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Ionisation as a Hilbert boundary value problem

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Abstract. The problem of single-photon ionisation of an atom is formulated as a Hilbert boundary value problem on a semi-infinite arc, and a solution is then presented.

1. Introduction

The problem of single-photon ionisation of an atom has been extensively studied over many years, and is now generally regarded as well understood. Normally, a solution to this problem is obtained in terms of time-dependent coefficients using eigenstates of the unperturbed system (Heitler 1954). We present here an alternative method of solution in which the decay (or ionisation) is envisaged as due to a spreading of an initially localised wave packet in the manner discussed by Khalfin (1958). Here, the coefficients are time independent, and the eigenstates are those of the complete atom–field coupled system. The qualitative difference in approach emphasises the physical mechanism of ionisation as due to an irreversible phase mixing of the eigenfunctions of the 'dressed continuum' system.

Viewed thus, a close similarity is noted between the apparently disparate topics of single-photon ionisation, and collisionless Landau damping of an initial disturbance in an isotropic, homogeneous electron plasma (Landau 1946), where the latter decay also arises from a phase mixing of continuum modes of the system—that is, of the Van Kampen–Case (Van Kampen 1955, Case 1959) normal modes supported by the plasma. Moreover, the exponential decay law normally associated with the Weisskopf–Wigner theory of ionisation (Agarwal 1974), and with Landau damping, stems in each case from a zero of the appropriate dispersion function located on the unphysical sheet of the complex frequency plane.

Our method utilises the formalism of pseudo-Fano autoionising states (Fano 1961, Beers and Armstrong 1975, Geller and Popov 1976), which are currently receiving much attention in connection with higher-order nonlinear processes involving multi-photon transitions into an atomic continuum (Armstrong *et al* 1975, Coleman and Knight 1982, Rzazewski and Eberly 1981, Coleman *et al* 1982); the application considered here constitutes the simplest (i.e. lowest order) non-perturbative application of such a formalism. The problem is then cast into the general form of a Hilbert boundary value problem on a semi-infinite arc (Muskhelishvili 1953), and techniques peculiar to this type of problem are used to obtain a solution.

The main purpose of this article is to demonstrate that the general Hilbert technique has a ready application in the study of the single-photon ionisation problem, and

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hence by extension may have a similarly useful application in the study of the higher-order processes mentioned above. Loosely speaking (see appendix for a more detailed discussion), the Hilbert technique is concerned with the branch-cut properties of a certain class of function. A branch cut in an appropriate dispersion function is a mathematical manifestation of the fact that the system in question can support a continuum of normal modes. Consequently, the Hilbert method would appear to be a most natural way to study problems where decay in a system results as a consequence of phase mixing of continuum modes.

For readers not acquainted with the Hilbert technique, an appendix is included in which the salient features of the technique are discussed; a complete discussion of the method is given in the standard text by Muskhelishvili (1953). In § 2, the problem is formulated as a Hilbert problem, and in § 3 a solution is presented.

2. Formulation as a Hilbert problem

2.1. Pseudo-Fano states

The coupling of a bound state with an overlapping continuum of states results in a non-trivial modification of the latter's structure. We briefly review the technique used by Fano (1961) in his original treatment of this problem.

Consider an atomic discrete state $|\phi\rangle$ which overlaps and is coupled to a continuum of states $|\psi_E\rangle$. The states $|\phi\rangle$ and $|\psi_E\rangle$ are zeroth-order approximation eigenstates of the atomic Hamiltonian H_A. With respect to these states, the matrix elements of H_A are taken as

$$\begin{aligned} &\langle\phi|H_A|\phi\rangle = E_\phi \\ &\langle\psi_E|H_A|\psi_{E'}\rangle = E'\delta(E'-E) \\ &\langle\psi_{E'}|H_A|\phi\rangle = V_{E'} \end{aligned} \qquad (1)$$

where $V_{E'}$ represents the coupling of the bound state to the continuum through configuration interaction, and δ is the Dirac δ function. The wavefunction which diagonalises H_A and has energy E is taken in the form

