

# Quantum ergodicity and localization of plasmon resonances

H. Ammari and Y. Chow and H. Liu

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Seminar für Angewandte Mathematik  
Eidgenössische Technische Hochschule  
CH-8092 Zürich  
Switzerland

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# QUANTUM ERGODICITY AND LOCALIZATION OF PLASMON RESONANCES

HABIB AMMARI, YAT TIN CHOW, AND HONGYU LIU

ABSTRACT. We are concerned with the geometric properties of the surface plasmon resonance (SPR). SPR is a non-radiative electromagnetic surface wave that propagates in a direction parallel to the negative permittivity/dielectric material interface. It is known that the SPR oscillation is topologically very sensitive to the material interface. However, we show that the SPR oscillation asymptotically localizes at places with high magnitude of curvature in a certain sense. Our work leverages the Heisenberg picture of quantization and quantum ergodicity first derived by Shnirelman, Zelditch, Colin de Verdiere and Helffer-Martinez-Robert, as well as certain novel and more general ergodic properties of the Neumann-Poincaré operator to analyse the SPR field, which are of independent interest to the spectral theory and the potential theory.

**Keywords:** surface plasmon resonance, localization, quantum ergodicity, high curvature, Neumann-Poincaré operator, quantization

**2010 Mathematics Subject Classification:** 58J50, 58J51, 35Q60, 82D80

## 1. INTRODUCTION

**1.1. Mathematical formulation.** In this work, we are mainly concerned with the plasmonic eigenvalue problem as follows. Let  $D$  be an open connected and bounded domain in  $\mathbb{R}^d$ ,  $d \geq 2$ , with a  $\mathcal{C}^{2,\alpha}$ ,  $0 < \alpha < 1$ , boundary  $\partial D$  and a connected complement  $\mathbb{R}^d \setminus \overline{D}$ . Let  $\gamma_c$  and  $\gamma_m$  be two real constants with  $\gamma_m \in \mathbb{R}_+$  given and fixed. Let

$$\gamma_D = \gamma_c \chi(D) + \gamma_m \chi(\mathbb{R}^d \setminus \overline{D}), \quad (1.1)$$

where and also in what follows,  $\chi$  stands for the characteristic function of a domain. Consider the following homogeneous problem for a potential field  $u \in H_{loc}^1(\mathbb{R}^d)$ ,

$$\nabla \cdot (\gamma_D \nabla u) = 0 \text{ in } \mathbb{R}^d; \quad u(x) = \mathcal{O}(|x|^{1-d}) \text{ when } d \geq 2 \text{ as } |x| \rightarrow \infty, \quad (1.2)$$

where the last asymptotics holds uniformly in  $\hat{x} := x/|x| \in \mathbb{S}^d$  and is known as the decay condition. Note that (1.2) is equivalent to the following transmission problem:

$$\begin{cases} \Delta u = 0 & \text{in } D \cup (\mathbb{R}^d \setminus \overline{D}), \\ u^+ = u^- & \text{on } \partial D, \\ \gamma_c \frac{\partial u^+}{\partial \nu} = \gamma_m \frac{\partial u^-}{\partial \nu} & \text{on } \partial D, \\ u \text{ satisfies the decay condition as } |x| \rightarrow \infty, \end{cases} \quad (1.3)$$

where  $\pm$  signify the traces taken from the inside and outside of  $D$  respectively. If there exists a nontrivial solution  $u$  to (1.3), then  $\gamma_c$  is referred to as a plasmonic eigenvalue and  $u$  is the associated plasmonic resonant field. It is apparent that a plasmonic eigenvalue must be negative, since otherwise by the ellipticity of the partial differential operator (PDO)  $\mathcal{L}_{\gamma_D} u := \nabla(\gamma_D \nabla u)$ , (1.3) admits only a trivial solution. The plasmonic eigenvalue problem

is delicately connected to the spectral theory of the Neumann-Poincaré (NP) operator as follows. Let  $\Gamma$  be the fundamental solution of the Laplacian in  $\mathbb{R}^d$  :

$$\Gamma(x-y) = \begin{cases} -\frac{1}{2\pi} \log|x-y| & \text{if } d=2, \\ \frac{1}{(2-d)\varpi_d} |x-y|^{2-d} & \text{if } d>2, \end{cases} \quad (1.4)$$

with  $\varpi_d$  denoting the surface area of the unit sphere in  $\mathbb{R}^d$ . The Neumann-Poincaré (NP) operator  $\mathcal{K}_{\partial D}^* : L^2(\partial D, d\sigma) \rightarrow L^2(\partial D, d\sigma)$  is defined by

$$\mathcal{K}_{\partial D}^*[\phi](x) := \frac{1}{\varpi_d} \int_{\partial D} \frac{\langle x-y, \nu(x) \rangle}{|x-y|^d} \phi(y) d\sigma(y), \quad (1.5)$$

where  $\nu(x)$  signifies the unit outward normal at  $x \in \partial D$ . It is remarked that the NP operator is a classical weakly-singular boundary integral operator in potential theory [5, 26]. Then a plasmonic resonant field to (1.3) can be represented as a single-layer potential:

$$u(x) = \mathcal{S}_{\partial D}[\phi](x) := \int_{\partial D} \Gamma(x-y) \phi(y) d\sigma(y), \quad x \in \mathbb{R}^d, \quad (1.6)$$

where the density distribution  $\phi \in H^{-1/2}(\partial D, d\sigma)$  satisfies

$$\mathcal{K}_{\partial D}^*[\phi] = \lambda(\gamma_c, \gamma_m) \phi, \quad \lambda(\gamma_c, \gamma_m) := \frac{\gamma_c + \gamma_m}{2(\gamma_c - \gamma_m)}. \quad (1.7)$$

That is, in order to determine the plasmonic eigenvalue  $\gamma_c$  of (1.3), it is sufficient to determine the eigenvalues of the NP operator  $\mathcal{K}_{\partial D}^*$ . On the other hand, in order to understand the peculiar behaviour of the plasmonic resonant field, one needs to study the quantitative properties of the NP eigenfunctions in (1.7) as well as the associated single-layer potentials in (1.6). In this paper, we are mainly concerned with the geometric properties of the plasmonic eigenmode  $u$ , namely its quantitative relationships to the geometry of  $\partial D$ . This leads us to establish more general quantum ergodic properties of the singularly integral operators  $\mathcal{K}_{\partial D}^*$  and  $\mathcal{S}_{\partial D}$ . The plasmonic eigenvalue problem is the fundamental basis to the so-called surface plasmon resonance as shall be described in the following. The quantitative understanding of the plasmonic eigenmodes would yield deep theoretical insights on the surface plasmon resonance as well as produce significant physical and practical implications.

**1.2. Physical relevance and connection to existing studies of our results.** Surface plasmon resonance (SPR) is the resonant oscillation of conducting electrons at the interface between negative and positive permittivity materials stimulated by incident light. It is a non-radiative electromagnetic surface wave that propagates in a direction parallel to the negative permittivity/dielectric material interface. The SPR forms the fundamental basis for an array of industrial and engineering applications, from highly sensitive biological detectors to invisibility cloaks [10, 17, 28, 31, 33, 38, 40, 43, 54] through the constructions of different plasmonic devices. The plasmonics was listed as one of the top ten emerging technologies of 2018 by the Scientific American, stating that “light-controlled nanomaterials are revolutionizing sensor technology”. Next, we briefly discuss the SPR in electrostatics and in Section 5 we shall extend all the results in the electrostatic case to the scalar wave propagation in the quasi-static regime.

Consider a medium configuration given in (1.1), where  $\gamma_c$  and  $\gamma_m$  respectively specify the dielectric constants of the inclusion  $D$  and the background space  $\mathbb{R}^d \setminus \overline{D}$ . Let  $u_0$  be a harmonic function  $\mathbb{R}^d$  which represents an incident field. The electrostatic transmission problem is given for an electric potential field  $u \in H_{loc}^1(\mathbb{R}^d)$  as follows,

$$\begin{cases} \nabla \cdot (\gamma_D \nabla u) = 0 & \text{in } \mathbb{R}^d, \\ u - u_0 & \text{satisfies the decay condition as } |x| \rightarrow \infty. \end{cases} \quad (1.8)$$

Denote the perturbed potential field  $u - u_0$  as a single-layer potential (1.6) with a density function  $\phi$  to be determined by the transmission conditions across  $\partial D$ . Using the following jump relation across  $\partial D$ :

$$\frac{\partial}{\partial \nu} (\mathcal{S}_{\partial D}[\phi])^\pm = (\pm \frac{1}{2} Id + \mathcal{K}_{\partial D}^*)[\phi], \quad (1.9)$$

one can show that

$$\frac{\partial u_0}{\partial \nu} = \left( \frac{\gamma_c + \gamma_m}{2(\gamma_c - \gamma_m)} Id - \mathcal{K}_{\partial D}^* \right) [\phi] \quad \text{on } \partial D. \quad (1.10)$$

Hence, formally there holds

$$u = u_0 + \mathcal{S}_{\partial D} \circ \left( \lambda(\gamma_c, \gamma_m) Id - \mathcal{K}_{\partial D}^* \right)^{-1} \left[ \frac{\partial u_0}{\partial \nu} \Big|_{\partial D} \right], \quad (1.11)$$

where  $\lambda(\gamma_c, \gamma_m)$  is defined in (1.7). Clearly, if  $\lambda(\gamma_c, \gamma_m)$  is an eigenvalue to  $\mathcal{K}_{\partial D}^*$ , then resonance occurs for the boundary integral equation (1.10). Consequently, due a proper incident field  $u_0$ , resonance can be induced for the electrostatic system (1.8). These exactly give the plasmonic eigenvalue problem (1.3) and the NP eigenvalue problem (1.7).

