

# Direct Determination of Crystal Structure

## III. Phase Determination Method

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### Abstract

Phase determination method is developed by improving sign determination method discussed by Takahashi (Bull. Kagoshima Univ. Fac. Educ. (Nat. Sci.). 1985, **37**, 5-9). The tested material is an organic crystal,  $C_{11}H_{13}NO_3$ , analyzed by Furusaki, Abe and Matsumoto (Bull. Chem. Soc. Japan. 1982, **55**, 611-612). The space group of the material is  $2_1$  and the number of independent atoms is 30. The number of correct phase sets obtained is 3 from the 10 initial sets whose phases are given randomly.

### 1. Introduction

The author has been engaged in phase determination for five years. The method to determine signs of the structure factors was published in 1985. In the paper, the author noted that the method was not efficient for the phase determination. The author suggested that the root of the failure may be the unsatisfactory of the structure factor convergence. After many trials and errors, it turned out that the method was essentially very effective, and that the origin of the failure was that the convergence of the phases was too rapid for the phase determination.

The slight improvement to make slow the convergence gave very satisfactory results, that is, 3 sets of phases which can be regarded to be correctly determined in this stage of phase determination development were obtained from 10 initial sets of phases which were given randomly, there were 2 sets of which R-value and Q-value were nearly the same for those of the 3 sets. The tested material was an organic crystal,  $C_{11}H_{13}NO_3$ , analyzed by Furusaki, Abe and Matsumoto (1982). The author's stance for the recent development of probability theoretical approach to the phase problem is very negative. The results suggest that the

author's way to solve phase problem is the right way.

## 2. Examination of basic concepts

The phases of structure factors of a crystal depend on the location of origin of the crystal structure. Hence the position of the origin for the phases obtained by the author's method must be determined together with the number of the correct phases. Since the space group of the tested material is  $2_1$ , the origins of the coordinates can be chosen as following four types,

$$(0, y, 0), (1/2, y, 0), (0, y, 1/2) \text{ and } (1/2, y, 1/2),$$

and the suitable value of  $y$  must be determined for the set of phases.

The unique determination of the value of  $y$  was very difficult and there was no good idea for the determination, the author adopted trial and error method. The phases obtained were compared with true ones with the four origins, where the value of  $y$  was determined by trial and error. When the absolute value of the difference between the both values was smaller than  $\pi/8$ , the obtained value was regarded as being equal to the true value. The meanings that the phase obtained is equal to the true phase in this paper is that the absolute value of the difference between the both phases is less than  $\pi/8$ .

### (a) Average structure factors

The possibility of the phase determination depends on the accuracy of the following average normalized structure factors obtained by Karle and Karle (1966),

$$E_h = \sigma_2^{3/2} \sigma_3^{-1} \langle E_k E_{h-k} \rangle_k. \quad (1)$$

Hereafter, normalized structure factors will be called simply structure factors.

To estimate the accuracy of the equation (1), the R-values, the standard deviations of the absolute values of the calculated structure factors from the true values and of the calculated phases from the true phases were calculated after one cycle calculation with the equation (1) for stronger 200 reflections, where one cycle calculation means that all the average structure factors are calculated by the equation (1) once. The set of the absolute values of the true structure factors is filed according to the magnitude of the values in this study. The R-value was 0.13954, the standard deviation of the absolute structure factors was 0.53903 and that of the phases was  $0.26639 \text{ rad} = 15.3 \text{ deg}$ . For these values, we can judge that the equation (1) is very good approximation for obtaining average structure factors.

To examine the description by Furusaki (1979) that "as tangent procedure is repeated, every one of these random phase sets approaches the correct phase set", the average structure factors of all reflections were calculated 10 cycles for the 200 reflections. The number of the calculated phases equal to the true phases was 157. When the structure factors of the strongest 30 reflections were fixed invariably and the other ones were put to be 0 at beginning of the calculation, the number became 161 for 10 cycles calculations. But, when the structure factors of the 30 reflections varied at each cycle of calculation according to the equation (1), the number became 49.

