Method for calculating Tr$\mathcal{H}^n$ in solid-state theory

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(Received 14 December 1973)

Expressing the quantum mechanics of Bloch electrons in a solid in terms of the Weyl transform instead of quantum operators and the Wigner function instead of state vectors, the method used by Wannier and Upadhyaya for calculating the trace of the nth power of the Hamiltonian operator is generalized as a series expansion in powers of $\hbar^2$ of a cosine function of a sum of Poisson-bracket operators. The expression is calculated explicitly to order $\hbar^2$. The result is applied to the derivation of the magnetic susceptibility of solids with substitutional impurities, to order $\hbar^2$, as well as to other problems where spatial inhomogeneity is present.

The need to evaluate the trace of the nth power of an operator such as the Hamiltonian operator in solid-state theory arises in many instances. One example is the calculation of the magnetic susceptibility of the system from the formulas

$$\chi = \lim_{B\to 0} -\frac{1}{V}\frac{\partial^2 F}{\partial B^2}, \tag{1}$$

$$F = N\mu + \mathrm{Tr}F(\mathcal{H}) , \tag{2}$$

$$F(\mathcal{H}) = -k_BT\ln(1+e^{(\mu-\mathcal{H})/k_BT}) \tag{3}$$

$$= \int_{c-i\infty}^{c+i\infty}\phi(s)\,e^{s\mathcal{H}}\,ds ,$$

$$e^{s\mathcal{H}} = \sum_n \frac{s^n}{n!}\mathcal{H}^n . \tag{4}$$

Wannier and Upadhyaya[1] (to be referred to as WU) evaluated Tr$\mathcal{H}^n$ up to the second order in the magnetic field strength for the case of Bloch electrons in a uniform magnetic field. Their method is very fascinating in that the result for Tr$\mathcal{H}^n$ assumes a particularly simple form such that one almost immediately knows χ once Tr$\mathcal{H}^n$ is obtained. Their method is essentially based on the existence of localized functions $A_\lambda(\vec{x},\vec{q})$ and extended functions $B_\lambda(\vec{x},\vec{p})$, where λ labels the energy band, $\vec{q}$ labels the lattice points, and $\vec{p}$ labels the crystal momentum $\hbar\vec{k}$. $A_\lambda(\vec{x},\vec{q})$ and $B_\lambda(\vec{x},\vec{p})$ are known as the magnetic Wannier functions and magnetic Bloch functions, respectively. The purpose of this paper is to generalize their method of evaluating Tr$\mathcal{H}^n$.

The urgent need to have a complete theory of the magnetic susceptibility of alloys, spurred on by the availability of extensive experimental data in the last two decades, has attracted many theorists.[2] Until now, however, only perturbative solutions in powers of the strength of the impurity potential exist and only in the special case of a free-electron band does the problem seem to have been formulated[2] by Kohn and Luming.

Here we extend the work in WU and find a general expression for Tr$\mathcal{H}^n$. The fundamental assumption made is that there exists a complete set of localized functions labeled by band index λ and lattice points $\vec{q}$, and a complete set of extended functions labeled by band index λ and crystal momentum $\vec{p}$. The two sets are connected by a unitary "lattice Fourier transformation" which is defined below.

Let $w_\lambda(\vec{x},\vec{q})$ be any localized state labeled by a band index λ and lattice point $\vec{q}$. Let $b_\lambda(\vec{x},\vec{p})$ be its lattice Fourier transform. Thus we can write

$$b_\lambda(\vec{x},\vec{p}) = (N\hbar^3)^{-1/2}\sum_{\vec{q}} e^{(i/\hbar)\vec{p}\cdot\vec{q}}\,w_\lambda(\vec{x},\vec{q}) , \tag{5}$$

$$w_\lambda(\vec{x},\vec{q}) = (N\hbar^3)^{-1/2}\sum_{\vec{p}} e^{-(i/\hbar)\vec{p}\cdot\vec{q}}\,b_\lambda(\vec{x},\vec{p}) , \tag{6}$$

$$\sum_{\vec{q}} e^{(i/\hbar)(\vec{p}-\vec{p}\,')\cdot\vec{q}} = N\hbar^3\delta_{\vec{p},\vec{p}\,'} , \tag{7}$$

$$\sum_{\vec{p}} e^{(i/\hbar)(\vec{q}-\vec{q}\,')\cdot\vec{p}} = N\hbar^3\delta_{\vec{q},\vec{q}\,'} , \tag{8}$$

where N is the total number of lattice points. For convenience, we introduce the Dirac ket and bra notation:

$$b_\lambda(\vec{x},\vec{p}) = |\vec{p},\lambda\rangle = |p\rangle , \tag{9}$$

$$w_\lambda(\vec{x},\vec{q}) = |\vec{q},\lambda\rangle = |q\rangle , \tag{10}$$

$$\langle\vec{p},\lambda'|\vec{q},\lambda\rangle = (N\hbar^3)^{-1/2}e^{-(i/\hbar)\vec{p}\cdot\vec{q}}\delta_{\lambda'\lambda} , \tag{11}$$

