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FOR RANDOM SYSTEMS

INCLUDING OFF-DIAGONAL DISORDER

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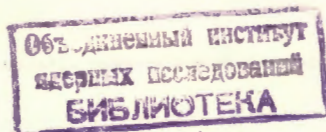
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**SOLVABLE MODEL
FOR RANDOM SYSTEMS
INCLUDING OFF-DIAGONAL DISORDER**

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Точно решаемая модель для неупорядоченных систем

Рассмотрена модель Андерсона для неупорядоченных систем в том случае, когда матричные элементы перекрытия распределены по закону Лоренца. Получено точное выражение для усредненной гриновской функции в предположении, что уровень энергии на каждом узле зависит линейно от интегралов перекрытия. С помощью этого результата обсуждается стабильность аморфных гейзенберговских ферромагнетиков. Изучается модель Уэйра в случае флуктуирующих матричных элементов. Вычисляется плотность состояний энергии.

Препринт Объединенного института ядерных исследований.
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John W., Schreiber J.

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Solvable Model for Random Systems Including
Off-Diagonal Disorder

Anderson's model for disordered systems is considered in the case in which the transfer matrix elements fluctuate according to a Lorentzian distribution. It is shown that the exact ensemble averaged Green function may be obtained if the energy level on each site depends linearly on the overlap integrals. Using this result the stability of amorphous Heisenberg ferromagnets is studied. The Weaire model of an amorphous covalent semiconductor with fluctuating matrix elements is considered. Numerical results for the density of states are given.

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Introduction

Lloyd¹ has shown, that the ensemble-averaged Green function for Anderson's disordered Hamiltonian² can be calculated exactly, if the potential at every site is a random variable with a Lorentzian distribution. Recently there have been obtained similar results for a system of muffin-tin potentials in which the strengths of the potentials are random variables^{3,4}. Also for the case of overlapping potentials with a definite sign the average Green function may be calculated⁵ if a Lorentzian distribution is used.

In the Lloyd model¹ only diagonal disorder is considered. However in a liquid or an amorphous solid off-diagonal disorder is essential. Also a more physical description of disordered alloys must include off-diagonal disorder in order to fulfill the Friedel sum rule^{6,7}. The main concern of this paper is the generalization of the model used by Lloyd to off-diagonal disorder and the multi-band case within a tight-binding description. Of course, in the models^{3,4,5} - in which tight-binding methods are not used - the fluctuation of overlap matrix elements are included automatically. However, recently the problem of extending the CPA to treat off-diagonal disorder has been discussed by several authors using simple tight-binding models⁸⁻¹⁶. We obtained an expression for the average Green function and for the density of states which can be compared with the CPA. Aside from this our tight-binding model can be applied to disordered magnetic systems¹⁷⁻²⁰.

The outline of the paper is as follows. In Section 2 a simple one-band model is considered with a Lorentzian distribution for the transfer matrix elements. It is shown that the model is solvable, if the potential at every site depends linearly on the overlap integrals. The results are discussed in Section 3. Unlike the Lloyd model¹ the off-diagonal disorder involves a state-dependent self energy and therefore a non-symmetric density of states. It is shown, that the model describes an amorphous Heisenberg magnet in the approximation of a lattice model. In Section 4 the Weaire model of an amorphous covalent semiconductor is considered. Using a Lorentzian distribution for the tight-binding parameters the density of states is calculated.

2. One-band model

We consider a system in which an electron moves between sites n , on which it has energy levels ϵ_n . The Hamiltonian describing the electron in this system has the form

$$H = \sum_n \epsilon_n |n\rangle\langle n| + \sum_{n \neq m} V_{nm} |n\rangle\langle m|. \quad (2.1)$$

It is the simplest way, but not necessary, to assume that the matrix elements V_{nm} for hopping between sites are nonzero for nearest neighbour sites only. We want to describe the structural disorder in a liquid or an amorphous solid within a lattice model. Accordingly we assume that the site positions form a regular lattice. The transfer matrix elements are taken to be statistical independent variables with a Lorentzian distribution function

$$P(V_{nm}) = \frac{\Gamma/\pi}{(V_{nm}-V)^2 + \Gamma^2}. \quad (2.2)$$

In order to solve the model in an exact manner, the energy level

ϵ_n on each site must be a function of the surrounding hopping integrals $\epsilon_n = f(V_{nm})$; the function $f(V_{nm})$ is given below.

