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# Local density of states $N(r, E)$ for central fields, with energy $E$ in the continuum: especially the Coulomb potential

I A Howard<sup>1</sup>, N H March<sup>1,2</sup> and Á Nagy<sup>3</sup>

<sup>1</sup> Department of Physics, University of Antwerp, Groenenborgerlaan 171, B-2020 Antwerp, Belgium

<sup>2</sup> Oxford University, Oxford, UK

<sup>3</sup> Department of Theoretical Physics, University of Debrecen, H-4010, Debrecen, Hungary

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## Abstract

A third-order linear homogeneous differential equation is first derived for the local density of  $s$ -states only,  $N_s(r, E)$ , for a general central potential  $V(r)$ . The major example presented is then for the bare Coulomb potential  $-Ze^2/r$ , for which analytical forms are obtained for both  $N_s(r, E)$  and for the total density of states  $N(r, E)$ . In the appendix, the repulsive linear potential case is also solved analytically.

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## 1. Background

For the Coulomb potential, available evidence strongly suggests that  $s$ -states are sufficient to solve the problem of the electron density  $\rho(r, E)$ , i.e., the number of electrons below energy  $E$  at a distance  $r$  from the nucleus. In particular, earlier work on bound states gave the spatial generalization of Kato's theorem as [1]

$$\frac{\partial \rho}{\partial r} = -\frac{2Z}{a_0} \rho_s(r) \quad (1)$$

and this was extended to the off-diagonal density, equivalent to the Dirac density matrix  $\gamma(\mathbf{r}, \mathbf{r}_0)$ , by Theophilou and March [2]. While equation (1) was derived specifically for a finite number of bound states, our concern here is with  $\rho(\mathbf{r}, E)$  when the energy  $E$  lies in the continuum.

## 2. Differential equation for local density of states

Starting then with the diagonal density  $\rho(r, E)$  and its  $s$ -state counterpart  $\rho_s(r, E)$ , let us define the corresponding local density of states as

$$N(r, E) = \frac{\partial \rho(r, E)}{\partial E} \quad N_s(r, E) = \frac{\partial \rho_s(r, E)}{\partial E} \quad (2)$$

respectively.

March and Murray in an early study [3] gave a differential equation for  $\rho_s(r, E)$  which reads, in atomic units in which  $m = 1$ ,  $\hbar = 1$ , and  $a_0 = 1$

$$\frac{1}{8} \frac{\partial^3}{\partial r^3} (r^2 \rho_s) - \frac{1}{2} \frac{\partial V}{\partial r} r^2 \rho_s - V \frac{\partial}{\partial r} (r^2 \rho_s) + \int_0^E E \frac{\partial^2}{\partial r \partial E} (r^2 \rho_s) dE = 0. \quad (3)$$

Therefore the  $s$ -state density of levels satisfies

$$\frac{1}{8} \frac{\partial^3}{\partial r^3} (r^2 N_s) + (E - V) \frac{\partial}{\partial r} (r^2 N_s) - \frac{V'}{2} r^2 N_s = 0. \quad (4)$$

It is readily verified that the solution of equation (4) with  $V = 0$  is

$$N_s^{(0)}(r, E) = \frac{1}{4\pi^2 k r^2} [1 - \cos(2kr)] \quad (5)$$

which is easily shown to be true by direct calculation for free electrons. In fact, equation (5) can be generalized for  $p$ ,  $d$ ,  $f$ , etc states, the results being set out in appendix A.

The main result, to be utilized below, is equation (4) giving the local density of  $s$ -states  $N_s(r, E)$  in the continuum at energy  $E$ , for a given central field represented by the potential energy  $V(r)$ .

### 3. Analytical solution of equation (4) for the bare Coulomb field

A general solution of equation (4) can be constructed in terms of Whittaker functions  $M$  and  $W$  (see [4]) for the case when  $V = -Z/r$ . The solution, having three arbitrary constants since equation (4) is third order, is

$$\begin{aligned} r^2 N_s(r, E) = & C_1 W \left( -\frac{iZ}{2} \sqrt{\frac{2}{E}}, \frac{1}{2}, 2i\sqrt{2Er} \right)^2 + C_2 M \left( -\frac{iZ}{2} \sqrt{\frac{2}{E}}, \frac{1}{2}, 2i\sqrt{2Er} \right)^2 \\ & + C_3 W \left( -\frac{iZ}{2} \sqrt{\frac{2}{E}}, \frac{1}{2}, 2i\sqrt{2Er} \right) M \left( -\frac{iZ}{2} \sqrt{\frac{2}{E}}, \frac{1}{2}, 2i\sqrt{2Er} \right). \end{aligned} \quad (6)$$

This can readily be verified by substitution of equation (6) into equation (4) and subsequent use of the differential equation satisfied by the Whittaker functions.