In addition, the cases that one third of the phases of the stronger (case 1) and weaker (case 2) reflections of the 30 strong reflections are replaced by the randomly given phases were examined. In the former case, the maximum number became 59 for 10 cycles calculations and 56 for 100 cycles, and in the latter case the number became 105 for 10 cycles and 132 for 100 cycles.

The author's interpretation for these results is : the contribution of  $E_k$  to structure factor  $E_h$  in the equation (1) is proportional to the absolute value of  $E_k$ , and when  $n$  cycles calculations are done, the contribution becomes proportional to the  $n$ th power of the absolute value if the phase is fixed invariably. This interpretation may be able to explain why the 161 phases in the 200 phases can be determined by only the 30 phases of strong reflections. Hence, if there are not correct phases with large absolute value of the structure factors, the negative effect becomes large and the phase determination becomes impossible. This can be understood by the fact that the phase determination is impossible even though by using the strongest 30 reflections, if the 30 phases are not fixed.

A speculation drawn from the above interpretation is as follows. When  $N_{ref}$  structure factors, by which the crystal structure can be determined, can be derived from the  $L$  strongest structure factors by the equation (1), and the  $N_{ref}$  structure factors becomes invariable after more than  $n$  cycle calculations with the fixed  $L$  phases, that is, the phase set converges to a certain set after the  $n$  cycle calculations, if the number of the correct structure factors in the  $L$  structure factors is  $L_c$  and the number of the incorrect structure factors is  $L_{ic}$ ,  $L = L_c + L_{ic}$ , the condition that most phases are correctly determined may be as follows

$$\sum_c |E_h|^n \gg \sum_{ic} |E_h|^n, \quad (2)$$

where  $\sum_c$  and  $\sum_{ic}$  means the summation over the correct and incorrect structure factors, respectively.

For the above test cases, the numbers of cycles for the convergence of the phase set are

determined if the phases of 85% agree with the after and before one cycle calculation, the number  $n$  become 9 for the case 1 and 6 for the case 2. The numbers for the equation (2) become

$$\sum_{i=1}^{10} |E_i|^9 = 4.07 \times 10^6 \quad \text{and} \quad \sum_{i=11}^{30} |E_i|^9 = 1.45 \times 10^6 \quad \text{for case 1,}$$

$$\sum_{i=1}^{20} |E_i|^6 = 71930.1 \quad \text{and} \quad \sum_{i=21}^{30} |E_i|^6 = 14714.9 \quad \text{for case 2.}$$

The phase set obtained in the case 2 was extended to the case of 400 reflections. The R-value was 0.11619, Q-value was 0.6783 and the number of phase equal to the the true phases was 370. The above values would indicate that the author's speculation is valid.

If the initial set consists of the 10 stronger correct phases and the 20 weaker incorrect phases, when the number of cycles  $n$  becomes very large, then the equation (2) is satisfied for  $n$ . After 100 cycle calculations of the set, the obtained set was very bad. Hence, we can conclude that the frame of the phase set is almost determined when the set converges, the additional calculations do only refine the set.

#### (b) Probability of phases

Probability of the phase  $S_h$  obtained by

$$S_h = \text{phase of } \left( \sum_k E_k E_{h-k} \right) \quad (3)$$

is

$$P(S_h) = \frac{1}{2} + \frac{1}{2} \tanh(\sigma_3 \sigma_2^{-3/2} |E_h| | \sum_k E_k E_{h-k} |). \quad (4)$$

As discussed in the previous paper, the probability is not the absolute probability for the phase of the  $h$  reflection being  $S_h$ , but the conditioned probability that when the structure factors of  $k$  and  $h-k$  reflections are  $E_k$  and  $E_{h-k}$ , then the phase of the  $h$  reflection becomes  $S_h$  with the probability  $P(S_h)$ . To show that the probability given by the equation (3) is not absolute probability but conditioned one is very easy. When we give randomly the phases to the structure factors and calculate the average structure factors by the equation (1), if we find the correlation coefficient between the differences of the phases of the calculated structure factors and the true phases and the probabilities to be nearly 0, then the probability given by the equatoin (4) becomes independent of the phase.

