Stark effect in finite-barrier quantum wells, wires, and dots

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Abstract
The properties of confined carriers in low-dimensional nanostructures can be controlled by external electric fields and an important manifestation is the Stark shift of quantized energy levels. Here, a unifying analytic theory for the Stark effect in arbitrary dimensional nanostructures is presented. The crucial role of finite potential barriers is stressed, in particular, for three-dimensional confinement. Applying the theory to CdSe quantum dots, finite barriers are shown to improve significantly the agreement with experiments.

1. Introduction

Nanostructures are characterized by carrier confinement due to potential barriers. Physically, the confinement may be in the form of a material discontinuity (hetero-junction) or an interface with vacuum. Hence, quantum wells, wires, and dots are fabricated through confinement in one, two, and three dimensions, respectively. The resulting quantized energy levels are essential for the functionality of nanostructured devices [1]. In many applications and characterization techniques, strong electric fields are applied in addition to the confinement. Hence, the bare energy levels are Stark-shifted by the field as demonstrated for both one-, two-, and three-dimensional nanostructures [2-4]. This leads to field-dependent functionalities but, also, the possibility of tuning properties via the applied field.

In order to interpret measurements and simulate device performance in strong electric fields, it is important to reliably predict the induced Stark shifts. For a symmetric structure, the Stark shift $E_2$ of an electronic state to lowest order in the field $F$ is determined by the polarizability $\alpha$ as $E_2 = -\frac{1}{2} \alpha F^2$. Hence, for many cases, the problem reduces to computing $\alpha$. Note that the term 'Stark shift' is used here generically for field-induced energy shifts. Hence, no grammatical distinction is made between, e.g., 'Stark effect' and 'quantum confined Stark effect'. In a highly simplified picture, the confinement barrier is taken to be infinitely high [5, 6]. Hence, electrons are completely confined to the interior of the structure no matter the strength of the applied field. Realistically, however, confinement barriers are always finite and typically of order several hundred meVs for hetero-junctions or several eVs for vacuum barriers [7]. A finite potential allows the electron to leak out into the barrier, thereby lowering the unperturbed energy level. Importantly, a finite potential also increases the polarizability as the field may drive the wave function further into the barrier. This increase can be very large for shallow and/or narrow quantum wells. A recent example of this is the case of MoS$_2$ monolayers encapsulated by dielectrics and subjected to perpendicular fields [8]. There, a simple 1D quantum well model shows that taking into account finite barriers enhances the (exciton) polarizability by a factor 3.7 assuming a quantum well thickness equal to the monolayer thickness of 6.5 Å [9]. For quantum wires and dots, in which more dimensions are restricted by confinement, the electronic states are even more sensitive to the barrier height.

In the present work, a unified solution to the Stark shift problem in quantum wells of arbitrary dimension and arbitrarily low confinement barriers is found. The solution is formulated in terms of the dimension $D$ that ranges from $D = 1$ (quantum wells) over $D = 2$ (wires) to $D = 3$ (dots). However, non-integer values are also, in principle, allowed and provide a smooth interpolation between integer-valued limits. Such non-integer or fractional dimensions are, in fact, relevant for models of structures that effectively exhibit confinement in-between strictly integer-dimensional limits. An example of this would be prolate (‘cigar shaped’) ellipsoidal

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quantum dots in a configuration with the field along the short axis. In such a structure, increasing the long to short axis ratio from unity to infinity would effectively mean interpolating between 3D and 2D confinement with intermediate cases described approximately by non-integer dimensions within this range. Hence, fractional-dimensional nanostructures have been studied in several previous works [10–13] covering also low-dimensional excitons.

