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Fractional dimensionality and fractional derivative spectra of interband optical transitions

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The model of fractional-dimensional space is used to study optical properties associated with electron interband transitions near van Hove critical points in anisotropic systems. Bloch electrons in an anisotropic solid are treated as an isotropic fractional-dimensional free gas, where the dimension is determined by the degree of anisotropy. Density of states and optical spectra are obtained as a function of spatial dimension. Fractional derivative spectra (FDS) are developed for analyzing the dimensionality of solids from the measured interband optical spectra in van Hove singularity regions. Using fractional differentiation, the dimension, as well as critical-point parameters, of a solid is straightforwardly determined from the derivative order that yields a symmetric line shape, Lorentzian or its derivative, in FDS. The fractional dimension determined by FDS is related to the anisotropic electron-lattice interactions and quantitatively describes the degree of anisotropy.

I. INTRODUCTION

Dimensionality has been a subject of considerable interest as it plays an important role in physical properties of condensed matter. Many novel phenomena observed in low dimensions revealed that the influence of restricted geometry is of paramount importance to our understanding, and this has stimulated recent theoretical and experimental achievements on low-dimensional physics.[1] One of the most interesting phenomena may be the continuous changing of dimensionality. However, although such transitions or crossovers have been observed experimentally, e.g., with the changes of monolayer coverage,[2] wire thickness,[3] or even temperature,[4] this dimensional behavior was described only qualitatively.

In this paper, a fractional-dimensional model is employed to study electron interband optical transitions near a van Hove critical point (CP). Bloch electrons in an anisotropic solid are treated as a fractional-dimensional free gas, where the dimension is determined by the degree of anisotropy. Density of states and optical spectra near a CP are obtained as a function of spatial dimension. It is shown that the dimension, in general being a fraction, as well as CP parameters of a solid can be determined directly from the measured optical spectra associated with electron interband transitions using the fractional derivative spectra (FDS) method. The FDS method provides a quantitative measure of the anisotropy of solids and it turns out that optical spectroscopy, such as ellipsometry or modulation measurements, is a potentially efficient technique for probing electron dimensional behavior in solids.

II. DENSITY OF STATES: FRACTIONAL-DIMENSIONAL SPACE MODEL

The model of fractional-dimensional space rests on the following assumption:[5] Excitations in an anisotropic solid can be treated as ones in an isotropic fractional-dimensional space, where the dimension is determined by the degree of anisotropy of the solid. In other words, the anisotropic interaction in a three-dimensional (3D) space becomes the isotropic one in a lower fractional-dimensional space.

To the first approximation, Bloch electrons in an anisotropic solid are considered as an isotropic α-dimensional (αD) gas, where $0 \le \alpha \le 3$. The density of states can then be calculated using αD mode counting.[6,7] For a minimum gap, the electron energy E is $E-E_g=(\hbar^2/2m_{vc})k^2$, where E_g is the band gap and m_{vc} and k are the reduced effective mass and wave vector of the electron, respectively. It can be demonstrated[5] that the corresponding joint density of states (JDOS) for a solid of unit volume $G_e(E)$ is given by

$$G_e(E)=\frac{2}{\Gamma(\alpha/2)}\left[\frac{m_{vc}}{2\pi\hbar^2}\right]^{\alpha/2}(E-E_g)^{\alpha/2-1}\ , \tag{1}$$

for $E \ge E_g$ and zero elsewhere, where $\Gamma(x)$ is a gamma function. Expressions for other types of CP's can also be obtained similarly. When $\alpha=0$, 1, 2, and 3, Eq. (1) yields the familiar dependencies obtained previously using integer dimension models, i.e., $G_e(E)\sim(E-E_g)^{-1}$ for 0 D (δ function), $G_e(E)\sim(E-E_g)^{-1/2}$ for 1 D, $G_e(E)\sim$const for 2D, and $G_e(E)\sim(E-E_g)^{1/2}$ for 3D systems.