$$\langle\vec{p},\lambda|\vec{p}\,',\lambda'\rangle = \delta_{\lambda\lambda'}\delta_{\vec{p}\vec{p}\,'} , \tag{12}$$

$$\langle\vec{q},\lambda|\vec{q}\,',\lambda'\rangle = \delta_{\lambda\lambda'}\delta_{\vec{q}\vec{q}\,'} , \tag{13}$$

$$\sum_p |p\rangle\langle p| = 1 , \tag{14}$$

$$\sum_q |q\rangle\langle q| = 1 . \tag{15}$$

By using the closure relation, Eqs. (14) and (15), the following identity holds for an arbitrary operator A:

$$A = \sum_{p',p'',q',q''} |p''\rangle\langle p''|q''\rangle\langle q''|A|q'\rangle\langle q'|p'\rangle\langle p'| . \tag{16}$$

Introducing the notation

$$\vec{p}\,'=\vec{p}+\vec{u}\ ,\quad \vec{q}\,'=\vec{q}+\vec{v}\ ,$$
$$\vec{p}\,''=\vec{p}-\vec{u}\ ,\quad \vec{q}\,''=\vec{q}-\vec{v}\ ,$$

and using Eq. (11), we obtain

$$A=(N\hbar^3)^{-1}\sum_{\vec{p},\vec{q},\lambda,\lambda'} A_{\lambda\lambda'}(\vec{p},\vec{q})\Delta_{\lambda\lambda'}(\vec{p},\vec{q})\ , \tag{17}$$

where

$$A_{\lambda\lambda'}(\vec{p},\vec{q})=\sum_{\vec{v}} e^{(2i/\hbar)\vec{p}\cdot\vec{v}}\langle\vec{q}-\vec{v},\lambda|A|\vec{q}+\vec{v},\lambda'\rangle, \tag{18}$$

$$\Delta_{\lambda\lambda'}(\vec{p},\vec{q})=\sum_{\vec{u}} e^{(2i/\hbar)\vec{q}\cdot\vec{u}}|\vec{p}-\vec{u},\lambda\rangle\langle\vec{p}+\vec{u},\lambda'|\ . \tag{19}$$

By applying Eqs. (5) and (6) it is easy to obtain the following equivalent expressions for $A_{\lambda\lambda'}(\vec{p},\vec{q})$ and $\Delta_{\lambda\lambda'}(\vec{p},\vec{q})$:

$$A_{\lambda\lambda'}(\vec{p},\vec{q})=\sum_{u} e^{(2i/\hbar)\vec{q}\cdot\vec{u}}\langle\vec{p}+\vec{u},\lambda|A|\vec{p}-\vec{u},\lambda'\rangle, \tag{20}$$

$$\Delta_{\lambda\lambda'}(\vec{p},\vec{q})=\sum_{v} e^{(2i/\hbar)\vec{p}\cdot\vec{v}}|\vec{q}+\vec{v},\lambda\rangle\langle\vec{q}-\vec{v},\lambda'|\ . \tag{21}$$

In Eq. (17) the operator nature of A is transferred to $\Delta_{\lambda\lambda'}(\vec{p},\vec{q})$. The reader who is familiar with the alternative formulation of quantum mechanics in terms of the Weyl transform and the Wigner function[3] will recognize that $A_{\lambda\lambda'}(\vec{p},\vec{q})$ and $\Delta_{\lambda\lambda'}(\vec{p},\vec{q})$ correspond to the Weyl transform of the operator A and to the Δ function, respectively, in that formalism. We will refer to $A_{\lambda\lambda'}(\vec{p},\vec{q})$ as the lattice Weyl transform of the operator A. [Our definition of $A_{\lambda\lambda'}(\vec{p},\vec{q})$ differs from that of the continuous $(\vec{p},\vec{q})$ formalism by a factor of 2 in the exponential and the absence of $\frac{1}{2}$ inside the ket and bra, and, of course, by the replacement of the integral with the summation.] Thus an alternative formulation of the quantum mechanics of solids is possible using a complete set of functions $w_\lambda(\vec{x},\vec{q})$ labeled by a band index λ and lattice point $\vec{q}$ and a complete set of functions $b_\lambda(\vec{x},\vec{p})$ labeled by a band index λ and crystal momentum $\vec{p}$, the two complete sets being connected by a unitary lattice Fourier transformation. Examples of these complete sets are the Wannier functions and Bloch functions, both with and without a magnetic field. Blount's "mixed representation"[4] and Wannier's formulation[5] of the dynamics of Bloch electrons in a solid both are an embryonic form of a discrete $(\vec{p},\vec{q})$ version of the statistical formulation of quantum mechanics when considered in terms of the Weyl transform instead of operators and the Wigner function instead of state vectors.[3] The power of this method of doing quantum mechanics in solid-state theory has recently been demonstrated by the author[6] in calculating the magnetic susceptibility of bismuth.

