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Position-dependent effective masses in semiconductor theory

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The motion of free carriers (electrons and holes) in semiconductors of nonuniform chemical composition is sometimes described by means of a Hamiltonian possessing a position-dependent effective mass. In previous work we have shown that position-dependent masses lead to inconsistencies on account of Bargmann's theorem, which postulates that a coherent superposition of states of different masses (wave packets) is forbidden. We have also shown how to circumvent this selection rule. We derive an extension of Bargmann's theorem to the effect that Hamiltonians with position-dependent masses are not Galilean invariant. Furthermore, it is also shown that the customary derivation of position-dependent effective-mass Hamiltonians is by no means unique. There exist, in general, many nonequivalent Hamiltonians within the same approximation, all derivable from the basic many-body Hamiltonian, as long as the concept of a position-dependent mass is maintained. Because of the lack of uniqueness and the lack of Galilean invariance of variable-effective-mass theories it seems appropriate to abandon the concept of a position-dependent mass. In previous work we have shown how to do this successfully.

I. INTRODUCTION

The importance of a theoretical understanding of transport phenomena in semiconductors of a variable, position-dependent chemical composition for modern device technology does not need to be emphasized. The Wannier-Slater theorem[1] valid for homogeneous semiconductors of a uniform chemical composition proved to be a natural starting point for an extension and generalization to nonuniform material. The Wannier-Slater theorem in its simplest form states that the envelope function $F_c(\vec{r},t)$ for the conduction band, for instance, obeys the following Schrödinger equation:

$$[E_c(-i\vec{\nabla})+U(\vec{r})]F_c(\vec{r},t)=i\hbar\dot{F}_c(\vec{r},t) , \tag{1}$$

where $E_c(\vec{k})$ signifies the conduction-band energy of the unperturbed crystal with $\vec{k}=-i\vec{\nabla}$ as the crystal momentum and $U(\vec{r})$ is the perturbation engendered by external agencies (applied voltages, etc.) or shallow impurities, as the case may be. In the effective-mass approximation in its simplest case (one minimum at $\vec{k}=0$ with a scalar effective mass m^*), Eq. (1) goes over into

$$\left[-\frac{\hbar^2}{2m^*}\nabla^2+E_c(0)+U(\vec{r})\right]F=i\hbar\dot{F} , \tag{2}$$

where now the bottom of the conduction band is given by $E_c(0)+U(\vec{r})$. For details, particularly in more complicated situations (several minima, degeneracy of energy eigenvalues, band mixing, etc.), the reader is referred to Ref. 2. An extension of the Wannier-Slater theorem to nonuniform semiconductors, material possessing a position-dependent varying chemical composition, has been attempted by a number of authors.[3-6] Since we are primarily interested in the consequences resulting from the concept of a position-dependent effective mass $m^*=m(\vec{r})$ (we omit the star from now on for simplicity), we shall concentrate only on the kinetic-energy term of the Schrödinger equation for the envelope function F as it emerges from the various and sundry theories proposed.[3-6] A brief review is indicated. Starting with the one-electron approximation of the many-body Hamiltonian for a binary alloy of a position-dependent composition, Gora and Williams, with the use of Slater's method,[1] derived the kinetic-energy operator[7]

$$T=-\frac{\hbar^2}{2m(\vec{r})}\nabla^2 , \tag{3}$$

which is manifestly non-Hermitian. Later, these authors amended their derivation of the pertinent Schrödinger equation for the envelope function F and postulated

$$T=-\frac{\hbar^2}{4}[m(\vec{r})^{-1}\nabla^2+\nabla^2 m^{-1}(\vec{r})] , \tag{4}$$

to be the correct kinetic-energy operator.[3] Expression (4) is indeed Hermitian and has also been adopted by Bastard *et al.*[3] Recently, van Vliet and Marshak[4] rederived Eq. (4), again with the use of the method of Slater.[1] Also quite recently, Zhu and Kroemer,[6] with the use of a variant of the tight-binding approximation, postulated

$$T=-\hbar^2[m(\vec{r})]^{-1/2}\nabla^2[m(\vec{r})]^{-1/2}\ , \tag{5}$$

as the correct kinetic-energy operator, applicable to situations which warrant the use of position-dependent effective masses.