According to our discussion above, the spectrum of the NP operator determines the plasmonic eigenvalues  $\gamma_c$  through the relationship given by  $\lambda(\gamma_c, \gamma_m)$ . That is, the spectra of the NP operator determine the negative dielectric constants which can induce the plasmon resonances. Such a connection has aroused growing interest on studying the spectral properties of the NP operator [2, 6, 7, 9, 13, 19, 27, 29, 32, 34, 35]. The NP operator  $\mathcal{K}_{\partial D}^*$  is compact and hence its eigenvalues are discrete, infinite and accumulating at zero. Moreover, one has  $\lambda(\mathcal{K}_{\partial D}^*) \subset (-1/2, 1/2]$ . It can be directly verified that if  $\gamma_c$  and  $\gamma_m$  are both positive, then  $|\lambda(\varepsilon_c, \varepsilon_m)| > 1/2$ . In such a case, the invertibility of the operator  $(\lambda(\varepsilon_c, \varepsilon_m)I - \mathcal{K}_{\partial D}^*)$  from  $L^2(\partial D, d\sigma)$  onto  $L^2(\partial D, d\sigma)$  and from  $L_0^2(\partial D, d\sigma)$  onto  $L_0^2(\partial D, d\sigma)$  is proved (cf. [5, 26]) via the Fredholm theory. This once again necessitates the negativity of the plasmonic eigenvalues  $\gamma_c$ . It is easily seen, from the properties of  $\mathcal{K}_{\partial D}^*$ , that the NP eigenvalues are invariant with respect to rigid motions and scaling. The spectrum of  $\mathcal{K}_{\partial D}^*$  can be explicitly computed for ellipses and spheres [24, 27]. It is worth pointing out that the convergence to zero of those eigenvalues is exponential for ellipses while it is algebraic for spheres. The exponential convergence is critical for the construction of plasmonic devices that can induce invisibility cloaks [3, 30]. Some other computations of Neumann-Poincaré eigenvalues as well as the corresponding plasmonic applications for different shapes can be found in [12, 13, 23]. More recently in [34, 35], it is derived in three dimensions a quantization rule of the NP eigenvalues, showing that the leading-order asymptotics of the  $j$ -th ordered NP eigenvalue with  $j \gg 1$  can be expressed in terms of two global geometric quantities of  $\partial D$ , namely the Euler characteristic and the Willmore energy.

It is clear to see that if plasmon resonance occurs, the peculiar behaviours of the resonant field critically depend on the quantitative properties of the NP eigenfunctions. Indeed, according to (1.10) and via the spectral resolution, the density distribution  $\phi$  can be expressed in terms of the NP eigenmodes, and this in turn gives the resonant modes via the single-layer potentials. The SPR field is the superposition of those resonant modes. Hence, in order to gain a thorough understanding of the resonant field, one should carefully study the quantitative properties of the NP eigenfunctions as well as the associated single-layer potentials. However, to our best knowledge, there is little study in the literature on this aspect. It is known the SPR propagation is topologically very sensitive to the material interface  $\partial D$ . That is, the SPR is sensitive to any change of the global geometry of  $\partial D$ . In fact, such a topological sensitivity forms the fundamental basis of the aforementioned bio-sensing application of SPRs. Nevertheless, it is speculated that the SPRs may possess certain invariant/robust property related to the local geometry of  $\partial D$ . Indeed, it is

observed in several numerics for some specific geometries [12] that the SPR waves reveal certain concentration/localisation phenomena at places where the magnitude of the associated curvature is relatively high. The aim of this paper is to rigorously establish the local geometric invariant property of the SPRs in a very general setup. In fact, we show that the SPR wave localizes in a certain sense at places where the magnitude of the associated extrinsic curvature (namely the second fundamental form) is relatively high. Since the SPR depends on  $\partial D$  globally, it is highly nontrivial to extract the local geometric information. Nevertheless, we establish a certain more general property of the SPR waves with the help of quantum ergodicity, with which the localization property is a natural consequence of the dynamics of an associated Hamiltonian. In addition to its theoretical significance, our result may have potential applications in generating SPRs that break the quasi-static limit [29] and produce plasmonic cloaks [2, 33], which are worth of investigation in our future study. Our study also leverages certain novel ergodic properties of the NP operator, which should be of independent interest to the spectral theory and potential theory.

**1.3. Discussion of the technical novelty.** Finally, we briefly discuss the mathematical strategies of establishing the quantum ergodicity and localization results. As discussed earlier, we need to analyze the geometric structures of the NP eigenfunctions as well as the associated single-layer potentials; that is, the quantitative behaviours of those distributions that are related to the boundary geometry of the underlying domain. Treating those layer-potential operators as pseudo-differential operators, we consider the Hamiltonian flows of the principle symbols of those operators. In particular, we obtain, via a generalized Weyl's law, that the asymptotic average of the magnitude of the NP eigenfunctions in a neighborhood of each point is directly proportional to a weighted volume of the characteristic variety at the respective point. Moreover, following the pioneering works of Shnirelman [41, 42], Zelditch [48–53], Colin de Verdiere [14] and Helffer-Martinez-Robert [20] (See also [16, 18, 44, 45]), by considering the Heisenberg picture and lifting the Hamiltonian flow of a principal symbol to a wave propagator, we generalize a result of quantum ergodicity via an application of the ergodicity decomposition theorem to include dynamics which is not uniquely ergodic. From that, we obtain a subsequence (of density one) of eigenfunctions such that their magnitude weakly converges to a weighted average of ergodic measures. This weighted average at different points relates to the volumes of the characteristic variety at the respective points. We also provide an upper and lower bounds of the volume of the characteristic variety as functions only depending on the principal curvatures. We therefore can characterize the localization of the plasmon resonance by the associated extrinsic curvature at a specific boundary point. From our result, we have also associated the understanding of plasmon resonances to the dynamical properties of the Hamiltonian flows. For instance, a Hamiltonian circle action will result in a parametrization of ergodic measures by a compact symplectic manifold of dimension  $2d - 4$  via a symplectic reduction. Our study opens up a new field with many possible developments on the quantitative properties of plasmon resonances as well as on the spectral properties of Neumann-Poincaré type operators.

The rest of the paper is organized as follows. In Section 2, we briefly discuss the principal symbols of the layer-potential operators. In Section 3, we recall the generalized Weyl's law, and generalize the argument of the quantum ergodicity to obtain a variance-like estimate. In Section 4, we apply the generalized Weyl's law and our generalization of the quantum ergodicity to obtain a comparison result of the magnitude of NP eigenfunction at different points with extrinsic curvature information at the respective points. These combine to give a description of localization of the plasmon resonance around points of high curvature. We present extensions to the plasmon resonance in the Helmholtz transmission problem in the quasi-static regime in Section 5. In Appendix A, we present further discussion upon geometric descriptions of the related Hamiltonian flows.

## 2. SYMBOLS OF POTENTIAL OPERATORS

In this section, we present the principle symbols of the Neumann-Poincaré operator (1.5) and the single-layer potential operator  $\mathcal{S}_{\partial D}$  in (1.6) associated with a shape  $D$  sitting inside a general space  $\mathbb{R}^d$  for any  $d \geq 2$ . The special three-dimensional case was first treated in [34, 35], and the general case was considered in [4]. Since this result is of fundamental importance for our future analysis, we shall briefly restate here for the sake of completeness.

We briefly introduce the geometric description of  $D \subset \mathbb{R}^d$ . Consider a regular parametrization of the surface  $\partial D$  as

$$\begin{aligned} \mathbb{X} : U \subset \mathbb{R}^{d-1} &\rightarrow \partial D \subset \mathbb{R}^d, \\ u = (u_1, u_2, \dots, u_{d-1}) &\mapsto \mathbb{X}(u). \end{aligned}$$

For notational sake, we often write the vector  $\mathbb{X}_j := \frac{\partial \mathbb{X}}{\partial u_j}$ ,  $j = 1, 2, \dots, d-1$ . For a given  $d-1$  vector  $\{v_j\}_{j=1}^{d-1}$ , we denote the  $d-1$  cross product  $\times_{j=1}^{d-1} v_j = v_1 \times v_2 \dots \times v_{d-1}$  as the dual vector of the functional  $\det(\cdot, v_1, v_2, \dots, v_{d-1})$ , i.e.,  $\langle w, \times_{j=1}^{d-1} v_j \rangle = \det(w, v_1, v_2, \dots, v_{d-1})$  for any  $w$ , whose existence is guaranteed by the Reisz representation theorem. Then, from the fact that  $\mathbb{X}$  is regular, we know  $\times_{j=1}^{d-1} \mathbb{X}_j$  is non-zero, and the normal vector  $\nu := \times_{j=1}^{d-1} \mathbb{X}_j / |\times_{j=1}^{d-1} \mathbb{X}_j|$  is well-defined. Next, we introduce the following matrix  $A_{ij}(x)$ ,  $x \in \partial D$ , defined as

$$A(x) := (A_{ij}(x)) = \langle \mathbf{II}_x(\mathbb{X}_i, \mathbb{X}_j), \nu_x \rangle,$$

where  $\mathbf{II}$  is the second fundamental form given by

$$\begin{aligned} \mathbf{II} : T(\partial D) \times T(\partial D) &\rightarrow T^\perp(\partial D), \\ \mathbf{II}(v, w) &= -\langle \bar{\nabla}_v \nu, w \rangle \nu = \langle \nu, \bar{\nabla}_v w \rangle \nu, \end{aligned}$$

with  $\bar{\nabla}$  being the standard covariant derivative on the ambient space  $\mathbb{R}^d$ . Moreover, we write  $H(x)$ ,  $x \in \partial D$  as the mean curvature satisfying

$$\text{tr}_{g(x)}(A(x)) := \sum_{i,j=1}^{d-1} g^{ij}(x) A_{ji}(x) := (d-1)H(x),$$

with  $(g^{ij}) = g^{-1}$  and  $g = (g_{ij})$  being the induced metric tensor. From now on, we shall always assume  $A(x) \neq 0$  for all  $x \in \partial D$  in this work. We are now ready to present the principle symbol of  $\mathcal{K}_{\partial D}^*$  (cf. [4, 34, 35]).