In this paper we will restrict our treatment to the derivation of the expression for the trace of the nth power of an arbitrary operator A, particularly the Hamiltonian operator $\mathcal{H}$. To the author's knowledge, this is not given in the continuous $(\vec{p},\vec{q})$ formalism.

From Eq. (17) we can see that, because of the way the operator Δ is defined, the only nontrivial way in which the lattice Weyl transform of Δ explicitly occurs is when one takes the lattice Weyl transform of at least a product of two operators A and B. Therefore we have as fundamental binary operations the taking of the trace and the lattice Weyl transform of products of two operators or, equivalently, the trace and lattice Weyl transform of products of two Δ's.

Let

$$\Delta_{\alpha\alpha'}(\vec{p}_1,\vec{q}_1)=\sum_{u_1} e^{(2i/\hbar)\vec{q}_1\cdot\vec{u}_1}|\vec{p}_1-\vec{u}_1,\alpha\rangle\langle\vec{p}_1+\vec{u}_1,\alpha'|, \tag{22}$$

$$\Delta_{\beta\beta'}(\vec{p}_2,\vec{q}_2)=\sum_{u_2} e^{(2i/\hbar)\vec{q}_2\cdot\vec{u}_2}|\vec{p}_2-\vec{u}_2,\beta\rangle\langle\vec{p}_2+\vec{u}_2,\beta'|\ . \tag{23}$$

Then we have

$$\begin{aligned}\mathrm{Tr}[\Delta_{\alpha\alpha'}(\vec{p}_1,\vec{q}_1)\Delta_{\beta\beta'}(\vec{p}_2,\vec{q}_2)] &=\sum_{\gamma,\vec{p}}\sum_{\vec{u}_1,\vec{u}_2}\exp\left[\frac{2i}{\hbar}(\vec{q}_1\cdot\vec{u}_1+\vec{q}_2\cdot\vec{u}_2)\right]\delta_{\vec{p}_1-\vec{u}_1,\vec{p}}\\ &\quad\times\delta_{\vec{p}_2+\vec{u}_2,\vec{p}}\delta_{\vec{p}_1+\vec{u}_1,\vec{p}_2-\vec{u}_2}\delta_{\gamma,\alpha}\delta_{\alpha',\beta}\delta_{\beta',\gamma} \qquad (24)\\ &=\sum_{\vec{u}_1,\vec{u}_2,\gamma,\vec{p}}\exp\left[\frac{2i}{\hbar}(\vec{q}_1\cdot\vec{u}_1+\vec{q}_2\cdot\vec{u}_2)\right]\delta_{\vec{u}_1,\vec{p}_1-\vec{p}}\\ &\quad\times\delta_{\vec{u}_2,\vec{p}-\vec{p}_2}\delta_{\vec{p}_1,\vec{p}_2}\delta_{\gamma,\alpha}\delta_{\alpha',\beta}\delta_{\beta',\gamma} \qquad (25)\\ &=\sum_{\gamma,\vec{p}}\exp\left[\frac{2i}{\hbar}[\vec{p}\cdot(\vec{q}_2-\vec{q}_1)+\vec{q}_1\cdot\vec{p}_1-\vec{q}_2\cdot\vec{p}_2]\right]\\ &\quad\times\delta_{\vec{p}_1,\vec{p}_2}\delta_{\gamma,\alpha}\delta_{\alpha',\beta}\delta_{\beta',\gamma}\ . \qquad (26)\end{aligned}$$

The last line is obtained by writing $\vec{q}_1\cdot(\vec{p}_1-\vec{p})+\vec{q}_2\cdot(\vec{p}-\vec{p}_2)$ as $\vec{p}\cdot(\vec{q}_2-\vec{q}_1)+\vec{q}_1\cdot\vec{p}_1-\vec{q}_2\cdot\vec{p}_2$. We thus obtain, using Eq. (17),

$$\begin{aligned}\mathrm{Tr}AB &=(N\hbar^3)^{-2}\sum_{\substack{\vec{p}_1,\vec{q}_1,\alpha,\alpha'\\ \vec{p}_2,\vec{q}_2,\beta,\beta',\gamma,\vec{p}}} A_{\alpha\alpha'}(\vec{p}_1,\vec{q}_1)B_{\beta\beta'}(\vec{p}_2,\vec{q}_2)\\ &\quad\times\exp\left[\frac{2i}{\hbar}\ \vec{p}\cdot(\vec{q}_2-\vec{q}_1)+\vec{q}_1\cdot\vec{p}_1-\vec{q}_2\cdot\vec{p}_2\right]\\ &\quad\times\delta_{\vec{p}_1,\vec{p}_2}\delta_{\gamma,\alpha}\delta_{\alpha',\beta}\delta_{\beta',\gamma} \qquad (27)\\ &=(N\hbar^3)^{-1}\sum_{\vec{p},\vec{q},\gamma,\alpha}A_{\gamma\alpha}(\vec{p},\vec{q})B_{\alpha\gamma}(\vec{p},\vec{q})\ . \qquad (28)\end{aligned}$$