This author has shown[5] how to avoid the use of position-dependent masses, arguing that Bargmann's theorem does not allow the superposition of states with different masses and that, therefore, wave-packet solutions to Schrödinger equations with kinetic-energy operators as given by Eqs. (4) or (5) are unphysical.

Bargmann's theorem, quoting Kaempffer,[8] states: "It is impossible to have in non-relativistic quantum mechanics states which are linear superpositions of states describing particles of different masses." In the next section we shall go a step further and show that *any* Hamiltonian with a position-dependent mass is *not* Galilean invariant. Therefore, an observer moving with a constant velocity with respect to a semiconductor sample of nonuniform composition, will in general measure *different* values for physical quantities as an observer at rest with respect to the sample. Clearly, this constitutes an unphysical state of affairs. It is, however, possible to view the lack of Galilean invariance in a different light with the concept of preferred coordinate systems. This will be shown in the Appendix.

In the third and last section we shall show that the approximation scheme of Gora and Williams,[3] as well as that of van Vliet and Marshak[4] leads to ambiguities which can only be resolved if the notion of a position-dependent mass is abandoned. This ambiguity or nonuniqueness of the Gora-Williams approximation scheme is a reflection of the lack of Galilean invariance of the ensuing Hamiltonians.

II. POSITION-DEPENDENT MASSES AND GALILEAN INVARIANCE

As in the Introduction we assume a scalar position-dependent mass, $m(\vec{r})$. The complications arising from a consideration of tensorial masses, being not germane to the issue, are ignored. We introduce the kinetic-energy operator

$$T=-\frac{\hbar^2}{4}(m^{\alpha}\vec{\nabla}m^{\beta}\vec{\nabla}m^{\gamma}+m^{\gamma}\vec{\nabla}m^{\beta}\vec{\nabla}m^{\alpha})\ , \tag{6}$$

where $m=m(\vec{r})$ is the position-dependent effective mass of an electron in the conduction band. The constants α, β, and γ, satisfying the constraint

$$\alpha+\beta+\gamma=-1\ , \tag{7}$$

are otherwise assumed to be arbitrary. Obviously, operator (6) is Hermitian. Gora and William's operator is retrieved from Eq. (6) by putting $\alpha=-1$ and $\beta=\gamma=0$, while Zhu and Kroemer's follows from Eq. (6) if we put $\alpha=\gamma=-\frac{1}{2}$ and $\beta=0$. Another kinetic-energy operator is obtained if we put $\alpha=\gamma=0$ and $\beta=-1$. We shall see later, in Sec. III how to derive this latter operator directly from Gora and William's approximation procedure, thus proving explicitly its ambiguity. The Schrödinger equation for the envelope function $F(\vec{r},t)$ reads

$$[T+V(\vec{r})]F=i\hbar\dot{F}\ . \tag{8}$$

$V(\vec{r})$ contains an externally applied potential and other terms due to a nonuniform chemical composition.[4,5] Performing the Galilei transformation

$$\begin{aligned}\vec{r}\,'&=\vec{r}+\vec{v}t\ ,\\ t'&=t\ ,\end{aligned} \tag{9}$$

where $\vec{v}$ is a constant but otherwise arbitrary velocity, Eq. (8) goes over into[9]

$$(T'+V')F'=i\hbar\dot{F}'+i\hbar\vec{v}\cdot\vec{\nabla}'F'\ . \tag{10}$$

Since the forces acting on the electron originate from force centers which are also displaced by transformation (9) we have[9]

$$V'(\vec{r}\,')=V'(\vec{r})=V(\vec{r})\ . \tag{11}$$

The same is true for the mass $m(\vec{r})$, since it originates ultimately from potentials which behave according to Eq. (11). Thus

$$m'(\vec{r}\,')=m'(\vec{r})=m(\vec{r})\ . \tag{12}$$

We now omit the primes in Eq. (10) for simplicity. The envelope function F then obeys, in the moving reference frame, the Schrödinger equation