**Theorem 2.1.** *The operator  $\mathcal{K}_{\partial D}^*$  is a pseudodifferential operator of order  $-1$  on  $\partial D$  if  $\partial D \in \mathcal{C}^{2,\alpha}$  with its symbol given as follows in the geodesic normal coordinate around each point  $x$ :*

$$\begin{aligned} p\mathcal{K}_{\partial D}^*(x, \xi) &= p\mathcal{K}_{\partial D, -1}^*(x, \xi) + \mathcal{O}(|\xi|^{-2}) \\ &= (d-1)H(x) |\xi|^{-1} - \langle A(x) \xi, \xi \rangle |\xi|^{-3} + \mathcal{O}(|\xi|^{-2}), \end{aligned} \tag{2.1}$$

where the asymptotics  $\mathcal{O}$  depends on  $\|\mathbb{X}\|_{\mathcal{C}^2}$ . Hence  $\mathcal{K}_{\partial D}^*$  is a compact operator of Schatten  $p$  class  $S_p$  for  $p > d-1$  for  $d > 2$ .

A remark is that the above result holds also for  $\mathcal{K}_{\partial D}$  instead of  $\mathcal{K}_{\partial D}^*$  when we only look at the leading-order term. Here,  $\mathcal{K}_{\partial D}$  signifies the  $L^2(\partial D, d\sigma)$ -adjoint of the NP operator  $\mathcal{K}_{\partial D}^*$ . We would also like to remark that if geodesic normal coordinate is not chosen, and for a general coordinate, we have instead

$$p\mathcal{K}_{\partial D}^*(x, \xi) = (d-1)H(x) |\xi|_{g(x)}^{-1} - \langle A(x) g^{-1}(x) \xi, g^{-1}(x) \xi \rangle |\xi|_{g(x)}^{-3} + \mathcal{O}(|\xi|_{g(x)}^{-2}).$$

From the fact that the Dirichlet-to-Neumann map  $\Lambda_0 : H^{1/2}(\partial D, d\sigma) \rightarrow H^{-1/2}(\partial D, d\sigma)$  of the Laplacian in the domain  $D \subset \mathbb{R}^d$  satisfies the following [46]:

$$p_{\Lambda_0}(x, \xi) = p_{\Lambda_0,1}(x, \xi) + \mathcal{O}(1) = |\xi|_{g(x)} + \mathcal{O}(1), \quad (2.2)$$

together with the jump relation (1.9), one can handily compute that:

$$p_{\mathcal{S}_{\partial D}}(x, \xi) = p_{\mathcal{S}_{\partial D},-1}(x, \xi) + \mathcal{O}(|\xi|_{g(x)}^{-2}) = \frac{1}{2}|\xi|_{g(x)}^{-1} + \mathcal{O}(|\xi|_{g(x)}^{-2}). \quad (2.3)$$

We recall the following well-known Kelley symmetrization identity:

$$\mathcal{S}_{\partial D} \mathcal{K}_{\partial D}^* = \mathcal{K}_{\partial D} \mathcal{S}_{\partial D}, \quad (2.4)$$

which indicates that  $\mathcal{K}_{\partial D}^*$  is symmetrizable on  $H^{-1/2}(\partial D, d\sigma)$  (cf., e.g., [8, 25]), i.e.  $\mathcal{K}_{\partial D}^*$  is a self-adjoint operator on  $L_{\mathcal{S}_{\partial D}}^2(\partial D) := (\overline{\mathcal{C}^\infty(\partial D)})^{\|\cdot\|_{\mathcal{S}_{\partial D}}}, \langle \cdot, \cdot \rangle_{\mathcal{S}_{\partial D}}$ , where, for any  $f \in L_{\mathcal{S}_{\partial D}}^2(\partial D)$ ,

$$\|f\|_{\mathcal{S}_{\partial D}}^2 := \langle f, f \rangle_{\mathcal{S}_{\partial D}} := -\langle \mathcal{S}_{\partial D} f, f \rangle_{H^{1/2}(\partial D, d\sigma), H^{-1/2}(\partial D, d\sigma)},$$

is a well-defined inner product for  $d \geq 3$  and with a minor modification for  $d = 2$  (see [6, 8]). We remark that there is an equivalence between the two norms  $\|\cdot\|_{\mathcal{S}_{\partial D}}$  and  $\|\cdot\|_{H^{-1/2}(\partial D, d\sigma)}$ .

Using the symmetrization identity (2.4) and by comparing the corresponding symbols, together with the fact that  $\mathcal{S}_{\partial D}$  is self-adjoint, we have

$$\begin{aligned} \mathcal{K}_{\partial D}^* &= |D|^{-1} \left\{ (d-1)H(x)\Delta_{\partial D} - \sum_{i,j,k,l=1}^{d-1} \frac{1}{\sqrt{|g(x)|}} \partial_i g^{ij}(x) \sqrt{|g(x)|} A_{jk}(x) g^{kl}(x) \partial_l \right\} |D|^{-2} \text{ mod } \Phi\text{SO}^{-2}, \\ \mathcal{S}_{\partial D} &= \frac{1}{2}|D|^{-1} \text{ mod } \Phi\text{SO}^{-2}. \end{aligned} \quad (2.5)$$

In (2.5),  $\Delta_{\partial D}$  is the surface Laplacian of  $\partial D$ , and  $|D|^{-1} := \text{Op}_{|\xi|_{g(x)}^{-1}}$  where  $\text{Op}_a = \mathcal{F}^{-1} \circ m_a \circ \mathcal{F}$  is the action given by the symbol without any large/small parameter, where  $\mathcal{F}$  is the Fourier transform (defined via a partition of unity, and is unique modulus  $\Phi\text{SO}^{m-1}$  if  $a \in \tilde{\mathcal{S}}^m(T^*(\partial D))$ ) that belongs to the symbol class of order  $m$ , and  $m_a$  is the action with multiplication by the symbol  $a$ . We notice that the operator in the curly bracket in (2.5) is itself symmetric. We therefore have

$$\mathcal{K}_{h,\partial D}^* := \frac{1}{h} |D|^{-\frac{1}{2}} \mathcal{K}_{\partial D}^* |D|^{\frac{1}{2}}$$

being self adjoint up to mod  $h\Phi\text{SO}_h^{-2}$ . In here,  $\Phi\text{SO}_h^{-m}$  is the pseudo-differential operator with action  $\text{Op}_{a,h} := \mathcal{F}_h^{-1} \circ m_a \circ \mathcal{F}_h$ , i.e., with a small parameter  $h$  (again uniquely defined modulus  $h\Phi\text{SO}_h^{m-1}$  if  $a \in \tilde{\mathcal{S}}^m(T^*(\partial D))$ ) belonging to the symbol class of order  $m$ . Here, we would like to clarify the following notations and definitions in our study,

$$\begin{aligned} \bigcup_i U_i &= \partial D, \quad F_i : \pi_i^{-1}(U_i) \rightarrow U_i \times \mathbb{R}^{d-1}, \quad \sum_i \psi_i = 1, \quad \text{supp}(\psi_i) \subset U_i; \\ \tilde{\mathcal{S}}^m(T^*(\partial D)) &:= \left\{ a : T^*(\partial D) \setminus \partial D \times \{0\} \rightarrow \mathbb{C}; a = \sum_i \psi_i F_i^* a_i, a_i \in \tilde{\mathcal{S}}^m(U_i \times \mathbb{R}^{d-1} \setminus \{0\}) \right\}; \\ \tilde{\mathcal{S}}^m(U_i \times \mathbb{R}^{d-1}) &:= \left\{ a : U_i \times (\mathbb{R}^{d-1} \setminus \{0\}) \rightarrow \mathbb{C}; \right. \\ &\quad \left. a \in \mathcal{C}^\infty(U_i \times (\mathbb{R}^{d-1} \setminus \{0\})), |\partial_\xi^\alpha \partial_x^\beta a(x, \xi)| \leq C_{\alpha,\beta} (|\xi|)^{m-|\alpha|} \right\}. \end{aligned}$$



Finally, we note that  $\left(\frac{\lambda_i^2}{h}, |D|^{-\frac{1}{2}}\phi_i\right)$  is an eigenpair of  $[\mathcal{K}_{h,\partial D}^*]^2$  if and only if  $(\lambda_i^2, \phi_i)$  is an eigenpair of  $\mathcal{K}_{\partial D}^*$  (cf. (1.7)). Throughout the rest of the paper, we denote

$$(\lambda_i^2(h), \phi_i(h)) := \left(\frac{\lambda_i^2}{h}, |D|^{-\frac{1}{2}}\phi_i\right). \quad (2.6)$$

### 3. GENERALIZED WEYL'S LAW AND QUANTUM ERGODICITY OVER THE NEUMANN-POINCARÉ OPERATOR

In this section, we recall the concept of quantum ergodicity following the pioneering works of Shnirelman [41, 42], Zelditch [48–53], Colin de Verdière [14] and Helffer-Martinez-Robert [20], (see also [16, 18, 44, 45]). Although it is a classical theorem, we would still sketch the proofs to some of the materials for the sake of completeness. Meanwhile, for our subsequent use, we would generalize it in a certain way via the ergodicity decomposition theorem.