The Green function G satisfies the equation

$$(E - \epsilon_n) G_{nm} - \sum_{n' \neq n} V_{nn'} G_{n'm} = \delta_{nm}. \quad (2.3)$$

The ensemble averaged Green function

$$\langle G \rangle = \int G(E, \epsilon_n, V_{nm}) \prod_{n,m} \{ P(V_{nm}) dV_{nm} \} \quad (2.4)$$

may be calculated by contour integration in the complex plane, if the Green function has no pole inside the chosen contour. Indeed, Lloyd¹ has shown, that in the case of diagonal disorder the averaged Green function may be obtained by this trick. Now we extend Lloyd's idea to the case of non-diagonal disorder.

Let us consider the Green function G as a function of the complex variables ϵ_n and V_{nm} . The singularities of the Green's function are given by the zeros of the determinant $\det(E-H)$. As is shown in the Appendix $\det(E-H)$ is certainly different from zero, if all eigenvalues λ of the imaginary part of $(E-H)$ are positive (negative). From the so-called Gershgorin criterion one gets for the eigenvalues λ the condition

$$|\operatorname{Im}(E - \varepsilon_n) - \lambda| \leq \sum_m |\operatorname{Im} V_{nm}| \quad (2.5)$$

for all n .

Let us first consider the case of the Lloyd model with only diagonal disorder. Consequently, we put $\operatorname{Im} V_{nm} = 0$

in equation (2.5) and the eigenvalues λ are given by

$\lambda_n = \operatorname{Im}(E - \varepsilon_n)$. Therefore, for $\operatorname{Im} E > 0$ and all $\operatorname{Im} \varepsilon_n \leq 0$ all eigenvalues λ are positive and the Green function cannot have a pole in the lower ε_n half-plane. For $\operatorname{Im} E < 0$ the same statement is valid for the upper ε_n half-plane.

Hence the average Green function may be evaluated by contour integration.

Now we consider the case of off-diagonal disorder. If ε_n and V_{nm} fluctuate independently, the condition $\lambda > 0$ (< 0) is not fulfilled. To make sure that the Green function has no poles in one half-plan of the complex variables V_{nm} we assume that the diagonal elements ε_n depend linearly on the transfer matrix elements V_{nm} :

$$\varepsilon_n = a \sum_m (V_{nm} - V) + \varepsilon, \quad |a| \geq 1. \quad (2.6)$$

Here a is an arbitrary parameter; ε and V denote the average site energy and the average hopping integral, respectively. Putting (2.6) into (2.5) it follows that the sign of the eigenvalues λ is determined by the sign of the diagonal elements $\operatorname{Im}(E - \varepsilon_n)$ if $|a| \geq 1$. Consequently, the Green function cannot have a pole in the V_{nm} half-plane defined by

$$S(aV_{nm}) = S(\varepsilon_n) = -S(E). \quad (2.7)$$

Here the function $S(E)$ is given by

$$S(E) = \operatorname{sgn}(\operatorname{Im} E). \quad (2.8)$$

Hence we should close the contours of integration in (2.4) in the V_{nm} half-plane (2.7). The only pole inside the contour of integration is $V_{nm} = V - iS(aE)\Gamma$. Thus we obtain for the averaged Green function the exact result

$$\langle G \rangle = G(E, \varepsilon - i|a|Z\Gamma S(E), V - iS(aE)\Gamma), \quad (2.9)$$

where Z is the number of the nearest neighbours. Introducing an effective Hamiltonian

$$H_{\text{eff}} = (\varepsilon + \sigma_0) \sum_n |n\rangle \langle n| + (V + \sigma_1) \sum_{n \neq m} |n\rangle \langle m|, \quad (2.10)$$

with

$$\sigma_0 = -i|a|Z\Gamma S(E), \quad \sigma_1 = -i\Gamma S(aE), \quad (2.11)$$

the average Green function may be expressed by

$$\langle G \rangle = (E - H_{\text{eff}})^{-1}. \quad (2.12)$$

By the way, the case of only diagonal disorder is included in these results in the limit $\Gamma \rightarrow 0$ and $a\Gamma = \text{constant}$.

