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Enhanced selective reflection from a thin layer of a dilute gaseous medium

T. A. Vartanyan* and D. L. Lin
Department of Physics, State University of New York at Buffalo, Buffalo, New York 14260-1500

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A theory of selective reflection and transmission is developed for a system consisting of a thin layer of a dilute atomic vapor sandwiched between two transparent solids with parallel interfaces. Strong effects of spatial dispersion due to the atomic motion and electronic quenching on gas-solid interfaces are accounted for. It is shown that both even and odd Doppler-free resonances may occur in selective reflection, depending upon the thickness of the vapor layer. It is also found that the amplitude of selective reflection is a result of the interference between reflections from the two boundaries of the vapor-solid interfaces and, hence, may be greatly enhanced under certain conditions.

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I. INTRODUCTION

The reflection of light within a transparent dielectric at the interface between the dielectric and an atomic vapor shows a sharp peak when the light frequency matches the transition energy of the atom. This phenomenon is known as the selective reflection (SR), and its study dates back to early this century. The SR observed by Wood [1] is accounted for by a conventional dispersion theory in which one assumes a local relation between the electric field and the induced polarization in a homogeneous medium.

In the SR experiments from a glass-mercury vapor interface, Cojan [2] observed evidence for spectral narrowing of reflection spectra relative to the Doppler broadening, while the conventional dispersion theory predicted a spectral width of the order of the Doppler width. Therefore, the conventional dispersion theory fails to describe the optical properties of rarefied atomic vapor when the Doppler broadening exceeds the homogeneous width of a resonant atomic transition. Similar spectral narrowing phenomenon was also observed by Woerdman [3] in his measurements of SR from a glass-sodium vapor interface with various vapor densities.

These experiments suggest that the transient polarization of vapor atoms associated with wall collisions is responsible for the spectral narrowing. A theory along this line of thought was then developed by Schuurmans [4] which described explicitly the transient polarization effects associated with collisions of atoms at the interface. Predictions of this theory have been verified in a number of experiments [5–7].

More recently, it is shown that Doppler-free selective reflection spectroscopy can be a powerful tool for the investigation of long-range atom-wall interactions [8]. The sub-Doppler resonance in selective reflection has also promising nonlinear optical properties [9,11]. On the other hand, the theory is far from complete even in the linear regime. The purpose of this paper is to investigate the linear optical properties of a resonant atomic vapor in a layer of thickness l of the order of the wavelength λ of the incident light. The boundaries of this layer of vapor are surfaces of transparent dielectric materials. Since both the selective reflection and ordinary nonselective reflection can take place at these surfaces simultaneously, the interference between them plays an important role in the shape of the selective line profile. As the reflectivity at these surfaces can be changed by different coatings, it is possible to simulate a condition that the vapor layer is surrounded by vacuum. We start with this condition in our calculation of selective reflection, and make necessary corrections due to the presence of dielectrics to the results later.

II. REVIEW OF THEORY

Consider a thin layer of gas consisting of two-level atoms with a transition frequency ω_{21} between levels $|1\rangle$ and $|2\rangle$. The gas is confined between two planes at $x=0$ and $x=l$. The space $x<0$ and $x>l$ is simulated vacuum. By neglecting the generation of harmonics, we can write in standard notation the field and polarization in the resonance region $\omega \gg |\omega_{21}-\omega|$, as

$$E(x,t)=\tfrac{1}{2}E(x)\exp(-i\omega t)+\text{c.c.} \tag{1}$$

and

$$P(x,t)=\tfrac{1}{2}P(x)\exp(-i\omega t)+\text{c.c.}\ , \tag{2}$$

respectively, for a monochromatic electromagnetic plane wave propagating along the positive x direction in the gas. It is straightforward to show that the spatial parts of E and P satisfy the equation

$$\frac{d^2E(x)}{dx^2}+k^2E(x)=-4\pi k^2P(x)\ . \tag{3}$$

The polarization can be expressed as

$$P(x)=2ND\langle\rho_{21}(x,v)\rangle\ , \tag{4}$$

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*Permanent address: State Optical Institute, St. Petersburg, 199034, Russia.