**3.1. Hamiltonian flows of principle symbols.** We begin by considering the following Hamiltonian

$$\begin{aligned} H : T^*(\partial D) &\rightarrow \mathbb{R} \\ H(x, \xi) &= [p_{\mathcal{K}_{\partial D}^*, -1}(x, \xi)]^2 \geq 0. \end{aligned} \quad (3.1)$$

Note that  $T^*(\partial D)$  is endowed with the standard symplectic form  $\omega := \sum_{i=1}^{d-1} dx_i \wedge d\xi_i = d\alpha$ , where  $\alpha := \sum_{i=1}^{d-1} x_i d\xi_i$  is the canonical 1-form. Now notice that  $H$  is only smooth outside  $\partial D \times \{0\} \hookrightarrow T^*(\partial D)$ . We now impose an assumption that we will take through our work.

**Assumption (A)** We assume  $\langle A(x)g^{-1}(x)\omega, g^{-1}(x)\omega \rangle \neq (d-1)H(x)$  for all  $x \in \partial D$  and  $\omega \in \{\xi : |\xi|_{g(x)}^2 = 1\} \subset T_x^*(\partial D)$ .

As we will discuss in Appendix A, this assumption is related to the regularity of the Hamiltonian flow generated by  $H$  on the set  $\overline{\{H=1\}}$ . In particular, Assumption (A) holds if and only if  $\overline{\{H=1\}} \cap (\partial D \times \{0\}) = \emptyset$ . In fact, we realize that this assumption is equivalent to the condition that the Hamiltonian  $H \neq 0$  everywhere. With this, gazing at (1.7), we immediately have that  $\phi$  in (1.7) actually sits in  $H^s(\partial D, d\sigma)$  for all  $s$ , and thus by the Sobolev embedding,  $\phi \in C^\infty(\partial D)$ .

In this work (up till the appendix), we always assume the validity of Assumption (A). We speculate that this assumption is not necessary for the conclusions of our theorems to hold, and that is subjected to future studies.

Now, let us consider the following auxiliary function

$$\begin{aligned} \rho : \mathbb{R}_+ &:= \{r \in \mathbb{R} : r \geq 0\} \rightarrow \mathbb{R}, \\ \rho(r) &= 1 - \exp(-r), \end{aligned}$$

which will be very helpful in our subsequent analysis. In particular, we realize that  $\rho(r) \geq 0$  and  $\rho'(r) > 0$  for all  $\mathbb{R}_+$ . Moreover, one realize that  $\rho(1/r^2) \in C^\infty(\mathbb{R})$ , with  $\partial_r^k|_{r=0} [\rho(1/r^2)] = 0$  for all  $k \in \mathbb{N}$  and

$$|\partial_r^k \rho(1/r^2)| \leq C_k (1 + |r|^2)^{-\frac{2-k}{2}}.$$

With this function, we define

$$\begin{aligned} \tilde{H} : T^*(\partial D) &\rightarrow \mathbb{R}, \\ \tilde{H}(x, \xi) &= \rho(H(x, \xi)). \end{aligned} \quad (3.2)$$



We may now handily verify, under Assumption (A), that we have  $\tilde{H} \in \mathcal{C}^\infty(T^*(\partial D))$ , and in fact,  $\tilde{H} \in S^{-2}(T^*(\partial D))$ , where  $S^m(T^*(\partial D))$  denotes the smooth symbol class of order  $m$  defined as

$$\begin{aligned} S^m(T^*(\partial D)) &:= \left\{ a : T^*(\partial D) \rightarrow \mathbb{C}; a = \sum_i \phi_i F_i^* a_i, a_i \in S^m(U_i \times \mathbb{R}^{d-1}) \right\}, \\ S^m(U_i \times \mathbb{R}^{d-1}) &:= \left\{ a : U_i \times \mathbb{R}^{d-1} \rightarrow \mathbb{C}; a \in \mathcal{C}^\infty(U_i \times \mathbb{R}^{d-1}) \right. \\ &\quad \left. |\partial_\xi^\alpha \partial_x^\beta a(x, \xi)| \leq C_{\alpha, \beta} (1 + |\xi|^2)^{\frac{m-|\alpha|}{2}} \right\}. \end{aligned}$$

With the above notations, let us consider the following solution under a Hamiltonian flow:

$$\begin{cases} \frac{\partial}{\partial t} a(t) &= -\frac{i}{\hbar} \{\tilde{H}, a(t)\}, \\ a_0(x, \xi) &\in S^m(T^*(\partial D)), \end{cases} \quad (3.3)$$

where  $\{\cdot, \cdot\}$  is the Poisson bracket given by

$$\{f, g\} := X_f g = -\omega(X_f, X_g),$$

with  $X_f$  being the symplectic gradient vector field given by

$$\iota_{X_f} \omega = df.$$

We notice that, away from  $\partial D \times \{0\}$ , we have

$$X_{\tilde{H}} = \rho'(H) X_{\tilde{H}},$$

where  $\rho'(H) > 0$ , whereas  $X_{\tilde{H}} = 0$  on  $\partial D \times \{0\}$ . With this notion in hand, we have  $\frac{\partial}{\partial t} a = X_{\tilde{H}} a$ , and it is clear that  $a(t) = a_0(\gamma(t), p(t))$  where

$$\begin{cases} \frac{\partial}{\partial t} (\gamma(t), p(t)) &= X_{\tilde{H}}(\gamma(t), p(t)), \\ (\gamma(0), p(0)) &= (x, \xi) \in T^*(\partial D). \end{cases}$$

To emphasize the dependence of  $a$  on the initial value  $(x, \xi)$ , we also sometimes write

$$a_{x, \xi}(t) = a(t) \quad \text{with} \quad (\gamma(0), p(0)) = (x, \xi).$$

Next we introduce the Heisenberg's picture and lift the above flow to the operator level via Egorov's theorem. Since this is a well-known theorem, we only provide a sketch of the proof for the sake of completeness.

**Proposition 3.1.** [16, 21, 22] *Under Assumption (A), the following operator evolution equation*

$$\begin{cases} \frac{\partial}{\partial t} A_h(t) = \frac{i}{\hbar} [\text{Op}_{\tilde{H}, h}, A_h(t)], \\ A_h(0) = \text{Op}_{a_0, h}, \end{cases} \quad (3.4)$$

defines a unique Fourier integral operator (up to  $h^\infty \Phi\text{SO}_h^{-\infty}$ )

$$A_h(t) = e^{-\frac{it}{\hbar} \text{Op}_{\tilde{H}, h}} A_h(0) e^{\frac{it}{\hbar} \text{Op}_{\tilde{H}, h}} + \mathcal{O}(h \Phi\text{SO}_h^{m-1})$$

for  $t < C \log(h)$ . Moreover,

$$A_h(t) = \text{Op}_{a_{x, \xi}(t), h} + \mathcal{O}(h \Phi\text{SO}_h^{m-1}),$$

or that  $p_{A_h(t)}(x, \xi) = a_{(x, \xi)}(t) + \mathcal{O}(|\xi|^{-1})$ .

*Proof.* The existence of the solution to the equation (3.4) comes from first constructing the symbol in the principle level by noting that  $[\text{Op}_a, \text{Op}_b] = \text{Op}_{\{a,b\}} + \mathcal{O}(h\Phi\text{SO}_h^{m+n-2})$  if  $a \in S^m(T^*(\partial D))$  and  $b \in S^n(T^*(\partial D))$ . Then one inductively constructs the full symbol, and bounds the error operator via the Calderón-Vaillancourt theorem repeatedly. By Beal's theorem, the operator is guaranteed as an FIO.

The proof of both expressions of  $A_h(t)$  comes from checking that the principle symbols coincide, and then using the Zygmund trick to bound the error operator.  $\square$

Let us consider  $\tilde{H}(x, \xi) = \rho \left( [p_{\mathcal{K}_{\partial D}^*}(x, \xi)]^2 \right)$ . Then we immediately have

$$\text{Op}_{H,h} = \rho \left( [\mathcal{K}_{h,\partial D}^*]^2 \right) \bmod (h\Phi\text{SO}_h^{-3}),$$

and hence there holds the following corollary.