In taking the lattice Weyl transform of $\Delta_{\alpha\alpha'}(\vec{p}_1,\vec{q}_1)\times\Delta_{\beta\beta'}(\vec{p}_2,\vec{q}_2)$ it is convenient to use the expression in Eq. (20) applied to the Δ operator in Eq. (19). We therefore have

$$\{\Delta_{\alpha\alpha'}(\vec{p}_1,\vec{q}_1)\Delta_{\beta\beta'}(\vec{p}_2,\vec{q}_2)\}_{\gamma\gamma'}(\vec{p},\vec{q})$$

$$=\sum_{\vec{u}} e^{(2i/\hbar)\vec{q}\cdot\vec{u}}\langle \vec{p}+\vec{u},\gamma|\Delta_{\alpha\alpha'}(\vec{p}_1,\vec{q}_1)\Delta_{\beta\beta'}(\vec{p}_2,\vec{q}_2) \times|\vec{p}-\vec{u},\gamma'\rangle \quad (29)$$

$$=\sum_{\vec{u},\vec{u}_1,\vec{u}_2}\exp\left[\frac{2i}{\hbar}(\vec{q}\cdot\vec{u}+\vec{q}_1\cdot\vec{u}_1+\vec{q}_2\cdot\vec{u}_2)\right]\delta_{\vec{p}+\vec{u},\vec{p}_1-\vec{u}_1} \times\delta_{\vec{p}_1+\vec{u}_1,\vec{p}_2-\vec{u}_2}\delta_{\vec{p}_2+\vec{u}_2,\vec{p}-\vec{u}}\delta_{\gamma\alpha}\delta_{\alpha'\beta}\delta_{\beta'\gamma'} \quad (30)$$

$$=\sum_{\vec{u},\vec{u}_1,\vec{u}_2}\exp\left[\frac{2i}{\hbar}(\vec{q}\cdot\vec{u}+\vec{q}_1\cdot\vec{u}_1+\vec{q}_2\cdot\vec{u}_2)\right]\delta_{\vec{u},\vec{p}_1-\vec{p}_2} \times\delta_{\vec{u}_1,\vec{p}_2-\vec{p}}\delta_{\vec{u}_2,\vec{p}-\vec{p}_1}\delta_{\gamma\alpha}\delta_{\alpha'\beta}\delta_{\beta'\gamma'} \quad (31)$$

$$=\exp\left[\frac{2i}{\hbar}[(\vec{p}_2-\vec{p})\cdot(\vec{q}_1-\vec{q})+(\vec{q}_2-\vec{q})\cdot(\vec{p}-\vec{p}_1)]\right] \times\delta_{\gamma\alpha}\delta_{\alpha'\beta}\delta_{\beta'\gamma'}\ , \quad (32)$$

where the last line is obtained by writing $\vec{q}\cdot(\vec{p}_1-\vec{p}_2)+\vec{q}_1\cdot(\vec{p}_2-\vec{p})+\vec{q}_2\cdot(\vec{p}-\vec{p}_1)$ as $(\vec{p}_2-\vec{p})\cdot(\vec{q}_1-\vec{q})+)\vec{q}_2-\vec{q})\cdot(\vec{p}-\vec{p}_1)$. Introducing $\hat{p}=\vec{p}_2-\vec{p}$ and $\hat{q}=\vec{q}_2-\vec{q}$, the lattice Weyl transform of a product of two operators A and B may now be written as

$$(AB)_{\lambda\lambda'}(\vec{p},\vec{q})=(N\hbar^3)^{-2}\sum_{\vec{p}_1,\vec{q}_1,\hat{p},\hat{q},\beta}\exp\left[\frac{2i}{\hbar}[\hat{p}\cdot(\vec{q}_1-\vec{q})-\hat{q} \times(\vec{p}_1-\vec{p})]\right]A_{\lambda\beta}(\vec{p}_1,\vec{q}_1)B_{\beta\lambda'}(\hat{p}+\vec{p},\hat{q}+\vec{q})\ . \quad (33)$$

