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Relativistic tight-binding approximation method for materials immersed in the magnetic field

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We develop a new tight-binding (TB) approximation method that enables us to calculate electronic structures of materials immersed in the uniform magnetic field. Since the present method is based on the Dirac equation, relativistic effects are inherently taken into account. Due to the Zeeman effect and the translational property of the wave function in the uniform magnetic field, matrix elements of the Hamiltonian explicitly depend on the magnetic field. Relativistic TB parameters, which are needed for actual calculations, can be evaluated by using results of relativistic energy-band calculations of zero magnetic field as the reference data.

Key words: tight-binding approximation, relativistic effect, magnetic field, silicon vacancy

1. INTRODUCTION

Recently, the softening in elastic constants of silicon, which is observed in ultrasonic experiments, has attracted much attention because the density of silicon vacancies can be evaluated from it [1]. It is also reported that the elastic softening is suppressed by an external magnetic field in the boron-doped silicon [1]. Several papers concerning such phenomena have been published so far, and it is pointed out that the spin-orbit interaction plays an important role on them as well as the Zeeman effect [2-5].

In order to reveal the mechanism of the suppression of the elastic softening, it is desirable to develop the energy-band calculation method that takes into account effects of both the relativity and magnetic field. However, the method considering such both effects has not yet been proposed so far. For example, in the previous work by Hofstadter [6], the tight-binding (TB) approximation method for Bloch electrons in the magnetic field is constructed by employing the so-called Peierls substitution. Relativistic effects are not taken into account in the formulation [6]. Furthermore, this method is applicable only to simple models such that the energy band of zero magnetic field is given in an analytical form [6].

In this paper, we propose the relativistic TB approximation method that is applicable to electronic structure calculations of materials immersed in the uniform magnetic field. By treating the Dirac equation for the electron that moves in both the uniform magnetic field and periodic potential of the crystal, the spin-orbit interaction and Zeeman effect are inherently included in the present method.

Organization of this paper is as follows. In Sec. 2, we present a new formulation of the relativistic TB approximation method, in which the relativistic atomic orbitals in the uniform magnetic field are used as basis functions. In Sec. 3, we explain how to calculate relativistic hopping integrals that appear in the expression for matrix elements of the Hamiltonian. Finally, some concluding remarks are given in Sec. 4.

2. RELATIVISTIC TIGHT-BINDING APPROXIMATION METOD IN THE UNIFORM MAGNETIC FIELD

The Dirac equation for the electron that moves in the uniform magnetic field and periodic potential of the crystal is given by [7]

$$H\Phi_\alpha(\boldsymbol{r}) = E_\alpha \Phi_\alpha(\boldsymbol{r}) \tag{1}$$

with

$$H = c\boldsymbol{\alpha}\cdot\left\{\boldsymbol{p} + e\boldsymbol{A}(\boldsymbol{r})\right\} + \beta mc^2 + \sum_i v_a\left(\boldsymbol{r} - \boldsymbol{R}_i\right), \tag{2}$$

where $\boldsymbol{A}(\boldsymbol{r})$ and $v_a\left(\boldsymbol{r} - \boldsymbol{R}_i\right)$ denote the vector potential of the uniform magnetic field and scalar potential of the nucleus that is located at $\boldsymbol{R}_i$, respectively. In this study, we suppose that the uniform magnetic field $\boldsymbol{B}$ is along the z-axis, and that the symmetric gauge is employed for $\boldsymbol{A}(\boldsymbol{r})$, i.e.,

$$\boldsymbol{A}(\boldsymbol{r}) = \frac{1}{2}\boldsymbol{B}\times\boldsymbol{r}\ . \tag{3}$$

In the present method, the four-component wave function $\Phi_\alpha(\boldsymbol{r})$ is expanded by using as basis functions relativistic atomic orbitals in the uniform magnetic field. If we denote the basis function by $\psi^{\xi}_{\boldsymbol{R}_n}(\boldsymbol{r})$, then we have

$$\Phi_\alpha(\boldsymbol{r}) = \sum_{\xi}\sum_i C^{\xi,\alpha}_{\boldsymbol{R}_i}\psi^{\xi}_{\boldsymbol{R}_i}(\boldsymbol{r}), \tag{4}$$

where $\psi^{\xi}_{\boldsymbol{R}_i}(\boldsymbol{r})$ obeys the following Dirac equation:

$$\left[c\boldsymbol{\alpha}\cdot\left\{\boldsymbol{p}+e\boldsymbol{A}(\boldsymbol{r})\right\}+\beta mc^2+v_a\left(\boldsymbol{r}-\boldsymbol{R}_i\right)\right]\psi^{\xi}_{\boldsymbol{R}_i}(\boldsymbol{r})=\varepsilon^{\xi}_{\boldsymbol{R}_i}\psi^{\xi}_{\boldsymbol{R}_i}(\boldsymbol{r}). \tag{5}$$

