

# Dynamic polarizability of Rydberg states in the Kramers approximation and their shifts in an external radiation field

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An analytic expression is obtained for the dynamic polarizability averaged over the angular quantum numbers of Rydberg atoms. Shifts of highly excited levels are considered as well as lines due to transitions between them in external Planck and nonthermal (power-law) radiation fields.

## 1. INTRODUCTION

The interaction between highly excited (Rydberg) atoms with external fields is of considerable interest from the general physics point of view (in view of the anomalously high sensitivity of Rydberg atoms to external perturbations) and also in the case of applications, particularly in astrophysics. Under conditions encountered in the interstellar medium there are rf recombination lines between states with the principal quantum numbers in the range  $n > 700$  (see, for example, Refs. 1 and 2). In applications of this kind there is special interest in the atomic characteristics averaged over the angular quantum numbers.

In the case of slowly varying electric fields (when the characteristic field frequency is  $\omega \rightarrow 0$ ) the dynamic polarizability reduces to the static one, the expression for which is given, for example, in Ref. 3. In the other limiting case ( $\omega \rightarrow \infty$ ) the problem reduces to a discussion of the motion of a free electron (see, for example, Ref. 4). A more detailed investigation of the dynamic polarizability in the limit  $\omega \rightarrow \infty$  using, in particular, an asymptotic expansion in reciprocal powers of the frequency, can be found in Refs. 5–9. Analytic expressions for the dynamic polarizability of the hydrogen atom in the states with  $n = 1$  and 2 in terms of hypergeometric functions can be found in Refs. 4, 10, and 11. The expression describing the dynamic polarizability of Rydberg states as a series of Bessel functions, asymptotic with respect to  $n$ , is given in Ref. 12. Calculations of the dynamic polarizability of Rydberg states and of their shift in a Planck radiation field (corresponding to a temperature 300 K) are reported in Ref. 13.

We use the familiar Kramers expression for the oscillator strength and find the dynamic polarizability of a Rydberg atom averaged over the angular quantum numbers. We use elementary functions and consider the shifts of Rydberg levels and of lines due to transitions between them in an external radiation field. A quite unexpected result is that in the limit  $n \rightarrow \infty$  for a smooth function describing the radiation density the expression for the polarizability is a differential operator acting on a function describing the radiation density.

If the spectrum of the external radiation obeys a power law, then the shifts of Rydberg levels (and of the lines due to transitions between them) are power-law functions of the principal quantum number  $n$ . In the case of a Planck spectrum the shift is a function of a parameter  $\beta = \delta/kT$  ( $\delta = 2Z^2 \text{ Ry}/n^3$  is the separation between the consecutive levels of  $n$  and  $n \pm 1$ ;  $Z$  is the spectroscopic symbol; for a

neutral atom  $Z = 1$ ;  $T$  is the radiation temperature) and it can be approximated by a rational expression.

## 2. DYNAMIC POLARIZABILITY OF RYDBERG STATES

The polarizability  $\kappa$  of an atom with the principal quantum number  $n$  in an external electric field of frequency  $\omega$  can be written in the form (see, for example, Ref. 14):

$$\kappa = \frac{4a_0^3}{Z^4} \sum_{n' \neq n} \frac{f_{n, n'}}{\Delta \varepsilon_{n, n'}^2 - (\hbar\omega/Z^2 \text{ Ry})^2}, \quad (1)$$

where  $a_0$  is the Bohr radius;  $\Delta \varepsilon_{n, n'} = \varepsilon_{n'} - \varepsilon_n$  is the difference between the energies of the levels  $n$  and  $n'$  in units of  $Z^2 \text{ Ry}$  (the energy of a level is  $E_n = \varepsilon_n Z^2 \text{ Ry}$ ,  $\varepsilon_n = 1/n^2$ );  $f_{n, n'}$  is the oscillator strength of the  $n \rightarrow n'$  transition. To within less than 25%, the oscillator strengths for Rydberg states are described by the Kramers expression