To compute polarizabilities, the traditional approach relies on the perturbation expansion [14], in which a sum-over-states expression is evaluated. To obtain exact results, the sum must cover all the infinitely many excited states. While this is possible for infinite barriers due to the discrete nature of the eigenvalues, it becomes highly nontrivial for a finite barrier potential. Thus, the powerful alternative proposal by Dalgarno and Lewis [15] is adopted instead in the present work. Here, the ground state Schrödinger equation is solved order-by-order in the electric field without explicitly involving excited states at all [14, 15]. It is demonstrated below that such an approach is actually reasonably straightforward even for problems of arbitrary dimension provided ‘spherical’ symmetry of the confinement is assumed. The polarizabilities of single particles, shallow donors/acceptors, and excitons in finite-barrier one-dimensional quantum wells have previously been studied using finite-field and variational approaches [16–18]. To the knowledge of the present author, the only semi-analytical result is that of [19], which is strictly limited to the 1D case and only provides maximally simplified expressions for the interior contribution to the polarizability. Below, fully simplified results are given for arbitrary dimensions. Thus, the strength of the present approach lies in the formulation of readily applicable analytical expressions, in contrast to purely numerical approaches that can clearly handle much more complicated problems such as, e.g. parallelepiped shapes [20]. In should be stressed, however, that high accuracy in sum-over-states numerical approaches requires summing over exceedingly many contributions, which is cumbersome, in particular, for structures featuring a continuum of states. Similarly, highly elaborate ansätze are required in numerical variational methods. In contrast, the present analytical approach offers exact results at minimal computational demand. To further stress the usefulness of the analytical approach, closed-form expressions for the frequency-dependent polarizability valid for arbitrary dimension are given.

2. D-dimensional Stark problem

In order to tackle the fully general case of arbitrary dimension and barrier, certain simplifying assumptions are made. Primarily, confinement is taken to be in the form of a square potential well with perfectly vertical (but finite) barriers. In addition, an isotropic, position-independent effective mass is assumed. Anisotropic effective masses have previously been included in the Stark problem for two-dimensional [21] and three-dimensional [22] excitons but this complication is not considered here. In this isotropic effective mass approximation, a low-dimensional semiconductor structure in a constant electric field \( \vec{F} \) is then described by the perturbed Schrödinger equation

\[
\left\{ -\frac{\hbar^2}{2m^*^\text{eff}} \nabla^2 + V(\vec{r}) + e\vec{F} \cdot \vec{r} \right\} \varphi(\vec{r}) = E \varphi(\vec{r}), \tag{1}
\]

where \( m^* \) is the effective mass. In order to describe nanostructures of arbitrary dimension, D-dimensional spherical coordinates [10–13] are now introduced. The dimension \( D \) refers to the number of confined dimensions, so that quantum wells, wires, and dots are characterized by \( D = 1, 2, \text{ and } 3 \), respectively. In such spherical coordinates, the confinement is in the ‘radial’ direction in all cases. The integer-dimensional cases are illustrated in figure 1. Furthermore, a polar angle \( \theta \) is introduced so that the electrostatic interaction is of the form \( \vec{F} \cdot \vec{r} = F r \cos \theta \). In the one-dimensional case, the angle is restricted to the values \( \theta = 0 \) and \( \theta = \pi \) by the singular nature of the angular part of the Laplacian [10]. A similar approach has been applied previously to find the polarizability of excitons in low-dimensional confinement, demonstrating a pronounced dependence on dimension [13, 23].

As argued above, it is essential to account for the finite barriers characteristic of realistic confinement potentials. Hence, the potential is taken to be of the form

\[
V(\vec{r}) = V_0 \Theta(\vec{r} - a) = \begin{cases} 0 & r < a \vspace{1ex} \\ V_0 & r > a. \end{cases} \tag{2}
\]

Here, \( a \) is the ‘radius’ of the confinement as shown in figure 1 and \( \Theta \) is the unit step function. It is convenient to use \( a \) as the unit distance and \( \hbar^2/(2m^*a^2) \) as the unit energy. Hence, introducing the normalized field \( E = e m^*a^2 F / \hbar^2 \) the D-dimensional eigenvalue problem becomes [10–13]