When the potential goes to zero ( $Z \rightarrow 0$ ), the Whittaker functions become

$$\begin{aligned} M(0, 1/2, i2\sqrt{2Er}) &= 2\Gamma(3/2) \sqrt{i2\sqrt{2Er}} I_{1/2}(i\sqrt{2Er}) \\ &= 2i \sin(\sqrt{2Er}) \end{aligned} \quad (7)$$

and

$$\begin{aligned} W(0, 1/2, i2\sqrt{2Er}) &= \left( \frac{i2\sqrt{2Er}}{\pi} \right)^{1/2} K_{1/2} \left( \frac{i2\sqrt{2Er}}{2} \right) \\ &= \exp \left( -i \frac{2\sqrt{2Er}}{2} \right) \end{aligned} \quad (8)$$

where the  $I$  and  $K$  are modified Bessel functions. Using equations (6)–(8), we can readily verify that in the limit  $Z \rightarrow 0$  equation (5) is regained, with appropriate choices of the constants  $C_1$ ,  $C_2$  and  $C_3$ . These are  $C_1 = C_3 = 0$  and  $C_2 = -1/8k\pi^2$ .

The next objective, having established the  $r$ -space dependence of the local density of  $s$ -states in equation (6), is to generalize this treatment to find the total density of states (essentially the weighted sum over all  $l$ -values from zero to infinity).

### 3.1. First-order calculation of the total density of states

Assuming the spatial generalization of Kato's theorem in equation (1) also applies in the continuum, a point we shall return to below, we have

$$\frac{\partial \rho(r, E)}{\partial r} = -\frac{2Z}{a_0} \rho_s(r, E) \quad (9)$$

and taking an energy derivative, it follows that

$$\frac{\partial N(r, E)}{\partial r} = -\frac{2Z}{a_0} N_s(r, E). \quad (10)$$

Hence

$$N(r, E) - N_0(E) = \frac{2Z}{a_0} \int_r^\infty N_s(r, E) dr \quad (11)$$

where  $N_0(E)$  is the total density of states for free electrons.

The considerable merit of equation (11) is that the right-hand side already has a factor  $Z$ , and hence to calculate  $N(r, E)$  to first order in  $Z$  we can insert for  $N_s(r, E)$  on the right-hand side the known free-electron result (5). Thus we find for  $N(r, E)$  to first order in  $Z$  the result

$$N(r, E) = N_0(E) + \frac{Z}{a_0} \frac{1}{2\pi k} \int_r^\infty \frac{1 - \cos(2kr)}{r^2} dr. \quad (12)$$

Now from [5]

$$\int \frac{\cos x}{x^2} dx = -\frac{\cos x}{x} - \text{Si}(x) \quad \text{Si}(x) = \int \frac{\sin x}{x} dx. \quad (13)$$

Hence we have that

$$N(r, E) = N_0(E) + \frac{Z}{a_0} \frac{1}{2\pi k} \left[ \frac{1}{r} - \left\{ \frac{\cos(2kr)}{r} - 2k \int_{2kr}^\infty \frac{\sin x}{x} dx \right\} \right]. \quad (14)$$

For selected values of energy  $E$ , a plot of this local density of states as a function of  $r$  is shown in figure 1 in the form of  $[N(r, E) - N_0(E)]/Z$ . Taking the limit  $r \rightarrow 0$  of this quantity from equation (14), the value  $1/2$  is obtained.