$$f_{n, n'} = \frac{32}{3^{3/2}\pi} \frac{1}{n^2} \left[ \frac{nn'}{s(n+n')} \right]^3, \quad (2)$$

where  $s \equiv n' - n$ . Since the quantities  $f_{n, n'}$  decrease proportionally to  $1/|s|^3$  as a function of the difference between the quantum numbers, we can assume that the main contribution to the sum in Eq. (1) is made by transitions with low values of  $s$ . Therefore, all the quantities occurring in Eq. (1) can be expanded in powers of  $s/n$ :

$$\begin{aligned} f_{n, n+s} &\approx \frac{4}{3^{3/2}\pi} \frac{n}{s^3} \left( 1 + \frac{3}{2} \frac{s}{n} \right), \\ \Delta \varepsilon_{n, n+s} &\approx \frac{2s}{n^3} \left( 1 - \frac{3}{2} \frac{s}{n} \right). \end{aligned} \quad (3)$$

Since  $f_{n, n+s} \approx -f_{n, n-s}$ , is accurate to within  $s/n$ , it follows that the first nonvanishing term of the expansion for Eq. (1) can be found by retaining at least two terms in the expansions of Eq. (3). Substituting Eq. (3) into Eq. (1), we have

$$\begin{aligned} \kappa &= \frac{4a_0^3 n^6}{3^{3/2} Z^4 \pi} \left\{ \sum_{s=1}^N \frac{1}{s^2 (s^2 - q^2)} \right. \\ &+ \left. \frac{n}{3} \sum_{s=1}^N \frac{1}{s^3} \left[ \frac{1}{s^2 [1 - 3/2 (s/n)]^2 - q^2} - \frac{1}{s^2 [1 + 3/2 (s/n)]^2 - q^2} \right] \right\}, \end{aligned} \quad (4)$$

where  $q = \hbar\omega/\delta$ . Strictly speaking, the upper limit of summation  $N$  in Eq. (4) should satisfy the condition  $N < n$ , but

since  $n \gg 1$  holds and all the sums converge rapidly, we can equally accurately assume that  $N = \infty$ .

We consider two cases in the subsequent transformations. The first corresponds to a situation in which the field frequency  $\omega$  is far from any resonance. The second, when the spectral density of the external field  $U(\omega)$  is a smooth function, which plays an important role in the vicinity of resonances.

In the first case we can expand the second sum in Eq. (4) in powers of  $s/n$ :

$$\kappa = \frac{4a_0^3 n^6}{3^{1/2} Z^4 \pi} \left\{ \sum_{s=1}^{\infty} \frac{1}{s^2 (s^2 - q^2)} + \sum_{s=1}^{\infty} \frac{2}{(s^2 - q^2)^2} \right\}. \quad (5)$$

Introducing now the functions

$$f_c(q) = \sum_{s=1}^{\infty} \frac{1}{(s^2 - q^2)} = \frac{1 - \pi q \operatorname{ctg}(\pi q)}{2q^2},$$

$$\Phi(q) = \sum_{s=1}^{\infty} \frac{1}{s^2 (s^2 - q^2)} = \frac{f_c(q) - f_c(0)}{q^2} \quad (6)$$

$$= \frac{1 - \pi q \operatorname{ctg}(\pi q) - (\pi q)^2/3}{2q^4},$$

and bearing in mind that

$$\sum_{s=1}^{\infty} \frac{1}{(s^2 - q^2)^2} = \frac{1}{2q} \frac{d}{dq} f_c(q),$$

we obtain

$$\kappa = \frac{4a_0^3 n^6}{3^{1/2} Z^4 \pi} \left\{ \Phi(q) + \frac{1}{q} \frac{d}{dq} f_c(q) \right\}. \quad (7)$$