3. Discussion

As an example let us consider a Bravais lattice with Z equivalent nearest neighbours. Without disorder ($\varepsilon_n = \varepsilon$, $V_{nm} = V$) the E versus \vec{k} relation is then

$$E(\vec{k}) = \varepsilon + Vf(\vec{k}), \quad (3.1)$$

with

$$f(\vec{k}) = \sum_n e^{i\vec{k}\vec{R}_n} \quad (3.2)$$

According to equation (2.12) the average Green function may be written as

$$\langle G_{\vec{k}}(E) \rangle = (E - E(\vec{k}) - \Sigma(\vec{k}))^{-1}, \quad (3.3)$$

where the exact self energy $\Sigma(\vec{k})$ is given by

$$\Sigma(\vec{k}) = \sigma_0 + \sigma_1 f(\vec{k}). \quad (3.4)$$

Thus for the density of states $D(E)$ we obtain

$$D(E) = \frac{1}{\pi} \sum_{\vec{k}} \frac{|\Sigma(\vec{k})|}{(E - E(\vec{k}))^2 + |\Sigma(\vec{k})|^2}. \quad (3.5)$$

Equation (3.5) shows that due to the disorder each state is smeared out by a Lorentzian distribution. Unlike the Lloyd model¹ where the width of the distribution is the same for all states the self energy $\Sigma(\vec{k})$ depends on \vec{k} and, therefore, the width of the Lorentzian distribution depends on \vec{k} . This effect involves an asymmetry in the density of states (3.5) even if the density of states in the ordered system is symmetrical.

Unfortunately, the applicability of our exact results is restricted by the condition (2.6), which relates the fluctuations of the site energies ϵ_n and the hopping matrix elements V_{nm} . A relation like (2.6) is appropriate for a liquid or an amorphous solid, where a change of the distance between the nearest neighbours n and m changes the transfer matrix

elements V_{nm} and the potentials on the sites n and m . In a first approximation the net change of the potential due to the fluctuations of all the nearest neighbours may be obtained by an expression like (2.6). However, we must note that the fluctuations of the energy levels ϵ_n are smaller than the fluctuations of the overlap integrals²¹. Thus for a liquid or amorphous solid we expect $|a| < 1$.

Pottier and Calecki²² have considered a model in which the off-diagonal elements are random variables and the diagonal elements are fixed. Using the CPA they derive an expression for the density of states which shows the same behaviour as that obtained in the Lloyd model. Recently, Herscovici²³ remarked that "at least in a certain approximation" - the average Green function may be obtained by contour integration also in the case of independently fluctuating diagonal and off-diagonal elements. However, from equation (3.3) follows that in this case the imaginary part of the average Green function may change the sign. Therefore, this approximation gives a density of states which is not positive definite.

An example, where our model describes the actual physical situation, is the case of an amorphous spin $-1/2$ Heisenberg ferromagnet. Within the Tjablikov-decoupling procedure the equation of motion for the Green function is given by²⁴

$$E/\sigma G_{nm} = \delta_{nm} + \sum_i J_{ni} (G_{nm} - G_{im}), \quad (3.6)$$

where σ is the magnetization and J_{nm} are the exchange integrals. Equation (3.6) is equivalent to (2.6) if we put $V_{nm} = -J_{nm}$ and $\epsilon_n = \sum_m J_{nm}$, i.e., $a = -1$. With the help

of our exact result for the density of states $D(E)$, we can try to calculate the magnetization $\bar{\sigma}$. For this purpose we must solve a self-consistent equation for $\bar{\sigma}$ ²⁴. We only find the solution $\bar{\sigma} = 0$, since for Bose like excitations it is not allowed that $D(E) > 0$ for $E \leq 0$. Consequently, the Lorentzian distribution gives no ferromagnetic solution. According to the discussion in¹⁸ the high part of negative exchange integrals destroys the ferromagnetic order. In contrast to this case the Gaussian distribution, which suppresses large fluctuation of the exchange integrals, yields a stable ferromagnetic solution¹⁸.

The exact determination of the average Green function can be used to check the quality of different approximations. We note that the single site CPA becomes exact for the Lloyd model²⁵. The same is true for our model including off-diagonal disorder. In a forthcoming paper we want to show that like Foo et al.⁸ and Morita and Chen¹⁶ a single bond CPA can be formulated for the model (2.6). This procedure becomes exact if a Lorentzian distribution is used.