**Corollary 3.2.** *Under Assumption (A), the symbol of*

$$A_h(t) = e^{-\frac{it}{h}\rho([\mathcal{K}_{h,\partial D}^*]^2)} A_h(0) e^{\frac{it}{h}\rho([\mathcal{K}_{h,\partial D}^*]^2)} + \mathcal{O}(h\Phi\text{SO}_h^{-1}) \quad (3.5)$$

is given by

$$p_{A_h(t)}(x, \xi) = a_{(x,\xi)}(t) + \mathcal{O}(|\xi|^{-1}). \quad (3.6)$$

**3.2. Trace formula and generalized Weyl's law.** We first state the Schwartz functional calculus as follows.

**Lemma 3.3.** [21, 22] *Let  $\mathcal{S}(\mathbb{R})$  denote the space of Schwartz functions on  $\mathbb{R}$ . Then for  $f \in \mathcal{S}(\mathbb{R})$ ,  $f(\text{Op}_{a,h}) \in \Phi\text{SO}_h^{-\infty}$  and*

$$f(\text{Op}_{a,h}) = \text{Op}_{f(a)} + \mathcal{O}(h\Phi\text{SO}_h^{-\infty}). \quad (3.7)$$

*Proof.* The theorem can be proved via an almost holomorphic extension of  $f$  to  $f^{\mathbb{C}}$  (e.g. by Hörmander [21, 22]) and the Helffer-Sjöstrand formula  $f(A) = \frac{1}{2\pi i} \int_{\mathbb{C}} \partial_{\bar{z}} f^{\mathbb{C}}(z - A)^{-1} dz \wedge d\bar{z}$ .  $\square$

We proceed to state the following trace theorem, and again give only a brief sketch of the proof for the sake of completeness.

**Proposition 3.4.** [14, 21, 22, 41, 45, 48, 49] *Given  $a \in S^m(T^*(\partial D))$ , if  $\text{Op}_{a,h}$  is in the trace class and  $f \in \mathcal{S}(\mathbb{R})$ , then*

$$(2\pi h)^{(d-1)} \text{tr}(f(\text{Op}_{a,h})) = \int_{T^*(\partial D)} f(a) d\sigma \otimes d\sigma^{-1} + \mathcal{O}(h),$$

where  $d\sigma \otimes d\sigma^{-1}$  is the Liouville measure given by the top form  $\omega^{d-1}/(d-1)!$ .

*Proof.* For notational convenience, let us first consider the Weyl quantization  $\text{Op}_{a,h}^w$  instead. We have from the Schwartz kernel theorem that

$$[\text{Op}_{f(a),h}^w(\phi)](x) = \int_{\partial D} K_h(x, y) \phi(y) d\sigma(y)$$

for some  $K_h(x, y) \in \mathcal{D}'(\partial D \times \partial D)$ , which actually has the following explicit expression

$$(2\pi h)^{(d-1)} K_h(x, y) = \mathcal{F}_h \left[ a \left( \frac{x+y}{2}, \cdot \right) \right] (x-y) + \mathcal{O}(h),$$

via the partition of unity and the local trivialisation (by an abuse of notation). Then from the functional calculus we have

$$\text{tr}(f(\text{Op}_{a,h}^w)) = \text{tr}(\text{Op}_{f(a),h}^w) = \int_{\partial D} K_h(x, x) d\sigma.$$

To conclude our theorem, we notice the Weyl quantization  $\text{Op}_{a,h}^w$  and left/right quantizations  $\text{Op}_{a,h}^{L/R}$  differ only in the higher order term after an application of the operator  $\exp(\pm i\frac{h}{2}\partial_x\partial_\xi)$ , and our choice of quantization here is  $\text{Op}_{a,h} := \text{Op}_{a,h}^R$ .  $\square$

Functional calculus and trace theorem combine to give the following generalized Weyl's law, together with the fact that  $(\lambda_i^2(h), \phi_i(h))$  is an eigenpair of  $[\mathcal{K}_{h,\partial D}^*]^2$  if and only if  $(\rho(\lambda_i^2(h)), \phi_i(h))$  is an eigenpair of  $\rho([\mathcal{K}_{h,\partial D}^*]^2)$ .

**Proposition 3.5.** [14, 21, 22, 41, 45, 48, 49] *Under Assumption (A), fixing  $r \leq s$ , for any  $a \in S^m(T^*(\partial D))$ , we have as  $h \rightarrow +0$ ,*

$$(2\pi h)^{(d-1)} \sum_{r \leq \lambda_i^2(h) \leq s} c_i \langle \text{Op}_{a,h} \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} = \int_{\{r \leq H \leq s\}} a d\sigma \otimes d\sigma^{-1} + o_{r,s}(1), \quad (3.8)$$

where  $c_i := |\phi_i|_{H^{-\frac{1}{2}}(\partial\Omega, d\sigma)}^{-2}$  is the  $H^{-\frac{1}{2}}$  semi-norm and the little- $o$  depends on  $r, s$ .

*Proof.* Take  $f_\varepsilon(\rho([\mathcal{K}_{h,\partial D}^*]^2))$  where  $f_\varepsilon \in (S)$  approximating  $\chi_{\{\rho(r), \rho(s)\}}$ . Then  $f_\varepsilon(\rho([\mathcal{K}_{h,\partial D}^*]^2)) \in \Phi\text{SO}_h^{-\infty}$  by the functional calculus with the trace formula

$$(2\pi h)^{(d-1)} \text{tr}(\varepsilon(\rho([\mathcal{K}_{h,\partial D}^*]^2)) \text{Op}_{a,h} \varepsilon(\rho([\mathcal{K}_{h,\partial D}^*]^2))) = \int_{T^*(\partial D)} a f_\varepsilon^2(\rho(H)) d\sigma \otimes d\sigma^{-1} + \mathcal{O}_{r,s,\varepsilon}(h), \quad (3.9)$$

where  $\mathcal{O}$  depends on  $r, s, \varepsilon$ . Passing  $\varepsilon$  to 0 in (3.9),  $f_\varepsilon(\rho([\mathcal{K}_{h,\partial D}^*]^2))$  converges to the spectral projection operator, and  $f_\varepsilon^2(\rho(H))$  converges to  $\chi_{\{\rho(r) \leq \rho(H) \leq \rho(s)\}} = \chi_{\{r \leq H \leq s\}}$ , which readily gives (3.8). At last we notice  $\|\phi_i(h)\|_{L^2(\partial D, d\sigma)} = \|\phi_i\|_{H^{-\frac{1}{2}}(\partial D, d\sigma)}$ .  $\square$

We note that if taking  $a = 1$  in (3.8), it leads us back to the well-known Weyl's law:

**Corollary 3.6.** [14, 21, 22, 41, 45, 48, 49] *Under Assumption (A), we have*

$$\sum_{r \leq \lambda_i^2(h) \leq s} 1 = (2\pi h)^{1-d} \int_{\{r \leq H \leq s\}} d\sigma \otimes d\sigma^{-1} + o_{r,s}(h^{1-d}). \quad (3.10)$$

**3.3. Ergodic decomposition theorem and quantum ergodicity.** Let us denote  $\sigma_H$  as the Riemannian measure on  $\{H = 1\} \subset T^*(\partial D)$ . Since  $X_H H = 0$  and  $\mathcal{L}_{X_H} \omega^{d-1} = 0$ , we have that  $\sigma_H := \lim_{\varepsilon \rightarrow 0} \varepsilon^{d-2} \chi_{\{|H-1| < \varepsilon\}} d\sigma \otimes d\sigma^{-1}$  is an invariant measure on  $\{H = 1\}$ . We also notice that, on  $\{H = 1\}$ ,

$$X_{\tilde{H}} = \rho'(1)X_{\tilde{H}} = e^{-1}X_{\tilde{H}}.$$

Let  $M_{X_H}(\{H = 1\})$  be the set of  $X_H$  invariant measures on  $\{H = 1\}$  and also  $M_{X_H, \text{erg}}(\{H = 1\})$  be the set of ergodic measures with respect to the Hamiltonian flow generated by  $X_H$  on  $\{H = 1\}$ . We realize that since  $X_{\tilde{H}} = e^{-1}X_{\tilde{H}}$ , we have  $M_{X_{\tilde{H}}}(\{H = 1\}) = M_{X_H}(\{H = 1\})$  and  $M_{X_{\tilde{H}}, \text{erg}}(\{H = 1\}) = M_{X_H, \text{erg}}(\{H = 1\})$ . Therefore, we do not distinguish between them. Now, since  $\{H = 1\}$  has a countable base, the weak-\* topology of  $M_{X_H}(\{H = 1\})$  is metrizable, and hence Choquet's theorem can be applied to obtain the following classical ergodic decomposition theorem.

**Proposition 3.7.** [47] *Given a probability measure  $\eta \in M_{X_H}(\{H = 1\})$ , there exists a probability measure  $\nu \in M(M_{X_H, \text{erg}}(\{H = 1\}))$  such that*

$$\eta = \int_{M_{X_H, \text{erg}}(\{H=1\})} \mu d\nu(\mu).$$

Applying Proposition 3.7 to  $\sigma_H/\sigma_H(\{H = 1\})$ , we have a probability measure  $\nu \in M(M_{X_H, \text{erg}}(\{H = 1\}))$  such that

$$\sigma_H = \sigma_H(\{H = 1\}) \int_{M_{X_H, \text{erg}}(\{H=1\})} \mu d\nu(\mu).$$

Note by rescaling  $\{H = E\} = E^{-1/2}\{H = 1\}$ , and therefore  $d\sigma \otimes d\sigma^{-1} = E^{1-\frac{d}{2}}dE \otimes d\sigma_H$ . For any  $\mu \in M_{X_H, \text{erg}}(\{H = 1\})$ , let  $\mu_E := m_{E^{-1/2}}\#\mu \in M_{X_H, \text{erg}}(\{H = E\})$  be the push-forward measure given by  $m_{E^{-1/2}} : T^*(\partial D) \rightarrow T^*(\partial D) (x, \xi) \mapsto (x, E^{-1/2}\xi)$ , then

$$\sigma \otimes \sigma^{-1} = \sigma_H(\{H = 1\}) \int_{(0, \infty] \times M_{X_H, \text{erg}}(\{H=1\})} \mu_E E^{1-\frac{d}{2}} (dE \otimes d\nu) (E, \mu).$$

Next, we aim to derive a more general version of quantum ergodicity, following the original argument in, e.g. [14, 18, 41, 42, 44, 45, 48–53] as follows. To start with, we have the following application from Birkhoff [11] and Von-Neumann's ergodic theorems [36].