In the second case, bearing in mind subsequent integration with respect to the external field frequency, we employ a relationship which is valid in the case of smooth functions  $f(x)$  and  $\xi \rightarrow 0$ :<sup>1)</sup>

$$\int_0^{\infty} \frac{f(x) dx}{a^2 (1 - \xi)^2 - x^2} - \int_0^{\infty} \frac{f(x) dx}{a^2 (1 + \xi)^2 - x^2} = \frac{1}{1 - \xi} \int_0^{\infty} \frac{f(x(1 - \xi)) dx}{a^2 - x^2}$$

$$- \frac{1}{1 + \xi} \int_0^{\infty} \frac{f(x(1 + \xi)) dx}{a^2 - x^2} \approx -2\xi \int_0^{\infty} x^2 \frac{d}{dx} \left( \frac{f(x)}{x} \right) \frac{dx}{a^2 - x^2}.$$

The interaction of an atom with a radiation field can then be described by

$$\hat{\kappa} U = \frac{4a_0^3 n^6}{3^{1/2} Z^4 \pi} \Phi(q) \left[ -q^3 \frac{d}{dq} \left( \frac{U(q)}{q^2} \right) \right], \quad (8)$$

i.e., in fact, the dynamic polarizability is a differential operator acting on a function describing the spectral density of the radiation field. The general form of the universal function  $\Phi(q)$  is shown in Fig. 1.

We now consider the behavior of the expressions given by Eqs. (4) and (7) in the limiting cases of high and low frequencies. If  $\omega \rightarrow \infty$ , we obviously have

$$\Phi(q) \xrightarrow{q \rightarrow \infty} -\frac{\pi^2}{6} \frac{1}{q^2},$$

$$\kappa \rightarrow -4a_0^3 \left( \sum_{n'} f_{n,n'} \right) \left( \frac{\operatorname{Ry}}{\hbar \omega} \right)^2 = -4a_0^3 \frac{2\pi}{3^{3/2}} \left( \frac{\operatorname{Ry}}{\hbar \omega} \right)^2.$$

The factor  $2\pi/3^{3/2} \approx 1.2$ , by which the sum of the oscillator strengths in the Kramers approximation differs from unity, is due to the error introduced by Eq. (2) at low values of  $s$ . In the limit  $\omega \rightarrow 0$ , the function  $\kappa(\omega)$  reduces to the usual polarizability. Averaging of the known expressions for the polarizability of a state with the parabolic quantum numbers  $n_1, n_2, m$  over all the numbers with the exception of  $n$  gives

$$\kappa(0) = \frac{a_0^3}{Z^4} \frac{15n^6 + 21n^4}{8}.$$

It follows from the expressions (4) and (7) that

$$\kappa(0) = \frac{2\pi^3}{15 \cdot 3^{3/2}} \frac{a_0^3}{Z^4} n^6,$$

where in the case of  $n \gg 1$  these expressions differ by a factor amounting to 1.27. If we write down the expression for the polarizability in the form

$$\kappa = Q \kappa^{\text{Kr}}, \quad (9)$$

where  $\kappa^{\text{Kr}}$  is the result obtained in the Kramers approximation and the correction factor  $Q$  is an analog of the Gaunt factor assumed to be  $Q = 3^{3/2}/2\pi$  (so as to ensure that the sum of the oscillator strengths is unity), we find that Eq. (9) differs from the exact limit when  $n \rightarrow \infty$  and  $\omega \rightarrow 0$  by a factor 1.05.

### 3. PLANCK RADIATION FIELD

The shift of a level in an external radiation field can be written down with the aid of Eq. (8):

$$\delta\omega = -\frac{2\pi}{\hbar} \int_0^{\infty} \hat{\kappa} U(\omega) d\omega$$

$$= \frac{8\pi^2}{3^{1/2} \hbar} \frac{a_0^3}{Z^4} n^6 Q \int_0^{\infty} \Phi(q) \left[ q^3 \frac{d}{dq} \left( \frac{U(q)}{q^2} \right) \right] d\omega. \quad (10)$$