III. INTERBAND OPTICAL SPECTRA

The complex dielectric function at photon energy $\hbar\omega$, $\epsilon(\hbar\omega)=\epsilon_1(\hbar\omega)+i\epsilon_2(\hbar\omega)$, due to direct allowed interband transitions near a CP can be expressed as,[8]

$$\epsilon(\hbar\omega)=\frac{C}{(\hbar\omega)^2}\int_{G_e(E)\neq 0}\frac{G_e(E)dE}{E-\hbar\omega-i\Gamma}\ , \tag{2}$$

where $C=|\langle v|\mathbf{a}\cdot\mathbf{p}|c\rangle|^2(e\hbar/m)^2/\epsilon_0$, $G_e(E)$ is the JDOS, and Γ is the phenomenological broadening parameter describing the finite lifetime effect. Using Eq. (1) and the relationship among optical responses of various types of CP's,[9] $i^{r-\alpha}$, where r is the type of CP's, the dielectric function of Eq. (2) becomes

$$\epsilon(\hbar\omega)=\frac{i^{r-\alpha}C_\alpha}{(\hbar\omega)^2}\int_0^{E_g}\frac{(E_g-E)^{\alpha/2-1}dE}{E-\hbar\omega-i\Gamma}, \tag{3}$$

where

$$C_\alpha=2|\langle v|\mathbf{a}\cdot\mathbf{p}|c\rangle|^2(e/m)^2(m_{vc}/2\pi)^{\alpha/2}\hbar^{2-\alpha}/\Gamma(\alpha/2)\epsilon_0 .$$

Comparing Eq. (3) with the definition of the Riemann-Liouville fractional integral gives[10] (see also the Appendix)

$$\epsilon(\hbar\omega)=\frac{i^{r-\alpha}M_\alpha}{(\hbar\omega)^2}\frac{d^{-\alpha/2}}{dE_g^{-\alpha/2}}\frac{1}{E_g-\hbar\omega-i\Gamma}, \tag{4a}$$

where the symbol $d^q/dx^q f(x)$ denotes the differintegral ($q>0$ for derivative and $q<0$ for integral) of function $f(x)$ between limit 0 and x (see the Appendix) and

$$M_\alpha=2|\langle v|\mathbf{a}\cdot\mathbf{p}|c\rangle|^2(e/m)^2(m_{vc}/2\pi)^{\alpha/2}\hbar^{2-\alpha}/\epsilon_0 .$$

The kth derivative of $\epsilon(\hbar\omega)$ ($k=1,2,3,\ldots$) can be obtained by differentiating Eq. (4a),

$$\epsilon^{(k)}(\hbar\omega)=\frac{i^{r-\alpha}M_{\alpha,k}}{(\hbar\omega)^2}\frac{d^{-\alpha/2}}{dE_g^{-\alpha/2}}\frac{1}{(E_g-\hbar\omega-i\Gamma)^{k+1}}, \tag{4b}$$

where $M_{\alpha,k}=k!M_\alpha$. For $\hbar\omega\approx E_g$, the fractional integrals of Eqs. (4a) and (4b) yield the following analytic forms:

$$\epsilon(\hbar\omega)=\frac{i^{r-\alpha}A_\alpha}{(\hbar\omega)^2}\int^{\hbar\omega-E_g+i\Gamma}t^{\alpha/2-2}dt \tag{5a}$$

and

$$\epsilon^{(k)}(\hbar\omega)=\frac{i^{r-\alpha}A_{\alpha,k}}{(\hbar\omega)^2}(\hbar\omega-E_g+i\Gamma)^{\alpha/2-(k+1)}, \tag{5b}$$

where $A_\alpha=\Gamma(2-\alpha/2)M_\alpha$ and

$$A_{\alpha,k}=(-1)^{k+1}\Gamma(k+1-\alpha/2)M_\alpha \quad (k=1,2,3,\ldots) .$$

In calculations, the prefactors $(\hbar\omega)^{-2}$ in Eqs. (5a) and (5b) can be treated as a constant, E_g^{-2}, to a fair approximation. Equation (5a) gives the dielectric function of an αD solid. When $\alpha=0$, Eqs. (5a) and (5b) are, respectively, Lorentzian line shapes and their derivatives, representing the optical spectra of 0D structure.[11]