$$|\Psi_E\rangle = a(E)|\phi\rangle + \int_0^\infty b_{E'}(E)|\psi_{E'}\rangle \, \mathrm{d}E' \qquad (2)$$

where the lower limit of the integral (zero) coincides with the beginning of the continuum, which then extends to infinity. Fano constructed eigenvalue problems for the coefficients $a(E)$ and $b_{E'}(E)$, and showed that these were related by

$$b_{E'}(E) = \left(\frac{\mathbb{P}}{E-E'} + Z(E)\delta(E-E')\right) V_{E'} a(E) \qquad (3a)$$

with

$$Z(E) = \frac{1}{|V_E|^2}\left(E - E_\phi + \mathbb{P}\int_0^\infty \frac{|V_{E'}|^2 \, \mathrm{d}E'}{E'-E}\right) \qquad (3b)$$

and $\mathbb{P}$ denotes 'principal part of'. By imposing δ-function normalisation on the states $|\Psi_E\rangle$,

$$\langle\Psi_E|\Psi_{E'}\rangle = \delta(E-E'). \tag{4}$$

Fano solved for the unknown $a(E)$

$$|a(E)|^2 = |V_E|^2 \Big/ \left\{\pi^2|V_E|^4 + \left[E - E_\phi + \mathbb{P}\int_0^\infty |V_E|^2/(E'-E)\,\mathrm{d}E'\right]^2\right\}. \tag{5}$$

Clearly, the Fano technique can also be applied to the case where the perturbation $V_{E'}$ describes coupling of a bound state to a continuum through an externally applied field rather than by configuration interaction (Armstrong *et al* 1975, Geller and Popov 1976). The unperturbated bound and continuum states then have the form $|n;\phi\rangle$, $|n-1;\psi_E\rangle$, where the first entry refers to the number of photons in the external (or applied) field mode. Here, the atomic states $|\phi\rangle$ and $|\psi_E\rangle$ are initially non-degenerate, but application of the external field couples these to produce the degenerate system considered above. Note that the eigenfunction $|\Psi_E\rangle$ (equation (2)) now refers to the dressed-atom–field system and, as before, has *real* eigenvalues since the system is self-adjoint.

If the atomic state $|\phi\rangle$ is not initially occupied, the continuum structure induced by the above coupling can be probed using a second laser to induce transitions from the initially occupied state (for example, the ground state) into the structured continuum; in this way, the usual Fano profile is recovered. Here, we wish to address the case where the state $|\phi\rangle$ is initially occupied, in which case ionisation results as a consequence of the interference of the diagonalised eigenstates $|\Psi_E\rangle$.

2.2. *Formulation as a Hilbert problem*

The state of the system at any time 't' may be written as a linear sum (or integral) over its eigenstates $|\Psi_E\rangle$:

$$|\Phi(t)\rangle = \int_0^\infty c_E|\Psi_E\rangle \exp(-\mathrm{i}Et/\hbar)\,\mathrm{d}E. \tag{6}$$

Here c_E is a time-independent quantity, and $|\Psi_E\rangle$ is given by equations (2) and (3), in which $|\phi\rangle$ and $|\psi_E\rangle$ are replaced by $|n;\phi\rangle$ and $|n-1;\psi_E\rangle$ respectively, as discussed earlier. The initial conditions are taken to be such that at time $t=0$, the state $|\Phi(t=0)\rangle = |n;\phi\rangle$; that is, the atom is in its ground state. Note that equation (6) emphasises the point made in the introduction, that subsequent ionisation of the atom is a consequence of the irreversible phase mixing of the eigenfunctions $|\Psi_E\rangle$. The unknown coefficients c_E are therefore to be determined from the integral equations

$$|n;\phi\rangle = \int_0^\infty c_E|\Psi_E\rangle\,\mathrm{d}E. \tag{7}$$

Since the continuum states $|\Psi_E\rangle$ form a complete set, a solution for the unknown c_E is obtained immediately on taking the inner product of equation (7) with $\langle\Psi_E|$, to give $c_E = a^*(E)$. However, as stated in the introduction, the main object of the present article is not so much to obtain a solution, as to discuss the application of the Hilbert technique to this type of problem; we therefore proceed as follows: taking the inner product of equation (7) with $\langle\phi\,;n|$ and $\langle\psi_E, n-1|$ respectively, and using equation (2), produces

$$\begin{aligned}&\int_0^\infty c_E a(E)\,\mathrm{d}E = 1\\ &\int_0^\infty c_E b_{E'}(E)\,\mathrm{d}E = 0.\end{aligned} \tag{8}$$

We now solve the second of these for the unknown c_E as follows.