\[
\{H_0 + 2E r \cos \theta\} \varphi(r, \theta) = E \varphi(r, \theta), \quad H_0 = -\frac{d^2}{dr^2} - \frac{D-1}{r} \frac{d}{dr} + \frac{\vec{r}^2}{r^2} + w^2 \Theta(r - 1). \tag{3}
\]
Here, the ‘normalized potential barrier’ $w$ is introduced via $w^2 \equiv 2m^2a^4V_0/h^2$. If a dimensionless polarizability is calculated in these units, conversion to physical units simply implies multiplication by a factor $\alpha_0 = e^{m^2a^4}/h^2$. The angular momentum operator $\hat{l}^2$ has eigenvalues $l(l + D - 2)$ with $l$ integer. In general, the eigenfunctions are Gegenbauer polynomials [10] but for the special case $l = 1$ these become proportional to $\cos \theta$ having the eigenvalue $D - 1$. This fact allows us to apply the Dalgarno–Lewis technique for arbitrary dimensions. To this end, energy and wave function are expanded in powers of the electric field, i.e., $E = E_0 + E_1 + \cdots$ and $\varphi(r, \theta) = \varphi_0(r) + \varphi_1(r, \theta) + \cdots$. Note that only even powers appear in the expansion of the energy due to the inversion symmetry of the unperturbed problem. Next, the first order terms of the eigenvalue problem equation (3) are collected. By writing the first order wave function in the form $\varphi_1(r, \theta) = 2\mathcal{E} \cos \theta \varphi_0(r)$ it is easily demonstrated that the first order equation becomes

$$-f''(r) = f'(r)\left\{2\frac{\varphi_0'(r)}{\varphi_0(r)} + \frac{D - 1}{r}\right\} + \frac{D - 1}{r^2}f(r) = -r.$$  \hspace{1cm} (4)

Moreover, the second order energy becomes

$$E_2 = -\frac{2\mathcal{E}^2}{D} \int_0^\infty \varphi_0^2(r)f(r)r^D dr,$$

where the factor $D^{-1}$ comes from the angular integration and the zero order solution is normalized according to $1 = \int_0^\infty \varphi_0^2(r)r^{D-1} dr$. It follows that the $D$-dimensional polarizability $\alpha_D$ is given by

$$\alpha_D = \frac{4}{D} \int_0^\infty \varphi_0^2(r)f(r)r^D dr.$$  \hspace{1cm} (6)

The unperturbed ground state depends on wave numbers $k$ and $q$ for the interior (well) and exterior (barrier) regions, respectively. Hence, writing $E_0 = k^2$ and $E_0 = w^2 - q^2$ it is found that

$$\varphi_0(r) = N r^{1-D/2} \begin{cases} J_{D/2-1}(kr)/J_{D/2-1}(k) & r < 1, \\ K_{D/2-1}(qr)/K_{D/2-1}(q) & r > 1, \end{cases}$$

where $J_n$ and $K_n$ are Bessel functions of the first and second kind, respectively, and the normalization factor is

$$N^{-2} = \frac{1}{2} \left\{ \frac{K_{D/2-1}(q)K_{D/2}(q)}{K_{D/2-1}(q)} - \frac{J_{D/2-1}(k)J_{D/2}(k)}{J_{D/2-1}(k)} \right\}.$$  \hspace{1cm} (8)

The wave function equation (7) is continuous across the barrier by construction and by requiring a continuous radial derivative as well, one finds the eigenvalue condition

$$qK_{D/2}(q) = \frac{kD/2}{J_{D/2}(k)}.$$  \hspace{1cm} (9)

The boundary condition for the first order correction is $f(0) = 0$. The most general solution to equation (4) is then

**Figure 1.** Schematic illustration of geometries and field orientation for quantum wells, wires, and dots.
\[ f(r) = \begin{cases} \frac{1 - 2}{4k^2} \left( (D - 2)r + \frac{k(A - r^2)J_{D/2}(kr)}{J_{D/2-1}(kr)} \right) & r < 1 \\ \frac{1}{4q^2} \left( (D - 2)r + \frac{q(B - r^2)K_{D/2}(qr)}{K_{D/2-1}(qr)} \right) & r > 1. \end{cases} \] (10)

