Solution of the Phase Problem in the Theory of Structure Determination of Crystals from X-Ray Diffraction Experiments

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We present a solution to a long-standing basic problem encountered in the theory of structure determination of crystalline media from x-ray diffraction experiments; namely, the problem of determining phases of the diffracted beams.

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In a well-known paper published almost 100 years ago, Laue laid the foundation for a method for determining the structure of crystalline media from x-ray diffraction experiment [1]. Since then the method has become of basic importance in solid state physics (see, for example, [2,3]) and in other fields, sometimes using neutrons or electrons rather than x rays. However, as is well known, the method suffers from a serious limitation due to the inability to measure phases of the diffracted beams.

In this Letter we show how the phases may be determined. Before doing so we point out that "the phase problem," as usually formulated, has no solution and is, in fact, rather meaningless. We will reformulate it and show that the reformulated problem has a solution which allows unambiguous determination of the crystal structure to be made [4].

We begin with the following observation: In usual treatments, the incident x-ray beam is assumed to be monochromatic. That is an idealization, because monochromatic beams are not realizable. Any beam which can be produced in a laboratory is, at best, quasimonochromatic; i.e., its spectral width $\Delta\omega$ is much smaller than its mean frequency $\bar{\omega}$. Both the amplitudes and the phases of the field oscillations are random variables and, hence, even if they out a three-dimensional domain D, i.e., if $|\ \ (\mathbf{r}_1, \mathbf{r}_2, \omega_0)| = 1$ for all $\mathbf{r}_1 \in D$, $\mathbf{r}_2 \in D$, then the cross-spectral density function of the field at that frequency has necessarily the factorized form

$$W(\mathbf{r}_1, \mathbf{r}_2, \omega_0) = u^*(\mathbf{r}_1, \omega_0) u(\mathbf{r}_2, \omega_0). \tag{6}$$

Moreover, throughout the domain D, $u(\mathbf{r}, \omega_0)$ satisfies the Helmholtz equation

$$(\nabla^2 + k_0^2) u(\mathbf{r}, \omega_0) = 0, \tag{7}$$

where $k_0 = \omega_0/c$, c being the speed of light in vacuum.

If we set

$$u(\mathbf{r}, \omega_0) = |u(\mathbf{r}, \omega_0)| e^{i\ \ (\mathbf{r}, \omega_0)}, \tag{8}$$

we readily find from Eqs. (5), (6), and (8) that

$$\ \ (\mathbf{r}_1, \mathbf{r}_2, \omega_0) = \exp\{i[\ \ (\mathbf{r}_2, \omega_0) - \ \ (\mathbf{r}_1, \omega_0)]\}. \tag{9}$$

In view of Eq. (7) the function $u(\mathbf{r}, \omega_0)$ may be identified with the space-dependent part of a monochromatic wave function $v(\mathbf{r}, t) = u(\mathbf{r}, \omega_0) \exp(-i\omega_0 t)$. Thus we have shown that one may associate with any wide-sense statis-

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