where N is the number density of gas atoms, D the transition dipole moment, and v the x component of the atomic velocity. The symbol $\langle\ \rangle$ means the average of the enclosed quantity over a Maxwellian distribution of all velocities. We have defined in Eq. (4) off-diagonal matrix elements ρ_{21} of the density matrix which can be written as

$$\rho_{21}(x,t,v)=\rho_{21}(x,v)\exp(-i\omega t)\ . \tag{5}$$

If the intensity of light is not strong enough to saturate the resonant transition, one finds that ρ_{21} satisfies the equation

$$v\frac{\partial\rho_{21}}{\partial x}+[\gamma+i(\omega_{21}-\omega)]\rho_{21}=\frac{i}{2\hbar}DE(x)\ . \tag{6}$$

For a very dense vapor, it may be assumed that the homogeneous width γ is much larger than the Doppler width kv_T where k is the wave number and v_T the most probable thermal velocity of atoms. Thus, the first term in Eq. (6) can be neglected and we have

$$\rho_{21}(x,v)=i\frac{DE(x)}{2\hbar}\frac{1}{\gamma+i(\omega_{21}-\omega)}\ . \tag{7}$$

Substituting Eq. (7) into Eq. (5) leads to the well-known results that inspired the classical experiment on selective reflection from mercury vapor as early as 1909.

For a dilute gas, the homogeneous width is small and $kv_T>\gamma$. Hence the first term in Eq. (6) is important. In the conventional dispersion theory, one still neglects the first term but assumes that its effect can be accounted for by simply replacing $\omega_{21}-\omega$ in Eq. (7) by $\omega_{21}-\omega+kv_T$. This means that only steady-state solutions are considered. In realistic cases, however, one has to consider the transient solution of Eq. (6) as well.

In order to find a particular solution to Eq. (6), we have to impose the boundary conditions. This is accomplished by requiring that atoms are in their ground state (or unpolarized) when they are at surfaces of dielectric material. As a matter of fact, we limit our discussion to the case that atoms leaving the dielectric surface have zero polarization. This choice has sound basis as has been experimentally demonstrated in the literature [10]. Thus, we set

$$\rho_{21}(x=0,v>0)=0 \tag{8a}$$

and

$$\rho_{21}(x=l,v<0)=0 \tag{8b}$$

as our boundary conditions. We shall solve Eqs. (3), (4), and (6) in the next section with boundary conditions (8).

III. PERTURBATION METHOD

In the limit of large Doppler broadening, or $kv_T\gg\gamma$, the optical density of the atomic vapor may be so small that we can apply the perturbation theory. We start with the corresponding homogeneous equation of Eq. (3), and find the unperturbed electric field as

$$E(x)=E_0\exp(ikx)\ . \tag{9}$$

Next, we insert Eq. (9) in Eq. (6) which is then solved with boundary conditions (8). This yields

$$\rho_{21}(x,v>0)=i\frac{DE_0}{2\hbar}\left\{\frac{\exp(ikx)-\exp\left[-\dfrac{\gamma+i(\omega_{21}-\omega)}{v}x\right]}{\gamma+i(\omega_{21}-\omega+kv)}\right\} \tag{10a}$$

for $v>0$ or atoms moving to the right, and

$$\rho_{21}(x,v<0)=i\frac{DE_0}{2\hbar}\left\{\frac{\exp(ikx)-\exp\left[ikl-\dfrac{\gamma+i(\omega_{21}-\omega)}{v}(x-l)\right]}{\gamma+i(\omega_{21}-\omega+kv)}\right\} \tag{10b}$$

for $v<0$ or atoms moving to the left.