Substituting for $b_{E'}(E)$ from equation (3) produces

$$\mathbb{P}\int_0^\infty \frac{a(E)c_E}{E-E'}\,\mathrm{d}E = -Z(E')a(E')c_{E'} \tag{9}$$

where, as before, $\mathbb{P}$ denotes 'principal part of'. Now define the sectionally holomorphic function (Muskhelishvili 1953)

$$\theta(U) = \frac{1}{2\pi\mathrm{i}}\int_0^\infty \frac{a(E)c_E}{E-U}\,\mathrm{d}E \tag{10}$$

so that

$$\theta^+(U)+\theta^-(U) = \frac{1}{\mathrm{i}\pi}\,\mathbb{P}\int_0^\infty \frac{a(E)c_E}{E-U}\,\mathrm{d}E \tag{11a}$$

$$\theta^+(U)-\theta^-(U) = a(E)c_E. \tag{11b}$$

Here, $\theta^\pm$ denote the limits of the function θ as (complex) U approaches the positive real axis from above (i.e. from $\mathrm{Im}\,U>0$) and below ($\mathrm{Im}\,U<0$) respectively. Note that $\theta(U)$ is sectionally holomorphic throughout the complex U plane, with a single branch cut which runs along the positive $\mathrm{Re}\,U$ axis. Substituting (11) into (9) produces

$$\theta^+(E) = \frac{Z(E)-\mathrm{i}\pi}{Z(E)+\mathrm{i}\pi}\,\theta^-(E) \equiv G(E)\theta^-(E) \qquad 0\leqslant E<\infty. \tag{12}$$

Equation (12) is in the standard form of a Hilbert boundary value problem on a semi-infinite arc which is to be solved for the unknown $\theta(E)$. As such, a formal solution could be written down immediately by using the method of solution outlined in the appendix. However, the particular form of the function $G(E)$ above permits a solution to be obtained by inspection; this simpler procedure is pursued in the next section, and a discussion of the formal solution of equation (12) is deferred until the appendix.

3. Solution of the Hilbert problem

Using equation (3*b*), it is readily shown that

$$G(E) = D^-(E)/D^+(E) \tag{13}$$

where $D^\pm(E)$ are the respective limits in the sense defined above of the *dispersion function*

$$D(E) = E - E_\phi + \int_0^\infty \frac{|V_{E'}|^2}{E'-E}\,\mathrm{d}E'. \tag{14}$$

Note that the integral here is not simply 'principal part of', as was the case in the definition of the quantity $Z(E)$. The Hilbert problem (12) then becomes

$$\theta^{+}(E)D^{+}(E)=\theta^{-}(E)D^{-}(E) \qquad 0\leqslant E<\infty. \tag{15}$$

The functions $\theta(E)$ and $D(E)$ are both, from their respective definitions, holomorphic in the cut plane with a single branch cut running along the positive Re E axis. From equation (15), it then follows that the product function $\theta(E)D(E)$ is entire in the whole of the complex E plane including the positive Re E axis. Moreover, as $|E|\rightarrow\infty$, it follows from equations (14) and (10), together with the first of equations (8), that $\theta D\rightarrow -1/2\pi\mathrm{i}$, and by Liouville's theorem, must equal this same constant value throughout the complex E plane. The unknown function $\theta(E)$ is then obtained as

$$\theta(E)=\frac{-1}{2\pi\mathrm{i}D(E)} \tag{16}$$

and, from equation (11*b*), the required c_E is

$$\begin{aligned} c_E &= \frac{-1}{2\pi\mathrm{i}a(E)}\left(\frac{1}{D^{+}}-\frac{1}{D^{-}}\right) \\ &= a^{*}(E). \end{aligned} \tag{17}$$