Further simplification of the above expression depends on our assumption that there exist two continuous functions of $\vec{q}$ having an infinite radius of convergence, which are equal to $A_{\lambda\beta}(\vec{p}_1,\vec{q}_1)$ and $B_{\beta\lambda'}(\hat{p}+\vec{p},\hat{q}+\vec{q})$, respectively, at all lattice points.[4] (We need not worry about the $\vec{p}$ dependence since, in the limit of infinite volume, $\vec{p}$ becomes continuous.) Therefore we can expand $B_{\beta\lambda'}(\hat{p}+\vec{p},\hat{q}+\vec{q})$ in a Taylor series around $B_{\beta\lambda'}(\vec{p},\vec{q})$ and obtain

$$(AB)_{\lambda\lambda'}(\vec{p},\vec{q})=(N\hbar^3)^{-2}\sum_{\vec{p}_1,\vec{q}_1,\hat{p},\hat{q},\beta}\exp\left[\frac{2i}{\hbar}[\hat{p}\cdot(\vec{q}_1-\vec{q})-\hat{q} \times(\vec{p}_1-\vec{p})]\right]A_{\lambda\beta}(\vec{p}_1,\vec{q}_1) \times\exp\left[\hat{p}\cdot\frac{\partial}{\partial\vec{p}}+\hat{q}\cdot\frac{\partial}{\partial\vec{q}}\right]B_{\beta\lambda'}(\vec{p},\vec{q}) \quad (34)$$

$$=(N\hbar^3)^{-2}\sum_{\vec{p}_1,\vec{q}_1,\hat{p},\hat{q},\beta}A_{\lambda\beta}(\vec{p}_1,\vec{q}_1) \times\exp\left[\frac{2i}{\hbar}[\hat{p}\cdot(\vec{q}_1-\vec{q})-\hat{q}\cdot(\vec{p}_1-\vec{p})]\right] \times\exp\left[\frac{\hbar}{2i}\left(\frac{\overleftarrow{\partial}}{\partial\vec{p}}\cdot\frac{\partial}{\partial\vec{q}}-\frac{\overleftarrow{\partial}}{\partial\vec{q}}\cdot\frac{\partial}{\partial\vec{p}}\right)\right]B_{\beta\lambda'}(\vec{p},\vec{q})\ , \quad (35)$$

where the arrows pointing to the left mean differential operator operating to the left. Summing over $\hat{p},\hat{q},\vec{p}_1,\vec{q}_1$ and using Eq. (8) we finally end up with

$$(AB)_{\lambda\lambda'}(\vec{p},\vec{q})=\sum_{\beta}\exp\left[\frac{\hbar}{2i}\left(\frac{\partial^{(a)}}{\partial\vec{p}}\cdot\frac{\partial^{(b)}}{\partial\vec{q}}-\frac{\partial^{(a)}}{\partial\vec{q}}\cdot\frac{\partial^{(b)}}{\partial\vec{p}}\right)\right] \times A_{\lambda\beta}(\vec{p},\vec{q})B_{\beta\lambda'}(\vec{p},\vec{q})\ , \quad (36)$$

where superscripts (a) and (b) indicate the objects of the differential operator.

In matrix notation we may write

$$(AB)(\vec{p},\vec{q})=\exp\left[\frac{\hbar}{2i}\left(\frac{\partial^{(a)}}{\partial\vec{p}}\cdot\frac{\partial^{(b)}}{\partial\vec{q}}-\frac{\partial^{(a)}}{\partial\vec{q}}\cdot\frac{\partial^{(b)}}{\partial\vec{p}}\right)\right] \times A(\vec{p},\vec{q})B(\vec{p},\vec{q})\ . \quad (37)$$

It is easy to see that the lattice Weyl transform of the anticommutator of A and B is

$$\{A,B\}(\vec{p},\vec{q})=\cos\left[\frac{\hbar}{2}\left(\frac{\partial^{(a)}}{\partial\vec{p}}\cdot\frac{\partial^{(b)}}{\partial\vec{q}}-\frac{\partial^{(a)}}{\partial\vec{q}}\cdot\frac{\partial^{(b)}}{\partial\vec{p}}\right)\right] \times[A(\vec{p},\vec{q})B(\vec{p},\vec{q})+B(\vec{p},\vec{q})A(\vec{p},\vec{q})] +i\sin\left[\frac{\hbar}{2}\left(\frac{\partial^{(a)}}{\partial\vec{p}}\cdot\frac{\partial^{(b)}}{\partial\vec{q}}-\frac{\partial^{(a)}}{\partial\vec{q}}\cdot\frac{\partial^{(b)}}{\partial\vec{p}}\right)\right] \times[A(\vec{p},\vec{q})B(\vec{p},\vec{q})-B(\vec{p},\vec{q})A(\vec{p},\vec{q})]\ . \quad (38)$$

Putting $A=B$ in the last expression we obtain

$$A^2(\vec{p},\vec{q})=\cos\left[\frac{\hbar}{2}\left(\frac{\partial^{(1)}}{\partial\vec{p}}\cdot\frac{\partial^{(2)}}{\partial\vec{q}}-\frac{\partial^{(1)}}{\partial\vec{q}}\cdot\frac{\partial^{(2)}}{\partial\vec{p}}\right)\right] \times\tfrac{1}{2}[A^{(1)}(\vec{p},\vec{q})A^{(2)}(\vec{p},\vec{q})+A^{(2)}(\vec{p},\vec{q})A^{(1)}(\vec{p},\vec{q})]\ . \quad (39)$$