Equations (5a) and (5b) with integer dimensions were used widely for analyzing the electronic structure of 1D, 2D, and 3D solids, e.g., in modulation or ellipsometry spectroscopy.[9,12] As the structure observed in optical spectra is attributed to the interband CP's related to the regions of large or singular JDOS, direct information on band gap E_g and broadening parameter Γ was obtained by fitting the measured derivative spectra to the theoretical expressions if the dimension of a solid was assumed.[13,14] It is not too surprising that the fractional-dimensional approach indicates that Eqs. (5a) and (5b) are also applicable to noninteger dimensionalities.

The fractional-dimensional expressions manifest themselves versatile in the treatment of optical properties. Many systems such as chalcogenides have dot structure, polymers have chainlike, and laminar compounds have layered structure.[15] The observed features of such systems were usually interpreted, respectively in terms of 0 D, 1D, or 2D models. From a microscopic point of view, low-dimensional materials are those with highly anisotropic structure and despite their weak effects, interactions among dots, chains, or layers should be taken into account. Such anisotropic effects now can be described by a fractional-dimension α and, consequently, the optical spectra at CP's are figured as easily as those of an isotropic one. By varying α in between 0, 1, 2, and 3 in Eqs. (5a) and (5b), one can examine the evolution of optical responses with the change of crystal anisotropy.

The interband dielectric functions near the minimum CP's of various dimensional solids were calculated using Eq. (5a) with α evolving from 0 to 3 and shown in Fig. 1. Basically, they are in agreement with those obtained using the tight-binding method[16] and effective-mass approximation.[17]

IV. FRACTIONAL DERIVATIVE SPECTRA

By differentiating Eqs. (4a) and (4b) or Eqs. (5a) and (5b) to an order of $\alpha/2$ with respect to $\hbar\omega$, the $(\alpha/2)$th derivative spectra of the interband dielectric function and its kth derivative near an α D CP were found to be

$$(\hbar\omega)^{-2}\frac{d^{\alpha/2}}{d(\hbar\omega)^{\alpha/2}}[(\hbar\omega)^2\epsilon(\hbar\omega)]=\frac{i^rM_\alpha}{(\hbar\omega)^2}\frac{1}{E_g-\hbar\omega-i\Gamma} \tag{6a}$$

and

$$(\hbar\omega)^{-2}\frac{d^{\alpha/2}}{d(\hbar\omega)^{\alpha/2}}[(\hbar\omega)^2\epsilon^{(k)}(\hbar\omega)]=\frac{i^rM_{\alpha,k}}{(\hbar\omega)^2}\frac{1}{(E_g-\hbar\omega-i\Gamma)^{k+1}} . \tag{6b}$$

For $\hbar\omega\approx E_g$, the prefactors $(\hbar\omega)^2$ and $(\hbar\omega)^{-2}$ in Eqs. (6a) and (6b) are relatively insensitive in a CP region and can be treated approximately as constants. Therefore the left-hand sides of Eqs. (6a) and (6b) are the $(\alpha/2)$th derivative of $\epsilon(\hbar\omega)$ and $\epsilon^{(k)}(\hbar\omega)$, respectively.

Presented in the right-hand sides of Eqs. (6a) and (6b) are standard Lorentzian line shapes and their kth derivatives, respectively, which are symmetric with respect to threshold E_g. This provides a direct means for determining the dimension, as well as CP parameters, of a solid from the measured dielectric functions or differential spectra.