When these density matrix elements are averaged over all velocities and substituted into Eq. (4), we can calculate the polarization $P(x)$. With the polarization known, the electric field can then be obtained to the first-order correction by solving the inhomogeneous equation (3). In terms of the Green's function, this solution is formally given by

$$E_1(x)=2\pi ik\int_0^l P(x')\exp(ik|x-x'|)dx'\ . \tag{11}$$

This equation holds for all x but has different physical meaning in different regions. For $x<0$, Eq. (11) gives the reflected field

$$E_1(x<0)=E_r\exp(-ikx)\ , \tag{12}$$

where we have defined

$$E_r=2\pi ik\int_0^l e^{ikx'}P(x')dx'\ . \tag{12a}$$

For $x>l$, we have the transmitted field

$$E_1(x>l)=E_t\exp(ikx)\ , \tag{13}$$

where

$$E_t=2\pi ik\int_0^l e^{-ikx'}P(x')dx'\ . \tag{13a}$$

The field inside the vapor is not of our concern, and we do not write down its explicit expression for the region $0<x<l$.

IV. SELECTIVE REFLECTION

We are now in the position to calculate the coefficient of selective reflection due to the resonance interaction of a thin vapor. This can be found in a straightforward manner from Eqs. (4), (10), (12), and (13) and the result is

$$\frac{E_r}{E_0}=\frac{2\sqrt{\pi}ND^2}{\hbar k v_T}\times\left\{\int_0^\infty\frac{\exp(-v^2)dv}{v+i\Gamma+\Omega}+e^{2i\phi}\int_0^\infty\frac{\exp(-v^2)dv}{v-i\Gamma-\Omega}-2e^{i\phi}\int_0^\infty\frac{v\,dv\exp[-v^2-(\Gamma-i\Omega)\phi/v]}{v^2+(\Gamma-i\Omega)^2}\right\}, \tag{14}$$

where we have introduced the dimensionless quantities $\Gamma=\gamma/kv_T$, $\Omega=(\omega-\omega_{21})/kv_T$, $\phi=kl$, and $v=v/v_T$.

It is interesting to remark that the wave number k is in general complex. The factor $\exp(i\phi)$ then vanishes in the limit of large l due to the imaginary part of k. Consequently, the last two terms in Eq. (14) disappear in the case of a thick layer of gases [4,8]. The factor of 2 in Eq. (14) has its origin in the fact that the integrals are equal whether they are taken from $-\infty$ to 0 or from 0 to ∞. This symmetry implies that atoms moving in opposite directions in the vapor make identical contributions to the coefficient of selective reflection just as in the case of thick vapor layer. On the other hand, both the redshifted and blueshifted denominators appear in Eq. (14). This is in contrast to the thick layer case in which only redshifted denominators make contributions.

The region we are interested in is $\Omega\sim\Gamma\ll1$ and $1\ll\phi\ll\Gamma^{-1}$. In this region, we evaluate approximately all three integrals involved in the Appendix. The resulting E_r is given by

$$\frac{\mathrm{Re}(E_r)}{E_0}=-\frac{2\sqrt{\pi}ND^2}{\hbar k v_T}\left\{(\cos\phi-1)[\ln(\Gamma^2+\Omega^2)\cos\phi+2\tan^{-1}(\Omega/\Gamma)\sin\phi]+\frac{\pi}{2}\sin2\phi-2\cos\phi\ln\phi+\frac{\gamma_E}{2}(1+\cos2\phi-6\cos\phi)\right\}, \tag{15}$$

$$\frac{\mathrm{Im}(E_r)}{E_0}=-\frac{2\sqrt{\pi}ND^2}{\hbar k v_T}\left\{(\cos\phi-1)[\ln(\Gamma^2+\Omega^2)\sin\phi-2\tan^{-1}(\Omega/\Gamma)\cos\phi]+\frac{\pi}{2}(1-\cos2\phi)-2\sin\phi\ln\phi+\frac{\gamma_E}{2}(\sin2\phi-6\sin\phi)\right\} \tag{16}$$