**Lemma 3.8.** *Under Assumption (A), for any  $r \leq s$  and all  $a_0 \in \mathcal{S}^m(T^*(\partial D))$ , we have*

$$\frac{1}{T} \int_0^T a_{(x, \xi)}(t) dt \xrightarrow{\text{a.e. } d\sigma \otimes d\sigma^{-1} \text{ and } L^2(\{r \leq H \leq s\}, d\sigma \otimes d\sigma^{-1})} \bar{a}(x, \xi) \text{ as } T \rightarrow \infty,$$

for some  $\bar{a} \in L^2(\{r \leq H \leq s\}, d\sigma \otimes d\sigma^{-1})$ , and a.e.  $E^{1-\frac{d}{2}}dE \otimes d\nu$ , we have

$$\bar{a}(x, \xi) = \int_{\{H=E\}} a_0 d\mu_E \quad \text{a.e. } d\mu_E.$$

*Proof.* By Birkhoff and Von-Neumann's ergodic theorems [11, 36] on  $\chi_{\{r \leq H \leq s\}} d\sigma \otimes d\sigma^{-1}$ , we have

$$\frac{1}{T} \int_0^T a_{(x, \xi)}(t) dt \xrightarrow{\text{a.e. } d\sigma \otimes d\sigma^{-1} \text{ and } L^2(\{r \leq H \leq s\}, d\sigma \otimes d\sigma^{-1})} \bar{a}(x, \xi) \text{ as } T \rightarrow \infty,$$

for some  $\bar{a} \in L^2(\{r \leq H \leq s\}, d\sigma \otimes d\sigma^{-1})$  invariant under the Hamiltonian flow. Let

$$\mathcal{E} := \left\{ (x, \xi) \in \{r \leq H \leq s\} : \limsup_T \left| \frac{1}{T} \int_0^T a_{(x, \xi)}(t) dt - \bar{a}(x, \xi) \right| > 0 \right\},$$

then  $\sigma \otimes \sigma^{-1}(\mathcal{E}) = 0$ . Now by Lemma 3.7, we have

$$\sigma_H(\{H = 1\}) \int_{[r, s] \times M_{X_H, \text{erg}}(\{H=1\})} \mu_E(\mathcal{E}) E^{1-\frac{d}{2}} dE \otimes d\nu(E, \mu) = \sigma \otimes \sigma^{-1}(\mathcal{E}) = 0,$$

and therefore, a.e.  $E^{1-\frac{d}{2}}dE \otimes d\nu$ , we have  $\mu_E(\mathcal{E}) = 0$ . Meanwhile, by the Birkhoff theorem [11],

$$\frac{1}{T} \int_0^T a_{(x, \xi)}(t) dt \xrightarrow{\text{a.e. } \mu_E \text{ and } L^2(\{H=E\}, d\mu_E)} \int_{\{H=E\}} a_0 d\mu_E \quad \text{as } T \rightarrow \infty.$$

Again let

$$\mathcal{E}_{\mu_E} := \left\{ (x, \xi) \in \{r \leq H \leq s\} : \limsup_T \left| \frac{1}{T} \int_0^T a_{(x, \xi)}(t) dt - \int_{\{H=E\}} a_0 d\mu_E \right| > 0 \right\},$$

we have  $\mu_E(\mathcal{E}_{\mu_E}) = 0$ . Therefore, a.e.  $E^{1-\frac{d}{2}}dE \otimes d\nu$ ,  $\mu_E(\mathcal{E} \cup \mathcal{E}_{\mu_E}) = 0$ . The lemma follows by the uniqueness of the limit.  $\square$

We can then show the following theorem by following the arguments in [14, 16, 18, 41, 42, 44, 45, 48–53] with some generalizations.

**Theorem 3.9.** *Under Assumption (A), fixing  $r \leq s$  and writing  $c_i := |\phi_i|_{H^{-\frac{1}{2}}(\partial D, d\sigma)}^{-2}$  to denote the  $H^{-\frac{1}{2}}$  semi-norm, we have the following (variance-like) estimate as  $h \rightarrow +0$ ,*

$$\frac{1}{\sum_{r \leq \lambda_i^2(h) \leq s} 1} \sum_{r \leq \lambda_i^2(h) \leq s} c_i^2 \left| \langle A_h \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} - \langle \text{Op}_{\bar{a}, h} \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} \right|^2 \rightarrow 0. \quad (3.11)$$

*Proof.* We lift the Birkhoff and Von-Neumann to the operator level, via the Hamiltonian flow of the principle symbol. Consider  $A_h(0) = A_h$ . From the definition of  $\phi_i(h)$ , we have for each  $i$

$$\begin{aligned} \langle A_h(t) \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} &= \langle A_h(0) e^{-\frac{it}{h} \rho([\mathcal{K}_{h, \partial D}^*]^2)} \phi_i(h), e^{-\frac{it}{h} \rho([\mathcal{K}_{h, \partial D}^*]^2)} \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} + \mathcal{O}_t(h) \\ &= \langle A_h \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} + \mathcal{O}_t(h), \end{aligned} \quad (3.12)$$

where the second equality comes from Corollary 3.2 and the definition of the NP eigenfunctions (cf. (2.6)), and the asymptotics  $\mathcal{O}$  depends on  $t$ . Averaging both sides of (3.12) with respect to  $T$ , we have

$$\left\langle \left( \frac{1}{T} \int_0^T A_h(t) dt \right) \phi_i(h), \phi_i(h) \right\rangle_{L^2(\partial D, d\sigma)} = \langle A_h \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} + \mathcal{O}_T(h),$$

where the asymptotics  $\mathcal{O}$  depends on  $T$ . Then, again by Corollary 3.2, it is handy to verify that

$$\frac{1}{T} \int_0^T A_h(t) dt - \text{Op}_{\bar{a}, h} = \text{Op}_{\frac{1}{T} \int_0^T a_{(x, \xi)}(t) dt - \bar{a}} + \mathcal{O}_T(h)$$

is a pseudo-differential operator. Next from the Cauchy-Schwarz inequality, we have

$$\begin{aligned} & \left| \frac{\langle \text{Op}_{\bar{a}, h} \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)}}{\langle \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)}} - \frac{\langle A_h \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)}}{\langle \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)}} \right|^2 \\ & \leq \frac{\left\langle \left( \frac{1}{T} \int_0^T A_h(t) dt - \text{Op}_{\bar{a}, h} \right)^* \left( \frac{1}{T} \int_0^T A_h(t) dt - \text{Op}_{\bar{a}, h} \right) \phi_i(h), \phi_i(h) \right\rangle_{L^2(\partial D, d\sigma)}}{\langle \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)}} + \mathcal{O}_T(h^2). \end{aligned} \quad (3.13)$$

Therefore, summing up  $i$  of (3.13) and applying (3.8) and (3.10), we have

$$\begin{aligned} & \frac{1}{\sum_{r \leq \lambda_i^2(h) \leq s} 1} \sum_{r \leq \lambda_i^2(h) \leq s} c_i^2 \left| \langle A_h \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} - \langle \text{Op}_{\bar{a}, h} \phi_i(h), \phi_i(h) \rangle_{L^2(\partial D, d\sigma)} \right|^2 \\ & \leq \frac{\int_{\{r \leq H \leq s\}} \left| \frac{1}{T} \int_0^T a_{(x, \xi)}(t) dt - \bar{a} \right|^2 d\sigma \otimes d\sigma^{-1}}{\int_{\{r \leq H \leq s\}} d\sigma \otimes d\sigma^{-1}} + o_{r, s, T}(1). \end{aligned} \quad (3.14)$$

Finally, (3.11) readily follows by noting that the first term at the right-hand side of (3.14) goes to zero as  $T$  goes to infinity.

The proof is complete.  $\square$

With Theorem 3.9, together with Chebyshev's trick and a diagonal argument, we have the following quantum ergodicity result [14, 18, 41, 42, 44, 45, 48–53] with some generalization.

**Corollary 3.10.** *Under Assumption (A), given  $r, s$ , there exists  $S(h) \subset J(h) := \{i \in \mathbb{N} : r \leq \lambda_i^2(h) \leq s\}$  such that for all  $a_0 \in S^m(T^*(\partial D))$ , we have as  $h \rightarrow +0$ ,*

$$\max_{i \in S(h)} c_i \left| \left\langle (A_h - \text{Op}_{\tilde{a},h}) \phi_i(h), \phi_i(h) \right\rangle_{L^2(\partial D, d\sigma)} \right| = o_{r,s}(1) \quad \text{and} \quad \frac{\sum_{i \in S(h)} 1}{\sum_{i \in J(h)} 1} = 1 + o_{r,s}(1) \quad (3.15)$$

A very important remark of the above corollary is that the set  $S(h)$  is chosen independent of the choice of  $a_0$ .

#### 4. LOCALIZATION/CONCENTRATION OF PLASMON RESONANCES IN ELECTROSTATICS

In this section, we are ready to present one of our main results on the localization/concentration of plasmon resonances in electrostatics.

