Thermodynamic Green's Function Methods in Neutron Scattering by Crystals*

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(Received August 5, 1960; revised manuscript received October 27, 1960)

Formulas are derived for the transition probabilities per unit time for both inelastic coherent scattering of neutrons by crystals, and resonant emission of photons and neutrons by nuclei bound in crystals, without making the assumption that the crystal is harmonic. In deriving these transition probabilities, the analytic structure of thermodynamic correlation or Green's functions, considered as functions of complex temperatures and times, is developed and used. In particular a spectral form is found for the phonon Green's function. Only one assumption is made about the crystal, namely that the displacement of the nuclei due to the forces exerted by the neutron in scattering are linear functions of these forces. This leads to an evaluation of the transition probabilities in terms of the exact thermodynamic displacement autocorrelation function. This evaluation obeys the detailed balancing condition, and Placzek's sum rule. A consequence of this evaluation is that the widths of the "one-phonon" peaks in the neutron scattering are exactly equal to the widths of the corresponding phonon states of the crystal.

I. INTRODUCTION

SEVERAL authors[1] have derived expressions for the transition probabilities for inelastic coherent scattering of neutrons by crystals, and for resonant absorption and emission of neutrons and photons by nuclei in crystals. These have been derived under the assumption that the crystal is harmonic, i.e., that the displacements of the nuclei from equilibrium may be described by a set of independent harmonic oscillators. In an actual crystal, however, the harmonic oscillators are not independent, and the phonon states therefore have finite lifetimes. The effects of these lifetimes on the cross sections are not seen in the harmonic approximation. Thus it would be desirable to avoid this assumption and to derive an expression that shows how the transition probabilities depend on the more exact structure of the phonon field in the crystal. It is the purpose of this paper to show how such an expression may be obtained.

By making the approximation that the crystal responds linearly to the forces exerted by the neutron in scattering, we shall derive an expression for the transition probabilities in terms of the exact thermodynamic displacement autocorrelation function, or phonon Green's function. To do this we shall use the analytic structure of thermodynamic correlation functions[2] considered as functions of complex temperatures and times, together with variational derivative techniques of quantum field theory.

We consider for simplicity a monatomic crystal described by an infinite Bravais lattice of fixed vectors, $\mathbf{g}$, $\mathbf{h}$, $\cdots$. Each of these lattice sites is taken to be the rest position of a nucleus with mass M. The small displacements of the nuclei from equilibrium will be described in the Heisenberg representation by the vector field $\boldsymbol{\phi}_{\mathbf{g}}(t)$; then the nucleus whose rest position is $\mathbf{g}$ will be at the point $\mathbf{g}+\boldsymbol{\phi}_{\mathbf{g}}$ instantaneously. The Hamiltonian of the crystal we call H, and we set $\hbar=1$.

First we outline the general aspects of the problem of inelastic coherent scattering of neutrons. The total transition probability $W(\omega)$ for inelastic coherent scattering of a neutron from initial energy E_n, initial momentum $\mathbf{k}$, to final energy $E_n-\omega$ and final momentum $\mathbf{k}'$, by a crystal with initial energy E_i, is given in the Born approximation by

$$W(\omega)=2\pi\langle E_i|\,V^\dagger\delta(E_i+\omega-H)V\,|E_i\rangle, \tag{1}$$

$|E_i\rangle$ is the initial energy eigenstate of the crystal. V is the matrix element between the initial and final neutron states of a pseudopotential with scattering length a appropriate to coherent scattering of neutrons by nuclei of the crystal:

$$V=\int e^{-i\mathbf{k}'\cdot\mathbf{r}}\sum_{\mathbf{g}}\frac{2\pi a}{m}\delta(\mathbf{r}-\mathbf{g}-\boldsymbol{\phi}_{\mathbf{g}})e^{i\mathbf{k}\cdot\mathbf{r}}d^3r$$
$$=\frac{2\pi a}{m}\sum_{\mathbf{g}}\exp[i\mathbf{K}\cdot(\mathbf{g}+\boldsymbol{\phi}_{\mathbf{g}})], \tag{2}$$

m is the neutron mass, and $\mathbf{K}=\mathbf{k}-\mathbf{k}'$ is the momentum lost by the neutron. The matrix element V still contains the displacements, $\boldsymbol{\phi}$, as operators. We then have for the transition probability per nucleus

$$W(\mathbf{K},\omega)=\left(\frac{2\pi a}{m}\right)^2\sum_{\mathbf{g}}\langle E_i|\exp[-i\mathbf{K}\cdot(\mathbf{g}+\boldsymbol{\phi}_{\mathbf{g}})]$$
$$\times 2\pi\delta(E_i+\omega-H)\exp[i\mathbf{K}\cdot(\mathbf{h}+\boldsymbol{\phi}_{\mathbf{h}})]\,|E_i\rangle. \tag{3}$$

The displacements $\boldsymbol{\phi}_{\mathbf{g}}$ and $\boldsymbol{\phi}_{\mathbf{h}}$ are at the same time in this expression. However, using $e^{iH(t-t')}\boldsymbol{\phi}(t')e^{-iH(t-t')}$

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* Work done as a National Science Foundation Predoctoral Fellow.

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[1] R. J. Glauber, Phys. Rev. **98**, 1692 (1955); A. Akhiezer and I. Pomeranchuk, J. Exptl. Theoret. Phys. U.S.S.R. **17**, 769 (1947); V. F. Turchin, J. Exptl. Theoret. Phys. U.S.S.R. **33**, 124 (1957) [translation: Soviet Phys.—JETP **6**, 96 (1958)]; L. Van Hove, Phys. Rev. **95**, 249 (1954); G. Placzek and L. Van Hove, Phys. Rev. **93**, 1207 (1954) give a list of other references. Also W. Lamb, Phys. Rev. **55**, 190 (1939).

[2] A general discussion of thermodynamic correlation functions is given by P. C. Martin and J. Schwinger, Phys. Rev. **115**, 1342 (1959).