The definition of Eq. (10) is such that a positive shift  $\delta\omega > 0$  denotes an upward shift of the level. Hence, if the Planck radiation density is

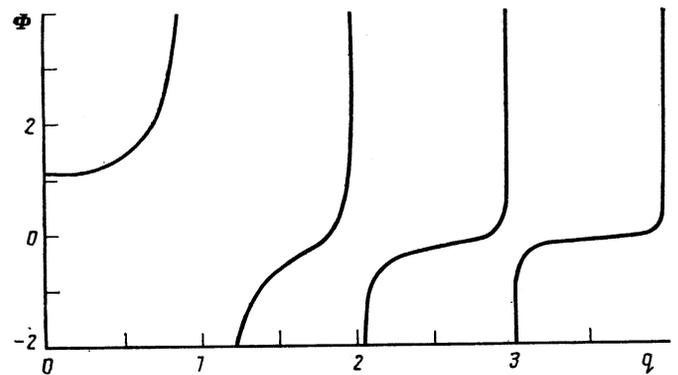


FIG. 1. Function  $\Phi(q)$  for the dynamic polarizability: the abscissa represents  $q = \hbar\omega/\delta$ .

$$U(\omega) = \frac{\hbar\omega^3}{\pi^2 c^3} \left[ \exp\left(\frac{\hbar\omega}{kT}\right) - 1 \right]^{-1},$$

we find that

$$\delta\omega = -\Delta_T Q \theta(\beta), \quad \Delta_T = \alpha^3 \frac{kT}{\hbar} \frac{kT}{Ry}, \quad \beta = \frac{\delta}{kT}, \quad \delta = \frac{2Z^2 Ry}{n^3}, \quad (11)$$

where  $\alpha = 1/137$  is the fine-structure constant and the function  $\theta(\beta)$  is given by

$$\theta(\beta) = \frac{4}{3^{3/2}\pi^2} \beta^2 \int_0^\infty \Phi(q) \left[ -q^3 \frac{d}{dq} \frac{q}{e^{-\beta q} - 1} \right] dq, \quad (12)$$

and has the following asymptotic forms:

$$\theta(\beta) \rightarrow \begin{cases} -\pi^2/3^{3/2} = -0,6331, & \beta \rightarrow 0 \\ (2\pi^6/25 \cdot 3^{3/2}) \beta^{-2} = 4,934\beta^{-2}, & \beta \rightarrow \infty \end{cases} \quad (13)$$

The limit  $\beta \rightarrow 0$  with the additional factor  $Q$  is equal to the exact result, given, for example, in Ref. 13. In general, the integral (12) can be found only by numerical methods. In view of the presence of the poles in the integrand, numerical integration is a nontrivial matter. This difficulty is bypassed by an approximation of the regular part of the function in the interval  $(x_k, x_{k+1})$  using the expression

$$f(x) = f_k + \frac{f_{k+1} - f_k}{h} \frac{\sin(\pi(x-x_k))}{\pi},$$

where  $h \equiv x_{k+1} - x_k$ ,  $f_k \equiv f(x_k)$ , and then  $\int f(x) \cot(\pi x) dx$  is calculated using the sum

$$\frac{1}{\pi} \sum f_k \ln \left| \frac{\sin(\pi x_{k+1})}{\sin(\pi x_k)} \right| + \frac{f_{k+1} - f_k}{\pi h} \left[ \sin(\pi h) - \sin(\pi x_k) \ln \left| \frac{\operatorname{tg}(\pi x_{k+1}/2)}{\operatorname{tg}(\pi x_k/2)} \right| \right].$$

The method of least squares can be used to approximate Eq. (12) with the expression

$$\theta(\beta) = \frac{-0,6331 + 0,336\beta^2 + 0,00077\beta^4}{1 + 0,1722\beta^2 - 0,028\beta^4 + 0,000156\beta^6}. \quad (14)$$