$$(T+V-i\hbar\vec{v}\cdot\vec{\nabla})F=i\hbar\dot{F}\ , \tag{13}$$

where T is given by Eq. (6). Since transformation (9) should induce a unitary transformation on the wave function F, or otherwise probability would not be preserved, we must have in the moving frame

$$F=e^{if}\phi\ , \tag{14}$$

where f is a real function of the coordinates $\vec{r}$ and time t. A little algebra shows that Eq. (13) goes over into

$$\left[T+\frac{\hbar^2}{2m}i\left[\frac{\vec{\nabla}m}{m}\cdot\vec{\nabla}f-\nabla^2 f\right]+\frac{\hbar^2}{2m}(\vec{\nabla}f)^2-i\left[\frac{\hbar^2}{m}\vec{\nabla}f+\hbar\vec{\mathrm{v}}\right]\cdot\vec{\nabla}+V\right]\phi=i\hbar\dot{\phi}-\hbar\dot{f}\phi\ . \tag{15}$$

We note, that although T depends explicitly on the coefficients α, β, and γ, Eq. (15) does otherwise not, provided Eq. (7) is satisfied.

If we now put tentatively

$$\frac{\hbar}{m}\vec{\nabla}f=-\vec{\mathrm{v}} \tag{16a}$$

and

$$\frac{m}{2}v^2=-\hbar\dot{f}\ , \tag{16b}$$

then formally Eq. (15) would go over into

$$(T+V)\phi=i\hbar\dot{\phi}\ , \tag{17}$$

an expression which is form invariant with Eq. (8). But in order that Eq. (16a) possesses a solution, the condition

$$\vec{\nabla}m\times\vec{\mathrm{v}}=0 \tag{18}$$

must be satisfied. This follows from equating mixed derivatives

$$\frac{\partial^2 f}{\partial x\partial y}=\frac{\partial^2 f}{\partial y\partial x}\ .$$

But since $\vec{\mathrm{v}}$ is arbitrary it follows by necessity that

$$\vec{\nabla}m=\vec{0}\text{ or }m=m_0=\text{const}\ , \tag{19}$$

and the solution of Eqs. (16) becomes

$$f=-\frac{m_0}{\hbar}\vec{\mathrm{v}}\cdot\vec{\mathrm{r}}-\frac{m_0}{2\hbar}v^2 t\ , \tag{20}$$

which leads to Galilean invariance in the customary fashion. However, if the mass m is position dependent, *no* solutions to Eqs. (16) exist and consequently Eq. (10) *does not possess Galilean invariance.*

The absurdity of the situation need not be emphasized as long as we insist on Galilean invariance. One small example suffices to show this. From Eq. (8) together with the kinetic-energy operator T given by Eq. (6) there follows the current-conservation law

$$\frac{\partial}{\partial t}FF^*+\frac{\hbar}{2i}\vec{\nabla}\cdot\left[\frac{1}{m}(F^*\vec{\nabla}F-F\vec{\nabla}F^*)\right]=0\ , \tag{21}$$

independently of the particular values the constants α, β, and γ may attain as long as they satisfy Eq. (7). In the moving frame characterized by transformation (9) and Eq. (13) for the motion of the electron we obtain

$$\frac{\partial}{\partial t}FF^*+\vec{\nabla}\cdot\left[\frac{\hbar}{2im}(F^*\vec{\nabla}F-F\vec{\nabla}F^*)\right]=-\vec{\mathrm{v}}\cdot\vec{\nabla}(FF^*)\ , \tag{22}$$

again omitting primes for simplicity. This clearly shows that the current is not conserved by merely looking at it from a moving frame, as it were. The unitary transformation (14) does not exist for position-dependent masses and therefore there is no possibility to transform away the offending term on the right-hand side (rhs) of Eq. (22), which is, however, always possible for a *constant* mass $m=m_0$ with the result that

$$\frac{\partial}{\partial t}\phi\phi^*+\frac{\hbar}{2im_0}\vec{\nabla}\cdot(\phi^*\vec{\nabla}\phi-\phi\vec{\nabla}\phi^*)=0 \tag{23}$$

in this case, clearly exhibiting current conservation as observed also from a moving frame of reference.