$=\phi(t)$, (3) may be written as

$$W(\mathbf{K},\omega)$$
$$=\left(\frac{2\pi a}{m}\right)^2\sum_{\mathbf{g}}\exp[i\mathbf{K}\cdot(\mathbf{h}-\mathbf{g})]\int_{-\infty}^{\infty}dt\,e^{i\omega(t-t')}$$
$$\times\langle E_i|\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{g}}(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{h}}(t')]|E_i\rangle. \quad (4)$$

Instead of an initial state of fixed energy for the crystal, we must take a thermal average of states of definite energy. The thermal average, for temperature T, of the expectation value of an operator X is given by

$$\langle X\rangle^{\beta}\equiv\mathrm{Tr}(e^{-\beta H}X)/\mathrm{Tr}(e^{-\beta H}), \quad (5)$$

where $\beta=1/KT$, and the trace is over a complete set of states of the crystal for which the number of nuclei is fixed. We shall assume that the presence of the decreasing exponential function of energy, $e^{-\beta H}$, is sufficient to guarantee that for any operator X, whose diagonal matrix element $\langle E_i|X|E_i\rangle$, between states of fixed energy, is not an exponentially increasing function of energy, $\mathrm{Tr}(e^{-\beta H}X)$ converges absolutely, i.e., $\sum_i e^{-\beta E_i}\times|\langle E_i|X|E_i\rangle|<\infty$.

After we take a thermal average W becomes

$$W^{\beta}(\mathbf{K},\omega)$$
$$=\left(\frac{2\pi a}{m}\right)^2\sum_{\mathbf{g}}\exp[-i\mathbf{K}\cdot(\mathbf{g}-\mathbf{h})]\int_{-\infty}^{\infty}dt\,e^{i\omega(t-t')}$$
$$\times\langle\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{g}}(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{h}}(t')]\rangle^{\beta}. \quad (6)$$

The transition probability is thus given in terms of the correlation function $\langle\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{g}}(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{h}}(t')]\rangle^{\beta}$ which depends on only the crystal dynamics and the momentum $\mathbf{K}$, transferred to the crystal by the neutron.

II. THE PHONON GREEN'S FUNCTION

Our principal object is to calculate the correlation function $\langle\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{g}}(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{h}}(t')]\rangle^{\beta}$ in terms of the phonon Green's function D. This is defined by

$$D_{ij}^{\beta}(\mathbf{g},\mathbf{h},t-t')\equiv-i\langle(\phi_{\mathbf{g},i}(t)\phi_{\mathbf{h},j}(t'))_+\rangle^{\beta}, \quad (7)$$

where $(\ )_+$ denotes the time-ordered product. For a crystal that is not acted on by time-dependent external forces, D is a function of $t-t'$. We may derive a spectral form for D by considering the function

$$D_{>,ij}^{\beta}(\mathbf{g},\mathbf{h},t)\equiv-i\langle\phi_{\mathbf{g},i}(t)\phi_{\mathbf{h},j}(0)\rangle^{\beta}, \quad (8)$$

in terms of which

$$D_{ij}^{\beta}(\mathbf{g},\mathbf{h},t)=\begin{cases}D_{>,ij}^{\beta}(\mathbf{g},\mathbf{h},t), & t>0\\ D_{>,ji}^{\beta}(\mathbf{h},\mathbf{g},-t), & t<0.\end{cases} \quad (9)$$

Let us introduce the variable $\tau=-i\beta$. From the assumption of absolute convergence of the traces for real positive β, it follows that $D_{>}{}^{i\tau}$, which may be written as

$$D_{>,ij}{}^{i\tau}(\mathbf{g},\mathbf{h},t)=\frac{1}{i}\frac{\mathrm{Tr}[e^{-i(\tau-t)H}\phi_{\mathbf{g},i}(0)e^{-itH}\phi_{\mathbf{h},j}(0)]}{\mathrm{Tr}(e^{-i\tau H})} \quad (10)$$

converges, and hence is an analytic function of τ for τ in the lower half plane. In addition, the traces still converge if we give t an imaginary part such that $0\geqslant\mathrm{Im}t>\mathrm{Im}\tau$. Therefore $D_{>}{}^{i\tau}(t)$ is analytic in the strip in the t plane defined by $0>\mathrm{Im}t>\mathrm{Im}\tau$.

This analyticity in τ and t holds for any ensemble average of the form:

$$F^{i\tau}(t-t')=\langle X(t)Y(t')\rangle^{i\tau}.$$

Later we shall evaluate

$$\langle\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{g}}(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_{\mathbf{h}}(t')]\rangle^{i\tau}$$

for complex τ and a limited time region, and then, by analytic continuation, infer the desired evaluation for both purely imaginary τ (i.e., real β) and the entire real time axis.

From the cyclic property of the trace,

$$\mathrm{Tr}(ABC)=\mathrm{Tr}(BCA),\ \text{etc.},$$

we deduce the boundary condition on $D_{>}$:

$$D_{>,ij}{}^{i\tau}(\mathbf{g},\mathbf{h},t)=D_{>,ji}{}^{i\tau}(\mathbf{h},\mathbf{g},\tau-t),\quad 0\geqslant\mathrm{Im}t>\mathrm{Im}\tau. \quad (11)$$

Equation (11) becomes a simple condition on the Fourier transform of $D_{>}$ in the strip $0\geqslant\mathrm{Im}t>\mathrm{Im}\tau$. Since $D_{>}$ is bounded in the strip, the Fourier transform exists in the generalized sense along every line in this strip parallel to the real axis and along the real axis. Hence

$$D_{>,ij}{}^{i\tau}(\mathbf{g},\mathbf{h},t)=\int_{-\infty}^{\infty}e^{-i\omega t}d_{>,ij}{}^{i\tau}(\mathbf{g},\mathbf{h},\omega)\frac{d\omega}{2\pi},$$
$$0\geqslant\mathrm{Im}t>\mathrm{Im}\tau, \quad (12)$$

and from (11) we have

$$d_{>,ij}{}^{i\tau}(\mathbf{g},\mathbf{h},\omega)=e^{i\omega\tau}d_{>,ji}{}^{i\tau}(\mathbf{h},\mathbf{g},-\omega). \quad (13)$$