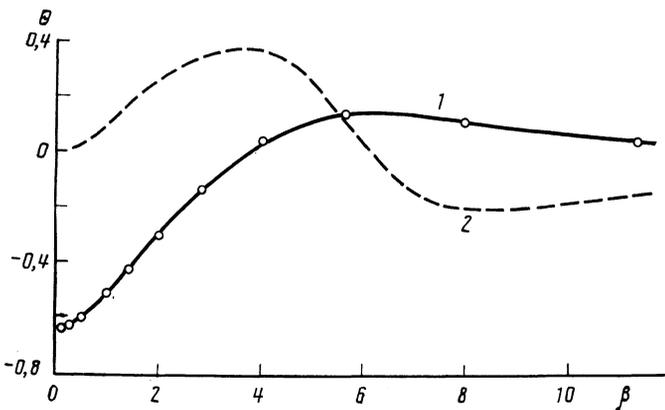


FIG. 2. Function  $\theta(\beta)$  [curve 1 represents the approximation by Eq. (14) and the points represent Eq. (12)] and the function  $\beta^{4/3} \theta'(\beta) 2^{1/3}$  (2) for the shift of Rydberg levels in an external Planck field; the abscissa represents  $\beta = \delta/kT$ .

Equation (14) has the same asymptotic forms as Eq. (12). The function  $\theta(\beta)$  and the errors of the approximation of Eq. (14) for intermediate values of  $\beta$  are shown in Fig. 2. It follows from Eqs. (11) and (14) (see also Fig. 2) that the maximum shift of the level on the scale of  $n$  is reached in the limit  $n \rightarrow \infty$  and it amounts to  $(\pi/6)\Delta_T$ . The minimum shift is negative and it corresponds to  $\beta \approx 6.2$  [ $n_{\min} \approx 0.7(Ry/kT)^{1/3}$ ] and amounts to  $-0.12\Delta_T$ . The shift vanishes for  $\beta \approx 4$  [ $n \approx 0.8(Ry/kT)^{1/3}$ ]. At room temperature (300 °K) and for  $Z = 1$ , we have  $n_{\min} \approx 5-6$ .

It is of interest to compare the shift of Eq. (11) with the additional radiation broadening of the level  $n$  due to absorption and stimulated emission. Using once again the Kramers approximation for these processes, we obtain

$$W_n = \frac{1}{n^2} \sum_{n'} \frac{2A_0(\epsilon\epsilon')^{1/2}}{|e-\epsilon'|} \frac{1}{\exp[(\Delta\epsilon)Z^2 Ry/kT] - 1} \approx \frac{2A_0}{n^5} \frac{1}{\beta} \sum_{s=1}^{\infty} \frac{\beta}{\exp(\beta s) - 1} \approx \alpha^3 \frac{1}{Z^2 n^2} \frac{kT}{\hbar} w(\beta), \quad (15)$$

where

$$A_0 = \alpha^3 \frac{16}{3^{3/2}\pi} Z^4 \frac{Ry}{\hbar}, \quad w(\beta) = \frac{16}{3^{3/2}} \sum_{s=1}^{\infty} \frac{\beta}{\exp(\beta s) - 1}.$$

The function  $w(\beta)$  has the following asymptotes:

$$w(\beta) = \frac{16}{3^{3/2}\pi} \begin{cases} \pi^2/6 = 1,645, & \beta \rightarrow 0 \\ \beta \exp(-\beta), & \beta \rightarrow \infty \end{cases} \quad (16)$$

In general, the sum in the above expression cannot be calculated analytically. Its approximation, relying on the asymptotic forms (16) and bearing in mind that in the limit  $\beta \rightarrow 0$  the derivative  $w(\beta)$  has a logarithmic singularity, can be written in the form

$$w(\beta) = \frac{1,612 + 4\beta^2}{1 + 4\beta(e^\beta - 1) + \beta \ln(1 + 0,4/\beta)}. \quad (17)$$