Of course, equation (14) has been derived assuming that equation (9) applies in the continuum. Though we have no general proof that this is so, we can alternatively calculate  $N(r, E)$  from the linear response theory of March and Murray [3], which did not invoke Kato's theorem. From the study of March and Murray, when  $V(r) = -Ze^2/r$ , we have, to first order in  $Z$ ,

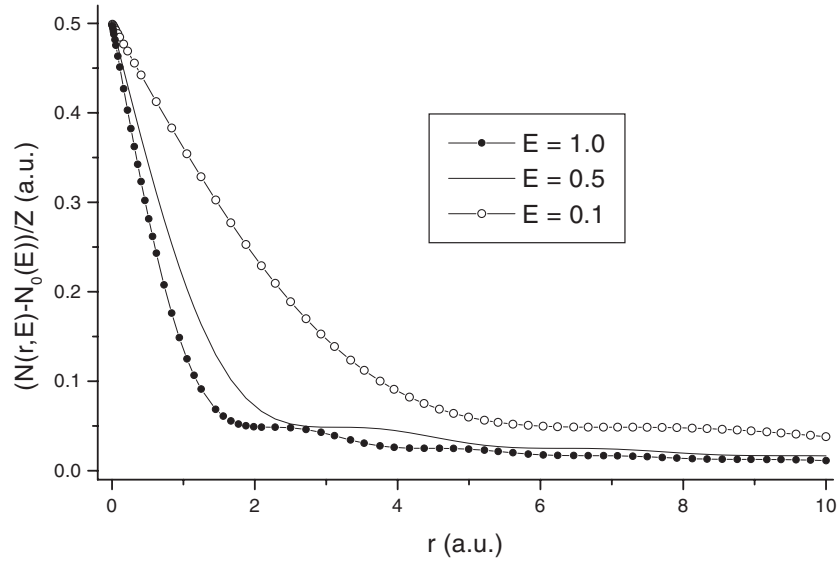
$$\rho(r, E) - \rho_0(E) = -\frac{mk^2}{4\pi^3 \hbar^2} \int \frac{j_1(2kr_1)}{r_1^2} \left( \frac{-Ze^2}{|\mathbf{r} - \mathbf{r}_1|} \right) d\mathbf{r}_1. \quad (15)$$

But the integral term has an electrostatic interpretation as the potential created by a 'charge density'  $j_1(2kr)/r^2$ . The total charge  $Q(r)$  enclosed by a sphere of radius  $r$  is then

$$\begin{aligned} Q(r) &= \int_0^r \frac{j_1(2kr_1)}{r_1^2} 4\pi r_1^2 dr_1 = -4\pi \int_0^{2kr} \frac{\partial}{\partial(2kr_1)} j_0(2kr_1) \frac{d(2kr_1)}{2k} \\ &= -\frac{2\pi}{k} [j_0(2kr) - 1]. \end{aligned} \quad (16)$$

But from Poisson's equation, the electrostatic potential  $V_{\text{es}}$  created by  $Q(r)$  is

$$V_{\text{es}} = -\int_r^\infty \frac{Q(r)}{r^2} dr \quad (17)$$



**Figure 1.** Plot of result for local density of states  $N(r, E)$  generated by a bare Coulomb potential  $-Ze^2/r$ , as given by equation (14). Note that, although there is some structure in the curves, which is different for the different continuum energies  $E$  chosen,  $\partial N(r, E)/\partial r$  is always  $\leq 0$ , in accord with the spatial generalization of Kato's theorem asserted in equation (21). Of course, this plot is valid only to first order in the Coulomb potential (see figure 3 below for a non-perturbative plot of the local density of states).

therefore, we can write

$$\rho(r, E) - \rho_0(E) = \frac{mk^2}{4\pi^3\hbar^2} Ze^2 \int_r^\infty \frac{Q(r)}{r^2} dr \quad (18)$$

or

$$\frac{\partial \rho}{\partial r} = -\frac{mk^2}{4\pi^3\hbar^2} Ze^2 \frac{Q(r)}{r^2} = \frac{mk^2}{4\pi^3\hbar^2} \frac{Ze^2}{r^2} \frac{2\pi}{k} [j_0(2kr) - 1]. \quad (19)$$

But the  $s$ -density for free electrons is given by (compare equation (5)):

$$\rho_0(r, E) = \frac{k}{4\pi^2 r^2} \left[ 1 - \frac{\sin(2kr)}{2kr} \right] \quad (20)$$

and therefore by comparing equations (19) and (20) it follows that

$$\frac{\partial \rho(r, E)}{\partial r} = -\frac{2Z}{a_0} \rho_0(r, E) \quad (21)$$

which proves the assumption of the spatial generalization of Kato's theorem to first order in  $Z$  for a bare Coulomb potential.