Since the probabilities became 1.0 for almost all reflections, so that the examination was carried out by using the following values,

$$\sigma_h = \sigma_3 \sigma_2^{-3/2} |E_h| \left| \sum_k E_k E_{h-k} \right|. \quad (5)$$

The correlation coefficient was very small and was 0.007 for the trial calculation. This indicates that the probability is quite of no correlation to the difference between the true phase and the calculated phase.

As seen easily from the equation (1), when structure factors of all reflections are known correctly, the equation (5) becomes

$$\alpha_h = N |E_h|^2, \quad (6)$$

where  $N$  is the number of pairs contributing to the  $h$  structure factor by the form of  $E_h E_{h-k}$ . This means that there is a suitable value of the probability and that the large probability does not necessarily mean that the phase of the  $h$  reflection is correctly given. This can be easily seen, since if we give all phase to be 0, then the probability becomes the largest value.

Germain and Woolfson (1979) recommended the use of the weight function for the calculation of the tangent of phase. The weight function is proportional to the second term in the right side of the equation (4). The present author's conclusion is that this weight function is useless to the calculation.

### (c) Q-value

The definition of Q-value is:

$$Q = 1.0 - \left( \sum_k E_h \left( \frac{|\sum_k E_k E_{h-k}|}{\sum_k |E_k E_{h-k}|} \right) \right) / \sum_h |E_h|. \quad (7)$$

Since the value  $\sum_k E_k E_{h-k}$  is proportional to the average structure factor given by the equation (1), we can understand the second term in the equation (7) analogously with structure factor that if  $E_k E_{h-k}$  is compared with scattering factor then  $|\sum_k E_k E_{h-k}|$  is the absolute value of the structure factor of the  $h$  reflection, then  $|\sum_k E_k E_{h-k}| / \sum_k |E_k E_{h-k}|$  is compared with the absolute value of the structure factor divided by the sum of scattering factor. Hence the second term becomes the average misfit of the phases with the weight of the absolute values of the structure factors.

Furusaki (1979) described that the correct phase set had to have not only a low R-value but also a moderately high Q-value. We can not know what value is moderate, but large Q-value seems to be inadequate for correct phase set.

### 3. Method and Results

The author already described the method to obtain the signs of structure factors from the set of the absolute values of the structure factors in the previous paper (Takahashi, 1985). The principles of the method are: (1) all signs are given randomly, that is, if the random number less than 1 and larger than 0 is less than 0.5, then the sign is given to be minus, and if the random number is more than 0.5, then the sign is given to be plus, (2) the average structure factors of all the reflections are calculated by using the signs and the observed absolute values of the structure factors (the true values were substituted in the study), (3) the difference of the absolute value of the calculated average structure factor and the absolute value of the observed (true) structure factor, divided by the true structure factor, is chosen as the measure of the fitness of the signs, (4) the contribution of several most unfitting reflections and those related to the reflections are put to be 0, this is done by putting  $\exp(iS_h)=0$ , (5) the average structure factors are calculated by using the remaining reflections, (6) the coincidence between the signs calculated in (2) and (5) are examined, and if all signs coincide, the calculation is stopped and results are printed out, if not, we return to (3) and continue the calculation.

A few technical improvement to save CPU time were done for the phase determination.

- (a) Absolute values of structure factors were previously calculated and filed.
- (b) The number of pairs of  $k$  and  $h-k$  reflections and the sequence numbers in the file were previously calculated and filed.
- (c) The limit of the number of phases in stage (4) are previously set, and the calculations of the structure factors were done successively until all the structure factors were not to be 0.

In addition, the reflections put to null contribution were chosen from the following two classes.

- (A) The most fitted reflections and their related reflections were chosen and  $\exp(iS_h)$ s of the other reflections were put to be 0.
- (B) The most unfitting reflection and their related reflections were chosen and their  $\exp(iS_h)$ s were put to be 0.

A trial calculation for the 200 reflections, where the number of reflections in the (A) was 100, gave the results that the R-value was 0.13482, Q-value was 0.54612 and the number of phases determined was 59. The CPU time was very large.