The quality of the approximation for intermediate values of  $\beta$  is illustrated in Fig. 3. The ratio of the shift of a level to its width is

$$\frac{\delta\omega_n}{W_n} = -\frac{kT}{E_n} \frac{\theta(\beta)}{w(\beta)}, \quad \frac{\theta(\beta)}{w(\beta)} \rightarrow \begin{cases} \pi/8, & \beta \rightarrow 0 \\ -(\pi^2/600) \exp \beta/\beta^3, & \beta \rightarrow \infty \end{cases} \quad (18)$$

Therefore, in the case of extremely high temperatures  $kT \gg \delta$  the ratio of the shift to the radiation width of a level is proportional to  $T$  and  $n^2$ , and then the level shifts upward. At low temperatures we must bear in mind that an exponential rise in Eq. (18) is related to an exponentially small broadening by an external field. The total width of a level is then governed by the mechanisms such as spontaneous decay and the ratio of the shift to the width decreases on increase in  $\beta$ . If the dominant mechanism is spontaneous decay, then at low temperatures it is proportional to  $(kT)^4$ .

The shift of the lines between the Rydberg states  $n+s$  and  $n$  is obviously

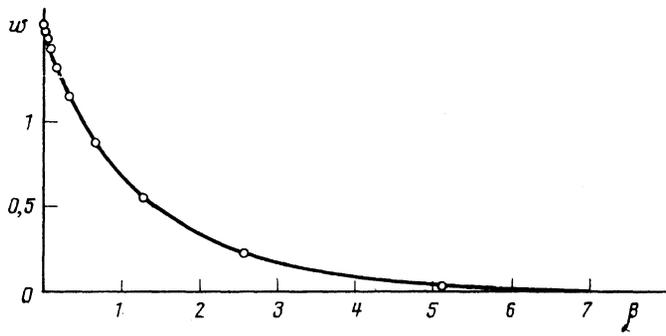


FIG. 3. Function  $w(\beta)$  [the continuous curve is the approximation by Eq. (17) and the points represent Eq. (15)] for the broadening of Rydberg levels in an external Planck field; the abscissa represents  $\beta = \delta/kT$ .

$$\delta\omega_{n,s} = \delta\omega_{n+s} - \delta\omega_n \approx \frac{\partial \delta\omega}{\partial n} s = 3\Delta_T \left( \frac{kT}{\text{Ry}} \right)^{1/3} Q\beta^{1/3} \frac{\theta'(\beta)}{2^{1/3}} s. \quad (19)$$

Using the approximation of Eq. (14), we obtain

$$\theta'(\beta) = 2\beta \left( \sum_{h=0}^4 a_h \beta^{2h} \right) / \left( \sum_{h=0}^6 b_h \beta^{2h} \right),$$

where

$$\begin{aligned} a_h &= (0,143; -0,203E-2; 0,524E-3; \\ &\quad -0,105E-4; -0,12E-6), \\ b_h &= (1,0; 0,345; 0,024; -0,659E-3; \\ &\quad 0,617E-4; -0,88E-6). \end{aligned}$$

In Eq. (19) the plus sign corresponds to an increase in the line frequency, i.e., it represents a blue shift. The function  $\beta^{4/3}\theta'(\beta)/2^{1/3}$  is shown in Fig. 2. The maximum blue shift corresponds to  $\beta = 4$  [ $n \approx 0.8(\text{Ry}/kT)^{1/3}$ ] and it amounts to  $1.8\Delta_T(kT/\text{Ry})^{1/3}s$ ; the maximum red shift corresponds to  $\beta = 8$  and it amounts to  $0.8\Delta_T(kT/\text{Ry})^{1/3}s$ . The shift vanishes for  $\beta = 6$ .