where $\gamma_E=0.577\ldots$ is Euler's constant. Both the real and imaginary parts of E_r may contribute to the selective reflection, depending on the properties of the surrounding media. As an example, we consider one of the most interesting cases, that is, the first boundary is uncoated and the second boundary has an antireflection coating. The ordinary nonselective reflection on the first boundary then gives rise to the reflected wave with an amplitude

$$E_{r0}=\frac{n_0-1}{n_0+1}E_0\ , \tag{17}$$

where n_0 is the refractive index of the dielectric in question. To calculate the intensity of the reflected beam, both the selective and nonselective reflection parts must be included. Thus, we have

$$I_r\propto|E_{r0}+E_r|^2=E_{r0}^2+2E_{r0}\mathrm{Re}(E_r)+|E_r|^2\ , \tag{18}$$

where E_r is the selective reflection. Usually, E_{r0} is much larger than E_r so that the last term in Eq. (18) may be neglected, and the spectral line profile of the selective reflection is solely determined by $\mathrm{Re}(E_r)$. We discuss in this paper mainly this case.

The most interesting feature of Eq. (15) is that the spectral profile of the reflected field depends strongly on the thickness of the vapor layer. As can be seen from Eq. (15), the sub-Doppler structure of the selective reflection is characterized by the term involving $\ln(\Gamma^2+\Omega^2)$, which vanishes when $\phi=2n\pi$, where n is an integer. Hence the selective reflection would not show any sub-Doppler feature when $l=n\lambda$, namely, when the thickness of the vapor layer is a multiple of the incident wave length. It is also worth noting that the spectral shape of reflectivity as given by Eq. (15) is dominated by the even term $\ln(\Gamma^2+\Omega^2)$ when $\phi=(2n+1)\pi$, and the odd term $\tan^{-1}(\Omega/\Gamma)$ dominates when $\phi=(n+1/2)\pi$. We emphasize that even in the former case, $\phi=(2n+1)\pi$, when the spectral line shape is similar to the reflectivity of a thick layer, the amplitude of reflected light as given by Eq. (15) is four times larger. Therefore, the present result cannot be obtained by simply adding contributions from the two boundaries.

To study the spectral line profile outside the region $|\Omega|\ll1$, numerical integration of Eq. (14) is necessary. We take $\Gamma=0.01$ in our computation, bearing in mind of alkali-metal atomic gases under normal experimental conditions. The Doppler broadening in such cases is about two orders of magnitude larger than the natural width. For the thickness of the gas layer, we choose $\phi=3\pi$, 3.5π, 4π, and 4.5π. Results are displayed in Fig. 1 for all the four cases. The narrow structure in the central part of each curve illustrates clearly the sub-Doppler feature of selective reflection resonances predicted by Eqs. (15) and (16). As we have discussed above, the profile of Doppler wings also depends on the layer thickness, but this dependence can be accounted for by the ordinary interference effects.

V. THE TRANSMITTED FIELD

We now turn our attention to the transmitted field. Following the same procedure with Eqs. (12) replaced by (13), we find within the framework of the first-order perturbation theory

$$\frac{E_t}{E_0}=-\frac{2\sqrt{\pi}ND^2}{\hbar k v_T}\left\{\phi\int_{-\infty}^{\infty}\frac{\exp(-v^2)dv}{\Gamma-i(\Omega-v)}+\int_{-\infty}^{0}\frac{v\exp(-v^2)dv}{[\Gamma-i(\Omega-v)]^2}-\int_{0}^{\infty}\frac{v\exp(-v^2)dv}{[\Gamma-i(\Omega-v)]^2}\right.$$
$$\left.-e^{i\phi}\int_{-\infty}^{0}\frac{v\exp[-v^2+(\Gamma-i\Omega)\phi/v]dv}{[\Gamma-i(\Omega-v)]^2}+e^{-i\phi}\int_{0}^{\infty}\frac{v\exp[-v^2-(\Gamma-i\Omega)\phi/v]dv}{[\Gamma-i(\Omega-v)]^2}\right\}. \tag{19}$$

In constrast with the reflected field, it is found that contributions to the transmitted field from atoms moving in opposite directions are no longer equal. They are separately represented in Eq. (19) by integrals with different integration limits except for the first term which is integrated over the whole range of velocity distribution.

