Dear Francis,

Here is a draft of our paper. I am not at all pleased with it, and I'm sure there is lots that you will want to change. I have tried to concentrate on making it as clean as possible what we are talking about. Now that I read through the whole thing as it is typed out, I have the impression that the actual results are lost, because the explanation takes up too much space. Anyway, I feel it's reached a stage now where some other's view would help.

Things continue very pleasant here. Freddy arrived a couple of days ago, equipped to counter people like Alex who show you photos of their children, with a photo of his new house. He and Alex + Jane and Benjy came to tea on New Year's Day and we gave them real English Christmas cake + mince pies, and Earl Grey tea.

The SDC business has turned out to be not so straightforward. The unit cell is very large, (probably 4 standard units/asymmetric unit) and getting a heavy atom in isn't easy. One of the things I miss around here is someone like Venon - I find myself doing a lot of chemistry and not much crystallography.
It looks as if we shall move about the beginning of April: we shall be 3 by then!

Best wishes,

David.
Dear David,

Many apologies for not having sent the paper before, but we seem to have nothing for a while since Christmas.

I have few comments. I will write more fully shortly, but may as well give you some idea of the main idea I suggest are sending a few remarks now.

The main thing held, in my is the relevance of the $E_d$ (I don't like the terminology for page 6), and we may try to use it. I think we should give a graph of $E_d$ from equation 13 and 17, $\alpha$ as a function of $2\pi (E-F_\infty)$ for the calculated dependence.

I think equation 19 should be clarified. Should not equation 17 be expanded, since the $P_1$ are uncorrected?

Don't also the factor 2 in equation 20 and 22? I usually consider that and the combined.

I fail to follow the derivation on page 4; I especially the derivation of $f$.
\[ r^+ = \left( F_0^+ + F_1^+ \right) (p_+ + p_-) - 2F_0 F_1 (p_+ - p_-) \]

\[ = \left( F_0^+ + F_1^+ \right) (p_+ - p_-) - 2 \frac{F_0 F_1}{p_+ + p_-} \]

\[ = \left( F_1^+ - F_0^+ \right) (p_+ + p_-) \]

\[ r^2 = \left[ \frac{(p_+ - p_-) - (p_+ + p_-)}{(p_+ + p_-)^2} \right]^2 \frac{F^2 p_+}{p_+ - p_-} + \left[ \frac{(p_+ - p_-) + (p_+ + p_-)}{(p_+ + p_-)^2} \right]^2 \frac{F^2 p_-}{p_+ - p_-} \]

\[ = \frac{4p_+^2 p_-}{(p_+ + p_-)^3} + \frac{4p_+^2 p_- F^2}{(p_+ + p_-)^3} \]

\[ = 4p_+ p_- F^2 \left( \frac{p_+ + p_-}{(p_+ + p_-)^2} \right) \]

\[ = \frac{4p_+ p_- F^2}{(p_+ + p_-)^2} \]

\[ = \frac{4R^2 F^2}{R(R+1)^2} \]

\[ p = \frac{R}{R+1} \]

\[ \frac{1}{2} F^2 + \frac{1}{2} F^2 = E^- \]
Dear Francis,

I'm sorry to hear that you've been ill again, and hope you are feeling now.

I have been working through the various points in your letter, and will go through them one by one.

1. Nomenclature. I'm not especially proud of this and look forward to improvements!

2. Equation (19). I knew you had worked very carefully through this whole section, and expected you would want to rewrite it. I just went through it for myself to see what points would arise. Then I got the impression the results looked different from what I remembered of yours, and left it.

   Now I have done the normalisation and end up with some results of a very simple form (see attached sheet). Are they right?

3. Equation (11). This is the old problem of the multiplicity factors which you raised about the P.R.S. paper. You are quite correct, and (12) and (13) should begin $2\sqrt{2}$ or $\frac{1}{2}\sqrt{2}$. (25) is also affected.