Using the translational property of the wave function in the uniform magnetic field [8], and approximating $\psi^{\xi}_{\boldsymbol{R}_i}(\boldsymbol{r})$ by the relativistic atomic orbital of zero magnetic field, we obtain matrix elements of the Hamiltonian as follows:

$$\begin{aligned}H_{i'n'l'J'M',inlJM}\approx&\left(\bar{\varepsilon}_{nlJM}+\frac{eB}{2m}\frac{2J+1}{2l+1}\mathrm{h}M\right)\delta_{\boldsymbol{R}_{i'},\boldsymbol{R}_i}\,\delta_{n'l'J'M',nlJM}\\&+e^{i\frac{e}{\mathrm{h}}\boldsymbol{B}\cdot(\boldsymbol{R}_{i'}\times\boldsymbol{R}_i)}(1-\delta_{\boldsymbol{R}_{i'},\boldsymbol{R}_i})t_{n'l'J'M',nlJM}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})\end{aligned} \tag{6}$$

with

$$\begin{aligned}t_{n'l'J'M',nlJM}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})=&\int\varphi_{n'l'J'M'}(\boldsymbol{r})^{\dagger}\,\frac{v_a(\boldsymbol{r})+v_a(\boldsymbol{r}-\boldsymbol{R}_i+\boldsymbol{R}_{i'})}{2}\\&\times\varphi_{nlJM}(\boldsymbol{r}-\boldsymbol{R}_i+\boldsymbol{R}_{i'})\mathrm{d}^3r.\end{aligned} \tag{7}$$

Here, $\varphi_{nlJM}(\boldsymbol{r})$ and $\bar{\varepsilon}_{nlJM}$ denote the relativistic atomic orbital and energy spectrum of the isolated atom of zero magnetic field, respectively. The subscripts n, l, J and M are the principal, azimuthal, total angular momentum and magnetic quantum numbers, respectively. It is found from Eq. (6) that matrix elements explicitly depend on the magnetic field. The diagonal elements (the first term of Eq. (6)) correspond to the energy spectrum of the isolated atom plus the Zeeman term. The phase factor, which appears in the second term of Eq. (6), also depends on the magnetic field. This dependence comes from the translational property of the wave function [8]. It is also found from Eq. (6) that relativistic effects are included in $\bar{\varepsilon}_{nlJM}$ and the relativistic hopping integral $t_{n'l'J'M',nlJM}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})$. By diagonalizing matrix elements Eq. (6), we can obtain the energy spectrum of materials immersed in the uniform magnetic field.

3. CALCULATIONS OF THE REALTIVISTIC HOPING INTEGRAL

In order to perform actual calculations on the basis of the present method, we need values of $\bar{\varepsilon}_{nlJM}$ and $t_{n'l'J'M',nlJM}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})$. In the case of the common TB approximation method (the non-relativistic and zero magnetic filed case), the hopping integral is calculated by using TB parameters such as $(ss\sigma)$, $(sp\sigma)$, $(pp\sigma)$ and $(pp\pi)$ [9]. In a similar way to this common method, $t_{n'l'J'M',nlJM}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})$ can be expressed in terms of relativistic TB parameters that are defined hereafter. For example, $t_{n0\frac{1}{2}\frac{1}{2},n0\frac{1}{2}\frac{1}{2}}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})$, $t_{n0\frac{1}{2}\frac{1}{2},n1\frac{1}{2}\frac{1}{2}}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})$ and $t_{n0\frac{1}{2}\frac{1}{2},n1\frac{1}{2}-\frac{1}{2}}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})$ are given by

$$t_{n0\frac{1}{2}\frac{1}{2},n0\frac{1}{2}\frac{1}{2}}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})=K(0\tfrac{1}{2},0\tfrac{1}{2})_{1/2}, \tag{8}$$

$$t_{n0\frac{1}{2}\frac{1}{2},n1\frac{1}{2}\frac{1}{2}}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})=q_zK(0\tfrac{1}{2},1\tfrac{1}{2})_{1/2}, \tag{9}$$

and

$$t_{n0\frac{1}{2}\frac{1}{2},n1\frac{1}{2}-\frac{1}{2}}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})=(q_x-iq_y)K(0\tfrac{1}{2},1\tfrac{1}{2})_{1/2}, \tag{10}$$