The constants \( A \) and \( B \) must be determined such that \( f(r) \) and its radial derivative are both continuous at \( r = 1 \). It is then found that

\[ A = 1 + \frac{D(2 - D)}{q^2} + \frac{Dk_J D/2(k)}{q^2 J_{D/2-1}(k)} = \frac{(D - 2)J_{D/2-1}(k)}{k_J D/2(k)}, \]

\[ B = 1 - \frac{D(2 - D)}{k^2} - \frac{Dk_J D/2(k)}{k_J D/2-1(k)} = \frac{(D - 2)J_{D/2-1}(k)}{k_J D/2(k)}. \] (11)

The integral in equation (6) is quite involved but after a tedious calculation the result can be expressed in terms of the Bessel function ratio \( R = J_{D/2-1}(k)/J_{D/2}(k) \) as

\[ \alpha_D = \frac{\alpha_0}{12Dk^4q^4[(D - 2)R - k]} \sum_{n=1}^{3} C_n R^n \] (12)

with the coefficients

\[ C_{-1} = 6D^2k^4q^4, \quad C_0 = -6(D - 2)k^2q^4, \]

\[ C_0 = |D(34D - 13D^2 - 24) - 4(2D - 3)(k^2 - q^2)k^2q^2 + (D - 6)(D - 4)Dk(k^4 + q^4), \]

\[ C_1 = 2(2D - 22D^2 + 7D^3) \left( \frac{1}{2}D^2 - D + k^2 - q^2 \right)k^2q^2 - (D - 6)(D - 4)(D - 2)D(k^4 + q^4), \]

\[ C_2 = 4(2D - 3)k^2q^4 - (D - 2)[24 + D(7D - 22)]kq^2(k^2 - q^2). \] (13)

### 3. Integer and fractional dimensional results

Before turning to the inclusion of finite barrier heights on the polarizability, it is instructive to consider the limit of infinite barriers, i.e. taking the limit \( w \to \infty \). The polarizability obtained in this limit will be denoted \( \alpha_D^\infty \). In the case of an infinite barrier, the exterior wave number becomes \( q = \infty \) while \( k \) is the first root of the Bessel function, i.e. \( k = \lambda_D \), with \( J_{D/2-1}(\lambda_D) = 0 \). In the integer-dimensional cases, one finds \( \lambda_1 = \pi/2 \)

\( \lambda_2 \approx 2.4048 \), and \( \lambda_3 = \pi \), respectively. It then follows that equation (12) becomes

\[ \alpha_D^\infty = \frac{\alpha_0}{12D \lambda_D^2} (D - 6)(D - 4)D + 4\lambda_D^2(2D - 3). \] (14)

The integer-dimensional limits for \( \alpha_D^\infty \) are

\[ \alpha_1^\infty = \alpha_0 \frac{4(15 - \pi^2)}{3\pi^4}, \quad \alpha_2^\infty = \alpha_0 \frac{4 + \lambda_2^2}{6\lambda_2^2}, \quad \alpha_3^\infty = \alpha_0 \frac{3 + 4\pi^2}{12\pi^4}. \] (15)

For \( D = 1 \), the result is in agreement with previous work \[15, 16, 19\]. The limiting cases as well as the general result equation (14) are illustrated in figure 2. It is seen that the polarizability is monotonously decreasing with dimension. Thus, upon increasing the dimension from \( D = 1 \) in the quantum well case to \( D = 3 \) for quantum dots, the infinite-barrier polarizability decreases by nearly a factor of two.

If a finite barrier is considered, the full result equation (12) is needed. The expressions for \( D = 1 \) and \( D = 3 \) are simplified by the fact that the eigenvalue condition equation (9) reduces to \( q = k \) and \( q = -k \cot k \), respectively, whereas the two-dimensional case cannot be similarly simplified. This is reflected in the final expressions, which read

\[ \alpha_1 = \alpha_0 \frac{k(15 - 4k^2) + 3 \cot k \{ 5 - 4k^2 + (4k^2 + 5\cot^2 k + 8k \cot k - 3)\cot^2 k - 4k \cot k \}}{12k^4(1 + \cot k)}, \] (16)