A. Analysis of dimensionality

To determine the dimensionality of a solid, fractional differentiation is applied to dielectric or differential spectra. With differentiation to various fractional orders, the symmetric profile, Lorentzian or its derivative, appears in

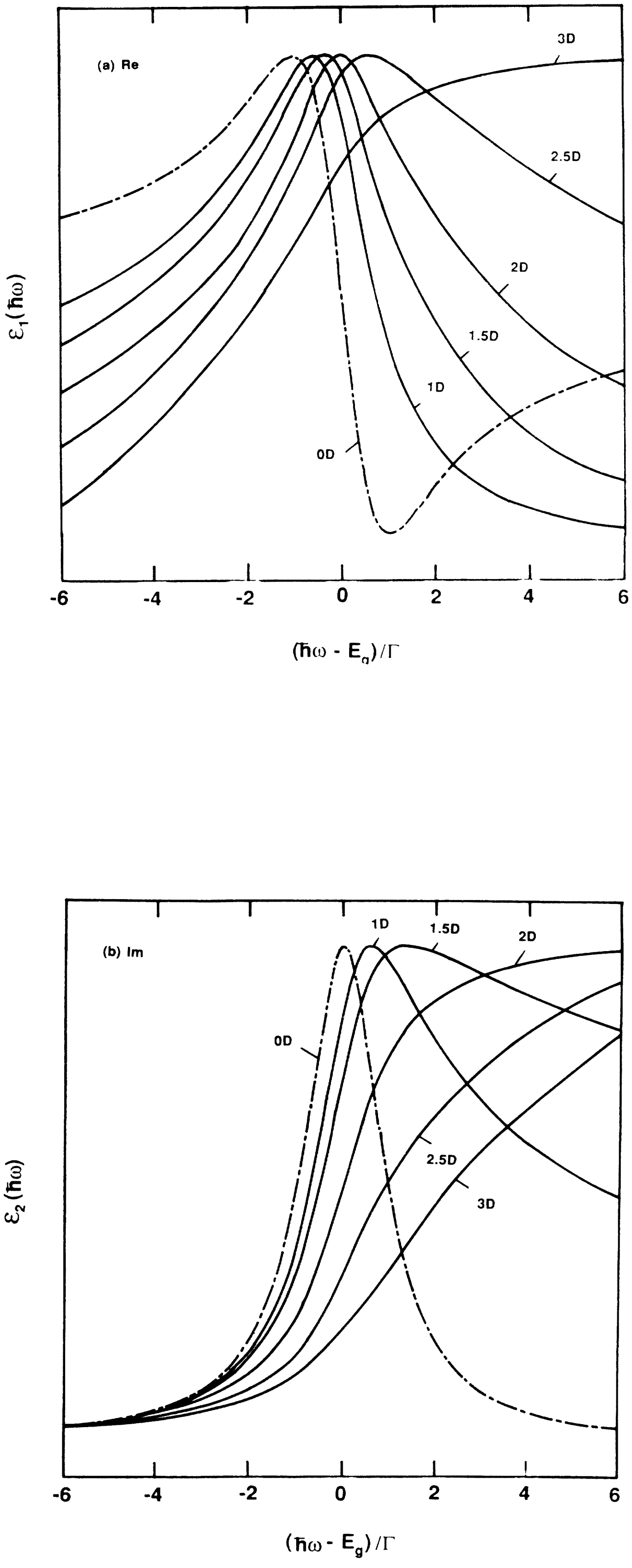

FIG. 1. Interband dielectric functions near the minimum CP's of various dimensional solids obtained using Eq. (5a). (a) Real parts; (b) imaginary parts.

FDS when the derivative order is equal to half of the dimension of a solid, $\alpha/2$. Therefore, the dimension is straightforwardly determined from the derivative order that yields a symmetric line shape in FDS.

Shown in Fig. 2 are the FDS near minimum CP's for various dimensional solids with different derivative orders. Firstly, the dielectric function or the kth differential spectrum of a 1D, 2D, 2.4D or 3D solid was obtained, respectively, by using Eq. (5a) or (5b). The $(x/2)$th differentiations were then calculated using a modified Grünwald formula (see the Appendix). From the line shapes one can clearly see that symmetric profiles appear only when $x=1$, 2, 2.4, or 3, respectively, are the assumed dimensions.