At another trial, where the number of reflections in (B) was 100, the R-value of 10 sets were

nearly 0.2 and the numbers of phases determined were less than 60. The CPU times was very small. Even the case of the set of (B) with the number of the reflections being 150, that is, the number of the remaining reflections with non-zero structure factors was 50, the convergence was too rapid, and the number of phases agreed between before and after one cycle calculation almost monotonously converged to 200.

Hence in order to moderate the convergence speed, the number of reflections in (A) was set

Table 1. NC is the number of the correct phases.

No.	R	Q	NC	R	Q	NC
1	0.19391	0.56130	49			
2	0.19417	0.56797	70			
3	0.14085	0.55281	146	0.11645	0.70578	369
4	0.13382	0.54474	157	0.11645	0.69674	373
5	0.19377	0.54978	51			
6	0.19823	0.57246	67			
7	0.14003	0.54737	61	0.11645	0.69096	112
8	0.13720	0.55226	61	0.11645	0.70580	108
9	0.188448	0.56985	56			
10	0.13956	0.55178	138	0.11645	0.67533	373
200	0.13171	0.54884	157			
400				0.11618	0.67534	371

130 and the number in (B) 60, and tried again. The results of the calculations are listed in Table 1. The total CPU time for the calculation could not be known since the calculation was done by TSS. But the time was probably less than 5 min. The program list is given in *Appendix*. The five sets with R-value nearly 0.14 were further extended to the case of the number of reflections being 400. The results are also listed in Table 1. It is interesting that the R-values of the five sets are the same and the numbers of the determined phases of the three set are about 370 but the numbers of the other two sets are about 110, which may be obtained from the set of phases given randomly.

The last two lines in the Table 1 are the results obtained from true phases with 200 reflections and 400 reflections by 10 cycle calculations.

#### 4. Discussions

In general, the convergence speed become slower if the number of non-zero structure factor is smaller in the stage (4) in the preceding section. The convergence speed depends also on

symmetries of crystal structures. When the symmetry is higher, the number of the related reflections to a reflection with respect to the equation (1) becomes larger, then the convergence speed becomes slower. The R-value for the set of phases calculated by the present author's method would converge to that for the set of true phases at any time, if convergence speed is very slow.

When convergence speed is very slow, we may have many set of phases which have nearly the same R-values and Q-values as those of the true structure but quite different from the set of true phases. The crystal structures analyzed by these sets may be homometric structures in practical sense. We can not discriminate these set of phases from the correct set, because we do not have any theoretical criteria to choose true set from many similar sets. Practically, we can decrease the number by adjusting convergence speed by using the sets (A) and (B).

The success of the symbolic addition method of Karle and Karle (1966), of the multiple solution method of Germain and Woolfson (1968, 1970) and of the Monte Carlo method of Furusaki (1979) depends on the choice of suitable initial set of phases. As seen in the preceding section, if we can choose comparatively small number of correct phases of strong reflections, the obtained phases by using the equation (1) are probably correct. But how can we choose the correct phases even though the number of the phases is small? At first, the multiple solution method is useless, since this depends on the probability of phases which is meaningless as discussed in the preceding section. The success of the Monte Carlo method depends on the chance where the structure factors satisfy the equation (2). The 30 strong reflections are grouped in the five groups where the reflections are related each other with respect to the equation (1). The largest group consists of 16 isolated reflections. The number of reflections in the next large group is only 5. We can not determine the phases of the 30 reflections by the equation (1). When the number of the reflections becomes 50, the largest number of reflections related each other becomes 18. However, we could not determine correctly the 18 phases by using the equation (1), since the set of phases obtained by using the 30 strong reflections where the phases were not fixed at each cycle of calculations were not good as described in the preceding section. The accuracy of the phase determination for the 18 reflections in symbolic addition method is naturally lower than that of the author's test calculation for the 30 reflections. Hence, even though the phases of the test material could be determined by symbolic addition method, it might be a very fortunate case. The author believes that the correct phases must be derived from the absolute values of structure factors of  $6N$  reflections at least, where  $N$  is the number of independent atoms in

a unit cell.