4. Generalized Weaire model

The results of Section 2 can be generalized to the multi-band case directly. As an example we consider in the following the Weaire model²⁶⁻²⁸ which describes an amorphous tetrahedrally bonded semiconductor like Si or Ge. It is assumed that the structure of these elemental amorphous semiconductors is that of a random network in which every atom is almost perfectly tetrahedrally coordinated with its nearest neighbours. The model Hamiltonian may be written as

$$H = \sum_{n,i} V_0^{ni} |ni\rangle\langle ni| + \sum_{n,i,j} V_1^n |ni\rangle\langle nj| + \sum_{n,i} V_2^{ni} |ni\rangle\langle n_i i|, \quad (4.1)$$

where the atoms are labelled by n and the sp^3 -orbitals by i, j . The first term in (4.1) determines the position V_0^{ni} of the energy level at the site n belonging to the orbital i . The second term describes the overlap between orbitals associated with the same atom. The last term involves the overlap along a bond i between neighbouring atoms n and n_i .

If we take into consideration only topological disorder—this means the matrix elements of the Hamiltonian (4.1) are the same everywhere in the structure but the connectivity is disordered—some exact results for the density of states can be obtained. First of all a gap persists for all topological structures²⁶⁻²⁸. The density of states for the diamond structure is shown in Fig.1. The delta functions at the top of the valence band and the conduction band are entirely p like and structure independent. The rest of the spectrum depends on the structure via a one-band Hamiltonian and connectivity matrix^{29,30}.

Experimental results^{31,32} for the density of states in the amorphous phase indicate that the p -like peak at the top of the valence band remains almost unchanged but the two lower peaks coalesce into a single broad peak. In contrast to these results the topological disorder of the Polk model turns out to be not sufficient to give a single peak³³.

Now let us consider the influence of fluctuations of the matrix elements in (4.1) on the density of states. Weaire and Thorpe²⁹ had shown that the gap remains if the fluctuations

of V_1 and V_2 are small. Recently Thorpe³⁴ considered the relationship between the structure and the gap and showed that the gap increases if one makes the structure more homogeneous. Streitwolf³⁵ calculated the density of states of the valence band in a single site CPA neglecting the interaction between bonding and antibonding states^x. Unfortunately, in³⁵ only the Hubbard form for the density of states is considered. Here we assume that the matrix elements V_1^{ni} and V_2^{ni} in the Hamiltonian (4.1) are distributed randomly according to Lorentzian distributions with mean values V_1 and V_2 and with widths Γ_1 and Γ_2 . From the considerations in Section 2 it follows that the average Green function may be obtained by contour integration if we assume, that the diagonal element V_0^{ni} in (4.1) depends linearly on V_2^{ni} :

$$V_0^{ni} = a(V_2^{ni} - V_2), \quad |a| \geq 1. \quad (4.2)$$

Introducing an effective Hamiltonian

$$H_{\text{eff}} = \tilde{V}_0 \sum_{n,i} |ni\rangle \langle ni| + \tilde{V}_1 \sum_{n,i,j} |ni\rangle \langle nj| + \tilde{V}_2 \sum_{n,i} |ni\rangle \langle n_i i|, \quad (4.3)$$

with

$$\tilde{V}_0 = -i|a|\Gamma_2 S(E), \quad \tilde{V}_1 = V_1 - i\Gamma_1 S(E), \quad \tilde{V}_2 = V_2 - i\Gamma_2 S(aE), \quad (4.4)$$

the average Green function may be written as

$$\langle G \rangle = (E - H_{\text{eff}})^{-1}. \quad (4.5)$$

^x We are grateful to Dr. Streitwolf for his results prior to publication.