To complete the formal solution of the problem, we now prove that the above form for c_E is consistent with the first of the integral equations (8). That is, it is required to show that

$$\frac{-1}{2\pi\mathrm{i}}\int_0^{\infty}\left(\frac{1}{D^{+}}-\frac{1}{D^{-}}\right)\mathrm{d}E=1. \tag{18}$$

More correctly, it is required to show that

$$-\frac{1}{2\pi\mathrm{i}}\lim_{t\rightarrow 0^{+}}\int_0^{\infty}\exp(-\mathrm{i}Et/\hbar)\left(\frac{1}{D^{+}}-\frac{1}{D^{-}}\right)\mathrm{d}E=1 \tag{18a}$$

where 0^{+} denotes the limit as $t\rightarrow 0$ from above. Recalling the definitions of the functions $D^{\pm}(E)$, equation (19) can be written in the equivalent form

$$\lim_{t\rightarrow 0^{+}}\int_{\mathrm{C}}\exp(-\mathrm{i}Et/\hbar)\frac{\mathrm{d}E}{D(E)}=-2\pi\mathrm{i} \tag{18b}$$

where C is the contour which runs around the branch cut of $D(E)$, as shown in figure 1. Moreover, since $D(E)$ is regular throughout the cut plane, and vanishes as $|E|$ at infinity, it is permissible, for $t>0$, to open up the contour C to give

$$\lim_{t\rightarrow 0^{+}}\int_{-\infty}^{\infty}\exp(-\mathrm{i}Et/\hbar)\frac{\mathrm{d}E}{D(E)}=-2\pi\mathrm{i} \tag{18c}$$

on the understanding that the contour now passes *above* the cut on the positive real axis. Equivalently, we may analytically continue the cut away from the positive real E axis by tilting it through an infinitesimal angle in a clockwise sense, and then bring the contour of integration down to lie along the entire real E axis; this is the sense in which equation (18*c*) is now understood to be defined.

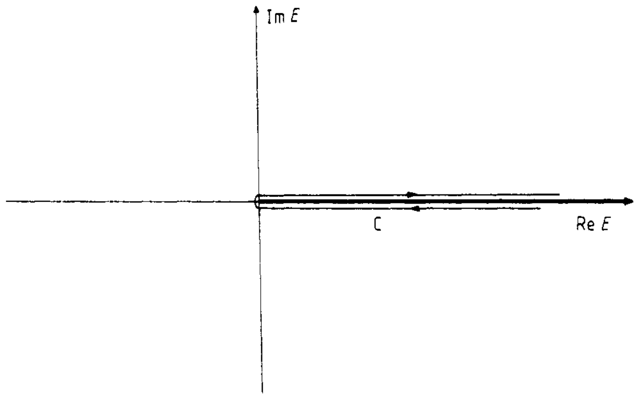

Figure 1. Contour C appearing in equation (18). The bold line denotes the branch cut of the function $D(E)$.

Defining

$$f(t)=\frac{-1}{2\pi \mathrm{i}}\int_{-\infty}^{\infty}\frac{\exp(-\mathrm{i}Et/\hbar)}{D(E)}\,\mathrm{d}E \tag{19}$$

we are required to prove that $f(t\to 0^{+})=+1$. This is easily done by Fourier inverting, and using the fact that $D(E)$ is regular in the upper half of the complex E plane including the real E axis, so that $f(t)=0$ for $t<0$. Then,

$$\frac{1}{D(E)}=\frac{\mathrm{i}}{\hbar}\int_{0}^{\infty}f(t)\exp(\mathrm{i}Et/\hbar)\,\mathrm{d}t. \tag{20}$$

Expanding both sides as power series in E^{-n}, taking the limit $|E|\to\infty$ and comparing coefficients of E^{-1} produces the desired result. This completes the formal solution to the problem.