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COMPLEX EPHS
DIMENSION IH(3,200),EPHS(200),MRL(200,20,2),
1AF(200),ANGM(200),AFC(200),NRM(200)
PAIT=6.283185307179586
PAI=PAIT/2.0
500 FORMAT(8I10)
IX=8735
READ(1,500)NRR
NREF=200
READ(1,510)((IH(J,I),J=1,3),I=1,NREF)
510 FORMAT(4(3I5,2X))
520 FORMAT(10I5)
READ(2,520)NRR
READ(2,530)F0
READ(2,530)(AF(I),I=1,NREF)
530 FORMAT(4F15.5)
DO 10 I=1,NREF
READ(3,500)NRM(I)
IF(NRM(I).EQ.0) GO TO 10
READ(3,540)(MRL(I,J,1),MRL(I,J,2),J=1,NRM(I))
10 CONTINUE
540 FORMAT(4(2I5,5X))
EPS=PAIT/16
NCAL=10
NTR=0
100 CONTINUE
NTR=NTR+1
IF(NTR.GT.NCAL) STOP
DO 20 I=1,NREF
ANGM(I)=10.0
EPHS(I)=0.0
CALL RANPHS(I,IH,EPHS(I),IX)
20 CONTINUE
DO 30 I=1,NREF
IED=NRM(I)
IF(IED.EQ.0)GO TO 30
CALL DETPHS(I,IED,IH,AF,EPHS,MRL,EPHS(I),AFC(I),ANGM(I),PAIT)
30 CONTINUE
CALL CORFAC(NREF,AF,AFC)
CALL FACCUM(NREF,IH,AF,MRL,NRM,EPHS,ANGM,AFC,PAIT,IX,EPS)
CALL RVALUE(NREF,AF,AFC,RI)
CALL QVALUE(NREF,MRL,NRM,AF,EPHS,QI)
NDTD=0
DO 60 I=1,NREF
IF(ANGM(I).EQ.10.0) GO TO 60
NDTD=NDTD+1
60 CONTINUE
WRITE(6,600)NTR,RI,QI,NDTD
600 FORMAT(5X,'TRIAL',I5,5X,'RI =',F10.5,
1/,5X,'QI =',F10.5,5X,'NDTD =',I5)
WRITE(11,550)NTR
550 FORMAT(I10)
WRITE(11,560)RI,QI
610 FORMAT(5X,'RI =',F10.5)
560 FORMAT(3F10.5)
WRITE(11,570)(ANGM(I),I=1,NREF)
570 FORMAT(4(5X,F10.5))
GO TO 100

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END
SUBROUTINE DETPHS(IJ, IED, IH, AF, EPHS, MRL, EPH, AC, ANG, PAIT)
COMPLEX EPHS, EPH, SUMEH, SM, REPS
DIMENSION IH(3,200), AF(200), EPHS(200), MRL(200,20,2), IK(3),
1 AFC(200)
SUMEH=0.0
NSM=0
DO 10 I=1, IED
J=MRL(IJ, I, 1)
K=MRL(IJ, I, 2)
DO 20 KA=1, 3
IK(KA)=IH(KA, IJ)-IH(KA, J)
20 CONTINUE
CALL REPHS(K, IH, IK, EPHS(K), REPS)
SM=EPHS(J)*REPS
IF(SM.NE.0.0)NSM=NSM+1
SUMEH=SUMEH+SM*AF(J)*AF(K)
10 CONTINUE
IF(NSM.EQ.0)AFC(IJ)=0.0
IF(NSM.EQ.0)ANG=10.0
IF(NSM.EQ.0)EPH=0.0
IF(NSM.EQ.0)RETURN
SUMEH=SUMEH/NSM
AA=CONJG(SUMEH)*SUMEH
AC=SQRT(AA)
CALL UNICMP(SUMEH, AC, EPH, ANG, PAIT)
IF(IH(2, IJ).EQ.0.AND.REAL(EPH).GE.0.0)ANG=0.0
IF(IH(2, IJ).EQ.0.AND.REAL(EPH).LT.0.0)ANG=PAIT/2.0
RETURN
END
SUBROUTINE UNICMP(FC, ABF, UNTV, ANG, PAIT)
COMPLEX FC, UNTV
IF(ABF.EQ.0.0)UNTV=0.0
IF(ABF.EQ.0.0)RETURN
A=REAL(FC)/ABF
B=AIMAG(FC)/ABF
UNTV=CMPLX(A, B)
CALL ARG(PAIT, UNTV, ANG)
RETURN
END
SUBROUTINE CORFAC(NREF, AF, AFC)
DIMENSION AF(200), AFC(200)
SUMF=0.0
SUMSC=0.0
DO 10 I=1, NREF
SUMF=SUMF+AF(I)
SUMSC=SUMSC+AFC(I)
10 CONTINUE
CK=SUMF/SUMSC
DO 20 I=1, NREF
AFC(I)=CK*AFC(I)
20 CONTINUE
RETURN
END
SUBROUTINE RVALUE(NR, AF, AFC, RV)
DIMENSION AF(200), AFC(200)
SUMF=0.0
DO 10 I=1, NR