The insistence on Galilean invariance for effective mass equations, although pleasing from a theoretical point of view, need not be maintained if one chooses to deal with preferred coordinate systems as shown in the appendix.

III. PROOF OF THE NONUNIQUENESS OF HAMILTONIANS WITH POSITION-DEPENDENT EFFECTIVE MASSES

Before we give a proof of ambiguities associated with theories dealing with position-dependent masses let us look at kinetic-energy operator (6) more closely. If we perform the indicated operations, we find that

$$TF=T_0F+T_1F\ , \tag{24}$$

where T_0 is independent of the constants α, β, and γ, and is given by

$$T_0=-\frac{\hbar^2}{2m}\nabla^2+\frac{\hbar^2}{2m^2}\vec{\nabla}m\cdot\vec{\nabla}\ , \tag{25}$$

and T_1 depends on α, β, and γ explicitly and is given by

$$T_1 = -\frac{\hbar^2}{4m}\{(\alpha+\gamma)m^{-1}\nabla^2 m + [\alpha(\beta+\alpha-1)+\gamma(\beta+\gamma-1)]m^{-2}(\vec{\nabla} m)^2\} \ . \tag{26}$$

For the particular choice $\alpha=-1, \beta=\gamma=0$ of Eq. (4) we have

$$T_1 = \frac{\hbar^2}{4m}[m^{-1}\vec{\nabla} m - 2m^{-2}(\vec{\nabla} m)^2] \ , \tag{27a}$$

and for $\alpha=\gamma=0, \beta=-1$, a choice which represents one of the many possibilities to be derived later, we obtain

$$T_1 = 0 \ . \tag{27b}$$

It could be argued that as long as variations of the mass with position are sufficiently small, so that $\nabla^2 m$ and $(\vec{\nabla} m)^2$ may be neglected, operator (6) is unique. Because T_1 becomes negligible in this case and T_0, being independent of the particular choices for α, β, and γ, becomes the unique representative of the kinetic-energy operator. This is, however, *not* true in general. The gradient of the wave function $\vec{\nabla} F$ depends on the rapidity of changes of the potential V with position. Sufficiently slowly varying external potentials cause small enough gradients of F, particular close to the band edge, so that, even in cases where second derivatives and squares of the first derivative of m are small, we have

$$m^{-1}\vec{\nabla} m \cdot \vec{\nabla} F \approx m^{-2}(\vec{\nabla} m)^2 F \ , \tag{28}$$

or, in other words, terms stemming from T_1 are of the same order of magnitude as the second term on the rhs of Eq. (25). Thus, in general, the kinetic-energy operator (6) represents *different* dynamical systems depending on the particular choices for the constants α, β, and γ.

The lack of uniqueness of theories with position-dependent effective masses may be proved most conveniently by using the formalism developed by Gora and Williams[3] and later expatiated on by van Vliet and Marshak.[4] The reader is referred to van Vliet and Marshak's paper for details.[4] Just as originally done by Slater[1] for homogeneous material, the wave function of the total system is expanded in Wannier functions representing solutions to a homogeneous system on which is imposed a "perturbation"

$$\mathscr{L}(\vec{r})H_1(\vec{r}) \ , \tag{29}$$

where $\mathscr{L}(\vec{r})$ is a measure of the nonuniformity and is supposed to be slowly varying, whereas H_1 is the difference of crystal-potential energies of the two constituents. As example a binary alloy was taken just as in Ref. 5. In the course of the derivation of an effective-mass—type equation there appears a matrix element [Eq. (2.9) of Ref. 4]

$$M = \int W_\beta^*(\vec{r}-\vec{l})\mathscr{L}(\vec{r})H_1(\vec{r})W_{\beta'}(\vec{r}-\vec{l}')d^3r \ . \tag{30}$$