4. Concluding remarks

The quantity $f(t)$ defined in equation (19) is the probability amplitude that the atom will be found in its ground state at any time $t\geqslant 0$. It is seen that this depends intimately on the analytic properties of the dispersion function $D(E)$. Recall that D has a branch cut running along the positive real E axis, and is nowhere zero on the physical sheet of the complex energy plane. However, $D(E)$ does have a zero lying just below the positive real axis on the unphysical sheet of the complex E plane, which may be exposed by the standard method of analytic continuation from the upper half-plane. Deforming the branch cut analytically in this manner (see figure 2), the exposed pole of the function D^{-1} is recognised as that which gives rise to the Weisskopf–Wigner theory of ionisation—that is, to the well known exponential decay law. The remaining branch-cut contribution represents deviation from this exponential decay law (e.g. Zakowicz and Rzazewski 1974, Mostowski and Wodkiewicz 1973).

The fact that the dispersion function $D(E)$ has a branch cut, and no other zeros or singularities, is a mathematical manifestation of the fact that the system supports a continuum of normal modes (or eigenstates), but no bound states. Decay is then a

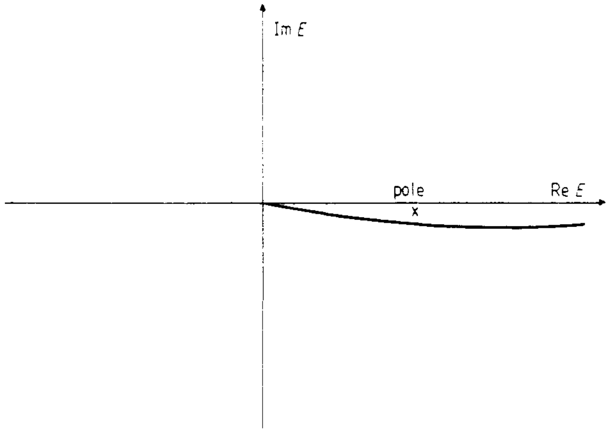

Figure 2. Exposed Weisskopf–Wigner pole on the unphysical sheet of the complex E plane; see text for details.

consequence of irreversible phase mixing of these modes. We have explored here the connection between the analytic properties of $D(E)$ and the decay mechanism, and have demonstrated that a useful mathematical technique—that based on the study of Hilbert boundary value problems—has ready application to this type of problem. It is believed that the general technique will also find useful application in the study of the higher-order processes alluded to in the introduction.

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Appendix

We present here a brief outline of the Hilbert boundary value problem; for a complete discussion the interested reader is referred to the standard text by Muskhelishvili (1953).

In its simplest form, the Hilbert problem may be stated thus: it is required to find the function $\Phi(Z)$ which satisfies the boundary condition

$$\Phi^+(Z) = G(Z)\Phi^-(Z) + g(Z) \qquad Z \in \mathrm{L} \tag{A.1}$$

on the contour L, where $G(Z)$ and $g(Z)$ are known functions. The superscripts $\pm$ on $\Phi(Z)$ denote the two limits of this function as Z approaches L from above and below respectively (cf figure 3). The functions $G(Z)$ and $g(Z)$ are required to satisfy

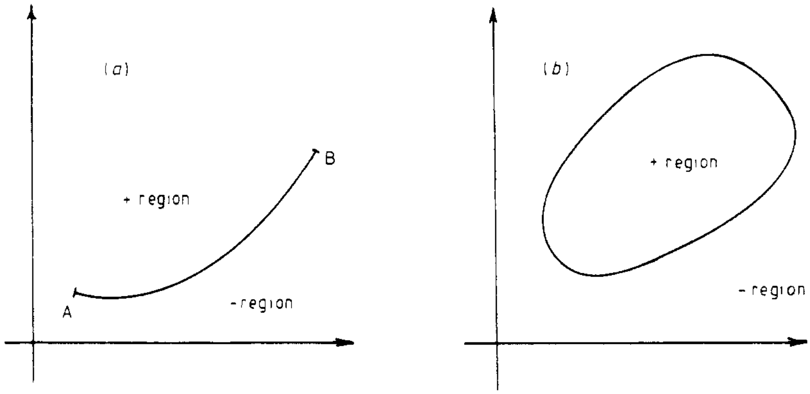

Figure 3. Open (a) and closed (b) contours; see text for details.