From (9), the Fourier transform $d(\omega)$ of D for real t is given by

$$d_{ij}{}^{i\tau}(\mathbf{g},\mathbf{h},\omega)=\int_0^{\infty}e^{i\omega t}dt\int\frac{d\omega'}{2\pi}d_{>,ij}{}^{i\tau}(\mathbf{g},\mathbf{h},\omega')e^{-i\omega' t}$$
$$+\int_{-\infty}^{0}e^{i\omega t}dt\int\frac{d\omega'}{2\pi}d_{>,ji}{}^{i\tau}(\mathbf{h},\mathbf{g},-\omega')e^{-i\omega' t}$$
$$=i\int\frac{d\omega'}{2\pi}\frac{d_{>,ij}{}^{i\tau}(\mathbf{g},\mathbf{h},\omega')}{\omega-\omega'+i\epsilon}$$
$$-i\int\frac{d\omega'}{2\pi}\frac{d_{>,ji}{}^{i\tau}(\mathbf{h},\mathbf{g},-\omega')}{\omega-\omega'-i\epsilon}. \quad (14)$$

Introducing the function

$$A_{ij}{}^{i\tau}(\mathbf{g},\mathbf{h},\omega)\equiv i(1-e^{-i\tau\omega})d_{>,ij}{}^{i\tau}(\mathbf{g},\mathbf{h},\omega), \quad (15)$$

we have, from (13),

$$d_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega)=\int\frac{d\omega'}{2\pi}\frac{A_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega')}{\omega+i\epsilon-\omega'}\frac{1}{1-e^{-i\omega'\tau}}+\int\frac{d\omega'}{2\pi}\frac{A_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega')}{\omega-i\epsilon-\omega'}\frac{1}{1-e^{i\omega'\tau}}. \quad (16)$$

Since $1/(\omega\pm i\epsilon)=P(1/\omega)\mp\pi i\delta(\omega)$, (16) is equivalent to

$$d_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega)=\frac{\Delta_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega+i\epsilon)}{1-e^{-i\omega\tau}}+\frac{\Delta_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega-i\epsilon)}{1-e^{i\omega\tau}}, \quad (17)$$

where the Green's function Δ of the complex variable z is defined by

$$\Delta_{ij}^{i\tau}(\mathbf{g},\mathbf{h},z)=\int\frac{d\omega}{2\pi}\frac{A_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega)}{z-\omega}. \quad (18)$$

The phonon Green's function $d(\omega)$ is thus a linear combination of boundary values of $\Delta(z)$.

We note that the spectral weight function $A(\mathbf{g},\mathbf{h},\omega)$ is just the Fourier transform of the displacement-displacement commutator:

$$A_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega)=\int_{-\infty}^{\infty}dt\,e^{i\omega(t-t')}\langle[\phi_{\mathbf{g},i}(t),\phi_{\mathbf{h},j}(t')]\rangle^{i\tau}. \quad (19)$$

From this form of A we may deduce two sum rules, and a symmetry relation. If we integrate both sides of (19) over all frequencies we find

$$\int\frac{d\omega}{2\pi}A_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega)=\langle[\phi_{\mathbf{g},i}(t),\phi_{\mathbf{h},j}(t)]\rangle^{i\tau}=0, \quad (20)$$

since the displacements commute at the same time. Also, using the equal-time commutation relations of momentum with displacement, we find

$$\int\frac{d\omega}{2\pi}M\omega A_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega)=i\left\langle\left[M\frac{d}{dt}\phi_{\mathbf{g},i}(t),\phi_{\mathbf{h},j}(t)\right]\right\rangle^{i\tau}=\delta_{\mathbf{gh}}\delta_{ij}, \quad (21)$$

since $M(d/dt)\phi_{\mathbf{g},i}(t)$ is just the momentum conjugate to $\phi_{\mathbf{g},i}(t)$.

The symmetry $A_{ij}(\mathbf{g},\mathbf{h},\omega)=-A_{ji}(\mathbf{h},\mathbf{g},-\omega)$ follows obviously from the antisymmetry of the commutator in (19). From the inversion symmetry of the lattice, and the fact that A depends only on the difference $\mathbf{g}-\mathbf{h}$, we then have the symmetry

$$A_{ij}(\mathbf{g},\mathbf{h},\omega)=-A_{ji}(\mathbf{g},\mathbf{h},-\omega). \quad (22)$$

In addition to Fourier transforms in time, we introduce spatial Fourier transforms:

$$D_{ij}^{i\tau}(\mathbf{k},t)=\sum_{\mathbf{g}}\exp[-i\mathbf{k}\cdot(\mathbf{g}-\mathbf{h})]D_{ij}^{i\tau}(\mathbf{g},\mathbf{h},t), \quad (23)$$

and

$$A_{ij}^{i\tau}(\mathbf{k},\omega)=\sum_{\mathbf{g}}\exp[-i\mathbf{k}\cdot(\mathbf{g}-\mathbf{h})]A_{ij}^{i\tau}(\mathbf{g},\mathbf{h},\omega). \quad (24)$$

To invert these transforms we use the formula

$$\Omega\int'\frac{d^3k}{(2\pi)^3}\exp(-i\mathbf{k}\cdot\mathbf{g})=\delta_{\mathbf{g}0}, \quad (25)$$

where Ω is the volume of a unit cell of the crystal, and the prime means that the integration is carried out only over the first Brillouin zone. Thus, for example,

$$D_{ij}^{i\tau}(\mathbf{g},\mathbf{h},t)=\Omega\int'\exp[i\mathbf{k}\cdot(\mathbf{g}-\mathbf{h})]D_{ij}^{i\tau}(\mathbf{k},t)\frac{d^3k}{(2\pi)^3}. \quad (26)$$

From the Green's function $\Delta(z)$, one may deduce the polarization and frequency versus wave number dependence of the low-frequency lattice oscillations, or phonons, and in addition the lifetime of these oscillations. The frequencies of the undamped phonons are found from the poles of $\Delta(\mathbf{k},z)$. We may see this in the following:

