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Coherent n^2 Scattering in Periodic Lattices

J. D. Franson
Applied Physics Laboratory, Johns Hopkins University, Laurel, Maryland 20723-6099
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The coherent scattering rate from n particles bound in a periodic lattice of N potential wells is considered. The quantum theory predicts a total coherent scattering rate proportional to n^2/N, whereas the conventional semiclassical approach predicts a rate proportional to n. These effects allow an experimental test of certain nonlocal assumptions inherent in quantum statistical mechanics and may be useful in analyzing the properties of crystalline materials.

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The conventional theory[1,2] of elastic scattering of incident particles, such as neutrons, by a perfect crystal is generally based on the semiclassical assumption that the atoms are bound to fixed sites in the crystal with occupation probabilities given by various correlation functions or order parameters. This paper is concerned, instead, with a more general situation in which n scattering particles are bound in a periodic lattice of N potential wells with $n \leq N$. The scattering particles will also be treated quantum mechanically, which introduces an intrinsic uncertainty as to the potential well in which a particle is actually bound. It will be found that the quantum-theory prediction for the total coherent scattering rate is quite different from the semiclassical result and is proportional to n^2/N.

The quantum-mechanical effects of interest here are neglected in the usual[1,2] semiclassical approach, which illustrates the observable difference between probabilities and probability amplitudes. An experimental investigation of these effects would test certain nonlocal assumptions inherent in quantum statistical mechanics, and they may be of practical use in analyzing the properties of crystalline samples. The extent to which this simple model can represent coherent scattering in actual crystals with various defects will be briefly discussed.

The term *coherent* is defined in may different ways in various applications, but here a coherently scattered wave will be defined as one that can be made to interfere with the incident wave. (Some comments on the experimental observability of coherent versus incoherent scattering will be made shortly.) It should be emphasized that the coherent scattering is generally a small fraction of the total scattering, and that the latter is simply proportional to n, as pointed out by a number of authors.[3]

It will be assumed for the moment that the n scattering particles are noninteracting, distinguishable, and labeled with an index μ, although these assumptions will be relaxed shortly. It will also be assumed that the N potential wells are identical, relatively deep and narrow, and centered on a periodic lattice of points $\mathbf{x}_i$, $i=1,N$. In that case, the lowest-energy eigenstate of a single particle μ bound in a particular well i can be written to a first approximation as

$$\psi(\mathbf{r}_\mu) \cong \psi_W(\mathbf{r}_\mu - \mathbf{x}_i)\,. \qquad (1)$$

Here $\psi_W(\mathbf{r})$ is defined as the lowest-energy solution to the Schrödinger equation for a single well at the origin $(\mathbf{x}=0)$ assuming that none of the other wells exist. An explicit solution for ψ_W could be calculated if we were to assume a specific form for the potential wells, but that will prove to be unnecessary.

Equation (1) is a reasonable approximation to the eigenstates of a particle to the extent that the overlap of the wave functions from two neighboring wells can be neglected. Even if the overlap of the wave functions were identically zero, however, the eigenstates of Eq. (1) would be degenerate, so that we can just as well consider eigenstates ψ_S that are superpositions of these

$$\psi_S(\mathbf{r}_\mu) = \sum_i \alpha_i \psi_W(\mathbf{r}_\mu - \mathbf{x}_i)\,, \qquad (2)$$

where the coefficients α_i are arbitrary in the degenerate case. It will be found that the scattering properties of Eq. (2) are very different from those of Eq. (1), and that any analysis of coherent scattering must take the actual wave function of the scattering particles into account.

Fortunately, the overlap of the wave functions from two neighboring wells splits the degeneracy and determines the form of the actual eigenstates. From Bloch's theorem,[4] the most general solution to the Schrödinger equation for a periodic potential is

$$\psi(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_k(\mathbf{r}) , \tag{3}$$

where $u_k(\mathbf{r})$ is a function periodic in the crystal lattice. Equation (3) constrains the allowed values of α_i, so that Eq. (2) must have the approximate form

$$\psi_k(\mathbf{r}_\mu) = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{i\mathbf{k}\cdot\mathbf{x}_i} \psi_W(\mathbf{r}_\mu - \mathbf{x}_i) . \tag{4}$$

Here the factor of $1/\sqrt{N}$ is required for normalization and we have used the fact that $\mathbf{r}_\mu \cong \mathbf{x}_i$ in the slowly varying exponential term, since the wells have been assumed to be very narrow. Although Eq. (4) is approximate, the results which follow could be derived directly from the form of Eq. (3). Equation (4) will be used instead to maintain an obvious connection with the usual view of particles bound in a specific well.

