4)}^{(0;1)} = G_{(4|1)}^{(0;1)} \odot {}^1\tilde{G}_{(1|4)}^{(0;1)} \n= \frac{1}{10} \epsilon_{r_1s'_1s'_2} \delta_{s'_3s'_4} (3\epsilon_{r_1s_1s_2} \delta_{s_3s_4} - \epsilon_{r_1s_1s_3} \delta_{s_2s_4} - \epsilon_{r_1s_1s_4} \delta_{s_2s_3} + \epsilon_{r_1s_2s_3} \delta_{s_1s_4} + \epsilon_{r_1s_2s_4} \delta_{s_1s_3} ,
$$

and subsequent use of Eq. (2.11) immediately leads to the results (3.4).

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