From (10) and (15) we may write $A(\mathbf{k},\omega)$ as

$$A_{ij}^{i\tau}(\mathbf{k},\omega)=\frac{1-e^{-i\tau\omega}}{\mathrm{Tr}e^{-i\tau H}}\sum_{E\zeta}\langle E\zeta|\phi_{\mathbf{g},i}2\pi\delta(H-E-\omega)\times\Big(\sum_{\mathbf{h}}e^{i\mathbf{k}\cdot\mathbf{h}}\phi_{\mathbf{h},j}\Big)|E\zeta\rangle e^{-i\tau E}, \quad (27)$$

where ζ represents the eigenvalues of the rest of a complete set of observables compatible with the energy. Then the absolute convergence of the trace, together with the positive definite structure of the operator $\phi_{\mathbf{g},i}\delta(H-E-\omega)\phi_{\mathbf{h},j}$, implies that the integral of $A(\mathbf{k},\omega)$ over all frequencies is absolutely convergent. Hence $\Delta(\mathbf{k},z)$ is analytic off the real z axis; consequently its possible poles are on the real axis and arise from the delta functions of $A(k,\omega)$.

It is clear from (27) that A will have a delta function whenever, for some $\mathfrak{A}$, and for all $|E\zeta\rangle$, $(\mathfrak{A}\cdot\sum_{\mathbf{h}}\exp(i\mathbf{k}\cdot\mathbf{h})\boldsymbol{\phi}_{\mathbf{h}})|E\zeta\rangle$ is a discrete energy eigenstate of the crystal. The energy difference between this state and $|E\zeta\rangle$ may then be regarded as due to the addition or removal of a single quantum of lattice oscillation. $\mathfrak{A}$ is the polarization vector of the phonon. For an actual system, one will not have sharp delta functions, but rather will have resonances, the widths of which correspond to the lifetimes of the phonon states.[3]

III. METHOD OF EVALUATION

Let us add to the Hamiltonian, H, the term

$$\sum_{\mathbf{g}}\mathbf{J}_{\mathbf{g}}(t)\cdot\boldsymbol{\phi}_{\mathbf{g}}(t),$$

which represents the interaction of each nucleus in the crystal with an external time-dependent force $\mathbf{J}_{\mathbf{g}}(t)$. We then define, for $\mathrm{Re}\tau>0$, an expectation value between states at time $t=0$, and $t=\mathrm{Re}\tau$:

$$\langle\tau|X|0\rangle_{\mathbf{J}}\equiv\sum_{E,\zeta}\langle E\zeta,t=\mathrm{Re}\tau|X|E\zeta,t=0\rangle e^{E\,\mathrm{Im}\tau}. \quad (28)$$

[3] A more complete account of the properties of the phonon Green's function, as well as an approximate evaluation of it for a model of a metal, will be published elsewhere.

The eigenvalues E, ζ are those of the unperturbed Hamiltonian and the rest of a complete set of observables compatible with the unperturbed energy. We shall assume that these observables do not depend explicitly on $\mathbf{J}$. The state $|E\zeta, t=\mathrm{Re}\tau\rangle$ is that into which $|E\zeta, t=0\rangle$ develops in the Heisenberg representation, under the dynamics of the full Hamiltonian, $H+\sum_g \mathbf{J}_g(t)\cdot\boldsymbol{\phi}_g(t)$. In the absence of the external force, i.e., $\mathbf{J}\equiv\mathbf{0}$, we have

$$\langle E\zeta, t=\mathrm{Re}\tau| = \langle E\zeta, t=0| e^{-iE\,\mathrm{Re}\tau}, \tag{29}$$

so that $\langle\tau|X|0\rangle_J$ reduces to $\mathrm{Tr}(e^{-i\tau H}X)$ when $\mathbf{J}\equiv\mathbf{0}$.

We will now show how to express thermal averages of time ordered functions of the nuclear displacement, $\boldsymbol{\phi}$, as variational derivatives of $\langle\tau|0\rangle$ with respect to $\mathbf{J}$. The variation $\delta_J\langle E_1\zeta_1 t_1|E_2\zeta_2 t_2\rangle$ in a transformation function $\langle E_1\zeta_1 t_1|E_2\zeta_2 t_2\rangle$ induced by the change in the dynamics due to a variation of $\mathbf{J}$ in the Hamiltonian is given by[4]

$$\delta_J\langle E_1\zeta_1 t_1|E_2\zeta_2 t_2\rangle = -i\left\langle E_1\zeta_1 t_1\left|\int_{t_2}^{t_1}\sum_g \delta\mathbf{J}_g(t)\cdot\boldsymbol{\phi}_g(t)\right|E_2\zeta_2 t_2\right\rangle. \tag{30}$$

The variational derivative, $[\delta/\delta\mathbf{J}_g(t)]\langle E_1\zeta_1 t_1|E_2\zeta_2 t_2\rangle$ is then the coefficient of $\delta\mathbf{J}_g(t)$ in the right side of (30). Thus

$$[\delta/\delta\mathbf{J}_g(t)]\langle E_1\zeta_1 t_1|E_2\zeta_2 t_2\rangle = \begin{cases} -i\langle E_1\zeta_1 t_1|\boldsymbol{\phi}_g(t)|E_2\zeta_2 t_2\rangle, & t_1>t>t_2 \\ 0, \text{ otherwise.}\end{cases} \tag{31}$$

In particular

$$\delta\langle\tau|0\rangle_J/\delta\mathbf{J}_g(t) = -i\langle\tau|\boldsymbol{\phi}_g(t)|0\rangle_J, \quad \mathrm{Re}\tau>t>0. \tag{32}$$