By reiterating the binary operation prescribed by Eq. (39) one can easily convince oneself that for any power n

$$A^n(\vec{p},\vec{q})=\cos\left[\frac{\hbar}{2}\sum_{\substack{j,k=1\\ j<k}}^{n}\left(\frac{\partial^{(j)}}{\partial\vec{p}}\cdot\frac{\partial^{(k)}}{\partial\vec{q}}-\frac{\partial^{(j)}}{\partial\vec{q}}\cdot\frac{\partial^{(k)}}{\partial\vec{p}}\right)\right] \times\tfrac{1}{2}[A^{(1)}(\vec{p},\vec{q})A^{(2)}(\vec{p},\vec{q})\cdots A^{(n)}(\vec{p},\vec{q}) +A^{(n)}(\vec{p},\vec{q})A^{(n-1)}(\vec{p},\vec{q})\cdots A^{(2)}(\vec{p},\vec{q})A^{(1)}(\vec{p},\vec{q})]\ . \quad (40)$$

Equations (28) and (40) are all we need to evaluate the trace of the nth power of an arbitrary operator A. Thus by using Eq. (28) we have

$$\mathrm{Tr}A^n=(N\hbar^3)^{-1}\,\mathrm{Tr_{band}}\sum_{\vec{p},\vec{q}}A^{n-1}(\vec{p},\vec{q})A(\vec{p},\vec{q})\ , \quad (41)$$

and with the aid of Eq. (40) we end up with

$$\mathrm{Tr}A^n=(N\hbar^3)^{-1}\,\mathrm{Tr_{band}}\sum_{\vec{p},\vec{q}} \times\cos\left[\frac{\hbar}{2}\sum_{\substack{j,k=1\\ j<k}}^{n-1}\left(\frac{\partial^{(j)}}{\partial\vec{p}}\cdot\frac{\partial^{(k)}}{\partial\vec{q}}-\frac{\partial^{(j)}}{\partial\vec{q}}\cdot\frac{\partial^{(k)}}{\partial\vec{p}}\right)\right]$$

$$\times \tfrac{1}{2}[A^{(1)}(\vec{p},\vec{q})A^{(2)}(\vec{p},\vec{q})\cdots A^{(n)}(\vec{p},\vec{q}) + A^{(n)}(\vec{p},\vec{q})\cdots A^{(2)}(\vec{p},\vec{q})A^{(1)}(\vec{p},\vec{q})] , \quad (42)$$

where $\mathrm{Tr}_{\mathrm{band}}$ means trace over band indices. To make explicit progress beyond Eq. (42), we expand the cosine function to order $\hbar^2$ where we have[6]

$$\mathrm{Tr}A^n = (N\hbar^3)^{-1}\,\mathrm{Tr}\sum_{\vec{p},\vec{q}}\Bigg\{[A(p,q)]^n - \frac{1}{2!}\left(\frac{\hbar}{2}\right)^2\Bigg[\frac{(n-1)(n-2)}{2}[A(\vec{p},\vec{q})]^{n-2}\left(\left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{p}};\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{q}}\right\} - \left\{\frac{\partial^2 A(p,q)}{\partial\vec{p}\partial\vec{q}};\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{p}}\right\}\right) + \left(\frac{(n-1)(n-2)(2n-3)}{6} - \frac{(n-1)(n-2)}{2}\right)[A(\vec{p},\vec{q})]^{n-3}\left(\left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{p}};\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{q}}\,\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{q}}\right\} + \left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{q}};\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{p}}\,\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{p}}\right\} - \left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{q}};\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{q}}\,\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{p}}\right\} - \left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{p}};\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{p}}\,\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{q}}\right\}\right) + 2\left(\frac{n(n-1)(n-2)}{6} - \frac{(n-1)(n-2)}{2}\right)[A(\vec{p},\vec{q})]^{n-3}\left(\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{p}}\cdot\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{p}}\cdot\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{q}} + \frac{\partial A(\vec{p},\vec{q})}{\partial\vec{q}}\cdot\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{q}}\cdot\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{p}} - \frac{\partial A(\vec{p},\vec{q})}{\partial\vec{q}}\cdot\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{p}}\cdot\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{q}} - \frac{\partial A(\vec{p},\vec{q})}{\partial\vec{p}}\cdot\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{q}}\cdot\frac{\partial A(\vec{p},\vec{q})}{\partial\vec{p}}\right)\Bigg]\Bigg\} + O(\hbar^4)\,; \quad (43)$$

$\{\,;\,\}$ is a symmetrized tensor contraction, i.e.,

$$\left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{p}};\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{q}}\right\} = \frac{\partial^2 A(\vec{p},\vec{q})}{\partial p_i\partial p_j}\,\frac{\partial^2 A(\vec{p},\vec{q})}{\partial q_i\partial q_j} + \frac{\partial^2 A(\vec{p},\vec{q})}{\partial q_i\partial q_j}\,\frac{\partial^2 A(\vec{p},\vec{q})}{\partial p_i\partial p_j}\,,$$

and moreover, the p's must never have identical indices as well as the q's where repeated indices are summed over. In evaluating the summation by an integral, $(1/N\hbar^3)\sum_{\vec{p},\vec{q}}$ goes over to $h^{-3}\int d^3p\,d^3q$. Equation (42) indeed will give us a systematic expansion of $\mathrm{Tr}A^n$ in powers of $\hbar^2$.