\[ \alpha_2 = \frac{\alpha_0}{6} \left\{ \frac{4}{k^4} + \frac{q^2 - 4}{k^2q^2} + \frac{6\lambda_2^2}{k^2q^2} + \frac{\lambda_2^2}{k^2q^2} + \frac{4(k^2 - q^2)J_0(k)}{k^2q^2J_1(k)} + \frac{4 - q^2}{k^4} \right\} \] (17)

and

\[ \alpha_3 = \frac{\alpha_0}{24k^4(\sin k - k \cos k)^2} \left\{ \frac{2(9 - 4k^2)k \tan k + 6(4k^2 - 3k \tan k - 4)\sec^2 k + 6\sec^2 k}{24k^4(\sin k - k \cos k)^2} \right\}. \] (18)
It is readily checked that taking the limits (i) $k \to \pi / 2$ in equation (16), (ii) $q \to \infty$ and $J_0(k) \to 0$ in equation (17), and (iii) $k \to \pi$ in equation (18) leads to the infinite-barrier results equation (15). In contrast to previous work [19], the 1D expression equation (16) shows that the total polarizability (interior plus exterior) can be expressed in a simple closed form.

To evaluate the polarizability for specific values of the normalized potential barrier $w$, the eigenvalue condition equation (9) combined with the restriction $k^2 + q^2 = w^2$ from the energy expressions must be solved numerically. This solution is readily found, however, and once $k$ and $q$ are determined the analytical expression equation (12) for the general polarizability is easily evaluated. The results for integer dimensions as well as representative fractional ones are illustrated in figure 3 using linear scales in the main panel and on logarithmic ones in the inset. In the figure, the infinite barrier limit equation (14) is included as well. As expected, the polarizability decreases monotonously with increased barrier height. Furthermore, as is clearly seen in the inset, this decrease is more pronounced in higher dimensional confinement, reflecting the greater sensitivity to the barrier in these cases. Thus, cross-over between curves for different dimensions are observed around $w = 2$ to 3. One may also observe that marked departures from $\alpha^\infty_D$ are found for $w \approx 10$ and below. Taking monolayer MoS$_2$ sandwiched between SiO$_2$ and Al$_2$O$_3$ barriers as an example [8, 9] the quantum well thickness is $2a = 6.5$ Å and the potential can be estimated as $V_0 \approx 3.5$ eV. Hence, using the free-electron mass $m_0$ for $m^*$ the normalized potential barrier becomes $w \approx 3.1$. The increase of $\alpha_i$ over $\alpha^\infty_1$ is then approximately 3.7 in agreement with [9]. It may be argued that this is an extreme case because of the very thin material. However, e.g., in GaAs/Al$_{0.3}$Ga$_{0.7}$As hetero-junctions, barriers of the order $V_0 \approx 0.2$ eV are typical and effective electron masses are as low as $m^* \approx 0.067 m_0$ [7]. Thus, even for a relatively thick quantum well of $2a \approx 100$ Å one finds $w \approx 3.0$. It follows that this case is also very far from the infinite-barrier limit.
The analytical expression equation (12) is easily evaluated but requires a prior numerical solution for the unperturbed eigenvalue problem equation (9). Ideally, however, a fully closed-form expression depending only on \( D \) and \( w \) would be preferable. While no such exact expression can be found it is possible to provide approximate closed-form expressions. To this end, the ‘deep quantum well approximation’ valid for \( w \gg 1 \) is considered. The objective is to improve upon the infinite barrier limit, for which \( q = \infty \) while \( k = \lambda_D \) with \( J_{D/2-1}(\lambda_D) = 0 \). Expanding equation (9) to first order in \( w^{-1} \) one still finds \( q = \infty \) while now \( k \approx \lambda_D (1 - w^{-1}) \). In turn, this implies a Bessel function ratio of \( R = J_{D/2-1}(k)/J_{D/2}(k) \approx \lambda_D/w \). When inserted into equation (12) and upon further reduction it then turns out that

\[
\alpha_D \approx \alpha_0 \frac{(D - 6)(D - 4)D + 4\lambda_D^2 (2D - 3)}{12D^2 \lambda_D^2} \left\{ 1 + 4 \frac{1}{w} \right\}.
\]

Hence, to lowest order, the finite-barrier correction is a universal factor independent of dimension \( D \). The approximate results based on this approximation are shown in the upper panel of figure 4.