respectively, where q_x, q_y and q_z denote the direction cosines of the vector $\boldsymbol{R}_i-\boldsymbol{R}_{i'}$ [10]. In Eqs. (8) - (10), $K(0\frac{1}{2},0\frac{1}{2})$ and $K(0\frac{1}{2},1\frac{1}{2})$ are relativistic TB parameters that are defined by

$$\begin{aligned}&K(l'J',lJ)_M\\&=\int\varphi_{nl'J'M}(\boldsymbol{r})^{\dagger}\,\frac{v_a(\boldsymbol{r})+v_a(\boldsymbol{r}-R\boldsymbol{e}_z)}{2}\varphi_{nlJM}(\boldsymbol{r}-R\boldsymbol{e}_z)\mathrm{d}^3r,\end{aligned} \tag{11}$$

where $R=|\boldsymbol{R}_i-\boldsymbol{R}_{i'}|$, and where $\boldsymbol{e}_z$ denotes the unit vector along the z-axis. Relativistic TB parameters can be determined by utilizing the results of relativistic energy-band calculations of zero magnetic field as the reference data, similarly to the case of the common TB approximation method.

The validity of the present method is confirmed by revisiting the Hofstadter butterfly diagram [6]. Namely, if we apply it to a two-dimensional square lattice immersed in the uniform magnetic field, and if we employ only nonrelativistic s-orbitals as $\varphi_{nlJM}(\boldsymbol{r})$ within nearest neighbor atoms, and if we take into consideration the hopping integrals between the nearest neighbor atoms, then matrix elements Eq. (6) coincide with those of Hofstadter's method rigorously [11]. This means that the present method can revisit the Hofstadter butterfly diagram [6].

4. CONCLUDING REMARKS

We develop the relativistic TB approximation method that is applicable to materials immersed in the uniform magnetic field. As compared with the previous work by Hofstadter [6], the present method has two advantages. One is that it is applicable to materials where relativistic effects emerge definitely. Another one is that the present method is applicable not only to a simple model but also to various kinds of realistic materials, while the application of Hofstadter's method is limited to a simple model such as the s-band model. Due to these desirable features, it is expected that the present method would be useful to not only the first-principles description of the magnetic Bloch electron but also the above-mentioned problem of the boron-doped silicon [1], which is the next issue.

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References

[1] T. Goto, H. Yamada-Kaneta, Y. Saito, Y. Nemoto, K. Sato, K. Kakimoto and S. Nakamura, *J. Phys. Soc. Jpn.*, **75**, 044602 (2006).
[2] H. Matsuura and K. Miyake, *J. Phys. Soc. Jpn.*, **77**, 043601 (2008).
[3] T. Yamada, Y Yamakawa and Y. Ono, *J. Phys. Soc. Jpn.*, **78**, 054702 (2009).
[4] T. Ogawa, K. Tsuruta and H. Iyetomi, *Solid State Comm.*, **151**, 1605 (2011).
[5] S. Baba, T. Goto, Y. Nagai, M. Akatsu, H. Watanabe, K. Mitsumoto, T. Ogawa, Y. Nemoto and H. Yamada-Kaneta, *J. Phys. Soc. Jpn.* ,**80**, 094601 (2011).
[6] D. R. Hofstadter, Phys. Rev. B., **14**, 2239 (1976).
[7] A. Messiah, "Quantum Mechanics", North-Holland, Amsterdam (1966) Chap. 20.
[8] The translational property can be obtained from the fact that $\boldsymbol{A}(\boldsymbol{r})$ and $\boldsymbol{A}(\boldsymbol{r}-\boldsymbol{R}_i)$ yield the same magnetic field $\boldsymbol{B}$. For example, see E. Brown, *Phys. Rev.*, **133**, A1038 (1964).
[9] J. C. Slater and G. F. Koster, *Phys. Rev.*, **94**, 1498 (1954).
[10] The expressions for $t_{n'l'J'M',nlJM}(\boldsymbol{R}_i-\boldsymbol{R}_{i'})$ such as Eqs. (8) - (10) can be derived with the aid of Table I of Ref. 9 (the Slater-Koster table).
[11] It should be noted that the Zeeman term in Eq. (6) vanishes in the nonrelativistic limit because only nonrelativistic s-orbitals are employed as $\varphi_{nlJM}(\boldsymbol{r})$. Furthermore, in the nonrelativistic limit, the hopping integrals can be calculated through Table I of Ref. 9. Thus, Eq. (6) corresponds to the first equation (unnumbered) of Hofstadter's paper[6].

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