The second variational derivative $[\delta/\delta\mathbf{J}_h(t')][\delta/\delta\mathbf{J}_g(t)]\times\langle E_1\zeta_1 t_1|E_2\zeta_2 t_2\rangle$ may be calculated from (30) and (31) by adding two complete sets of intermediate states at time t, i.e.,

$$[\delta/\delta\mathbf{J}_g(t)]\langle E_1\zeta_1 t_1|E_2\zeta_2 t_2\rangle = -i\sum_{E_3\zeta_3 E_4\zeta_4}\langle E_1\zeta_1 t_1|E_3\zeta_3 t\rangle\langle E_3\zeta_3 t|\boldsymbol{\phi}_g(t)|E_4\zeta_4 t\rangle \times\langle E_4\zeta_4 t|E_2\zeta_2 t_2\rangle,$$

and using the product rule for differentiation. Thus, varying $\mathbf{J}$ in the Hamiltonian, we find

$$\begin{aligned}\delta_J\{[\delta/\delta\mathbf{J}_g(t)]\langle E_1\zeta_1 t_1|E_2\zeta_2 t\rangle\} &= -i\sum_{E_3\zeta_3}(\delta_J\langle E_1\zeta_1 t_1|E_3\zeta_3 t\rangle)\langle E_3\zeta_3 t|\boldsymbol{\phi}_g(t)|E_2\zeta_2 t_2\rangle \\ &\quad -i\sum_{E_4\zeta_4}\langle E_1\zeta_1 t_1|\boldsymbol{\phi}_g(t)|E_4\zeta_4 t\rangle(\delta_J\langle E_4\zeta_4 t|E_2\zeta_2 t_2\rangle) \\ &= (-i)^2\left\langle E_1\zeta_1 t_1\left|\int_{t_2}^{t_1}\sum_h(\boldsymbol{\phi}_g(t)\boldsymbol{\phi}_h(t'))_+\cdot\delta\mathbf{J}_h(t')dt'\right|E_2\zeta_2 t_2\right\rangle.\end{aligned}$$

We have used $\delta_J\langle E_3\zeta_3 t|\boldsymbol{\phi}(t)|E_4\zeta_4 t\rangle=0$, since all the times in this matrix element are equal, so that it does not depend on the dynamics, and neither $\boldsymbol{\phi}$ nor the operators whose eigenvalues define the states depend explicitly on $\mathbf{J}$. Hence

$$[\delta/\delta\mathbf{J}_h(t')][i\delta/\delta\mathbf{J}_g(t)]\langle\tau|0\rangle = -i\langle\tau|(\boldsymbol{\phi}_g(t)\boldsymbol{\phi}_h(t'))_+|0\rangle, \quad \mathrm{Re}\tau>t, t'>0. \tag{33}$$

When the external force is set equal to zero, the right side of (33) clearly becomes the Green's function $D^{i\tau}(\mathbf{g},\mathbf{h}, t-t')$ times $\mathrm{Tr}e^{-i\tau H}$.

By successive applications of the procedure used to derive (33), we may prove that

$$(\prod_{i=1}^{n} i\delta/\delta J_{g,i}(t_i))\langle\tau|0\rangle = \langle\tau|(\prod_{i=1}^{n}\boldsymbol{\phi}_{g,i}(t_i))_+|0\rangle, \quad \mathrm{Re}\tau>t_i>0. \tag{34}$$

This may be further generalized from polynomials to arbitrary time-ordered functions of $\boldsymbol{\phi}$. Thus, to evaluate the thermal expectation value for complex β, of a time-ordered function of the displacement coordinate, $\boldsymbol{\phi}$, we need only evaluate $\langle\tau|0\rangle$ in terms of J, then apply the appropriate variational derivatives, and finally set J equal to zero. In particular, if $\mathbf{J}_l'(t)$ is a c-number function,

$$\left\langle\left[\exp\left(-i\sum_l\int_0^{\mathrm{Re}\tau}dt\,\boldsymbol{\phi}_l(t)\cdot\mathbf{J}_l'(t)\right)\right]_+\right\rangle^{i\tau} = \left\{\left[\exp\sum_l\int_0^{\mathrm{Re}\tau}dt\,\mathbf{J}_l'(t)\cdot[\delta/\delta\mathbf{J}_l(t)]\right]\langle\tau|0\rangle_J\right\}_{J\equiv 0} \times(\langle\tau|0\rangle_0)^{-1}. \tag{35}$$

We can easily carry out the variational differentiations in (32). This is because the effect of the exponentiated $\delta/\delta\mathbf{J}_l$ is to induce a translation of $\mathbf{J}_l(t)$ by the amount $\mathbf{J}_l'(t)$ in the functional, $\langle\tau|0\rangle_J$, to its right. Once we have carried out the translation, $\mathbf{J}$ may be set equal to zero. Thus we find

$$\left\langle\left[\exp\left(-i\sum_l\int_0^{\mathrm{Re}\tau}dt\,\boldsymbol{\phi}_l(t)\cdot\mathbf{J}_l(t)\right)\right]_+\right\rangle^{i\tau} = \langle\tau|0\rangle_J/\langle\tau|0\rangle_0. \tag{36}$$

Read backwards, (36) is readily recognized as the usual formula for the transformation function $\langle\tau|0\rangle_J$ in the interaction representation.

For the case of neutron scattering, we want to evaluate (36) with the external force J taken to be

$$\mathbf{J}_l(t'') = \mathbf{K}[\delta_{gl}\delta(t-t'') - \delta_{hl}\delta(t'-t'')], \quad 0<t, t'<\mathrm{Re}\tau, \tag{37}$$

since with this force, the left side of (36) becomes

$$\langle(\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_g(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_h(t')])_+\rangle^{i\tau}.$$

[4] See J. Schwinger, Phys. Rev. **91**, 713 (1953), for further details.

We see then that what we must calculate is the transformation function that describes the response of the crystal to a force that imparts momentum $\mathbf{K}$ to the nucleus at $\mathbf{g}$ at time t and momentum $-\mathbf{K}$ to the nucleus at $\mathbf{h}$ at time $\mathbf{t}'$. This force, of course, is just that exerted by the neutron when forward scattering in the second Born approximation. We turn then to the problem of finding an explicit evaluation of the transformation function $\langle\tau|0\rangle_J$.