From a practical point of view, FDS are performed preferably on differential spectra, obtained experimentally or numerically. One of the reasons is that the ease of data extrapolation for the differentiation to a noninteger order needs all the data between lower and higher limits, in the case of Eqs. (6a) and (6b), between 0 and $\hbar\omega$. For differential spectra this is easy to be manipulated because contributions far away from E_g can be neglected without significant errors. The other reason is sensitivity, which significantly improves as k increases.

B. Determination of critical point parameters

FDS also provide a direct method for measuring CP parameters, which is the main goal of modulation spectroscopy.[18] In modulation spectra, e.g., wavelength modulation absorption, $\Delta\epsilon \sim d\epsilon d(\hbar\omega)$, or electroreflectance spectra, $\Delta\epsilon \sim d^3\epsilon/d(\hbar\omega)^3$, the line shapes generally are not symmetric. Therefore determining E_g and Γ is somewhat complicated and special techniques, such as the three-points method or Fourier transformation were required.[13,19] Using FDS, however, E_g is easily determined for a single CP for it corresponds to either the extremum or zero point in the resulting symmetric line shape. Because of the symmetric profiles, the broadening parameter is also well estimated even without a line-shape-fitting procedure. Our recent work[20,21] has shown that the FDS method is a more efficient method for analyzing electronic structure compared with conventional technique.[22]

V. DISCUSSION

Fractional dimensionality is a consequence of restrained motion. Electrons in a solid are subject to the forces introduced by the lattice and restricted dynamical behavior may result from the anisotropic electron-lattice interaction. The dimension measured by the FDS method reflects the average dimensionality the electrons experience and provides a quantitative measure of the degree of anisotropy. It is different from that determined based on phonon density of states. The latter is a measure of anisotropic lattice-lattice interaction as reflected in the vibrational properties.[4,23,24] Clearly, the dimensionality depends on the kind of interaction being probed in the measurement and is the intrinsic property of the physical interactions. This manifests that one has to distinguish such space, termed *dynamic space* here, from