An estimate of these integrals can be made in a similar fashion as what is outlined in the Appendix. We find that in the region $\Omega\sim\Gamma\ll 1$ and $1\ll\phi\ll\Gamma^{-1}$, the first term in (19) dominates the others, and it is equal to $\phi\pi$. This implies that our perturbation approach is justified for

$$\phi\ll\frac{\hbar k v_T}{2\pi^{3/2}ND^2}\ . \tag{20}$$

It is of interest to mention that the conventional disper-

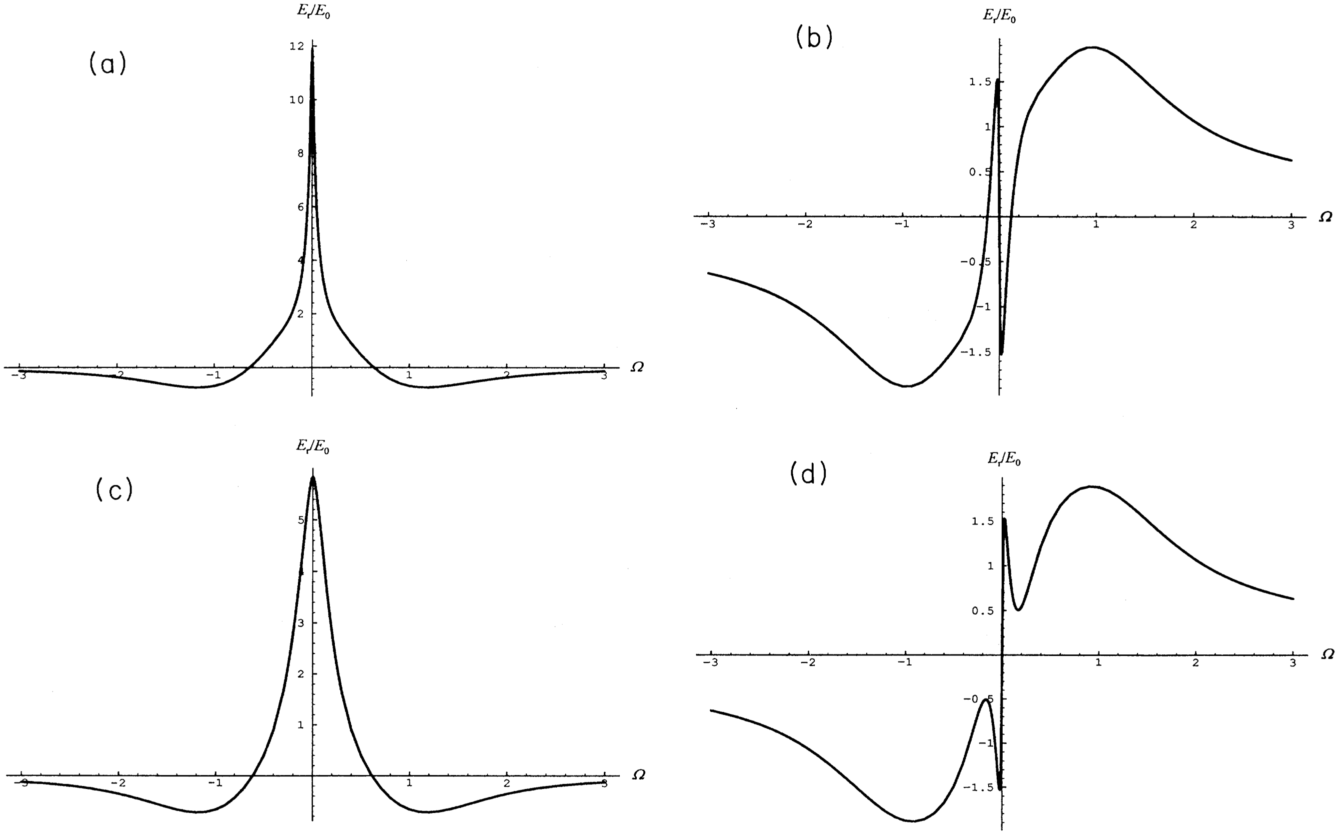

FIG. 1. Reflectivity (in arbitrary units) of a thin layer of resonant vapor vs the dimensionless detuning for four choices of the vapor thickness. (a) $l=3\lambda/2$, (b) $l=7\lambda/4$, (c) $l=2\lambda$, and (d) $l=9\lambda/4$. The vapor layer is confined between two interface boundaries, with the first interface uncoated and the second antireflection coated. The detuning is normalized to Doppler width which is chosen to be 100 times larger than the homogeneous width.

sion theory yields exactly the first term in Eq. (19) appears to be correct for calculations of the transmitted field in this particular case, as has been already pointed out in Ref. [4].

VI. DISCUSSION AND CONCLUSION

We have seen from the above that selective reflection from interfaces between a dielectric and a thin layer of dilute vapor exhibits unexpected optical properties due to the spatial dispersion and atom-wall collisions. When the Doppler width is larger than the homogeneous width of an optical transition which is the sum of the natural and collisional widths, the mean free path of the atom becomes larger than the wavelength. As a consequence, the vapor response on the external field becomes nonlocal. An atom desorbing from the wall in its ground state cannot adopt the external field before it travels a distance of many wavelengths.

From this point of view, it is not difficult to understand the main feature of the Doppler-free selective reflection from a thick layer. Indeed, when light is in resonance with desorbing atoms, and the detuning is positive, both the transient and steady-state solutions of Eq. (7) have almost the same spatial dependence. This means that the polarization of desorbing atoms remains to be zero for a very large distance from the wall. Therefore, these atoms do not contribute to the reflected field on their own resonance frequency despite of the fact that amplitudes of both the transient and steady-state solutions are large.