a Hölder condition on L; that is, if Z_1 and Z_2 are two points on L, then

$$|G(Z_2)-G(Z_1)| \leqslant A|Z_2-Z_1|^{\mu} \tag{A.2}$$

where A and μ are positive constants, and $0<\mu\leqslant 1$. When $\mu=1$ the Hölder condition becomes the Lipschitz condition.

When $g(Z)$ is identically zero, equation (A.1) is a statement of the homogeneous Hilbert problem; when $g(Z)\neq 0$, it is generalised to the inhomogeneous problem.

It is also necessary to distinguish between 'open' contours, as in figure $3(a)$, and 'closed' contours, where 'A' and 'B' meet and in addition the first derivatives of the functions G and g change continuously on passing through the point of contact (figure $3(b)$). It will be seen from the statements above that the ionisation problem considered in the main text (cf equation (12)) can be cast as a homogeneous Hilbert problem on an open arc (the positive real energy axis).

Properties of the inhomogeneous problem will not be considered here. The homogeneous problem on a *closed* arc can be solved by first taking the logarithm of both sides of equation (A.1),

$$\ln \Phi^{+}(Z)-\ln \Phi^{-}(Z)=\ln G(Z) \qquad Z\in \mathrm{L} \tag{A.3}$$

then comparing with the Plemelj formula

$$\Psi(Z)=\frac{1}{2\pi \mathrm{i}}\int_{\mathrm{L}}\frac{\Psi(U)}{U-Z}\,\mathrm{d}U. \tag{A.4}$$

In the limit $Z\in \mathrm{L}$, equation (A.4) can be written as

$$\begin{aligned}\Psi^{+}(Z)&=\tfrac{1}{2}\Psi(Z)+\frac{1}{2\pi \mathrm{i}}\mathbb{P}\int_{\mathrm{L}}\frac{\Psi(U)}{U-Z}\,\mathrm{d}U\\ \Psi^{-}(Z)&=-\tfrac{1}{2}\Psi(Z)+\frac{1}{2\pi \mathrm{i}}\mathbb{P}\int_{\mathrm{L}}\frac{\Psi(U)}{U-Z}\,\mathrm{d}U\end{aligned} \tag{A.5}$$

where $\Psi^{\pm}(Z)$ are the two limiting forms of the function $\Psi(Z)$ in the sense defined above, and $\mathbb{P}$ is 'principal part of'. Subtracting the two equations in (A.5) and comparing with (A.4) gives the required result:

$$\begin{aligned}\Phi(Z) &= \exp\left(\frac{1}{2\pi i}\int_{L}\frac{\ln G(U)}{U-Z}\,dU\right)\\ &\equiv X(Z), \text{ say.}\end{aligned} \tag{A.6}$$

This method of obtaining the solution is valid provided the quantity $\ln(G(Z))$ is single valued in the '+' and '−' regions, in which case $\ln(\Phi(Z))$ is also single valued. The particular solution $\Phi(Z)=X(Z)$ is referred to as the fundamental solution by Muskhelishvili. It is not difficult to see that a more general solution to this same problem is

$$\Phi(Z)=p(Z)X(Z) \tag{A.7}$$

where $p(Z)$ is an arbitrary polynomial, chosen to give Φ a specified asymptotic behaviour.