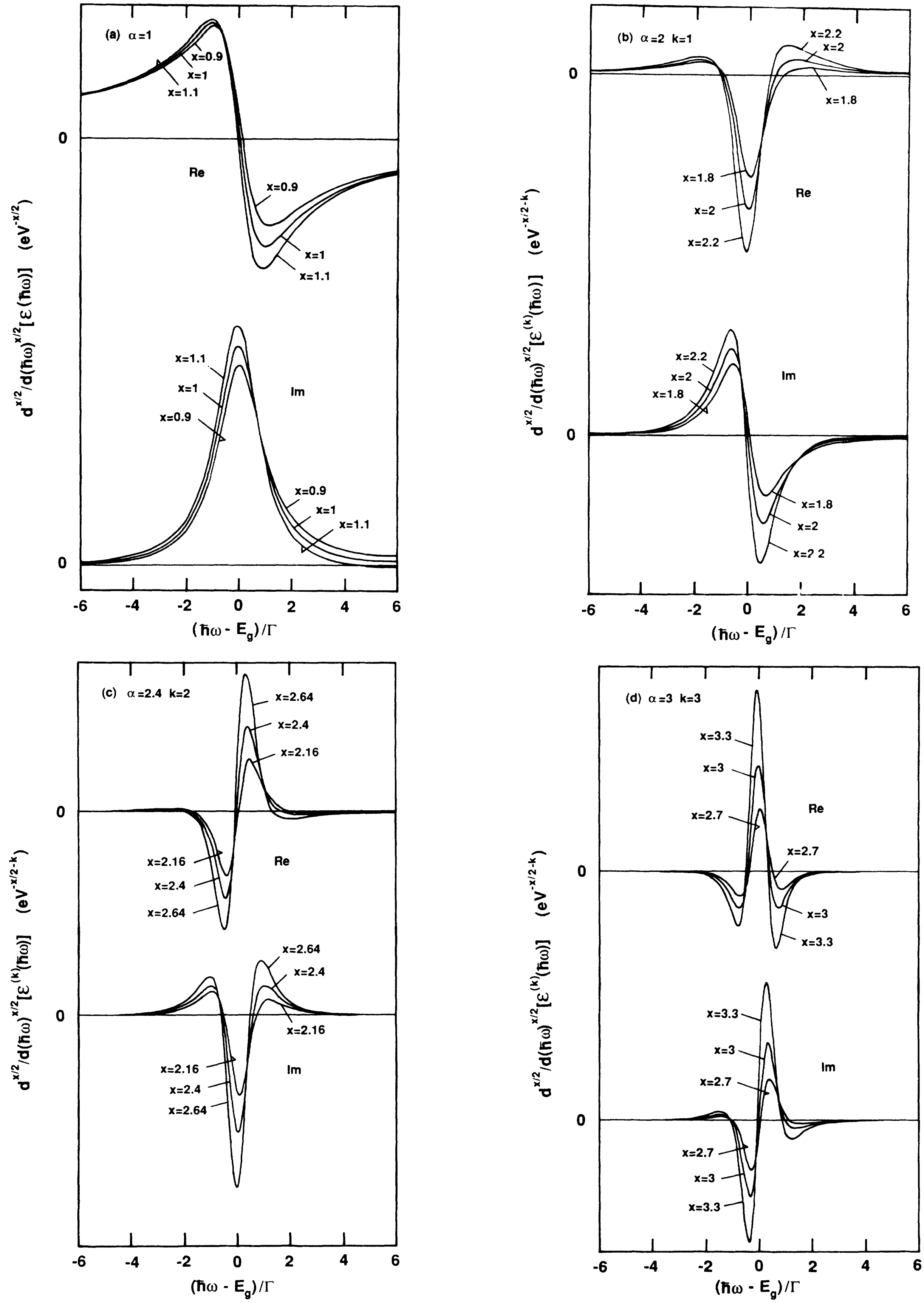

FIG. 2. The $(x/2)$th derivative spectra of the dielectric function or its kth derivative in the vicinities of minimum αD CP's calculated using Eqs. (6a) and (6b). (a) $\alpha=1$; (b) $\alpha=2$, $k=1$; (c) $\alpha=2.4$, $k=2$, and (d) $\alpha=3$, $k=3$. Obviously only when $x=\alpha$, the FDS have symmetric profiles, which are Lorentzians or their kth derivatives as predicted.

that embedding geometric bodies. The latter is the space the geometric volume of a body occupies while the former is the one the motion of a body possesses. In addition, fractional-dimensional behavior discussed here is associated with the extended state, therefore 0D systems imply electron localization.

For low-lying excited states close to the ground state, a solid is well described as the assembly of elementary excitations, which have definite energies and wave vectors. This approach has greatly simplified many-body problems and reduced a complicated interacting system to a near-ideal gas. Using the fractional-dimensional model, the elementary excitation picture becomes more versatile, as an anisotropic solid can be treated, to the first approximation, as an isotropic near-ideal gas in a fractional-dimensional space and statistics is easily employed to derive physical properties of interests, e.g., density of states and the related properties. On the other hand, differential equations can be performed in a fractional-dimensional space, in which one may expect an isotropic potential form for an anisotropic system. We have shown recently that excitons or impurities in an anisotropic solid can be described by the Schrödinger equation in a fractional-dimensional space and eigenvalues and wave functions can be obtained as a function of dimension, as will be discussed elsewhere.[25] All of these would in turn provide physical insights into the concept of spatial dimensionality.

VI. CONCLUSION

Using the fractional-dimensional space model, Bloch electrons in an anisotropic solid are treated as a fractional-dimensional free gas, where the dimension is determined by the degree of anisotropy. Density of states and optical spectra near a CP of an anisotropic solid are obtained as a function of spatial dimension. With fractional differentiation it has been shown that the dimensionality, as well as CP parameters, of a solid can be straightforwardly determined from the derivative order that yields a symmetric Lorentzian or its derivative line shape in FDS associated with interband optical transitions. The fractional dimension measured by FDS reflects *anisotropic electron-lattice interactions* and quantitatively described the degree of anisotropy.