If $A(\vec{p},\vec{q})$ is of the functional form $A(\vec{p}-(e/c)\vec{A}(\vec{q}); f(\vec{p},\vec{q}))$, where $f(\vec{p},\vec{q})\to 0$ as $|\vec{q}|\to\infty$, then integration by parts permits the reduction of the $[A(\vec{p},\vec{q})]^{n-3}$ term to the form of the $[A(\vec{p},\vec{q})]^{n-2}$ term plus gradient terms which do not contribute after integration with respect to $\vec{p}$ and $\vec{q}$. To prove this, one has to make use of the periodicity in $\vec{p}$ space and also the fact that the result of $\vec{p}$ integration of an integrand, which is a function of $[\vec{p}-(e/c)\mathrm{A}(\vec{q})]$ only, does not depend on $\vec{q}$.

The final result can be written

$$\mathrm{Tr}A^n = \mathrm{Tr}_{\mathrm{band}}h^{-3}\int d^3p\,d^3q\Bigg[[A(\vec{p},\vec{q})]^n - \frac{1}{2!}\left(\frac{\hbar}{2}\right)^2\left(\frac{n(n-1)}{6}\right)[A(\vec{p},\vec{q})]^{n-2} \times\left(\left\{\frac{\partial^2 A(p,q)}{\partial p\partial p};\frac{\partial^2 A(p,q)}{\partial q\partial q}\right\} - \left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{q}};\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{p}}\right\}\right) + O(\hbar^4)\Bigg]\,, \quad (44)$$

and the trace of an arbitrary function of operator A defined in terms of power series can thus be expressed as

$$\mathrm{Tr}F(A) = \mathrm{Tr}_{\mathrm{band}}h^{-3}\int d^3p\,d^3q\Bigg[F(A(p,q)) - \frac{1}{2!}\left(\frac{\hbar}{2}\right)^2\frac{1}{6}F''(A(\vec{p},\vec{q})) \times\left(\left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{p}};\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{q}}\right\} - \left\{\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{p}\partial\vec{q}};\frac{\partial^2 A(\vec{p},\vec{q})}{\partial\vec{q}\partial\vec{p}}\right\}\right) + O(\hbar^4)\Bigg]\,. \quad (45)$$

We wish to point out that $\mathrm{Tr}\mathcal{H}^n$ in WU exactly follows from Eq. (44), where the lattice Weyl transform $H(\vec{p},\vec{q})$ of the Hamiltonian is $H(\vec{p},\vec{q}) = W_\lambda(\vec{p}-(e/c)\vec{A}(\vec{q});B)\delta_{\lambda\lambda}$, W being referred to in WU as the renormalized-energy band function. (For the case where spin-orbit coupling is included, for each band index λ, W is a 2×2 matrix, and this is given by Roth.[7]

Because of the particular combination of $\vec{p}$ and $\vec{q}$ in $W_\lambda(\vec{p}-(e/c)\vec{A}(\vec{q});B)$, $\vec{q}$ did not explicitly appear in the problem considered in WU. In the presence of a substitutional-impurity potential, $\vec{q}$ will explicitly appear in the problem. The virtue of Eq. (42) is that it allows inhomogeneity in direct space.

Using the above trace formulas, we have extended the work in WU to the derivation of the magnetic susceptibility of dilute alloys to order $\hbar^2$. The result given for χ is valid for general Bloch bands to all orders in the impurity potential[8] and reduces to all known limiting cases discussed by

Hebborn and Scannes.[2] In the free-electron-band model the expression for $\Delta\chi$ per solute atom gives a firm theoretical foundation to the formula used by Henry and Rogers,[9] as pointed out by Kohn and Luming,[2] which accounts quite well for their experimental results on dilute alloys of Zn, Ga, Ge, and As with Cu. This is given as

$$\Delta\chi = -\frac{e^2}{6mc^2}\int d^3q\,\Delta\rho(\vec{q})\,|\vec{q}\,|^2 + \chi_{\rm orb}(0)\Delta g_1/g + \chi_{\rm spin}(0)\Delta g_2/g\ , \tag{46}$$

where

$$\Delta\rho(\vec{q}) = \frac{2h^{-3}}{V}\int d^3p\left\{\left[f(\Sigma^0) + \tfrac{1}{24}f''(\Sigma^0)\frac{\hbar^2\nabla^2}{m}V_{\rm I}(\vec{q}) - f\left(\frac{p^2}{2m}\right)\right] + \left[f'(\Sigma^0) + \tfrac{1}{24}f'''(\Sigma^0) \times \frac{\hbar^2\nabla^2}{m}V_{\rm I}(\vec{q}) - f'\left(\frac{p^2}{2m}\right)\right]\frac{p^2}{3m}\right\}\ , \tag{47}$$