We shall evaluate $\langle\tau|0\rangle_J$ by making the approximation that the nuclear displacement [that is defined by $\langle\tau|\boldsymbol{\phi}_g(t)|0\rangle/\langle\tau|0\rangle$] induced by an external force $\mathbf{J}_h(t')$, is a linear functional of that force. The statement of linear response is just the statement that if one force produces a certain vibrational excitation, and a second force produces a second excitation, then the excitation produced by the sum of the two forces is just the sum of the two individual excitations. What we are assuming then is that the crystal possesses a spectrum of vibrational excitations, or phonons, that may be additively excited or de-excited by the external force. In other words, the individual phonons created or absorbed by the external force are taken to develop in time in the same way as would a single phonon interacting with the crystal at temperature $i\tau$. We make no assumptions about this time development—it is described by the phonon Green's function D, or alternatively, by the spectral weight function $A(\omega)$. It should also be pointed out that the assumption that the crystal responds linearly to external forces does not imply that the internal interparticle forces in the crystal are themselves linear, or harmonic, forces. On the other hand, for a harmonic crystal our approximation is exact.

As we have seen, the validity of the approximation of linear response in the case of neutron scattering depends on there not being significant interaction between the particular phonons created or absorbed by the neutron in passing through the crystal. Furthermore, the approximation may be expected to break down when the energy transferred to the crystal by the neutron is comparable to the characteristic binding energies of the crystal.

The linearity assumption is equivalent to the assumption that $[\delta/\delta\mathbf{J}_h(t')][\langle\tau|\boldsymbol{\phi}_g(t)|0\rangle/\langle\tau|0\rangle]$ is independent of $\mathbf{J}$, or alternatively, that it is satisfactorily approximated by its value for $\mathbf{J}\equiv 0$. Now, on the one hand,

$$[\delta/\delta\mathbf{J}_h(t')][\langle\tau|\boldsymbol{\phi}_g(t)|0\rangle/\langle\tau|0\rangle]$$
$$=-i\langle\tau|(\boldsymbol{\phi}_g(t)\boldsymbol{\phi}_h(t'))_+|0\rangle/\langle\tau|0\rangle$$
$$-i[\langle\tau|\boldsymbol{\phi}_g(t)|0\rangle/\langle\tau|0\rangle][\langle\tau|\boldsymbol{\phi}_h(t')|0\rangle/\langle\tau|0\rangle],\qquad 0<t,\,t'<\mathrm{Re}\tau,$$

and the right side for $\mathbf{J}\equiv\mathbf{0}$ is just $D^{i\tau}(\mathbf{g},\mathbf{h},t-t')$. On the other hand, from (32)

$$\langle\tau|\boldsymbol{\phi}_g(t)|0\rangle/\langle\tau|0\rangle=i[\delta/\delta\mathbf{J}_g(t)]\ln\langle\tau|0\rangle, \tag{39}$$

so that we find the approximate equation for $\langle\tau|0\rangle$:

$$i[\delta/\delta\mathbf{J}_h(t')][\delta/\delta\mathbf{J}_g(t)]\ln\langle\tau|0\rangle=D^{i\tau}(\mathbf{g},\mathbf{h},t-t'). \tag{40}$$

This has the immediate solution

$$\langle\tau|0\rangle_J=\langle\tau|0\rangle_0\exp\left[\frac{1}{2i}\sum_{gh}\int_0^{\mathrm{Re}\tau}\mathbf{J}_g(t)\cdot D^{i\tau}(\mathbf{g},\mathbf{h},t-t')\cdot\mathbf{J}_h(t')dtdt'\right]. \tag{41}$$

The linear coefficient of $\mathbf{J}$ in $\ln\langle\tau|0\rangle$, which from (39) equals $\langle\boldsymbol{\phi}\rangle_{J\equiv 0}{}^{i\tau}$, vanishes because the $\boldsymbol{\phi}$'s are the displacements from the nuclear equilibrium positions.

IV. NEUTRON SCATTERING

From (41) and (36), using the force given in (37), we find

$$\langle(\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_g(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_h(t')])_+\rangle^{i\tau}$$
$$=\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^{i\tau}+i\mathbf{K}\cdot D^{i\tau}(\mathbf{g},\mathbf{h},t-t')\cdot\mathbf{K}]. \tag{42}$$

Let us consider $t'\to 0$. Then for $\mathrm{Re}\tau>t>0$, we have the relation

$$\langle\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_g(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_h(0)]\rangle^{i\tau}$$
$$=\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^{i\tau}+\mathbf{K}\cdot\langle\boldsymbol{\phi}_g(t)\boldsymbol{\phi}_h(0)\rangle^{i\tau}\cdot\mathbf{K}]. \tag{43}$$

Both sides of (43) are boundary values of functions of t analytic in the strip $0>\mathrm{Im}t>\mathrm{Im}\tau$. Hence (43) must hold everywhere in the strip, since an analytic function is determined uniquely by its boundary values in an open set of a line segment of its boundary. This latter theorem is a consequence of the Schwarz reflection principle.

We now take the boundary values of both sides of (43) everywhere along the real t axis; hence (43) holds everywhere along the real t axis. Analytically continuing in τ to $\tau=-i\beta$, we find

$$\langle\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_g(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_h(t')]\rangle^{\beta}=\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^{\beta}+\mathbf{K}\cdot\langle\boldsymbol{\phi}_g(t)\boldsymbol{\phi}_h(t')\rangle^{\beta}\cdot\mathbf{K}]$$
$$=\exp\left[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^{\beta}+\int e^{-i\omega'(t-t')}\frac{\mathbf{K}\cdot A(\mathbf{g},\mathbf{h},\omega')\cdot\mathbf{K}}{1-e^{-\beta\omega'}}\frac{d\omega'}{2\pi}\right]. \tag{44}$$

This evaluation is basic to both coherent inelastic neutron scattering and resonant emission and absorption of neutrons and photons.