While the deep quantum well approximation is a clear improvement over the \( \alpha_D^{\infty} \) limit, marked deviations from the exact results are still observed, in particular, for \( w \leq 10 \). Hence, it is tempting to improve further upon equation (19) by regarding the correction \( 1 + 4w^{-1} \) as a first order expansion of a more complicated factor. Surely, this factor will depend on dimension \( D \) as well as \( w \) but, taking a heuristic approach, the \( D \)-dependence can be ignored. Still, a factor \( 1 + 4w^{-1} \) represents the limiting behavior of many plausible functions. An obvious choice, however, is the exponential \( \exp \left\{ 4/w \right\} = 1 + 4/w + 8/w^2 + \ldots \) so that

\[
\alpha_D \approx \alpha_0 \frac{(D - 6)(D - 4)D + 4\lambda_D^2 (2D - 3)}{12D^2 \lambda_D^2} \exp \left\{ 4/w \right\}.
\]

Polarizability curves based on this heuristic ‘improved deep quantum well approximation’ are displayed in the lower panel of figure 4. In comparison to the exact curves, very good agreement over a wide range of \( w \)-values is found, in particular, for the low-dimensional cases. For \( D = 1 \) and 1.5, the agreement is practically perfect. Hence, this approximate expression provides an easily evaluated alternative to the exact, but more complicated one equation (12).
4. Frequency-dependent polarizability

All of the above results were restricted to the static regime. In the present section, it is demonstrated how this may actually be extended to the more general time-dependent case. This situation is typically somewhat more involved that the static one since the full time-dependent Schrödinger equation must be solved rather than the eigenvalue problem equation (3). Nevertheless, for the $D$-dimensional quantum well, such a solution can be found. Thus, the static electric field of equation (3) is replaced by a time-dependent monochromatic field of frequency $\omega$ so that

$$\{H_0 + E(e^{-i\omega t} + e^{i\omega t})r \cos \theta\} \psi(r, \theta, t) = \frac{i}{\hbar} \frac{\partial}{\partial t} \psi(r, \theta, t).$$

Again, the first step of the solution is collecting first order terms. In the presence of the field, the wave function is modified by first-order terms $\varphi_+^2$ and $\varphi_-^2$ proportional to the positive ($e^{-i\omega t}$) and negative ($e^{i\omega t}$) frequency perturbations, respectively. These terms are found by writing the full wave function as

$$\psi(r, \theta, t) = \varphi_0^2(r) e^{-i\omega t} + \frac{1}{2} \varphi_+^2(r, \theta) e^{-i(\omega_0 + \omega)t} + \frac{1}{2} \varphi_-^2(r, \theta) e^{-i(\omega_0 - \omega)t} + \cdots.$$  

Here, $E_0$ is the unperturbed ground state energy. Inserting this into equation (21) and upon collecting positive and negative frequency first-order terms, it is found that

$$\{H_0 - E_0 \mp \omega\} \varphi_\pm(r, \theta) + 2E \cos \theta \varphi_0(r) = 0.$$  

This is the generalization of the Dalgarno–Lewis inhomogeneous problem to the frequency-dependent case. It is seen to follow from the replacement $E_0 \rightarrow E_0 \pm \omega$ representing states shifted in energy by the frequency $\omega$. This seemingly modest modification, however, complicates the solution quite dramatically. For this reason, only the infinite-barrier case will be pursued. Hence, writing $\varphi_\pm(r, \theta) = 2E \cos \theta \varphi_0(r)$ in analogy with the static case, it can be demonstrated that

$$f_\pm(r) = \frac{2^{3/2} \lambda_0^{D-1/2}}{\omega^2} \left\{ \pm \frac{\omega r}{2\lambda_0^2} \left( \frac{\lambda_0}{\lambda_0^2} \right) - \frac{I_{2D-1}^{(2)}(\lambda_0)}{I_{2D}^{(2)}(\lambda_0)} - \frac{I_{2D-1}^{(2)}(\lambda_0)}{I_{2D}^{(2)}(\lambda_0)} \right\},$$

where $\lambda_0^\pm = \sqrt{\lambda_0^2 \pm \omega}$. Generalizing equation (6), the frequency-dependent polarizability is then found to follow from the expression

$$\alpha_D(\omega) = \frac{2}{D} \int_0^\infty \varphi_0(r) \left\{ f_+(r) + f_-(r) \right\} r^D dr.$$  