4. Page 4. There is a diagram in my thesis (included in the list of figures) which shows what $\varphi$ is. It is the phase of $f_1$, relative to the projection's centre of symmetry; i.e. the heavy atom contribution of one half of the crystal unit cell, relative in projection by a centre of symmetry to the other half. The argument about what happens out of the centrosymmetric zone should, I agree, be expanded slightly, so the whole argument was
runs as follows:

In a centrosymmetric zone

\[ |E|^2 = 2 |E|^2 \cos \phi \]

\[ \langle E^2 \rangle = \frac{4 |E|^2 \cos^2 \phi}{|V|^2} = 2 \langle E \rangle^2 \]

Outside of this zone we assume \( E_1, E_2 \) follow the same distribution. But now

or if you like, they are uncorrelated, and the same result follows.

\[ |E|^2 = \left( \langle E_1^2 \rangle + \langle E_2^2 \rangle - 2 \langle E_1 \rangle \langle E_2 \rangle \cos \phi \right)^{1/2} \]

whence

\[ \langle E^2 \rangle = 2 \langle E_1^2 \rangle \] as before.

5. **Equation (1)** Transposed, as you say.

6. **Equation (22)** Perhaps it would be best to give appendix 5 of my thesis as an appendix to the paper. In view of the revision of (10) some factors of 2 need to be added. (A factor of \( \frac{1}{2} \) seems to have slipped out of (22) already in the typing!) For the first section of (22) with some slight adaptation into account (22) should read

\[ \langle E^2 \rangle = \langle E_1^2 \rangle + \exp \left( \frac{1}{4} \right) \langle E_2^2 \rangle \]  

Although the way these things are stated in my thesis are formally incorrect, I'm pleased to say it all comes out in the wash and the results are unaltered.

You will see that (22) contains the extra term \( \delta^2 \) in addition to the expression from equation (13.10), to account for the Gaussian *broadth* of the distribution. One minor problem is that I'm not sure whether this is an accurate correction or merely an approximation because of the peculiar coordinate system. I've worked it so as to avoid stating whether it's accurate or not.

I've redrafted the relevant bits, but as you say...
are on the point of writing again with more detailed comments, I'll wait to hear from you: there are a number of minor points of English I would like to amend.

You'll realise the purpose of this rapid reply is to try to get on with it while it's fresh in our minds; I would certainly like to get all this stuff out of the way as soon as possible. If you like to let me have the rest of your comments, I'll try and get a semi-final version typed out and send back to you. I think I have negatives from which I can get prints made of all the figures, except figure 84 which was done in manuscript on p. 76 of my thesis. I'll leave drawing this until I've had your comments, as I'm not sure that this part of the argument is adequately clear yet. One other point about figure 6. You will remember some discussion about the scale on the ordinate of this figure. Because of the difficulty about normalisation, I think it is best to put no scale on the ordinate: the alternative seems to be to include the normalisation factor explicitly, i.e. label it \( \frac{P(0)}{N} \) (eq. 21).

We were all very sad to see the news about Rosalind in the New York Times. Not being given to asking questions about people's illnesses, I hadn't even known she had cancer. It's a great pity from many points of view, especially the TMV work. I've just had a letter from Aaron saying they will have to try and carry on without her and will therefore be needing some more people, e.g. me.

The move to M.I.T. is at least definite, and fixed for the first week in June. I still foresee trouble getting Alex to move, but we've now handed in notice on our apartment here for the
end of May, so we shall be going in any case!

Elizabeth is a healthy, noisy but fairly placid American citizen, daily taking more and more interest in her surroundings. Her powers of ingestion and excretion are phenomenal. Mary is fine, too. She had to wait 3 weeks longer than the textbook date, and during this period began to wonder whether she wouldn't rather have a puppy dog after all, but all those doubts are dispelled now.

Please give my best wishes to everyone.

Yours

David.

* P.S. I find that $r$ has never been properly defined in the paper, and this adds to the confusion about equation (22). I want to define $r$ as the integral in (13), i.e. the radius of gyration of the diagram for a single reflection in $F$ space. In the appendix, instead of defining the quantity $M$ called $M$, we use $r^2$ which equals $2M$.

(in the appendix to my thesis)
The probabilities of the two cases are then as follows:

**Probability $F$ is positive:**

$$P_+ = N \left[ \exp \left\{ -\frac{(F_c - [F_h - F])^2}{2\langle E \rangle^2} \right\} + \exp \left\{ -\frac{(F_c - [F_h + F])^2}{2\langle E \rangle^2} \right\} \right] \quad (15a)$$

**Probability $F$ is negative:**

$$P_- = N \left[ \exp \left\{ -\frac{(F_c + [F_h - F])^2}{2\langle E \rangle^2} \right\} + \exp \left\{ -\frac{(F_c + [F_h + F])^2}{2\langle E \rangle^2} \right\} \right] \quad (15b)$$

where $N$ is a normalising factor such that $P_+ + P_- = 1$.