At this point we have considered only the eigenstates for a single particle in a periodic lattice. For noninteracting, distinguishable particles, the wave function ψ_n describing all n scattering particles can be defined by assigning a value of $\mathbf{k}$ to each:

$$\psi_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n) = \psi_{k_1}(\mathbf{r}_1)\psi_{k_2}(\mathbf{r}_2) \cdots \psi_{k_n}(\mathbf{r}_n) . \tag{5}$$

A thermal distribution at some relatively low temperature T will be assumed, in which case the excitation of higher-energy states in the wells can be ignored. It will be found that all states of this form have the same scattering properties, so that we need not be concerned with any averaging over the thermal distribution. (More formally, this corresponds to the use of a diagonal density matrix, as will be discussed below.) We see that the actual form of the wave function and thus the scattering properties are determined by quantum mechanics and thermodynamics, and that it cannot simply be assumed that each particle is bound in a specific well.

Having determined the appropriate wave function, we would now like to calculate the corresponding scattering rate. One approach would be to assume some specific form for the scattering interaction and lattice properties, for which the scattering rate could be computed; such a result would not be general in nature and would require the evaluation of a number of integrals. That will be avoided here by first computing the total elastic-scattering rate R_N for the case of N fixed scattering centers in a periodic lattice, for which the result is known.[3,5] The total coherent scattering rate from the wave function of Eq. (5) will then be shown to differ from R_N by a factor of $(n/N)^2$.

We therefore consider a wave function ψ_F in which each of N particles is bound to a fixed site in the lattice, which can just as well be chosen to be

$$\psi_F(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) = \psi_W(\mathbf{r}_1 - \mathbf{x}_1) \times \psi_W(\mathbf{r}_2 - \mathbf{x}_2) \cdots \psi_W(\mathbf{r}_N - \mathbf{x}_N) . \tag{6}$$

An incident particle, such as a neutron, will be assumed to have momentum $\hbar\mathbf{p}$ and the total elastic-scattering rate into momentum $\hbar\mathbf{p}'$ will be computed. The spin of the incident particle will be neglected for simplicity.

The exact form of the interaction between the incident and the scattering particles is of no importance and will simply be taken to be

$$H' = \sum_\mu U(\mathbf{r}_\nu - \mathbf{r}_\mu) , \tag{7}$$

where $U(\mathbf{r})$ is some short-range potential and $\mathbf{r}_\nu$ is the position of the incident particle. Conventional perturbation theory then gives

$$R_N = \frac{2\pi}{\hbar} \sum_{p'} |\langle p'\psi_F | H' | p\psi_F \rangle|^2 \delta(E_{p'} - E_p) , \tag{8}$$

where E_p is the energy of the corresponding scattered particle state. ψ_F is the same in the initial and final states since the scattering is elastic and the short-range nature of U as well as the negligibly small overlap of the wave functions ensures that the matrix elements are zero for any of the scattering particles to be found in a different potential well. Inserting Eq. (6) for ψ_F and plane-wave states for the incident particle gives

$$R_N = \frac{2\pi}{\hbar} \sum_{p'} \left| \sum_{\mu=1}^{N} \int d^3 r_\nu \int d^3 r_\mu \, e^{i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}_\nu} \psi_W^*(\mathbf{r}_\mu - \mathbf{x}_\mu) U(\mathbf{r}_\nu - \mathbf{r}_\mu) \psi_W(\mathbf{r}_\mu - \mathbf{x}_\mu) \right|^2 \delta(E_{p'} - E_p) . \tag{9}$$

By making a suitable change of variables ($\mathbf{r} = \mathbf{r}_\mu - \mathbf{x}_\mu$, $\mathbf{r}'_\nu = \mathbf{r}_\nu - \mathbf{x}_\mu$), all the integrals can be cast in the same form, aside from the exponential factor, so that Eq. (9) can be reduced to

$$R_N = \frac{2\pi}{\hbar} \sum_{p'} \left| \sum_{i=1}^{N} e^{i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{x}_i} M_{pp'} \right|^2 \delta(E_{p'} - E_p) , \tag{10}$$

where

$$M_{pp'} = \int d^3 r'_\nu \int d^3 r \, e^{i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}'_\nu} \psi_W^*(\mathbf{r}) U(\mathbf{r}'_\nu - \mathbf{r}) \psi_W(\mathbf{r}) . \tag{11}$$

The sum over μ in Eq. (9) has been relabeled with the index i in Eq. (10) for reasons that will become apparent shortly.