As in^{29,30} we obtain from equation (4.5) for the density of states the expression

$$D(E) = -\frac{1}{\pi} \text{Im} \left\{ \frac{1}{E - \tilde{V}_0 - \tilde{V}_2} + \frac{1}{E - \tilde{V}_0 + \tilde{V}_2} - \int d\varepsilon g(\varepsilon) F(\varepsilon, E) \right\}, \quad (4.6)$$

with the density of states $g(\varepsilon)$ of the one-band Hamiltonian and the definition

$$F(\varepsilon, E) = 2(E - \tilde{V}_0 - 2\tilde{V}_1) \left\{ (E - \tilde{V}_0 - 2\tilde{V}_1)^2 - \tilde{V}_2^2 - 4\tilde{V}_1 \tilde{V}_2 \varepsilon \right\}^{-1}. \quad (4.7)$$

(In equation (4.6) we put $\text{Im } E \rightarrow +0$).

The numerical results for the density of states for the diamond structure are shown in Figs. 2 to 5. In all cases the parameters V_1 and V_2 are the same ($V_1 = -1$, $V_2 = -3$). Of course, due to the Lorentzian distribution the bands have infinite tails and the gap is smeared out. However, our model allows to study the influence of quantitative disorder in a simple manner. In Fig. 2 only the fluctuations of V_0 is taken into account ($\Gamma_1 = 0$, $\Gamma_2 \rightarrow 0$, $|a|\Gamma_2 = 0.11$). As in the Lloyd model the fluctuations of the site energy V_0 provide an overall broadening of the density of states. In contrast to this case the fluctuations of V_1 influence only the bottom of the bands (Fig. 3). Since the term in the Hamiltonian (4.1) which involves V_1 is a projection operator for s-states, the p-like states at the top of the valence band and the conduction band remain unchanged.

In Fig. 4 the density of states is considered if V_0 and V_2 fluctuate ($\Gamma_2 = 0.1$, $|a| = 1.1$). For a < 0 the fluctuations of V_2 compensate the fluctuations.

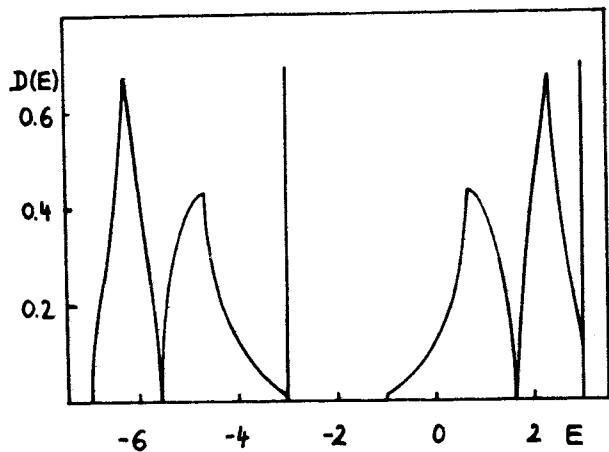


Fig. 1. Density of states for the diamond structure with $V_1 = -1$ and $V_2 = -3$.

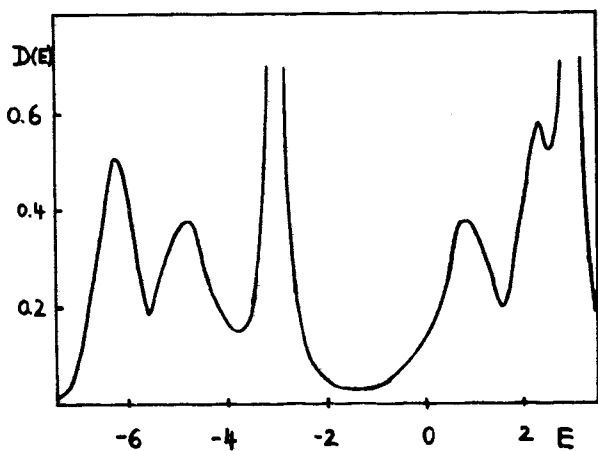


Fig. 2. Density of states for the diamond structure ($\Gamma_1 = \sigma$, $\Gamma_2 \rightarrow \sigma$, $|a|\Gamma_2 = \sigma.11$).