To evaluate the integrals, use is made of the result

$$\int_0^\infty I_{\nu}(\alpha r) J_{\nu}(\beta r) r^\nu dr = \frac{\alpha I_{\nu+1}(\alpha)}{\alpha^2 - \beta^2}$$

$$\times \left\{ -2\beta I_{\nu}(\beta) + (\alpha^2 - \beta^2) J_{\nu+1}(\beta) \right\},$$

which is valid for arbitrary $\nu$ provided $I_{\nu}(\alpha) = 0$. Hence, from equations (24) and (25) one finds

$$\alpha_D(\omega) = -\frac{4\alpha_0}{\omega^2} \left\{ 1 + \frac{4\lambda_0^2 \lambda_0^D J_{2D-1}^{(2)}(\lambda_0) - 4\lambda_0^2 \lambda_0 J_{2D-1}^{(2)}(\lambda_0)}{4\lambda_0^2 J_{2D}^{(2)}(\lambda_0)} \right\}.$$  

This rather compact expression is a general result valid for arbitrary dimension $D$ and frequency $\omega$. To the knowledge of the present author, similar closed-form results for non-trivial models have only previously been obtained for the Hydrogen atom [24]. To demonstrate the correct $\omega \rightarrow 0$ limit, one uses the fourth order expansion

$$\frac{\lambda_0^D J_{2D-1}^{(2)}(\lambda_0)}{J_{2D}^{(2)}(\lambda_0)} = \frac{D \omega^2}{4\lambda_0^2} - \frac{\omega^4(D(D-4)(D-6) + 4\lambda_0^2(2D-3))}{192\lambda_0^6} + O(\omega^6).$$

Using this limit, equation (27) agrees perfectly with equation (14). Similarly, $\alpha_D(\omega) \rightarrow -4\alpha_0/\omega^2$ as $\omega \rightarrow \infty$. Converting to ordinary units this limit is $\lim_{\omega \rightarrow \infty} \alpha_D(\omega) \rightarrow -e^2/mr^2 \omega^2$ in agreement with the Thomas–Reiche–Kuhn sum rule [25]. For the integer-dimensional cases, the explicit expressions are

$$\alpha_1(\omega) = -\frac{4\alpha_0}{\omega^2} \left\{ 1 + \frac{\pi^2}{\omega^2} \left( \frac{\pi^2}{2} + \omega \cot \left( \frac{\pi}{2} \right) \right) + \frac{\pi^2}{\omega^2} \left( \frac{\pi^2}{2} - \omega \cot \left( \frac{\pi}{2} \right) \right) \right\},$$

$$\alpha_2(\omega) = -\frac{4\alpha_0}{\omega^2} \left\{ 1 + \frac{2\lambda_0^2 \lambda_0^2 J_{2}^{(2)}(\lambda_0^2) + \omega J_2(\lambda_0^2 + \omega)}{\omega^2 J_2(\lambda_0^2 + \omega)} + \frac{2\lambda_0^2 \lambda_0^2 J_{-2}^{(2)}(\lambda_0^2 - \omega)}{\omega^2 J_2(\lambda_0^2 - \omega)} \right\}.$$
These results as well as those for $D = 1.5$ and $2.5$ are illustrated in figure 5. A small imaginary part of magnitude unity has been added to the frequency to ensure a finite line width at the resonances. The upper and lower panels of the figure show the imaginary and real parts of the polarizability multiplied by $\omega^3$ and $\omega^2$, respectively. This scaling has been applied in order to increase visibility of the higher resonances. Also, the low-frequency part of the unscaled polarizability is shown in the inset. This shows that, in the DC limit $\omega \to 0$, agreement with the previous results for the static case found in figures 2 and 3 is obtained. Figure 5 also illustrates the universal behavior of the high-frequency response. Upon increasing the dimension $D$, all resonances shift to higher frequencies as expected due to the increased degree of confinement. It is remarkable that all these characteristics including the infinitely many resonances are captured in the simple and compact expression equation (27).