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APPENDIX: FRACTIONAL CALCULUS

1. Symbolism

The Oldham-Spanier terminology[10] is used in this work to denote the fractional differintegral of a function $f(y)$ to an arbitrary order q ($q>0$ for derivative and $q<0$ for integral),

$$\frac{d^q f(x)}{d(x-a)^q} \equiv q\text{th differintegral of } f(y) \text{ with limits } a \text{ and } x \ , \tag{A1}$$

where y is a dummy variable and $f(y)$ is defined on the interval $a \le y \le x$. The lower limit a is a constant so that $d^q f(x)/d(x-a)^q$ is a function of the independent variable x. (For FDS discussed above, $a=0$ and $x=\hbar\omega$).

If q is an integer n or $-n$, (A1) gives an ordinary derivative or integral,

$$\frac{d^n f(x)}{d(x-a)^n} \equiv \frac{d^n f(x)}{dx^n} \quad (\text{when } q=n) \ , \tag{A2}$$

$$\frac{d^{-n} f(x)}{d(x-a)^{-n}} \equiv \int_a^x dx_{n-1} \int_a^{x_{n-1}} dx_{n-2} \cdots \int_a^{x_2} dx_1 \int_a^{x_1} f(x_0) dx_0 \quad (\text{when } q=-n) \ , \tag{A3}$$

where n is a positive integer. When q is a positive integer (A1) becomes a local operator.

2. Definitions

Grünwald gave the most fundamental definition of the differintegral of arbitrary order q (either positive or negative) by the formula

$$\frac{d^q f(x)}{d(x-a)^q} = \lim_{N\to\infty} \left[\frac{[(x-a)/N]^{-q}}{\Gamma(-q)} \sum_{j=0}^{N-1} \frac{\Gamma(j-q)}{\Gamma(j+1)} f\left[x - j\left[\frac{x-a}{N}\right]\right]\right] \ , \tag{A4}$$

where $\Gamma(x)$ is a gamma function. Equation (A4) involves only evaluations of the function itself. Another definition frequently used is the Riemann-Liouville integral for $q<0$,

$$\frac{d^q f(x)}{d(x-a)^q} = \frac{1}{\Gamma(-q)} \int_a^x \frac{f(y)dy}{(x-y)^{q+1}} \quad (q<0) \ . \tag{A5}$$

To replace Eq. (A5) for $q > 0$ one can use

$$\frac{d^q f(x)}{d(x-a)^q} = \frac{d^n}{dx^n}\left[\frac{1}{\Gamma(n-q)}\int_a^x \frac{f(y)dy}{(x-y)^{q-n+1}}\right] \quad (n > q)\ , \tag{A6}$$

where n is again a positive integer.

3. Calculation

The definition of Eq. (A4) also gives a formula for numerical calculation. However, the following modified Grünwald formula permits a more efficient differintegration algorithm:

$$\frac{d^q f(x)}{d(x-a)^q} = \lim_{N\to\infty}\left[\frac{[(x-a)/N]^{-q}}{\Gamma(-q)}\sum_{j=0}^{N-1}\frac{\Gamma(j-q)}{\Gamma(j+1)} f\left\{x-\left[j-\frac{q}{2}\right]\left[\frac{x-a}{N}\right]\right\}\right]\ , \tag{A7}$$

which is based on central difference formulation. One will find that Eq. (A7) is a very useful formula for all operations of fractional calculus. Only for a limited number of functions can their fractional differintegrals be expressed as analytic forms.

It should be noted that when the order is a fraction differentiation is no longer a local operator and it needs all values of $f(y)$ between limits a and x in the calculation. In practice, evaluation at the point $x = a$ should be excluded because of its complexity.

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