$$g = \frac{2h^{-3}}{V}\int d^3p\,d^3q\,f'\left(\frac{p^2}{2m}\right)\ , \tag{48}$$

$$\Delta g_1 = \frac{2h^{-3}}{V}\int d^3p\,d^3q\left[f'(\Sigma^0) - f'\left(\frac{p^2}{2m}\right)\right] \tag{49}$$

$$\Delta g_2 = \Delta g_1 + \frac{2}{24}\frac{h^{-3}}{V}\int d^3p\,d^3q f'''(\Sigma^0)\frac{\hbar^2\nabla^2}{m}V_{\rm I}(\vec{q})\ , \tag{50}$$

$$\Sigma^0 = \frac{p^2}{2m} + V_{\rm I}(\vec{q})\ , \tag{51}$$

$$\chi_{\rm spin}(0) = \frac{-2h^{-3}}{V}\int d^3p\,d^3q\,f'\left(\frac{p^2}{2m}\right)\mu_B^2\ , \tag{52}$$

$$\chi_{\rm orb}(0) = \frac{2}{3}\frac{h^{-3}}{V}\int d^3p\,d^3q f'\left(\frac{p^2}{2m}\right)\mu_B^2\ . \tag{53}$$

The derivation of the above result and detailed discussion is given in a separate paper.[8] The general result for χ can also be applied to bismuth, where the effect of the impurity potential should be large.[2,10] Using the Lax $\vec{k}\cdot\vec{p}$ model for bismuth, which is equivalent to the Dirac Hamiltonian by a simple velocity scale factor[10] one may make use of the result of Blount[11] and Suttorp[12] for $H(\vec{p},\vec{q})$ in the form given for Dirac particles.

The potentiality of the formalism presented here cannot be underestimated. It is applicable to a vast number of quantum-mechanical problems. Indeed the formalism yields a rigorous quantum-mechanical basis of the distribution-function method in potential screening, the Thomas-Fermi method[13] in the absence of the magnetic field, and the quasiclassical approximation for nonzero magnetic field.[14] By means of the formula

$$N = -\frac{\partial}{\partial\mu}\,{\rm Tr}F(\mathcal{H}) \tag{54}$$

$$= \int d^3q\,n(\vec{q})\ , \tag{55}$$

the local particle density $n(q)$ can thus be obtained as a series expansion in powers in $\hbar^2$, which can, in principle, be obtained to all orders in $\hbar^2$. Then the self-consistent potential due to an impurity charge Ze located at the origin must satisfy the Poisson equation in electrostatics:

$$\nabla^2 V(\vec{q}) = 4\pi e n(\vec{q}) - 4\pi e n_0 - 4\pi Ze\,\delta(\vec{q})\ . \tag{56}$$

The result for the potential screening at zero field, low field, and very high magnetic field is given by the author in a separate paper[15] and agrees with the general result of Horing,[14] to order $\hbar^2$, obtained by the Green's function method and random-phase approximation using linear-response theory. The present formalism has the advantage that it can be applied to particles of arbitrary dispersion law. Thus it finds an immediate extension to potential screening by a relativistic quantum plasma using $H(\vec{p},\vec{q})$ for the Dirac particle given by Blount[11] and Suttorp.[12] The result of this calculation will be reported elsewhere.

Finally we would like to mention the calculation of the density of states $D(\epsilon)$, which is just equal to $dN(\epsilon)/d\epsilon$ for $T=0$, where ϵ plays the role of the Fermi energy. We find for the Hamiltonian $\mathcal{H} = \vec{P}^2/2m - eV(\vec{Q})$,

$$D(\epsilon) = \frac{{\rm Tr}_{\rm band}}{h^3}\int d^3p\,d^3q\left[\delta(H(\vec{p},\vec{q}) - \epsilon) + \frac{\hbar^2 e}{24m}\delta''(H(\vec{p},\vec{q}) - \epsilon)\nabla^2 V(\vec{q})\right] + O(\hbar^4)\ . \tag{57}$$

The above result leads to a necessary, though not sufficient, general condition for the existence of bound states for a given potential function $V(\vec{q})$. To the author's knowledge nothing of this sort of generality has been given in the literature. Further discussion will again be given in another communication. Another foreseeable application is perhaps in the theory of disordered systems approached along the lines given by Kane.[16]

The author would like to thank Professor N. H. March for his critical reading of the manuscript and for some very helpful comments; also Dr. U. N. Upadhyaya for checking some of the results and the conclusion. He is grateful to Professor Abdus Salam and Professor Paolo Budini, the International Atomic Energy Agency, and UNESCO for hospitality at the International Center for Theoretical Physics, Trieste.

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