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SUMF=SUMF+AF(I)
10 CONTINUE
SUM=0.0
DO 20 I=1,NR
SUM=SUM+ABS(AF(I)-AFC(I))
20 CONTINUE
RV=SUM/SUMF
RETURN
END
SUBROUTINE QVALUE(NREF,MRL,NRM,AF,EPHS,QV)
COMPLEX EPHS,SUMA,AA
DIMENSION MRL(200,20,2),NRM(200),AF(200),EPHS(200)
SUM=0.0
SUMF=0.0
DO 10 I=1,NREF
SUMA=0.0
SUMB=0.0
IED=NRM(I)
IF(IED.EQ.0) GO TO 10
DO 20 J=1,IED
JA=MRL(I,J,1)
JB=MRL(I,J,2)
AA=AF(JA)*AF(JB)*EPHS(JA)*EPHS(JB)
BB=ABS(AA)
SUMA=SUMA+AA
SUMB=SUMB+BB
20 CONTINUE
SUM=SUM+AF(I)*ABS(SUMA)/SUMB
SUMF=SUMF+AF(I)
10 CONTINUE
QV=1.0-SUM/SUMF
RETURN
END
SUBROUTINE REPHS(K,IH,IK,EPH,REPS)
COMPLEX EPH,REPS
DIMENSION IK(3),IH(3,200)
IJK=0
NN=0
NA=0
DO 10 I=1,3
IF(IK(I).NE.IH(I,K))IJK=1
IF(IK(I)*IH(I,K).LT.0)NN=NN+1
IF(IK(I).NE.0)NA=NA+1
10 CONTINUE
IF(IJK.EQ.0)REPS=EPH
IF(IJK.EQ.0)RETURN
IF(NN.EQ.NA)REPS=CONJG(EPH)
IF(NN.EQ.NA)RETURN
AKO=0.5*IK(2)
CKO=AKO-AINT(AKO)
EPS=0.001
IF(ABS(CKO).LT.EPS) GO TO 30
IF(ABS(CKO).GE.EPS) GO TO 40
30 CONTINUE
IF(IK(2).LT.0)REPS=CONJG(EPH)
IF(IK(2).LT.0)RETURN
REPS=EPH
RETURN