The coherent scattering rate R corresponding to the wave function of Eq. (5) can now be computed in a similar manner by inserting ψ_n instead of ψ_F into Eq. (8):

$$R=\frac{2\pi}{\hbar}\sum_{k'p'}\left|\sum_{\mu=1}^{n}\sum_{i=1}^{N}\int d^3r_\nu\int d^3r_\mu e^{i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}_\nu}\frac{1}{N}e^{-i\mathbf{k}'_\mu\cdot\mathbf{x}_i}e^{i\mathbf{k}_\mu\cdot\mathbf{x}_i}\psi_W^*(\mathbf{r}_\mu-\mathbf{x}_i)U(\mathbf{r}_\nu-\mathbf{r}_\mu)\psi_W(\mathbf{r}_\mu-\mathbf{x}_i)\right|^2\delta(E_p-E_{p'})\,. \quad (12)$$

A sum over $\mathbf{k}'_\mu$ has been included to indicate that, in general, the values of k_μ need not be the same in the initial and final states. Nearly all of the terms with $\mathbf{k}'_\mu\neq\mathbf{k}_\mu$ correspond to an exchange of energy between the incident and scattering particles and must therefore be omitted in a calculation of the elastic-scattering rate. More generally, all terms with $\mathbf{k}'_\mu\neq\mathbf{k}_\mu$ must be omitted when computing the coherent scattering rate, since they correspond to orthogonal quantum states and cannot interfere with the incident wave.

Retaining only the $\mathbf{k}'_\mu=\mathbf{k}_\mu$ terms, Eq. (12) differs from Eq. (9) only by the addition of the sum over μ and the factor of $1/N$. In particular, it should be noted that the factors of $\exp(\pm i\mathbf{k}_\mu\cdot\mathbf{x}_i)$ cancel out, so that no thermal averaging over the $\mathbf{k}$'s is necessary, as mentioned earlier. Making a similar change of variables, Eq. (12) can be written as

$$R=\frac{2\pi}{\hbar}\sum_{p'}\left|\frac{1}{N}\sum_{\mu=1}^{n}\sum_{i=1}^{N}e^{i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{x}_i}M_{pp'}\right|^2\delta(E_{p'}-E_p)\,. \quad (13)$$

The sum over μ can be performed immediately and is simply equal to n, so that

$$R=\frac{2\pi}{\hbar}\left(\frac{n}{N}\right)^2\sum_{p'}\left|\sum_{i=1}^{N}e^{i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{x}_i}M_{pp'}\right|^2\delta(E_{p'}-E_p) \quad (14)$$

or

$$R=(n/N)^2R_N \quad (15)$$

as asserted earlier. The total elastic-scattering rate from an array of N fixed scattering centers is proportional to N for small wavelengths, as one might expect,[3,5] so that $R_N=\beta N$, where β is a constant. Thus, we have finally that

$$R=\beta n^2/N \quad (16)$$

for the total coherent scattering rate.

A brief comment regarding the experimental observability of coherent versus incoherent scattering may be relevant. The conventional theory[1,2,5] gives a large diffuse scattering rate for $n\ll N$ which has a pseudorandom phase compared to the incident wave due to the random (but fixed) locations of the n atoms. This diffuse scattering is coherent (in the semiclassical theory) in that it can produce an interference pattern if the incident and scattered waves are superimposed, as would be the case using a thin crystal and a photographic plate close behind it, for example. The pseudorandomness of the phase would cause the interference pattern to vary with position along the photographic plate similar to a speckle interference pattern from a laser. In the quantum-theory treatment, however, this diffuse scattering is intrinsically incoherent and no such speckle interference pattern would occur.

It can be shown that the results obtained above hold approximately in the limit of $n\ll N$ even for indistinguishable particles or particles with a short-range interaction. This seems reasonably apparent and the details will not be given here.