On the other hand, when light is in resonance with atoms traveling towards the surface with a velocity v, not only these atoms contribute but those moving with the velocity v in the opposite direction will also make resonant contribution to reflection as well. To understand this unexpected phenomenon, one has to notice the fact that the spatial dependence of the transient and steady-state solutions are very different in this case according to Eq. (14). The net polarization of desorbing atoms becomes nonzero already in the immediate proximity of the surface. On the other hand, we note from Eq. (10a) that the spatial dependence of the transient solution is very similar to that of the reflected wave $\exp(-ikx)$. Therefore, the nonresonant recoiling atoms contribute to the reflection spectrum with the same amplitude as resonant atoms traveling towards the surface.

It is also important to point out that atoms traveling against the incident direction are not in the steady state if the vapor layer is thin. This is because they remain in the ground state right after leaving the back surface, and there is simply not enough time for them to adopt the external field. Nevertheless, atoms moving in opposite directions with the same speed contribute the same amount to the reflected field just as in the case of a thick layer of vapor.

Furthermore, we remark that the reflection from a thin atomic layer cannot be obtained by a simple superposition of reflections from two similar boundaries. Due to transient terms in the atomic polarization, the medium becomes nonuniform, and moreover the optical properties of the medium at any point depend on the overall thickness of the vapor layer. Consequently, the reflected field as given by Eq. (14) cannot reproduce the classical interference pattern $E_r \sim [\exp(2i\phi)-1]$ with a sequence of nodes at $\phi = n\pi$. Instead, the interference pattern due to selective reflection is given by Eq. (15), which implies that only the sequence $\phi = 2n\pi$ remain to be the nodes. When $\phi = (2n+1)\pi$, the amplitude of the selective reflection in the case of a thin layer is four times stronger than that of a thick layer although it has the same even counter $\ln(\Gamma^2+\Omega^2)$. When $\phi = (n+1/2)\pi$, the selective reflection is described by the odd function $\tan^{-1}(\Omega/\Gamma)$. Even and odd parts mix for any other thickness.