**4.1. Consequences of generalized Weyl's law and quantum ergodicity.** We first derive the following theorem to characterise the local behaviour of the NP eigenfunctions and their relative magnitude. In what follows, we let  $(\lambda_i, \phi_i)$ ,  $i = 1, 2, \dots$ , be the ordered eigenpairs to (1.7). We denote  $\sigma_{x,H}$  as the Riemannian measure on  $\{H(x, \cdot) = 1\} \subset T_x^*(\partial D)$ . By the generalized Weyl's law in Section 3, we can first show the following key result in our study.

**Theorem 4.1.** *Given any  $x \in \partial D$ , we consider  $\{\chi_{x,\delta}\}_{\delta>0}$  being a family of smooth nonnegative bump functions compactly supported in  $B_\delta(x)$  with  $\int_{\partial D} \chi_{p,\delta} d\sigma = 1$ . Under Assumption (A), fixing  $r \leq s$ ,  $\alpha \in \mathbb{R}$  and  $p, q \in \partial D$ , there exists a choice of  $\delta(h)$  depending on  $r, s, p, q$  and  $\alpha$  such that, as  $h \rightarrow +0$ , we have  $\delta(h) \rightarrow 0$  and*

$$\frac{\sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{p,\delta(h)}(x) |D|^\alpha |\phi_i(x)|^2 d\sigma(x)}{\sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{q,\delta(h)}(x) |D|^\alpha |\phi_i(x)|^2 d\sigma(x)} = \frac{\int_{\{H(p,\cdot)=1\}} |\xi|_{g(p)}^{1+2\alpha} d\sigma_{p,H}}{\int_{\{H(q,\cdot)=1\}} |\xi|_{g(q)}^{1+2\alpha} d\sigma_{q,H}} + o_{r,s,p,q,\alpha}(1), \quad (4.1)$$

where  $c_i := |\phi_i|_{H^{-\frac{1}{2}}(\partial D, d\sigma)}^{-2}$  is the  $H^{-\frac{1}{2}}$  semi-norm and the little- $o$  depends on  $r, s, p, q$  and  $\alpha$ . In particular, if  $\alpha = -\frac{1}{2}$ , the right-hand side of (4.1) is the ratio between the volumes of the two varieties at the respective points.

*Proof.* Taking  $p \in \partial D$ , we consider  $a(x, \xi) := \chi_{p,\delta}(x) |\xi|_{g(x)}^{1+2\alpha}$  in (3.8). With this, together with the fact that  $\text{Op}_{a,h} = h^{1+2\alpha} |D|^{1/2+\alpha} \text{Op}_{\chi_{p,\delta}(x),h} |D|^{1/2+\alpha} - h \text{Op}_{\tilde{a}_{p,\delta},h}$  for some  $\tilde{a}_{p,\delta} \in S^{2\alpha}(T^*(\partial D))$ , we have

$$\begin{aligned} & (2\pi h)^{(d+2\alpha)} \sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{p,\delta}(x) |D|^\alpha |\phi_i(x)|^2 d\sigma(x) \\ &= \int_{\{r \leq H \leq s\}} \chi_{p,\delta}(x) |\xi|_{g(x)}^{1+2\alpha} d\sigma \otimes d\sigma^{-1} + h \int_{\{r \leq H \leq s\}} \tilde{a}_{p,\delta} d\sigma \otimes d\sigma^{-1} + o_{r,s,\alpha}(1) \end{aligned} \quad (4.2)$$

after applying (3.8) once more upon  $\tilde{a}_{p,\delta}$ . With (4.2), we have, after choosing another point  $q \in \partial D$  and taking a quotient between the two, that

$$\begin{aligned} & \frac{\sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{p,\delta}(x) |D|^\alpha |\phi_i(x)|^2 d\sigma(x)}{\sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{q,\delta}(x) |D|^\alpha |\phi_i(x)|^2 d\sigma(x)} \\ &= \frac{\int_{\{r \leq H \leq s\}} \chi_{p,\delta}(x) |\xi|_{g(x)}^{1+2\alpha} d\sigma \otimes d\sigma^{-1} + h \int_{\{r \leq H \leq s\}} \tilde{a}_{p,\delta} d\sigma \otimes d\sigma^{-1}}{\int_{\{r \leq H \leq s\}} \chi_{q,\delta}(x) |\xi|_{g(x)}^{1+2\alpha} d\sigma \otimes d\sigma^{-1} + h \int_{\{r \leq H \leq s\}} \tilde{a}_{q,\delta} d\sigma \otimes d\sigma^{-1}} + o_{r,s,\alpha}(1). \end{aligned}$$

Now, for any given  $h$ , we can make a choice of  $\delta(h)$  depending on  $r, s, p, q, \alpha$  such that as  $h \rightarrow +0$ , we have  $\delta(h) \rightarrow 0$  (much slower than  $h$ ) and

$$\left| h \int_{\{r \leq H \leq s\}} \tilde{a}_{p,\delta(h)} d\sigma \otimes d\sigma^{-1} \right| + \left| h \int_{\{r \leq H \leq s\}} \tilde{a}_{q,\delta(h)} d\sigma \otimes d\sigma^{-1} \right| \rightarrow 0.$$

We also realize as  $h \rightarrow +0$ , with this choice of  $\delta(h)$  that  $\delta(h) \rightarrow 0$ , one in fact has for  $y = p, q$  that

$$\int_{\{r \leq H \leq s\}} \chi_{y,\delta(h)}(x) |\xi|_{g(x)}^{1+2\alpha} d\sigma \otimes d\sigma^{-1} \rightarrow \int_{\{r \leq H(y,\cdot) \leq s\}} |\xi|_{g(y)}^{1+2\alpha} d\sigma^{-1}.$$

Therefore, we have

$$\frac{\sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{p,\delta(h)}(x) ||D|^\alpha \phi_i(x)|^2 d\sigma(x)}{\sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{q,\delta(h)}(x) ||D|^\alpha \phi_i(x)|^2 d\sigma(x)} = \frac{\int_{\{r \leq H(p,\cdot) \leq s\}} |\xi|_{g(p)}^{1+2\alpha} d\sigma^{-1}}{\int_{\{r \leq H(q,\cdot) \leq s\}} |\xi|_{g(q)}^{1+2\alpha} d\sigma^{-1}} + o_{r,s,p,q,\alpha}(1).$$

Now to conclude our theorem, we realize that for all  $y = p, q$ ,

$$\int_{\{r \leq H(y,\cdot) \leq s\}} |\xi|_{g(y)}^{1+2\alpha} d\sigma^{-1} = \left( \int_r^s E^{-\frac{1}{2}-\alpha-\frac{d}{2}} dE \right) \left( \int_{\{H(y,\cdot)=1\}} |\xi|_{g(y)}^{1+2\alpha} d\sigma_{y,H} \right).$$

The proof is complete.  $\square$

Theorem 4.1 states that, given  $p, q \in \partial D$ , the relative magnitude between a  $c_i$ -weighted sum of a weighed average of  $||D|^\alpha \phi_i|^2$  over a small neighborhood of  $p$  to that of  $q$  asymptotically depends on the ratio between the weighted volume of  $\{H(p,\cdot) = 1\}$  and that of  $\{H(q,\cdot) = 1\}$ . This is critical for our subsequent analysis since it reduces our study to analyzing the aforementioned weighted volumes.

**Theorem 4.2.** *Under Assumption (A), there is a family of distributions  $\{\Phi_\mu\}_{M_{X_H, \text{erg}}} \in \mathcal{D}'(\partial D \times \partial D)$  as the Schwartz kernels of  $\mathcal{K}_\mu$  such that they form a partition of the identity operator  $Id$  as follows:*

$$Id = \int_{M_{X_H, \text{erg}}(\{H=1\})} \mathcal{K}_\mu d\nu(\mu), \quad (4.3)$$

which holds in the weak operator topology satisfying that for any given  $r, s$ , there exists  $S(h) \subset J(h) := \{i \in \mathbb{N} : rh \leq \lambda_i^2 \leq sh\}$  such that for all  $\varphi \in \mathcal{C}^\infty(\partial D)$  and as  $h \rightarrow +0$ ,

$$\max_{i \in S(h)} \left| \int_{\partial D} \varphi(x) \left( c_i ||D|^{-\frac{1}{2}} \phi_i(x)|^2 - \int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(x) g_i(\mu) d\nu(\mu) \right) d\sigma(x) \right| = o_{r,s}(1). \quad (4.4)$$

In (4.3),

$$g_i(\mu) := c_i \left\langle \mathcal{K}_\mu |D|^{-\frac{1}{2}} \phi_i, |D|^{-\frac{1}{2}} \phi_i \right\rangle_{L^2(\partial D, d\sigma)}, \quad (4.5)$$

$$\int_{M_{X_H, \text{erg}}(\{H=1\})} g_i(\mu) d\nu(\mu) = 1, \quad \frac{\sum_{i \in S(h)} 1}{\sum_{i \in J(h)} 1} = 1 + o_{r,s}(1),$$

and moreover,

$$\mu(p) \geq 0, \quad \int_{\partial D} \mu(p) d\mu(p) = 1, \quad (4.6)$$

$$\frac{\int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(p) d\nu(\mu)}{\int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(q) d\nu(\mu)} = \frac{\int_{\{H(p,\cdot)=1\}} d\sigma_{p,H}}{\int_{\{H(q,\cdot)=1\}} d\sigma_{q,H}} \text{ a.e. } (d\sigma \otimes d\sigma)(p, q).$$