Let us now compare our expressions with the results reported in Ref. 13. For the radiation temperature of 300 K assumed in Ref. 13 the shift of the Rydberg levels in the limit  $kT \rightarrow \infty$  amounts to 2.417 kHz. The majority of the calculations reported in Ref. 13 applies to states with small angular momenta. A calculation of the shift of the levels with all possible values of the momenta  $l$  for a given value of  $n$  is given only for  $n = 30$ . For this value of  $n$  ( $\beta = 0.039$ ) the shift of a level with the maximum possible momentum  $l = 29$  exceeds the limit by 0.13% (which the authors of Ref. 13 regard as a measure of the error). The average shift is 3% less than the limit. Equations (11) and (14) give shifts which are 0.05% smaller than the limit.

#### 4. EXTERNAL FIELD WITH A POWER-LAW SPECTRUM

Let us assume that the radiation field has an effective temperature  $T_r$  at a frequency  $\omega_r$  and that its power exponent is  $\nu$ . The radiation density can then be written in the form

$$U(\omega) = kT_r \frac{\omega_r^2}{\pi^2 c^3} \left( \frac{\omega_r}{\omega} \right)^\nu. \quad (20)$$

Using Eqs. (15) and (10), we obtain (in this section we assume that  $Z = 1$ )

$$\delta\omega = -\alpha^3 \frac{kT_r}{\hbar} \left( \frac{\hbar\omega_r}{\text{Ry}} \right)^{2+\nu} n^{3(\nu-1)} \theta(\nu), \quad (21)$$

where the function  $\theta(\nu)$  is given by

$$\theta(\nu) = \frac{2(2+\nu)}{2^\nu \cdot 3^{1/2} \pi^2} \int_0^\infty \Phi(q) \frac{dq}{q^\nu} = \frac{(2+\nu)}{2^\nu \cdot 3^{1/2} \pi} \text{tg} \left( \frac{\pi\nu}{2} \right) \zeta(3+\nu), \quad (22)$$

and  $\zeta(\nu) - \zeta$  is the Riemann  $\zeta$  function. The integral in  $\theta(\nu)$  can be calculated by expanding  $\Phi(q)$  into elementary fractions and then separating in each term of the series the following integral as a factor:

$$\int_0^\infty \frac{dx}{(1-x^2)x^\nu}.$$

This integral is extended over the whole real axis and calculated by the standard method of the theory of functions of a complex variable. The integral in Eq. (17) converges if  $0 < \nu < 1$ . In the opposite case it is necessary to cut off the spectrum at the lower (or upper) end.

The width of a level, deduced under the same assumptions as for a Planck field, can be written in the form

$$\begin{aligned} W_n &\approx 2 \frac{A_0}{n^5} \frac{kT_r}{\hbar\omega_r} \left( \frac{\hbar\omega_r}{\delta} \right)^{3+\nu} \sum_{s=1}^\infty \frac{1}{s^{4+\nu}} \\ &= \alpha^3 \frac{kT_r}{\hbar} \left( \frac{\hbar\omega_r}{\text{Ry}} \right)^{2+\nu} n^{4+3\nu} w(\nu), \end{aligned}$$

$$w(\nu) = \frac{4}{2^\nu \cdot 3^{1/2} \pi} \zeta(4+\nu).$$

If we take  $\nu = 0.6$  and the frequency is  $\omega_r = 2\rho \times 100$  MHz, Eq. (23) gives the result obtained in Ref. 15.

The relative shifts of levels and lines are then described by

$$\begin{aligned} \frac{\delta\omega_n}{W_n} &= \frac{1}{n} \frac{\theta(\nu)}{w(\nu)} = (2+\nu) \frac{\zeta(3+\nu)}{\zeta(4+\nu)} \frac{\text{tg}(\pi\nu/2)}{4n}, \\ \frac{\delta\omega_{n,n+s}}{W_n + W_{n+s}} &= -3s(1+\nu) (2+\nu) \frac{\zeta(3+\nu)}{\zeta(4+\nu)} \frac{\text{tg}(\pi\nu/2)}{8n^2} \end{aligned}$$

(the lines shift in the red direction).

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<sup>1</sup> Here and later  $f$  denotes the principal value of the integral.

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