Substituting (44) in (6), we find for the transition probability per nucleus:

$$W^\beta(\mathbf{K},\omega)=\left(\frac{2\pi a}{m}\right)^2\sum_{\mathbf{g}}\exp[i\mathbf{K}\cdot(\mathbf{h}-\mathbf{g})]\int_{-\infty}^{\infty}dt\exp\left[i\omega(t-t')-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^\beta+\int e^{-i\omega'(t-t')}\frac{d\omega'}{2\pi}\frac{\mathbf{K}\cdot A(\mathbf{g},\mathbf{h},\omega')\cdot\mathbf{K}}{1-e^{-\beta\omega'}}\right]. \quad (45)$$

This is the same formula as that derived by Glauber for a harmonic crystal,[5] only here the spectral weight function $A(\omega)$ may be perfectly general, and not just that for a harmonic crystal. The actual calculation of $A(\omega)$ is a separate problem, and one need not make the same assumption in calculating $A(\omega)$ as was made in deriving (45). The important point is that (45) expresses the neutron scattering probabilities in terms of the structure of the phonon field, without it being necessary to specify that structure. If one expands the term $\exp[\int e^{-i\omega'(t-t')}\mathbf{K}\cdot A\cdot\mathbf{K}/(1-e^{-\beta\omega'})]$ on the right of (45) in a power series, then the term in $W^\beta(\mathbf{K},\omega)$ containing

$$\frac{1}{n!}\left[\int e^{-i\omega'(t-t')}\frac{d\omega'}{2\pi}\frac{\mathbf{K}\cdot A(\mathbf{g},\mathbf{h},\omega')\cdot\mathbf{K}}{1-e^{-\beta\omega'}}\right]^n$$

may be interpreted as the transition probability, $W_n^\beta(\mathbf{K},\omega)$, for the process in which the neutron suffers an energy loss by exciting phonons, and gains energy by absorbing other phonons, such that the total number of phonons involved is n. The transition probability per nucleus for a one-phonon process is then

$$W_1^\beta(\mathbf{K},\omega)=\left(\frac{2\pi a}{m}\right)^2\sum_{\mathbf{g}}\exp[i\mathbf{K}\cdot(\mathbf{h}-\mathbf{g})]\times\frac{\mathbf{K}\cdot A(\mathbf{g},\mathbf{h},\omega)\cdot\mathbf{K}}{1-e^{-\beta\omega}}\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^\beta]$$
$$=\left(\frac{2\pi a}{m}\right)^2\frac{\mathbf{K}\cdot A(\mathbf{K},\omega)\cdot\mathbf{K}}{1-e^{-\beta\omega}}\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^\beta]. \quad (46)$$

In an actual experiment, for $\mathbf{K}$ fixed, one observes peaks in the energy spectrum of the outgoing neutron, above a diffuse background. These peaks come from the ω resonances in $A(\mathbf{K},\omega)$, for $\mathbf{K}$ fixed, in (46). The center of a peak is at the energy of a phonon with wave number $\mathbf{K}$, and we see clearly from (46) that the width of the peak is exactly the inverse of the phonon lifetime.[6] In principle, then, one may infer $A(\mathbf{K},\omega)$ and hence the structure of the phonon field in the crystal from such an experiment. The diffuse background is caused by the multiphonon processes. The quantity $\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^\beta]$ is the Debye-Waller factor which multiplies each $W_n^\beta(\mathbf{K},\omega)$. Since the average squared displacement increases with temperature, the Debye-Waller factor is a decreasing function of temperature.

It is of interest to note that the expression for $W^\beta(\mathbf{K},\omega)$ that we have calculated obeys Placzek's exact sum rule[7]:

$$\int_{-\infty}^{\infty}W^\beta(\mathbf{K},\omega)\omega\frac{d\omega}{2\pi}=\left(\frac{2\pi a}{m}\right)^2\frac{K^2}{2M}. \quad (47)$$

Multiplying both sides of (45) by ω and integrating over all frequencies, holding $\mathbf{K}$ fixed, we find:

$$\int W^\beta(\mathbf{K},\omega)\omega\frac{d\omega}{2\pi}=\left(\frac{2\pi a}{m}\right)^2\sum_{\mathbf{g}}\exp[i\mathbf{K}\cdot(\mathbf{h}-\mathbf{g})]\times\exp\{-\tfrac{1}{2}\langle[\mathbf{K}\cdot(\boldsymbol{\phi}_{\mathbf{g}}-\boldsymbol{\phi}_{\mathbf{h}})]^2\}\rangle\times\int\frac{d\omega'}{2\pi}\omega'\frac{\mathbf{K}\cdot A(\mathbf{g},\mathbf{h},\omega')\cdot\mathbf{K}}{1-e^{-\beta\omega'}}. \quad (48)$$

The numerator of the last integral is even in ω', from (22), so the last integral may be written as

$$\frac{1}{2}\int\frac{d\omega'}{2\pi}\omega'\mathbf{K}\cdot A(\mathbf{g},\mathbf{h},\omega')\cdot\mathbf{K}[(1-e^{-\beta\omega'})^{-1}+(1-e^{\beta\omega'})^{-1}]=\frac{1}{2}\int\frac{d\omega'}{2\pi}\omega'\mathbf{K}\cdot A(\mathbf{g},\mathbf{h},\omega')\cdot\mathbf{K}=\frac{K^2}{2M}\delta_{\mathbf{gh}}, \quad (49)$$

using (21). Equation (47) then follows from substituting (49) in (48). It is also seen from (49) and (46) that the one-phonon peak obeys the sum rule,

$$\int W_1^\beta(\mathbf{K},\omega)\omega d\omega=\left(\frac{2\pi a}{m}\right)^2\frac{K^2}{2M}\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^\beta], \quad (50)$$

i.e., the total intensity under the one-phonon peak for fixed $\mathbf{K}$ is just the Debye-Waller factor times the total scattered intensity for fixed $\mathbf{K}$.

Finally, expression (42) obeys the detailed balancing condition,

$$W^\beta(-\mathbf{K},-\omega)=e^{-\beta\omega}W^\beta(\mathbf{K},\omega).$$

[5] R. J. Glauber, reference 1.