Physically, the incident particles are scattering off the spatial variations in the density $\psi^*\psi$ induced by the periodic potential, which is the same for all the scattering particles and gives a coherent factor of n^2. Although it will not be shown here, similar effects can be expected from the spatial modulation of $\psi^*\psi$ for propagating states as well as bound states.

One of the interesting features of these results is the fact that, for a single-scattering particle ($n=1$), the total coherent scattering rate is a factor of $1/N$ less than it would be if the scattering particle were assumed to be found in a specific but unknown potential well. More generally, it can be shown[5] that a semiclassical treatment in which n particles are bound in specific but randomly chosen locations gives a coherent scattering rate proportional to n, as opposed to the n^2/N rate obtained quantum mechanically. These results illustrate the nonclassical nature of the quantum-mechanical uncertainty in the position of a particle and may not be too surprising in view of the recently derived form of Bell's inequality[6] in the position domain.[7]

Equation (3) (Bloch's theorem) is valid regardless of the magnitude of the overlap of the wave functions, but these results are also dependent on the thermodynamic postulate[8] that the energy eigenstates, which are not localized but are distributed throughout the crystal, are populated with random phases, which corresponds to a diagonal density matrix. Very different results would be obtained if an incoherent population of some localized set of states, such as the fixed-site ψ_F's of Eq. (6), were assumed instead. The essence of quantum statistical mechanics is that it considers a set of unperturbed energy eigenstates, which are nonlocalized in this example, and then assumes that the introduction of some small, random perturbations will ensure a diagonal density matrix in that set of basis states. Classical physics (and perhaps our intuition) suggests, instead, that the particles are really bound in specific but unknown locations, which corresponds to a density-matrix diagonal in a set of localized basis states. The question then arises as to whether or not the true effect of local perturbations may be to give a density-matrix diagonal in a set of localized states, a possibility that could be investigated experimentally by means of these effects.

In principle, these results can be applied directly to neutron scattering in a crystal containing n atoms of an isotope that scatters strongly due to resonance effects and $N-n$ atoms of a different isotope with negligible scattering. As a practical matter, scattering of this kind may be limited by impurities, dislocations, thermal phonons, mechanical deformations, and other crystal defects, all of which are beyond the intended scope of this paper. In particular, as the density of defects increases one might expect that the nonlocalized eigenstates would eventually be replaced by localized states due to Anderson localization.[9] Below the threshold for such localization, however, the energy eigenstates remain nonlocalized and the results obtained above should hold.

In summary, n^2 coherent scattering is predicted by the quantum theory for n particles bound in a periodic potential, whereas the semiclassical approach predicts a total coherent rate proportional to n. An experimental investigation of these inherently quantum-mechanical effects would test certain nonlocal assumptions inherent in quantum statistical mechanics. The observability of such effects in actual crystals may be limited by imperfections of various kinds, which are beyond the intended scope of this paper.

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[1]For example, see J. M. Cowley, *Diffraction Physics* (North-Holland, New York, 1981), 2nd ed.

[2]See, for example, *Atomic Transport and Defects in Metals by Neutron Scattering,* edited by C. Janot, W. Petry, D. Richter, and T. Springer (Springer-Verlag, New York, 1986); M. A. Krivoglaz, *Theory of X-Ray and Thermal Neutron Scattering* (Plenum, New York, 1969); *Small Angle X-Ray Scattering,* edited by H. Brumberger (Gordon and Breach, New York, 1967).

[3]Y. Aharonov, F. T. Avignone, A. Casher, and S. Nussinov, Phys. Rev. Lett. **58**, 1173 (1987); H. J. Lipkin, Phys. Rev. Lett. **58**, 1176 (1987).

[4]C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1963).

[5]Although a more general derivation can be given, this can be obtained directly from Eq. 7-15 of Ref. 1, for example, by interchanging the role of vacancies and occupied sites and then summing the two terms to obtain the total scattering rate, which is coherent as defined here.

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[7]J. D. Franson, Phys. Rev. Lett. **62**, 2205 (1989).

[8]K. Huang, *Statistical Mechanics* (Wiley, New York, 1963), p. 185.

[9]P. W. Anderson, Phys. Rev. **109**, 1492 (1958).