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40  CONTINUE
    A=REAL(EPH)
    B=AIMAG(EPH)
    IF(IH(1,K).LT.0) GO TO 50
    IF(IK(2).LT.0)REPS=CMPLX(-A,B)
    IF(IK(2).LT.0)RETURN
    IF(IK(2).GT.0)REPS=CMPLX(-A,-B)
    IF(IK(2).GT.0)RETURN
50  CONTINUE
    IF(IK(2).GT.0)REPS=CMPLX(-A,-B)
    IF(IK(2).GT.0)RETURN
    IF(IK(2).LT.0)REPS=CMPLX(-A,B)
    IF(IK(2).LT.0)RETURN
    END
    SUBROUTINE NUMBR(NREF,RH,MRH,MHR)
    DIMENSION RH(200),MRH(200),RHH(200),MHR(200),IM(200)
    DO 10 I=1,NREF
    RHH(I)=RH(I)
10  CONTINUE
    IA=NREF-1
    DO 20 I=1,IA
    JA=I+1
    DO 30 J=JA,NREF
    IF(RHH(I).GE.RHH(J)) GO TO 40
    SRS=RHH(I)
    RHH(I)=RHH(J)
    RHH(J)=SRS
40  CONTINUE
30  CONTINUE
20  CONTINUE
    DO 50 I=1,NREF
    IM(I)=0
50  CONTINUE
    DO 60 I=1,NREF
    IJK=0
    DO 70 J=1,NREF
    IF(IJK.EQ.1) GO TO 70
    IF(IM(J).EQ.1) GO TO 70
    IF(RHH(I).NE.RH(J)) GO TO 70
    MRH(J)=I
    IJK=1
    IM(J)=1
70  CONTINUE
60  CONTINUE
    DO 80 I=1,NREF
    IM(I)=0
80  CONTINUE
    DO 90 I=1,NREF
    IJK=0
    DO 100 J=1,NREF
    IF(IJK.EQ.1) GO TO 100
    IF(IM(J).EQ.1) GO TO 100
    IF(RH(I).NE.RHH(J)) GO TO 100
    MHR(J)=I
    IJK=1
    IM(J)=1
100 CONTINUE
90  CONTINUE

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RETURN
END
SUBROUTINE FACCOM(NREF, IH, AF, MRL, NRM, EPHS,
1 ANGM, AFC, PAIT, IX, EPS)
  COMPLEX EPHS
  DIMENSION IH(3,200), AF(200), MRL(200,20,2), NRM(200),
1 EPHS(200), AFC(200), ANGM(200), IAM(200), HANGM(200),
2 NPO(200), NPM(200), PM(200), IBM(200)
20 CONTINUE
  LREF=1
  CALL DIFEH(NREF, AF, AFC, PM)
  CALL NUMBR(NREF, PM, NPM, NPO)
30 CONTINUE
  DO 40 I=1, NREF
    IAM(I)=0
40 CONTINUE
  NN=0
  DO 50 I=1, LREF
    II=NREF+1-I
    IA=NPO(II)
    IAM(IA)=1
    NN=NN+1
    IED=NRM(IA)
    IF(IED.EQ.0) GO TO 50
    DO 60 J=1, IED
      JA=MRL(IA, J, 1)
      JB=MRL(IA, J, 2)
      IAM(JA)=1
      IAM(JB)=1
      NN=NN+2
60 CONTINUE
50 CONTINUE
    IF(NN.LT.130) LREF=LREF+1
    IF(NN.LT.130) GO TO 30
    LREF=1
70 CONTINUE
  NN=0
  DO 80 I=1, NREF
    IBM(I)=0
80 CONTINUE
  DO 90 I=1, LREF
    IA=NPO(I)
    IBM(IA)=1
    NN=NN+1
    IED=NRM(IA)
    DO 100 J=1, IED
      JA=MRL(IA, J, 1)
      JB=MRL(IA, J, 2)
      IBM(JA)=1
      IBM(JB)=1
      NN=NN+2
100 CONTINUE
90 CONTINUE
    IF(NN.LT.60) LREF=LREF+1
    IF(NN.LT.60) GO TO 70
    DO 110 I=1, NREF
      IF(IAM(I).EQ.0) EPHS(I)=0.0
      IF(IBM(I).EQ.1) EPHS(I)=0.0
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110 CONTINUE
    NC=0
120 CONTINUE
    NC=NC+1
    IF(NC.GT.3)GO TO 130
    DO 140 I=1,NREF
        IED=NRM(I)
        AFC(I)=0.0
        CALL DETPHS(I, IED, IH, AF, EPHS, MRL, EPHS(I), AFC(I),
1 HANGM(I), PAIT)
        IF(AFC(I).LE.0.01)NC=0
140 CONTINUE
        GO TO 120
130 CONTINUE
    CALL CORFAC(NREF, AF, AFC)
    CALL CRITLM(NREF, ANGM, HANGM, ACC, PAIT, EPS)
    DO 170 I=1, NREF
        ANGM(I)=HANGM(I)
170 CONTINUE
    WRITE(6,510)ACC
510 FORMAT(5X, 'ACC=', F10.5)
    IF(ACC.LE.0.99)GO TO 20
    RETURN
    END
    SUBROUTINE DIFEH(NREF, AF, AFC, PM)
    DIMENSION AF(200), AFC(200), PM(200)
    DO 10 I=1, NREF
        PM(I)=0.0
        PM(I)=ABS(AF(I)-AFC(I))/AF(I)
10 CONTINUE
    RETURN
    END
    SUBROUTINE ARG(PAIT, UNTV, ANG)
    COMPLEX UNTV
    AA=REAL(UNTV)
    BB=AIMAG(UNTV)
    IF(AA.EQ.0.0.AND.BB.EQ.0.0)ANG=10.0
    IF(AA.EQ.0.0.AND.BB.EQ.0.0) RETURN
    IF(AA.EQ.0.0.AND.BB.EQ.1.0)ANG=PAIT/4.0
    IF(AA.EQ.0.0.AND.BB.EQ.-1.0)ANG=PAIT*3.0/4.0
    IF(AA.EQ.0.0)RETURN
    D=BB/AA
    C=ATAN(D)
    IF(AA.GT.0.0.AND.BB.GE.0.0)ANG=C
    IF(AA.GT.0.0.AND.BB.GE.0.0)RETURN
    IF(AA.GT.0.0.AND.BB.LT.0.0)ANG=PAIT+C
    IF(AA.GT.0.0)RETURN
    ANG=PAIT/2.0+C
    RETURN
    END
    SUBROUTINE RANPHS(I, IH, EPH, IX)
    COMPLEX EPH
    DIMENSION RAN(1), IH(3,500)
    A=0.707107
    CALL RANU2(IX, RAN, 1, ICON)
    IF(RAN(1).LT.0.125)EPH=CMPLX(1.0,0.0)
    IF(RAN(1).GE.0.125.AND.RAN(1).LT.0.25)EPH=CMPLX(A,A)
    IF(RAN(1).GE.0.25.AND.RAN(1).LT.0.375)EPH=CMPLX(0.0,1.0)