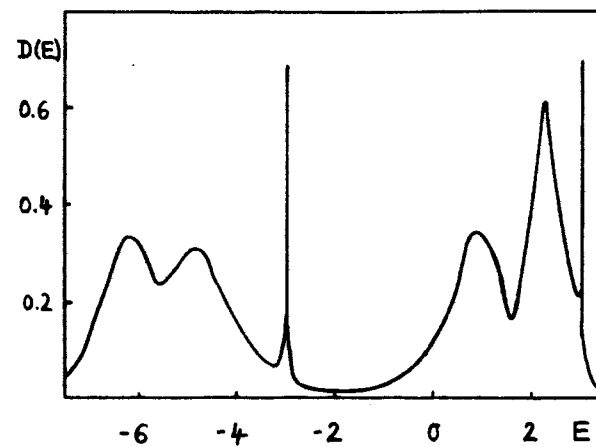


Fig. 3. Density of states for the diamond structure ($\Gamma_1 = 0.1$, $\Gamma_2 \rightarrow \sigma$, $|a|\Gamma_2 = 0.002$).

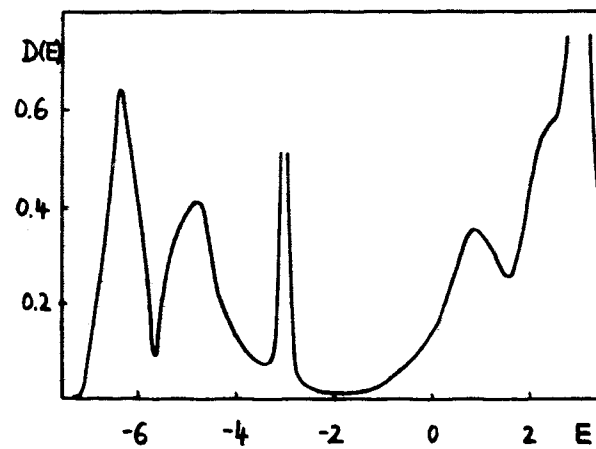


Fig. 4. Density of states for the diamond structure ($\Gamma_1 = 0$, $\Gamma_2 = 0.1$, $a = -1.1$).

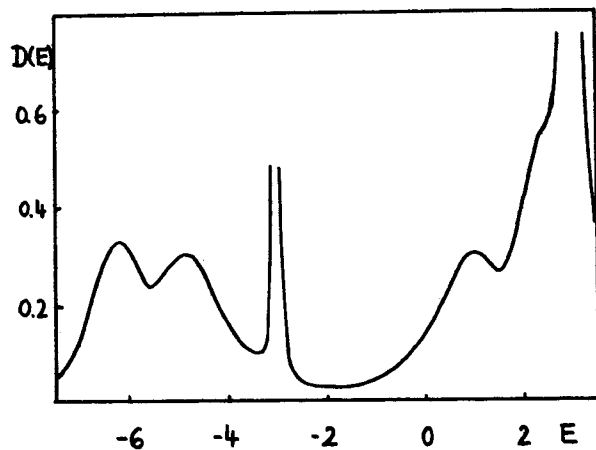


Fig. 5. Density of states for the diamond structure
 ($\Gamma_1 = 0.1$, $\Gamma_2 = 0.1$, $a = -1.1$).

of V_0 for the bonding states and increases the broadening of the antibonding states. Hence we expect a large broadening of the conduction band. Fig.5 shows the density of states for the case of fluctuating matrix elements V_0 , V_1 and V_2 . The valence band and the conduction band are smeared out. However, the delta-peak at the top of the valence band is only slightly changed. The density of states for any structure can be calculated from equation (4.6) if the density of states $q(\epsilon)$ of the one-band Hamiltonian is known. However, as have been shown by Weaire and Thorpe²⁷ the s- and bonding-like fraction of the density of states does not depend on the structure. Hence the general behaviour of the density of states due to quantitative disorder is the same for all structures.

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APPENDIX

We want to prove that

$$\det|A \pm iB| \neq 0, \quad (\text{A.1})$$

where A and B are hermitean matrices and B is a positive definite one. Let b a hermitean matrix which satisfies $b^2 = B$. Then it follows

$$\det|A \pm iB| = \det|B| \det|b^{-1}Ab^{-1} \pm iI|, \quad (\text{A.2})$$

where I is the unit matrix. Using the real eigenvalues $\tilde{\alpha}_n$ of the hermitean matrix $b^{-1}Ab^{-1}$, (A.1) may be written as

$$\det(A+iB) = \det|B| \prod_n (\tilde{\alpha}_n \pm i) \neq 0 \quad (\text{A.3})$$

and the proof follows.

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