Finally, we just mention that the influence of the second boundary on selective reflection can be quite dramatic if nonselective contributions from both the back and front interfaces are eliminated by some experimental techniques such as different kind of coatings. Such conditions have never been exploited in experiments thus far because only weak signals can be expected. Our analysis indicates that the presence of the second boundary enhances the selective reflection signals up to sixteen times under particular conditions and hence an attempt of experimental observation becomes quite feasible. It is of particular interest to note that this strong enhancement may be realized without making use of any antireflection coatings. As a matter of fact, the nonselective reflection from the front and rear interfaces interfere destructively whenever the layer thickness satisfies the condition $\phi = n\pi$. Under the condition, however, selective reflection from the two boundaries interfere constructively for odd n and destructively for even n.

As a conclusion, we emphasize that it should be of great interest to carry out experimental investigations of selective reflection from thin films of vapor. The most attractive features of such experiments are the nontrivial dependence of the line shape on the vapor layer thickness and the extraordinarily enhanced signals.

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APPENDIX

The first two integrals in Eq. (14) are of the same type

$$A = \int_0^\infty \frac{\exp(-x^2)dx}{x+a} , \tag{A1}$$

where $a = \pm(\Omega + i\Gamma)$. For $|a| \ll 1$, the integral is already evaluated in Ref. [4] and the result is

$$A = -\frac{\gamma_E}{2} - \ln a + O(|a|) , \tag{A2}$$

where $\gamma_E = 0.577\ldots$ is Euler's constant and $\ln(z)$ is defined for $|\arg(z)| < \pi$ and $\ln 1 = 0$.

The third integral takes the form

$$B = \int_0^\infty \frac{x\exp(-x^2 - b\phi/x)}{x^2+b^2} dx , \tag{A3}$$

where $b=\Gamma-i\Omega$ so that $\mathrm{Re}(a)>0$. We are only interested in the region $|\mathrm{Im}(b)|\sim|\mathrm{Re}(b)|\ll 1$, $1\ll\phi\ll[\mathrm{Re}(b)]^{-1}$. The integral can be carried out approximately by dividing the range of integration from 0 to $x_1=\phi^{1/3}\mathrm{Re}(b)$, x_1 to x_2, and finally from x_2 to ∞ where x_2 satisfies the inequality $\phi\,\mathrm{Re}(b)\ll x_2\ll 1$. Thus $B=B_1+B_2+B_3$.

For the first interval of integration, we note that $\phi\,\mathrm{Re}b\gg x_1$. Therefore, we have

$$|B_1|\le\frac{1}{(\mathrm{Re}b)^2}\int_0^{x_1}x\exp\left[-\frac{\phi\,\mathrm{Re}b}{x}\right]dx$$
$$\approx\frac{x_1^3}{\phi(\mathrm{Re}b)^3}\exp\left[-\frac{\phi\,\mathrm{Re}b}{x_1}\right]=\exp(-\phi^{2/3})\ll 1\ . \tag{A4}$$

In the second interval, $x\gg|b|$ so that b^2 can be neglected in the denominator. Accordingly, we have for $|b|\phi\gg x_1$ and $|b|\phi\ll x_2$.

$$B_2\approx\int_{x_1}^{x_2}x^{-1}\exp(-b\phi/x)dx\approx-\gamma_E+\ln(x_2/b\phi)\ . \tag{A5}$$

For $x>x_2$, the integral becomes

$$B_3\approx\int_{x_2}^{\infty}x^{-1}\exp(-x^2)dx\approx-\frac{\gamma_E}{2}-\ln x_2\ . \tag{A6}$$

Combining the last three equations, we find a reasonable estimate

$$B\approx-\tfrac{3}{2}\gamma_E-\ln b\phi\ . \tag{A7}$$

As it is expected, this result does not depend on the exact positions of x_1 and x_2 which are arbitrarily chosen.

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