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IF(RAN(1).GE.0.375.AND.RAN(1).LT.0.5)EPH=CMPLX(-A,A)
IF(RAN(1).GE.0.5.AND.RAN(1).LT.0.625)EPH=CMPLX(-1.0,0.0)
IF(RAN(1).GE.0.625.AND.RAN(1).LT.0.75)EPH=CMPLX(-A,-A)
IF(RAN(1).GE.0.75.AND.RAN(1).LT.0.875)EPH=CMPLX(0.0,-1.0)
IF(RAN(1).GE.0.875.AND.RAN(1).LT.1.0)EPH=CMPLX(A,-A)
IF(IH(2,I).EQ.0.AND.RAN(1).LT.0.5)EPH=CMPLX(1.0,0.0)
IF(IH(2,I).EQ.0.AND.RAN(1).GE.0.5)EPH=CMPLX(-1.0,0.0)
RETURN
END
SUBROUTINE CRITLM(NR,ANGM,HANGM,
1ACC,PAIT,EPS)
DIMENSION ANGM(200),HANGM(200)
PAI=PAIT/2.0
NN=0
DO 10 I=1,NR
DA=ANGM(I)-HANGM(I)
IF(DA.GE.0.0)DA=DA-PAIT*AIN(T(DA/PAIT))
IF(DA.LT.0.0)DA=DA-PAIT*(AIN(T(DA/PAIT))-1.0)
IF(DA.GE.PAI)DA=DA-PAIT
ADA=ABS(DA)
IF(ADA.GT.EPS)NN=NN+1
CONTINUE
ACC=FLOAT(NR-NN)/NR
RETURN
END
```