[6] Van Hove has noted this result, without derivation. L. Van Hove, Solid-State and Molecular Theory Group, Technical Report No. 11, Massachusetts Institute of Technology, Cambridge, Massachusetts, March, 1959 (unpublished).

[7] G. Placzek, Phys. Rev. 86, 377 (1952).

This is an immediate consequence of the boundary condition (11) on $D_>$.

V. EMISSION OF NEUTRONS AND PHOTONS

We will treat at the same time both the cases of emission of neutrons and of photons, since they differ only in the structure of the internal nuclear matrix element. The total transition probability $W_e(\omega)$ for emission of a neutron or photon with energy by a single excited nucleus, is, to within a constant factor,

$$W_e(\omega)=\sum_{E\zeta}\frac{|\langle E\zeta|V|E_0+\omega_0,\zeta_0\rangle|^2}{(E+\omega-E_0-\omega_0)^2+\Gamma^2/4}. \tag{51}$$

$|E_0+\omega_0,\zeta_0\rangle$ is the initial state of the crystal; ω_0 is the excitation energy of the excited nucleus (not including the rest mass of the neutron, in the case of nonrelativistic neutron emission), and E_0 is the initial crystal energy. We assume that the internal degrees of freedom of the nucleus are dynamically separable from those of the crystal. $|E\zeta\rangle$ is the final state of the crystal, and $1/\Gamma$ is the total lifetime of the excited nucleus bound in the crystal. V is the matrix element between internal nuclear states of the emission interaction. It still contains the center-of-mass coordinate of the nucleus as an operator. As in the case of neutron scattering, this dependence enters in a factor $\exp[-i\mathbf{K}\cdot(\mathbf{g}+\boldsymbol{\phi}_g)]$, where now $\mathbf{K}$ is the momentum of the emitted neutron or photon.

Using

$$(E^2+\Gamma^2/4)^{-1}=(1/\Gamma)\int_{-\infty}^{\infty}e^{-itE-\Gamma|t|/2}dt,$$

$W_e(\mathbf{K},\omega)$ becomes, again to within a constant factor:

$$\begin{aligned}W_e(\mathbf{K},\omega)&=\int_{-\infty}^{\infty}dt\,\exp[-i(\omega-\omega_0)t-\Gamma|t|/2]\langle E_0\zeta_0|e^{itE_0}\exp[-i\mathbf{K}\cdot(\mathbf{g}+\boldsymbol{\phi}_g)]e^{-itH}\exp[i\mathbf{K}\cdot(\mathbf{g}+\boldsymbol{\phi}_g)]|E_0\zeta_0\rangle\\&=\int_{-\infty}^{\infty}dt\,\exp[-i(\omega-\omega_0)t-\Gamma|t|/2]\langle E_0\zeta_0|\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_g(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_g(0)]|E_0\zeta_0\rangle.\end{aligned} \tag{52}$$

Averaging over a canonical ensemble of initial crystal states, at temperature $T=1/K\beta$, and neglecting the variation of Γ with crystal energy, we find

$$\begin{aligned}W_e^{\beta}(\mathbf{K},\omega)&=\int_{-\infty}^{\infty}dt\,\exp[-i(\omega-\omega_0)t-\Gamma|t|/2]\\&\quad\times\langle\exp[-i\mathbf{K}\cdot\boldsymbol{\phi}_g(t)]\exp[i\mathbf{K}\cdot\boldsymbol{\phi}_g(0)]\rangle^{\beta}.\end{aligned} \tag{53}$$

The thermally averaged matrix element in (53) is exactly the same one that we evaluated for neutron scattering. Thus from (44)

$$\begin{aligned}W_e^{\beta}(\mathbf{K},\omega)&=\int_{\infty}^{\infty}dt\,\exp[-i(\omega-\omega_0)t-\Gamma|t|/2]\\&\quad\times\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^{\beta}+\mathbf{K}\cdot\langle\boldsymbol{\phi}_g(t)\boldsymbol{\phi}_g(0)\rangle^{\beta}\cdot\mathbf{K}].\end{aligned} \tag{54}$$

Once more we may expand $\exp[\mathbf{K}\cdot\langle\boldsymbol{\phi}(t)\boldsymbol{\phi}(0)\rangle\cdot\mathbf{K}]$ in a power series and identify multiphonon terms. Of particular interest is the zero-phonon term, i.e.,

$$\begin{aligned}W_{e,0}^{\beta}(\mathbf{K},\omega)&=\int_{-\infty}^{\infty}dt\,\exp[-i(\omega-\omega_0)t-\Gamma|t|/2]\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^{\beta}]\\&=[\Gamma/((\omega-\omega_0)^2+\Gamma^2/4)]\exp[-\langle(\mathbf{K}\cdot\boldsymbol{\phi})^2\rangle^{\beta}].\end{aligned} \tag{55}$$

This term gives rise to a peak in the energy distribution of the emitted neutron or photon, with a width that is just the inverse of the lifetime of the excited nucleus bound in the crystal. Since the distribution is centered about the actual excitation energy of the nucleus,[8] there is no energy of recoil associated with the emission. It is this peak that is responsible for the Mössbauer effect.[9] The temperature dependence of the height of the peak enters through the Debye-Waller factor, which, as we have mentioned, is a decreasing function of temperature.

ACKNOWLEDGMENT

I would like to express my appreciation to Professor Julian Schwinger and to Dr. Kurt Gottfried for several valuable discussions about this work.

[8] There is actually a slight second order "Doppler" shift of the center of this peak; the excitation energy of the nucleus leads to a decrease in the kinetic energy of the excited nucleus due to its increased mass. This energy shift is approximately $\delta m/m$ times the average nuclear kinetic energy, where m is the mass of the unexcited nucleus, and $m+\delta m$ the mass of the excited nucleus. See R. V. Pound and G. A. Rebka, Jr., Phys. Rev. Letters **4**, 274 (1960).

[9] R. L. Mössbauer, Z. Physik **151**, 124 (1958); Naturwissenschaften **45**, 538 (1958); Z. Naturforsch. **14a**, 211 (1959).