Here $W_\beta(\vec{r}-\vec{l})$ are Wannier functions of band β localized at the lattice site $\vec{l}$. Since $\mathscr{L}(\vec{r})$ is slowly varying, it can be taken out of the integral. The choice made in Refs. 3 and 4 was to put

$$M \approx \tfrac{1}{2}[\mathscr{L}(\vec{l})+\mathscr{L}(\vec{l}')] \times \int W_\beta^*(\vec{r}-\vec{l})H_1(\vec{r})W_{\beta'}(\vec{r}-\vec{l}')d^3r \ . \tag{31}$$

If we now expand the Wannier functions into Bloch functions, following Ref. 4 closely, we obtain

$$M \approx \frac{1}{2N}[\mathscr{L}(\vec{l})+\mathscr{L}(\vec{l}')]\sum_k e^{i\vec{k}\cdot(\vec{l}-\vec{l}')}\delta_{\beta\beta'}S_\beta(\vec{k}) \ . \tag{32}$$

With the use of the Fourier series over the direct lattice,

$$S_\beta(\vec{k}) = \sum_L S_\beta(\vec{L})e^{i\vec{k}\cdot\vec{L}} \ , \tag{33}$$

we find for the matrix element M,

$$M \approx \tfrac{1}{2}[\mathscr{L}(\vec{l})+\mathscr{L}(\vec{l}')]\delta_{\beta\beta'}S_\beta(\vec{l}'-\vec{l}) \ . \tag{34}$$

The reader is referred to Ref. 4 for details. One of the terms in Schrödinger's equation for the envelope function F_β as derived in Ref. 4 is given by

$$\sum_{\beta',l'} F_{\beta'}(\vec{l}')M = \sum_{l'} S_\beta(\vec{l}')\tfrac{1}{2}[\mathscr{L}(\vec{l})+\mathscr{L}(\vec{l}+\vec{l}')]F_\beta(\vec{l}+\vec{l}') \ . \tag{35}$$

The rhs of Eq. (35) follows with the aid of Eq. (34). Equation (35) can, in turn be written, making $\vec{l}$ the continuous variable $\vec{r}$,

$$\sum_{\beta',l'} F_{\beta'}(\vec{l}')M = \tfrac{1}{2}\sum_{\vec{l}} S_\beta(\vec{l})[\mathscr{L}(\vec{r})e^{i\vec{l}\cdot(-i\vec{\nabla})} + e^{i\vec{l}\cdot(-i\vec{\nabla})}\mathscr{L}(\vec{r})]F_\beta(\vec{r}) \ . \tag{36}$$

Expanding the exponentials in Eq. (36) up to second order, the term (36) leads ultimately to a kinetic-energy operator of the type exhibited by Eq. (4) as shown in detail by van Vliet and Marshak.[4]

After these preliminaries it is fairly simple to demonstrate that the approximation scheme sketched on these pages leads to ambiguities. We apologize to the reader for the necessarily rather short review of the formalism involved. The reader is urged to consult Ref. 4 for details.

The transition from Eq. (30) to Eq. (31) is ambiguous. In fact, in their first paper on this subject Gora and Williams approximated matrix element (30) by[7]

$$M \approx \mathscr{L}(\vec{l}) \int W^*_\beta(\vec{r}-\vec{l}) H_1(\vec{r}) W_{\beta'}(\vec{r}-\vec{l}\,')d^3r \,, \tag{37}$$

which led to a non-Hermitian, position-dependent effective-mass equation. But there are many other ways to approximate matrix element (30) with equal justification as with the choice of (31). Just to give one other example, we might think that

$$M \approx \mathscr{L}(\tfrac{1}{2}(\vec{l}+\vec{l}\,')) \times \int W^*_\beta(\vec{r}-\vec{l}) H_1 W_{\beta'}(\vec{r}-\vec{l}\,')d^3r \tag{38}$$

constitutes another approximation as good as the choice of (31). Here the vector $\frac{1}{2}(\vec{l}+\vec{l}\,')$ lies midway between the lattice sites $\vec{l}$ and $\vec{l}\,'$, and since $\mathscr{L}$ is assumed to be slowly varying the choice of (38) seems to be a viable alternative to the choice of (31) and should therefore lead to the same result. But if we now retrace the steps leading from Eq. (31) to Eq. (36) with the new approximate matrix element (38), we find instead of Eq. (36) that

$$\sum_{\beta',l'} F_{\beta'}(\vec{l}\,')M = \sum_{\vec{l}} S_\beta(\vec{l})\exp\{i\vec{l}\cdot[-(i/2)\vec{\nabla}]\}\mathscr{L}(\vec{r})\exp\{i\vec{l}\cdot[-(i/2)\vec{\nabla}]\}F_\beta(\vec{r}) \,. \tag{39}$$

