Reply to "Comment on 'Existence of Wannier-Stark localization'"

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(Received 10 July 1989)

As Kleinman claims in the preceding Comment, our work indicates that the staircase potential mixes *Bloch states* of different energy bands. However, Wannier-Stark states are superpositions of Bloch states. There is no contradiction with the argument of our work that coherence effects result in each electric-field-dependent energy band forming its own set of Wannier-Stark states.

The nature of the eigenstates of an electron in a one-dimensional periodic potential under the application of a spatially and temporally constant electric field is a long-standing controversial question. It is widely accepted that the application of an electric field leads to the formation of bulk eigenstates that are localized, "Wannier-Stark states," if the basis functions upon which the eigenstates are constructed are limited to the field-free Bloch eigenstates of a single energy band. The controversy centers on whether or not the existence of Wannier-Stark eigenstates transcends this single-band approximation.

The approach of Ref. 1 is based on the observation that the application of a constant electric field to a periodic potential has two distinct effects: The electric field both (i) equivalently alters the shape of each potential well and (ii) inequivalently shifts the energy of each potential well. The first effect retains the periodicity of the potential while the second effect does not. Thus, the first effect only produces a translationally invariant intrasite polarization. However, the second effect leads to the shifting of charge between sites (intersite polarization). An eigenstate with charge accumulated about a given site is a Wannier-Stark localized state.

The eigenstates of a deformed potential well are generally mixtures of all of the eigenstates of an undeformed potential well. Thus, the intrasite polarization is associated with matrix elements of the electric field energy $-qEx$ between states of all electric-field-free energy bands. Here q and x are the carrier's charge and position, respectively, and E is the electric field strength.

Reference 1 adopts an approach to the study of Wannier-Stark localization that incorporates intrasite polarization into the zeroth-order Hamiltonian. In particular, the electric field perturbation $-qEx$ is written as a sum of a periodic sawtooth function and a nonperiodic staircase like function with length scales equal to the primative lattice constant (Fig. 2 of Ref. 1). This sawtooth function produces an equivalent electric-field-induced deformation of each potential well. The staircase function inequivalently shifts the potential energy of each potential well. The eigenstates of an electron in the periodic potential comprising the solid's periodic potential plus the sawtooth periodic potential are electric-field-dependent Bloch states of energy $\varepsilon(E;n,k)$ denoted by $|E;n,k\rangle$, where n is the band index and k is the wave vector.

An eigenstate of energy ε_m of an electron in a periodic potential under the influence of a constant electric field is then expressed as

$$\psi_m(E;x)=\sum_{n',k'} A_m(E;n',k')|E;n',k'\rangle \ , \tag{1}$$

where

$$[\varepsilon(E;n,k)-\varepsilon_m]A_m(E;n,k) = qEa\sum_{n',k'}\langle E;n,k|S(x,a)|E;n',k'\rangle A_m(E;n',k') \ , \tag{2}$$

and $S(x,a)$ is the staircase function with step length equal to the lattice constant a and step height equal to unity. For comparison with Eq. (2), it is noted that in the single-band ($n=n'$) treatment of Wannier-Stark localization with electric-field-free Bloch states as a basis, the expansion coefficients obey[2]

$$[\varepsilon(0;n,k)-\varepsilon_m]A_m(0;n,k)=iqE\,\partial A_m(0;n,k)/\partial k \ . \tag{3}$$

In obtaining this result, matrix elements of the operator x between Bloch states of the same band are expressed in the crystal momentum representation as $-i\partial(\delta_{k,k'}\delta_{n,n'})/\partial k$ with $\delta_{k,k'}$ and $\delta_{n,n'}$ being Kronecker deltas.[3,4] With appropriate boundary conditions, solution of Eq. (3) yields the Wannier-Stark eigenstates.[2,5] Each Wannier-Stark state is a localized state in which charge accumulates about a different site. An analogous equation to Eq. (3) will be obtained for each electric-field-dependent band in the multiband problem if each such band forms its own set of Wannier-Stark states.

The matrix element of the staircase potential between Bloch states of a linear chain of N sites is evaluated in Ref. 1 [Eqs. (16) and (18)]:

$$\langle E;n,k|S(x,a)|E;n',k'\rangle = I(n,k;n',k')\sum_{s=0}^{N-1} s\exp[i(k'-k)sa] \ , \tag{4}$$

where s is an integer, and $I(n,k;n',k')$ is the overlap integral of two Bloch states within the first primitive cell $\{=\int_0^a dx \exp[i(k'-k)x]u^*_{E;n,k}(x)u_{E;n',k'}(x)\}$. The interband matrix elements vanish when $k=k'$ since $I(n,k;n',k)=\delta_{n,n'}/N$. However, the s summation on the right-hand side of Eq. (4) does not generally vanish for $k\neq k'$ since the preexponential factor of s in the s summation destroys the coherence that would otherwise cause the s summation to yield $\delta_{k,k'}$. Thus, the staircase potential generally has both interband and intraband matrix elements *between Bloch states.* These matrix elements vary strongly with $k'-k$. In fact, by itself, the factor arising from the s summation of Eq. (4) displays singular behavior: $\propto(k'-k)^{-1}$ for $(k'-k)a \ll 1$.

We now consider whether the electronic eigenstates in the presence of the electric field are superpositions of Wannier-Stark states of different electric-field-dependent energy bands. That is, we examine the solutions of Eq. (2). We observe that matrix elements of the staircase potential between Bloch states enter as terms in summations on the right-hand side of Eq. (2). Because these matrix elements enter in summations over k', the existence of interband matrix elements of the staircase potential between Bloch states does not necessitate the eigenstates being superpositions of Wannier-Stark states of different electric-field-dependent energy bands.