Rearranging these gives

$$P_+ = 2N \exp \left\{ -\frac{(F_c)^2 + H^2}{2\langle E \rangle^2} \right\} \sinh \left\{ \frac{H (F_c + F)}{\langle E \rangle^2} \right\},$$

$$P_- = 2N \exp \left\{ -\frac{(F_c)^2 + H^2}{2\langle E \rangle^2} \right\} \sinh \left\{ \frac{H (F - F_c)}{\langle E \rangle^2} \right\},$$

$$\frac{P_+}{P_-} = \exp \left\{ -\frac{2F_c}{\langle E \rangle^2} \right\} \frac{\sinh \left\{ \frac{H (F_c + F)}{\langle E \rangle^2} \right\}}{\sinh \left\{ \frac{H (F - F_c)}{\langle E \rangle^2} \right\}}. \quad (16)$$

The expressions may be simplified if $F_c$ and $E$ are small compared to $(F + F_h)$. In this case the possibility of $F$ and $F_h$ having different signs may be neglected and the second terms in (15) disappear. Under these conditions

$$N^{-1} = 2 \exp \left\{ -\frac{(F - F_h)^2 + F_c^2}{2\langle E \rangle^2} \right\} \cosh \left\{ \frac{F_c (F - F_h)}{\langle E \rangle^2} \right\},$$

and if we write

$$t = \frac{F_c (F - F_h)}{\langle E \rangle^2},$$

$P_+$, $P_-$ may be written

$$P_+ = \frac{e^t}{2} \cosh t \quad (17a)$$

$$P_- = \frac{e^{-t}}{2} \cosh t \quad (17b)$$
Thus when $F + F_H$ is large, the value $F_0$ which must be used to give the best Fourier is

$$F_0 = (P_+ - P_-)F = F \tanh t$$

(18)

and the mean square error $<F - F_0>^2$ is taken over the whole probability distribution

$$\gamma^2 = (F - F_0)^2 P_+ + (F + F_0)^2 P_- + <\delta^>'^2$$

$$= F^2 \text{sech}^2 t + <\delta^>'^2.$$

(19)
Some comments.

"No way of refinement from a trial structure"

When I left Hopke was working on a very promising-looking least-squares method of fitting phases to observations. Perhaps the above statement should be modified. A

after approximate
phases have been
found.

p. 7

Application to centrosymmetric case

This is the section I am most certain you will want to rewrite. However, just to get an idea of exactly what I remember in it, I have written it out in my own way. (I'm convinced the results look entirely different from yours!)

p. 9

This page has given me the greatest trouble. I have had repeated second thoughts about the treatment in my thesis, and in particular this point about what effect it might have on the phase error if the observational error happens to be entirely in F and not F*. I have also spent a long time trying to treat the problem analytically - it is full of snags because you have to be very careful what you neglect - without reaching any useful results.
However, I think all is in order.

Notation:

Not quite consistent, I'm afraid; I had hoped not to have to explain the convention about using F for $|E|$, but when I came to fill in the algebra I discovered things became quite unwieldy if $|I|$ signs were used everywhere, but not till about halfway through.

p. 11:

The integrals are done in one of the appendices to my thesis, and seemed awfully boring to put into the paper. We could put them into an appendix if you like.
Figures.
Rita has photos of all the figures except figure 4, which has never been drawn out properly. Some of these photos were taken by me and may not be of best quality. The original drawings of figures 2, 3, and 7 are here; the remainder (1, 5, 6) should be in Cambridge. The photos of 2, 3, and 7 are certainly good, as slides have been made of them.

There are also photos of the $\Delta^2$ slide rule (fig 6.8 in my thesis) which could also be included, though they are not terribly good.

Figure 6: we have discussed via Max the problem of normalising this thing, and in particular whether there should be a $\frac{1}{10}$ in front of the ordinate scale or not. I think it would be best not to mark any numbers on the ordinate. $\exists$

Figure 5 To conform with the text the top left vertex needs to be marked 0.