Again expanding the exponentials to second order we are led to an effective-mass equation with a kinetic-energy operator of type

$$T = -\frac{\hbar^2}{2}\vec{\nabla}(m^{-1}\vec{\nabla}) \,, \tag{40}$$

and we know from analysis given in the begininng of this section that operator (40) is *not* equivalent to operator (4), hence the approximation method of Gora and Williams leads to ambiguous results.

On the preceding pages we have shown that (a) Hamiltonians representative of carriers possessing position-dependent masses are not Galilean invariant, and (b) approximation schemes which lead to position-dependent mass equations are not unique. Therefore, the concept of a position-dependent mass should be abandoned. Previously we have shown how to circumvent the difficulties associated with position-dependent masses. The reader is referred to those papers for details.[5]

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APPENDIX

Here we wish to show that although position-dependent effective-mass equations are not Galilean invariant, nevertheless it is possible to give a unique meaning to such approximations. Suppose that we start out adopting a particular coordinate system $\vec{r},t$. In this coordinate system the Schrödinger equation for an electron within the one-electron approximation is given by

$$H\psi(\vec{r},t) = i\hbar\dot{\psi}(\vec{r},t) \,, \tag{A1}$$

where H signifies the Hamiltonian consisting of the kinetic energy, the potential energy of the unperturbed periodic lattice, and a point-symmetry—breaking perturbation, due to an external electric field, for instance.

Expanding the wave function ψ, either into Bloch functions or Wannier functions,[1,2] it becomes possible to derive an effective-mass equation by the method of Gora and Williams[3] which exhibits a kinetic-energy operator of the type of Eq. (4) of the text. We know from Sec. II that the effective-mass equation is not Galilean invariant, whereas the original equation [Eq. (A1)] is. The approximation leading to position-dependent effective masses broke the symmetry of Galilean invariance. It is therefore not possible to go on to another equivalent coordinate system via the transformation (9) of the text without encountering inconsistencies as far as the position-dependent effective-mass equation is concerned. However, if we first perform transformation (9), now applied to Eq. (A1), we obtain

$$\psi(\vec{r},t)\rightarrow\psi'(\vec{r}\,',\vec{t}\,')\ , \tag{A2}$$

where

$$\psi'(\vec{r}\,',t')=e^{if(\vec{r}\,',t')}\phi(\vec{r}\,',t')\ , \tag{A3}$$

with f given by Eq. (20) of the text. The wave function ϕ now satisfies

$$H'\phi(\vec{r}\,',t')=i\hbar\dot{\phi}(\vec{r}\,',t')\ . \tag{A4}$$

The Hamiltonian H' is obtained from the Hamiltonian H of Eq. (A1) merely replacing $\vec{r}$ and t with $\vec{r}\,'$ and t'. We may now again use the approximation scheme of Gora and Williams,[3] but this time starting with Eq. (A4) rather than Eq. (A1), and we are thus lead to a position-dependent effective-mass equation which is identical with the equation previously obtained, where $\vec{r}$ and t are replaced by $\vec{r}\,'$ and t'. In this sense, position-dependent effective-mass equations are indirectly Galilean invariant. But once a coordinate system is chosen from all possible equivalent systems, connected via transformations of the type of Eq. (9), the choice of another seemingly equivalent coordinate system is not possible anymore. In this sense, the adopted coordinate system may be called a preferred coordinate system.

In view of this disagreeable feature and in view of the ambiguities associated with position-dependent effective-mass equations, the very concept of position-dependent masses should be avoided.

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