To address the solution of Eq. (2), the right-hand side of Eq. (4) is first manipulated to a more useful form:

$$\begin{aligned}\langle E;n,k|S(x,a)|E;n',k'\rangle &= (-i/a)I(n,k;n',k')\left[\partial\sum_{s=0}^{N-1}\exp[i(k'-k)sa]/\partial k'\right]\\ &= (-i/a)\partial\left[\sum_{s=0}^{N-1}\exp[i(k'-k)sa]I(n,k;n',k')\right]\partial k'\\ &= (-i/a)\partial\langle E;n,k|E;n',k'\rangle/\partial k' \ . \end{aligned} \tag{5}$$

To obtain the second equality of Eq. (5), it is noted that

$$\sum_{s=0}^{N-1}\exp[i(k'-k)sa]=N\delta_{k,k'} \tag{6a}$$

and

$$\partial I(n,k;n',k')/\partial k'|_{k'=k}=0 \ , \tag{6b}$$

where Eq. (6b) is established in Appendix A of Ref. 1 with the arbitrary phase factor of the Bloch state being chosen so that Eq. (6b) is satisfied when $n=n'$. The final quality of Eq. (5) is obtained by expressing the overlap of different Bloch states as a sum of the contributions from each cell of the periodic structure.

To understand Eq. (5), note that the product of the lattice constant a and our staircase potential is the operator that locates the primitive cell occupied by the carrier. Thus, Eq. (5) restates the result that the cellular-position operator (R of Ref. 5) is $-i\partial/\partial k'$.[6]

The issue of Wannier-Stark localization is addressed by inserting Eq. (5) into Eq. (2). Treating k' as a continuous variable and integrating by parts over k', we obtain

$$\begin{aligned}[\varepsilon(E;n,k)-\varepsilon_m]A_m(E;n,k) &= -iqE\sum_{n',k'}(\partial\langle E;n,k|E;n',k'\rangle/\partial k')A_m(E;n',k')\\ &= iqE\sum_{n',k'}\langle E;n,k|E;n',k'\rangle\partial A_m(E;n',k')/\partial k'\\ &= iqE\sum_{n',k'}\delta_{k,k'}\delta_{n,n'}\partial A_m(E;n',k')/\partial k' = iqE\,\partial A_m(E;n,k)/\partial k \ . \end{aligned} \tag{7}$$

The third equality of Eq. (7) results from the orthonormality of the Bloch states. Thus, comparison with Eq. (2) indicates that the states of each electric-field-dependent energy band form their own Wannier-Stark ladder. In other words, as noted previously,[1,7] the eigenstates that result when the staircase potential is added to a periodic potential are sets of Wannier-Stark states with each set being associated with a different band of eigenstates in the absence of the staircase potential.

Thus, there is no disagreement between our work[1] and Kleinman's claim that there are matrix elements of the staircase potential between *Bloch* states of different energy bands. In particular, Wannier-Stark states are superpositions of Bloch states. Here and in Ref. 1 we find that coherence effects result in each eigenstate of the sum of a periodic potential and the staircase potential being a Wannier-Stark state that is associated with a single band of eigenstates of the periodic potential alone.

Here and in the crystal momentum representation k and k' are treated as if they were continuous.[2-6] In particular, derivatives are written with respect to k and k'. However, k and k' represent discrete points with the

minimum separation between successive values being $2\pi/Na$. Thus, one cannot always write $\partial f(k)/\partial k = \lim_{\Delta k \to 0}\{[f(k+\Delta k)-f(k)]/\Delta k\}$ since Δk is not permitted to vanish. Nontheless, Δk can become arbitrarily small as $N \to \infty$. However, this condition is not sufficient to justify treating k as continuous. For example, if $f(k) = \exp(ikx)$ one has that $[f(k+\Delta k)-f(k)]/\Delta k = f(k)[\exp(i\,\Delta k\, x)-1]/\Delta k$ is only equal to $ixf(k)$ if $x\,\Delta k \ll 1$. This condition cannot generally be met if x is unbounded, since then x can reach a value $\simeq Na$ for which $x\,\Delta k$ is not $\ll 1$. However, if x is a bounded function, as would be the case with a localized eigenstate, this differentiation is justified. Thus, since the Wannier-Stark states are localized, our treatment is self-consistent.

Finally, we note that, since the Wannier-Stark states are localized, dc charge transport requires transitions (e.g., via phonon scattering) between Wannier-Stark states.[5,6,8] When, as is usually the case, the lifetime broadening of the Wannier-Stark states exceeds the energy separation between adjacent Wannier-Stark levels of a given band qEa, the Bloch states, rather than the Wannier-Stark states, are the convenient zeroth-order states for the study of electronic transport. In particular, the theory of interband transitions between Wannier-Stark states (Zener tunneling) is carried out under the presumption that the lifetime broadening for a carrier in a Wannier-Stark state exceeds qEa.[2]

This work was supported by United States Department of Energy Contract No. DE-AC04-76DP00789.

[1]D. Emin and C. F. Hart, Phys. Rev. B **36**, 7353 (1987).
[2]E. O. Kane, J. Phys. Chem. Solids **12**, 181 (1959).
[3]R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).
[4]E. I. Blount, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1962), Vol. XIII, p. 305.
[5]G. H. Wannier, *Elements of Solid State Theory* (Cambridge University, Cambridge, London, 1959), pp. 190–193.
[6]P. W. Anderson, *Concepts in Solids* (Addison-Wesley, New York, 1963), pp. 82–95.
[7]J. Zak. Phys. Lett. **76A**, 287 (1980).
[8]D. Emin and C. F. Hart, Phys. Rev. B **36**, 2530 (1987).