### 3.2. Total density of states to all orders in nuclear charge

Evidently, but now more formally, we can use equation (11) in conjunction with the exact form of  $N_s(r, E)$  in terms of the Whittaker functions  $M$  and  $W$ . Since the 'constants' of integration  $C_1$ ,  $C_2$  and  $C_3$  ( $C_1$  and  $C_3$  zero for  $Z = 0$ ) depend only on  $E$  and  $Z$  (at most), we

have immediately

$$\begin{aligned} \frac{a_0}{2Z} [N(r, E) - N_0(E)] = & C_1 \int_r^\infty \frac{W^2(-iZ/k, 1/2, 2ikr)}{r^2} dr \\ & + C_2 \int_r^\infty \frac{M^2(-iZ/k, 1/2, 2ikr)}{r^2} dr \\ & + C_3 \int_r^\infty \frac{M(-iZ/k, 1/2, 2ikr)W(-iZ/k, 1/2, 2ikr)}{r^2} dr. \end{aligned} \quad (22)$$

Having established equations (6) and (15) for  $N_s(r, E)$  and the total density of states, let us return to the explicit evaluation of  $N_s(r, E)$ , given in equation (26) immediately below.

Let us employ first the physical boundary condition that  $N_s(r \rightarrow 0, E)$  tends to a nonzero function of  $E$ . Then we must isolate the parts on the right-hand side of equation (6) which tend to zero as  $r^2$ . The limiting behaviour of  $W^2$  is found from [4] to be

$$W^2(-iZ/k, 1/2, 2ikr) \rightarrow [\Gamma(1 + iZ/k)]^2 \quad r \rightarrow 0. \quad (23)$$

Such behaviour from the term multiplying  $C_1$  cannot be cancelled by the terms involving  $C_2$  and  $C_3$  since the result corresponding to equation (23) for the function  $M^2$  is

$$M^2(-iZ/k, 1/2, 2ikr) \rightarrow 0 \quad r \rightarrow 0. \quad (24)$$

Thus the first conclusion is that  $C_1$  must be put equal to zero, as was found above to be the case also for the free-electron limit  $Z \rightarrow 0$ .

The next question to resolve is whether  $C_3$  must also be put to zero, as in the free-electron case discussed above. To decide the answer, one needs to extend the result (24) to determine the manner in which  $M^2 \rightarrow 0$  as  $r \rightarrow 0$ . A hint is already available from equation (7) for free electrons, which shows that

$$M_{Z=0}^2(r, E) \rightarrow -8Er^2. \quad (25)$$

If equation (25), giving  $M^2(r, E) \rightarrow f(E, Z)r^2$  applies for all  $Z$ , then evidently  $C_3$  must be put to zero as for free electrons. This is in fact the case, from properties of the Whittaker functions, for arbitrary  $Z$ . Then the desired physical result for the local density of  $s$ -states is given by

$$N_s(r, E) = C_2 \frac{M^2\left(-\frac{iZ}{2}\sqrt{\frac{2}{E}}, \frac{1}{2}, 2i\sqrt{2Er}\right)}{r^2}. \quad (26)$$

This result for  $N_s/C_2$  is plotted versus  $r$  in figure 2 for a few values of the energy  $E$ , for the case  $Z = 10$ . Equation (26) reduces to the free-electron limit  $N_s^{(0)}(r, E)$  in equation (5) when  $C_2(Z = 0) = -1/8k\pi^2$ , with  $k = \sqrt{2E}$ , using the limiting form of  $M$  as  $Z \rightarrow 0$  in equation (7). The form for small  $r$  is