The above technique needs to be modified slightly when considering the homogeneous Hilbert problem on an *open* arc, as follows. Assume, as before, that the function $\Phi(Z)$ is continuous on L from above and below, with the possible exception of the end-points A and B (figure 3(*a*)), but near these end-points, it is required to satisfy

$$\begin{gathered}|\Phi(Z)| \leqslant \frac{A}{|Z-c_k|^{\alpha}}\\ k=1,2 \qquad c_1 \equiv a \qquad c_2 \equiv b\end{gathered} \tag{A.8}$$

with A and α real constants and $\alpha<1$. Such functions are called sectionally holomorphic functions, with the line of discontinuity coincident with L as before. Clearly, a solution for $\Phi(Z)$ for the homogeneous problem on an open arc will again be given by equation (A.6), as can be verified by direct substitution into equation (A.1) (with $g(Z)=0$). However, it remains to show that this particular solution also satisfies equation (A.8); if not, then the solution must be suitably amended to find a form which does. Defining

$$\Gamma(Z) \equiv \frac{1}{2\pi i}\int_{L}\frac{\ln G(U)}{U-Z}\,dU \tag{A.9}$$

a study of the function $\exp(\Gamma(Z))$ in the vicinity of either of the end-points A and B reveals that it takes the form (Muskhelishvili 1953, § 79)

$$\exp(\Gamma(Z)) = (Z-c_k)^{\alpha_k+i\beta_k}\Omega(Z) \tag{A.10}$$

where α_k and β_k are real constants given by

$$\alpha_k + i\beta_k = \mp \ln(G(c_k))/2\pi i \tag{A.11}$$

(upper sign for $c_k=a$, lower for $c_k=b$), and $\Omega(Z)$ is a non-vanishing bounded function which assumes a definite value at the points c_k. Now select integers λ_k satisfying the condition

$$-1<\alpha_k+\lambda_k<+1 \tag{A.12}$$

and put

$$\Pi(Z)=(Z-a)^{\lambda_1}(Z-b)^{\lambda_2}. \tag{A.13}$$

Then, the function

$$X(Z)=\Pi(Z)\exp\Gamma(Z) \tag{A.14}$$

obviously satisfies all the conditions of the problem and is therefore a particular solution of the homogeneous problem on the open arc L. The ends c_k for which α_k is an integer—i.e. for which $G(c_k)$ is a real positive quantity—are called special ends.

We conclude this appendix by presenting a formal solution of the Hilbert problem represented by equation (12) in the main text. As stated earlier, this is identified as a homogeneous Hilbert problem on an open arc, the latter being the real positive energy axis. The function $D^-(E)/D^+(E)$ satisfies the Hölder condition on the semi-infinite arc; hence, using the method presented above, a particular solution is

$$\theta(E)=E^{\lambda}\exp\left(\frac{1}{2\pi i}\int_0^{\infty}\ln\left(\frac{D^-(E')}{D^+(E')}\right)\frac{dE'}{E'-E}\right) \tag{A.15}$$

where $D^{\pm}(E)$ are the plus and minus parts of the dispersion function $D(E)$ given by equation (14). Also, from equation (A.11), we have

$$\alpha+i\beta=-\frac{1}{2\pi i}\ln\left(\frac{D^-(0)}{D^+(0)}\right)=-\frac{1}{2\pi i}\ln(\exp(-2\pi i)) \tag{A.16}$$

which implies that $\alpha=1$, $\beta=0$, and hence from equation (A.12), that $\lambda=-1$. Note that $E=0$ is a special end, in the sense defined above. The above finding is conditional on the assumption that the principal part of the integral appearing in equation (14) converges in the limit $E\to 0$, and that its value is less than E_ϕ; we assume this to be the case without further comment here. Defining $d(E)=D(E)/E$, and noting—from inspection of the analytic properties of the function $D(E)$—that the integral in equation (A.15) can be written as an equivalent integral of the function $\ln(1/d(E))$ around the contour 'C' shown in figure 1, the asymptotic behaviour of the ln function then permits this contour to be opened up capturing the single pole at $E'=E$, giving

$$\theta(E)=\frac{1}{E}\exp\left\{\frac{-1}{2\pi i}2\pi i\ln d(E)\right\}=\frac{1}{D(E)} \tag{A.17}$$

as found previously. The particular solution (A.17) is generalised by multiplying by an arbitrary polynomial, as in equation (A.7), whose form is deduced by considering the behaviour of the respective functions at infinity; in this way, the result shown in equation (16) in the text is obtained again.

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