5. Application to CdSe quantum dots

Quantum dots with three-dimensional confinement are highly sensitive to the finite-barrier potential as demonstrated above. Hence, it is interesting to compare the present theory to experiments for such structures. The measurements by Wang et al [26] of exciton polarizabilities in CdSe nanocrystals are well suited for this purpose. These nanocrystals are sufficiently small ($a \approx 1.4–2.4$ nm) that Coulomb effects can be regarded as free electrons and holes to a good approximation. This also means that the total quantum dot polarizability is simply the sum of electron and hole contributions. The effect of electron–hole interaction on the Stark shift in CdSe quantum dots has been included in purely numerical studies [27, 28] but are minor corrections whenever $a < a_B$ with $a_B \approx 5$ nm the effective Bohr radius [26]. While the effective electron mass is almost isotropic, the hole mass of Wurzite CdSe has a pronounced dependence on direction [7]. This complication will be ignored, however, and as spherical averages the values $m_e^* \approx 0.13m_0$ and $m_h^* \approx 0.45m_0$ will be adopted [27–29]. Furthermore, the actual nanocrystals in [26] are capped by a thin ZnS shell and suspended in an organic solvent. Since no reliable estimates of the confinement for such structures are available, silicate glass will be used as a substitute for the insulating surroundings. Following Einevoll [30], a band
A gap of 7 eV in the barrier material combined with the CdSe band gap of 1.84 eV [29] yields potential barriers of $V_0 \approx 2.6$ eV assuming identical electron and hole values. Thus, for a nanocrystal radius of $a \approx 1.5$ nm the normalized potential barriers $w$ are as low as 4.5 and 8.3 for electrons and holes, respectively. It is quite clear from the discussion above that this is in the regime, where finite barriers are expected to be of importance, in particular, for the electron contribution.

To compute the quantum dot polarizability, equation (18) is evaluated using both electron and hole parameters and adding the results. The individual contributions as well as their sum are illustrated in figure 6, in which experimental data from [26] are also reproduced. In order to compare directly with experiments, the scaled quantity $\alpha / 4\pi\varepsilon_0$, having units of volume is displayed. The results of infinite-barrier confinement are included for comparison. For the electron contribution, in particular, the infinite-barrier assumption leads to a significant underestimation of the polarizability due to the low effective mass. Hence, the error amounts to approximately 50% and 35% at particle radii of 2 and 3 nm, respectively. For holes with a much larger mass, the error is smaller but still significant. At $a = 2$ nm the summed electron and hole contributions are $\sim 640 \, \text{Å}^3$ and $1000 \, \text{Å}^3$ for finite and infinite barriers, respectively, thus indicating an error of 35%. It is noted, however, that even including a finite barrier the calculated values are somewhat smaller than the experimental results. This discrepancy is probably a consequence of the ignored mass anisotropy and ZnS capping, which should lower the effective barrier.

6. Summary

Summarizing, a theory of the single-particle polarizability applicable to nanostructures in various dimensions has been developed and explored. By working in fractional-dimensional coordinates, a completely general framework valid for any dimension has been constructed. However, complications such as anisotropic masses, ‘non-spherical’ confinement, and electron–hole interaction have been ignored. The important role of finite barriers on the polarizability has been stressed, in particular, for higher-dimensional confinement. An approximate but extremely simple closed-form expression for the dimension- and barrier-dependent polarizability has been proposed. In addition, a compact expression for the frequency dependence of the infinite-barrier polarizability is obtained. As an application, the theory has been compared to experimental results for CdSe quantum dots. Here, the inclusion of finite barriers is shown to improve agreement with experiments significantly.

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