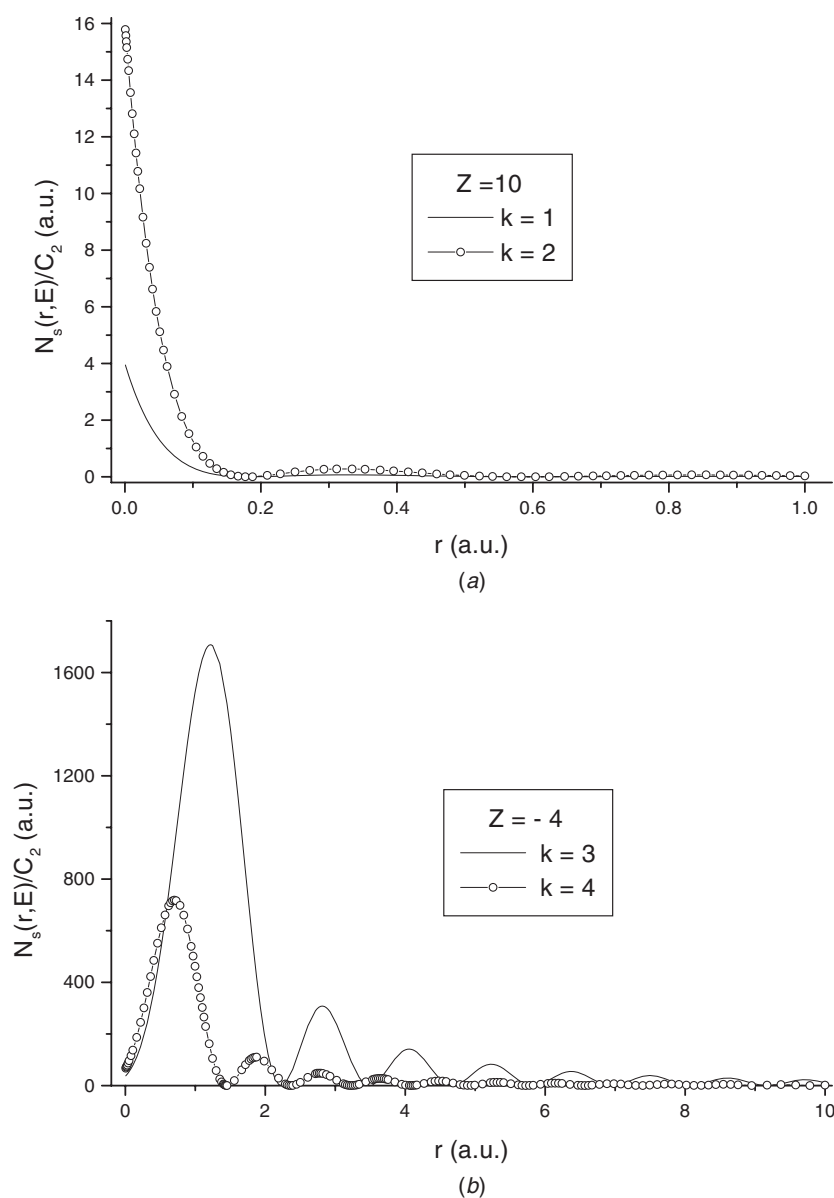
$$N_s(r, E) = C_2 \left[ -8E + 16EZr - \frac{8}{3}E(-2E + 5Z^2)r^2 + O(r^3) \right]. \quad (27)$$

Finally, we have inserted the result (26) into equation (11) to obtain  $[N(r, E) - N_0(E)]/C_2$  and this quantity is displayed as a function of  $r$ , for a few values of  $E$ , and  $Z = 10$ , in figure 3.

#### 4. Summary and future directions

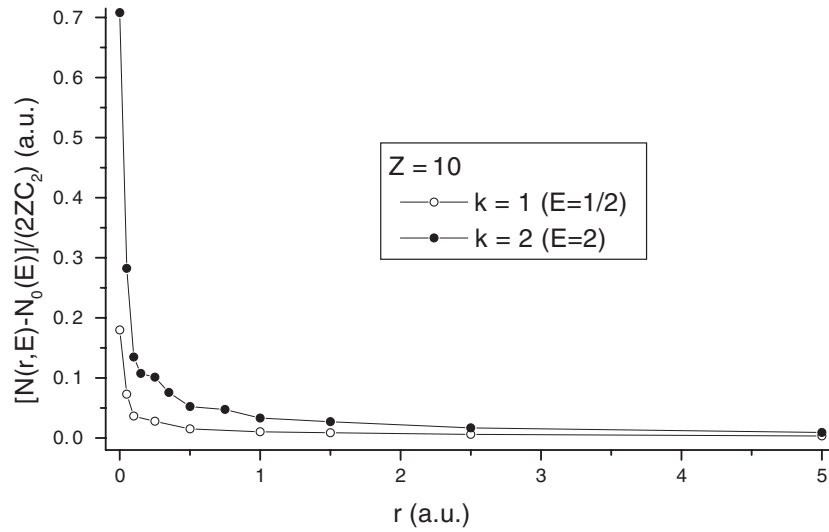
The main results of the present study are as follows:

- (i) The third-order linear homogeneous differential equation (4) for the local density of states  $N_s(r, E)$  for  $s$ -waves only with  $E$  in the continuum, for the central potential  $V(r)$ ;
- (ii) Its solution (5) for free electrons;



**Figure 2.** (a) Shows plot of local density of  $s$ -states,  $N_s(r, E)$  versus  $r$  for two energies  $E = k^2/2$ , and for  $Z = 10$ . The ratio of the values of the two curves is correctly given in the plots shown. (b) For contrast with the attractive potential corresponding to  $Z = 10$  in figure 2(a), the plot shown is for a repulsive potential corresponding to  $Z = -4$ . Again the ratio of the values of the two curves shown gives the ratio of the  $s$ -state (only) density of states  $N_s(r, E)$ , with  $E = k^2/2$ . Note that, as in figure 2(a), there is a finite slope at the origin (see equation (27)).

- (iii) The general solution (6), with three arbitrary quantities  $C_i(Z, E)$ ,  $i = 1, 2$  and  $3$ , for the Coulomb potential  $-Z/r$ ;
- (iv) The physical solution for  $N_s(r, E)$  in equation (26) for arbitrary  $Z$ , i.e., for both an attractive potential with an infinite number of bound states, plus continuum states, and for negative  $Z$ , for which, of course, there are only states with  $E > 0$ .



**Figure 3.** Local density of states obtained non-perturbatively, by assuming that the spatial generalization of Kato's theorem applies for energies  $E$  lying in the continuum. Note that this result (21) has been proved to first order in the perturbing charge  $Ze$  in section 3.1. It also follows at  $r = 0$  for all  $Z$  from equation (27) and the results of appendix A. The plot in this figure is to be compared with the perturbative results in figure 1, the value of  $Z = 10$  being adopted in the non-perturbative calculations.

It would be of interest for future work if physical solutions of equation (4) could be found for other, more general, potentials, such as the screened Coulomb form  $V(r) = -(Z/r) \exp(-qr)$ . Presently it is our impression that this case would need numerical solution of the basic equation (4). However, analytical solution is possible for the linear potential  $V \propto r$ , and the results are recorded in appendix B.

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### Appendix A. General solution of free particle and Coulomb local density of states for arbitrary orbital angular momentum quantum number $l$

The general solution for the density of states for an arbitrary quantum number  $l$  for free particles (limit  $Z \rightarrow 0$ ) is given by

$$N_l(r) = C_1 \frac{J_{l+1/2}^2(kr)}{r} + C_2 \frac{Y_{l+1/2}^2(kr)}{r} + C_3 \frac{J_{l+1/2}(kr)Y_{l+1/2}(kr)}{r} \quad (\text{A1})$$

or in terms of spherical Bessel and spherical Neumann functions (written as  $j_l(z)$  and  $\bar{n}_l(z)$  respectively):

$$N_l(r) = \frac{2C_1 k}{\pi} j_l^2(kr) + \frac{2C_2 k}{\pi} \bar{n}_l^2(kr) + \frac{2C_3 k}{\pi} j_l(kr)\bar{n}_l(kr). \quad (\text{A2})$$



March and Murray gave for the integrated density of states for  $l = 1$  the form

$$\rho_1(r, E) = \frac{1}{4\pi r^2} \left\{ k + \frac{\sin(2kr)}{2r} + \frac{\cos(2kr) - 1}{kr^2} \right\}.$$

Hence,

$$\frac{\partial \rho_1}{\partial E} \equiv \frac{1}{k} \frac{\partial \rho_1}{\partial k} = \frac{1}{4\pi k r^2} \left[ \left( 1 + \frac{1}{k^2 r^2} \right) + \left( 1 - \frac{1}{k^2 r^2} \right) \cos(2kr) - 2 \frac{\sin(2kr)}{kr} \right]. \quad (\text{A3})$$

Thus, for  $l = 1$ , this must be equated to equation (A2) to yield

$$\begin{aligned} N_1(r, E) = \frac{2C_1 k}{\pi} \left[ \frac{\sin^2 kr}{(kr)^4} - 2 \frac{\sin kr \cos kr}{(kr)^3} + \frac{\cos^2 kr}{(kr)^2} \right] \\ + \frac{2C_2 k}{\pi} \left[ \frac{\cos^2 kr}{(kr)^4} + 2 \frac{\sin kr \cos kr}{(kr)^3} + \frac{\sin^2 kr}{(kr)^2} \right] \end{aligned} \quad (\text{A4})$$

where  $C_3$  has already been put equal to zero because of the presence of a term more singular at the origin than any terms involving  $C_1$  and  $C_2$ . But on closer inspection,  $C_2$  must also equal zero because of different singular behaviour at  $r = 0$ . Thus, equation (A4) with  $C_2$  put to zero has to be compared with equation (A3), and to do so we rearrange (A4) to read

$$\begin{aligned} N_1(r, E) = \frac{2C_1 k}{\pi} \left[ \frac{1 - \cos 2kr}{2(kr)^4} - \frac{\sin 2kr}{(kr)^3} + \frac{1 + \cos 2kr}{2(kr)^2} \right] \\ = \frac{C_1}{\pi k r^2} \left[ \left( 1 + \frac{1}{k^2 r^2} \right) + \left( 1 - \frac{1}{k^2 r^2} \right) \cos 2kr - 2 \frac{\sin 2kr}{kr} \right] \end{aligned} \quad (\text{A5})$$

which agrees with equation (A3) if  $C_1$  is chosen to equal  $1/4$ .

For the Coulomb potential, the general solution for the density of states for an arbitrary quantum number  $l$  is given by

$$\begin{aligned} r^2 N_l(r, E) = C_1 W \left( -\frac{iZ}{2} \sqrt{\frac{2}{E}}, l + \frac{1}{2}, 2i\sqrt{2Er} \right)^2 + C_2 M \left( -\frac{iZ}{2} \sqrt{\frac{2}{E}}, l + \frac{1}{2}, 2i\sqrt{2Er} \right)^2 \\ + C_3 W \left( -\frac{iZ}{2} \sqrt{\frac{2}{E}}, l + \frac{1}{2}, 2i\sqrt{2Er} \right) M \left( -\frac{iZ}{2} \sqrt{\frac{2}{E}}, l + \frac{1}{2}, 2i\sqrt{2Er} \right) \end{aligned} \quad (\text{A6})$$

with  $C_1$ ,  $C_2$  and  $C_3$  being constants. Considerations similar to those discussed in section 3 lead one to set  $C_1 = C_3 = 0$ . It is not difficult to expand the expression for  $r^2 N_l(r, E)$  above to low order in  $r$ , and then to sum (multiplying by the appropriate degeneracy factor  $2l + 1$ ) over all  $l$  from zero to infinity to find a total density of states  $N_\infty(r, E)$  going as

$$r^2 N_\infty(r, E)/C_2 \simeq \frac{-16E^4 r^2}{(16E^4 r^2 + 1)} + 2rZ \ln(16E^4 r^2 + 1). \quad (\text{A7})$$

for small  $r$ .

## Appendix B. Solution for local density of states for the linear potential $V(r) \propto r$

For a potential  $V(r) = cr$ , with  $c$  being a (negative) constant corresponding to a repulsive potential, a solution of equation (6) can be found in terms of the Airy functions Ai and Bi as

$$\begin{aligned} N_s(r, E) = C_1 \frac{\text{Ai}\left(-\frac{(-2c)^{1/3}(-E+cr)}{c}\right)^2}{r^2} + C_2 \frac{\text{Bi}\left(-\frac{(-2c)^{1/3}(-E+cr)}{c}\right)^2}{r^2} \\ + C_3 \frac{\text{Bi}\left(-\frac{(-2c)^{1/3}(-E+cr)}{c}\right) \text{Ai}\left(-\frac{(-2c)^{1/3}(-E+cr)}{c}\right)}{r^2} \end{aligned} \quad (\text{B1})$$

as can be verified by substitution and subsequent use of the differential equation satisfied by the Airy functions. Since the Airy functions can be converted to the modified Bessel functions  $I$  and  $K$ , with

$$\text{Ai}(z) = \frac{1}{\pi} \sqrt{\frac{z}{3}} K\left(\frac{1}{3}, \frac{2z^{3/2}}{3}\right) \quad (\text{B2})$$

and

$$\text{Bi}(z) = \sqrt{\frac{z}{3}} \left( I\left(-\frac{1}{3}, \frac{2z^{3/2}}{3}\right) + I\left(\frac{1}{3}, \frac{2z^{3/2}}{3}\right) \right) \quad (\text{B3})$$

the solution can equally well be written in terms of Bessel functions of order  $\pm 1/3$ .

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