*Proof.* Let  $f, \varphi \in C^\infty(\partial D)$  be given. Let us consider  $a(x, \xi) := \varphi(x)$ . Then we have

$$\int_{\{H=E\}} \varphi d\mu_E = \int_{\{H=1\}} \varphi d\mu.$$

Take a partition of unity  $\{\chi_i\}$  on  $\{U_i\}$ . With an abuse of notation via identification of points with the local trivialisation  $\{F_i\}$ , we have by Lemmas 3.7 and 3.8 that

$$\begin{aligned} & [\sigma_H(\{H=1\})]^{-1} [\text{Op}_{\bar{\varphi}, h} f](y) \\ &= \sum_l [\sigma_H(\{H=1\})]^{-1} [\text{Op}_{\bar{\varphi}\chi_l, h} f](y) \\ &= \int_{(0, \infty) \times M_{X_H, \text{erg}}(\{H=1\})} \sum_l \left( \int_{\{H=E\}} \exp(x-y, \xi/h) \bar{a}(x, \xi) \chi_l(x) f(x) d\mu_E \right) \\ & \quad \times E^{1-\frac{d}{2}} (dE \otimes d\nu)(E, \mu) \\ &= \int_{(0, \infty) \times M_{X_H, \text{erg}}(\{H=1\})} \sum_l \left( \int_{\{H=1\}} \varphi d\mu \right) \left( \int_{\{H=E\}} \exp(x-y, \xi/h) \chi_l(x) f(x) d\mu_E \right) \\ & \quad \times E^{1-\frac{d}{2}} (dE \otimes d\nu)(E, \mu). \end{aligned} \tag{4.7}$$

On the other hand, considering  $Id = \text{Op}_{1, h} = \text{Op}_{\bar{1}, h}$  (which is independent of  $h$ ), we can show that

$$\begin{aligned} & [\sigma_H(\{H=1\})]^{-1} [\text{Op}_{1, h} f](y) \\ &= \int_{(0, \infty) \times M_{X_H, \text{erg}}(\{H=1\})} \sum_l \left( \int_{\{H=E\}} \exp(x-y, \xi/h) \chi_l(x) f(x) d\mu_E \right) E^{1-\frac{d}{2}} (dE \otimes d\nu)(E, \mu). \end{aligned}$$

If we define  $\mathcal{K}_\mu$  (which is again independent of  $h$ ) to be such that

$$[\sigma_H(\{H=1\})]^{-1} [\mathcal{K}_\mu f](y) = \int_{(0, \infty)} \sum_l \left( \int_{\{H=E\}} \exp(x-y, \xi/h) \chi_l(x) f(x) d\mu_E \right) E^{1-\frac{d}{2}} dE(E),$$

then we have by definition that

$$Id = \int_{M_{X_H, \text{erg}}(\{H=1\})} \mathcal{K}_\mu d\nu(\mu)$$

in the weak operator topology. That is,

$$\langle f, f \rangle_{L^2(\partial D, d\sigma)} = \int_{M_{X_H, \text{erg}}(\{H=1\})} \langle \mathcal{K}_\mu f, f \rangle_{L^2(\partial D, d\sigma)} d\nu(\mu),$$

and

$$\langle \text{Op}_{\bar{\varphi}, h} f, f \rangle_{L^2(\partial D, d\sigma)} = \int_{M_{X_H, \text{erg}}(\{H=1\})} \int_{\{H=1\}} \varphi \langle \mathcal{K}_\mu f, f \rangle_{L^2(\partial D, d\sigma)} d\mu d\nu(\mu).$$

Recall that  $d\sigma_H(x, \xi)/\sigma_H(\{H=1\}) = d\mu(x, \xi) d\nu(\mu)$  is a probability measure. We now apply the disintegration theorem to the measure  $d\mu(x, \xi) d\nu(\mu)$  and obtain a disintegration  $d\mu_p(x, \xi) d\nu(\mu) \otimes d\sigma(p)$ , where the measure-valued map  $(\mu, p) \mapsto \mu_p$  is a  $d\nu \otimes d\sigma$  measurable function together with  $\mu_p(\{H=1\} \setminus (\{H(p, \cdot) = 1\} \cap \text{spt}(\mu))) = 0$  a.e.  $d\nu \otimes d\sigma$ . Therefore, we obtain

$$\langle \text{Op}_{\bar{\varphi}, h} f, f \rangle_{L^2(\partial D, d\sigma)} = \int_{\partial D} \int_{M_{X_H, \text{erg}}(\{H=1\})} \int_{\{H=1\}} \varphi \langle \mathcal{K}_\mu f, f \rangle_{L^2(\partial D, d\sigma)} d\mu_p (d\nu \otimes d\sigma)(\mu, p).$$

We next observe that

$$\int_{\{H=1\}} \varphi d\mu_p = \int_{\{H(p,\cdot)=1\}} \varphi d\mu_p = \varphi(p) \mu_p(\{H=1\}).$$

If we denote

$$\mu(p) := \mu_p(\{H=1\}) \geq 0,$$

then a.e.  $d\nu(\mu)$ , the function  $\mu(p) \in L^1(\partial\Omega, d\sigma)$ . As a result of the disintegration, we have a.e.  $d\nu(\mu)$ ,

$$\int_{\partial\Omega} \mu(p) d\sigma(p) = \mu(\{H=1\}) = 1.$$

Furthermore, we have

$$\langle \text{Op}_{\bar{\varphi},h} f, f \rangle_{L^2(\partial D, d\sigma)} = \int_{\partial D} \int_{M_{X_H, \text{erg}}(\{H=1\})} \varphi(x) \mu(x) \langle \mathcal{K}_\mu f, f \rangle_{L^2(\partial D, d\sigma)} (d\nu \otimes d\sigma)(\mu, x).$$

Now, we choose  $f = \phi_i(h) = |D|^{-\frac{1}{2}} \phi_i$  and apply (3.15) to obtain the conclusion of our theorem. It is noted that the choice of  $S(h)$  is independent of  $\varphi \in C^\infty(\partial D)$ . The ratio in the last line of the theorem comes from the fact that a.e.  $d\sigma(p)$  we have by definition

$$\int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(p) d\nu(\mu) = \int_{M_{X_H, \text{erg}}(\{H=1\})} \mu_p(\{H(p,\cdot)=1\}) d\nu(\mu) = \frac{\int_{\{H(p,\cdot)=1\}} d\sigma_{p,H}}{\sigma_H(\{H=1\})}.$$

The proof is complete.  $\square$

Theorem 4.2 indicates that most of the function  $c_i |D|^{-\frac{1}{2}} \phi_i^2$  weakly converges to a  $g_i(\mu) d\nu(\mu)$ -weighted average of  $\mu(p)$ , where the ratio between a  $d\nu(\mu)$ -weighted average of  $\mu(p)$  and that of  $\mu(q)$  depends on the ratio between the volume of  $\{H(p,\cdot)=1\}$  and that of  $\{H(q,\cdot)=1\}$ .

For the sake of completeness, we also give the original version of the quantum ergodicity:

**Corollary 4.3.** *Under Assumption (A), if the Hamiltonian flow given by  $X_H$  is furthermore uniquely ergodic on  $\{H=1\}$ , then given  $r, s$ , there exists  $S(h) \subset J(h) := \{i \in \mathbb{N} : rh \leq \lambda_i^2 \leq sh\}$ , such that for all  $\varphi \in C^\infty(\partial D)$  and as  $h \rightarrow +0$ ,*

$$\begin{aligned} \max_{i \in S(h)} \left| \int_{\partial D} \varphi(x) \left( c_i |D|^{-\frac{1}{2}} \phi_i(x)^2 - \frac{\int_{\{H(x,\cdot)=1\}} d\sigma_{x,H}}{\sigma_H(\{H=1\})} \right) d\sigma(x) \right| &= o_{r,s}(1), \\ \frac{\sum_{i \in S(h)} 1}{\sum_{i \in J(h)} 1} &= 1 + o_{r,s}(1). \end{aligned} \tag{4.8}$$

*Proof.* The conclusion follows by noting that the unique ergodicity of  $X_H$  implies that it is ergodic with respect to  $\sigma_H$ , and in such a case

$$\sigma_H(x) = \int_{\{H(x,\cdot)=1\}} d\sigma_{x,H} / \int_{\{H=1\}} d\sigma_H.$$

$\square$

By Corollary 4.3, we see that if  $X_H$  is uniquely ergodic, most of the function  $c_i |D|^{-\frac{1}{2}} \phi_i^2$  weakly converges to the volume of the characteristic variety  $\{H(x,\cdot)=1\}$  up to a constant. We remark that we expect the above argument can be extended to the comparison between  $c_i |D|^\alpha \phi_i^2$ , and we choose to investigate along that direction in our future study.

#### 4.2. Localization/concentration of plasmon resonance at high-curvature places.

From Theorems 4.1 and 4.2 in the previous subsection, it is clear that the relative magnitude of the NP eigenfunction  $\phi_i$  at a point  $x$  depends on the (weighted) volume of the characteristic variety  $\{H(x, \cdot) = 1\}$ . Therefore, in order to understand the localization of plasmon resonance, it is essential to obtain a better description of this volume. It turns out that this volume heavily depends on the magnitude of the second fundamental forms  $A(x)$  at the point  $x$ . As we will see in this subsection, in general, the higher the magnitude of the second fundamental forms  $A(x)$  is, the larger the volume of the characteristic variety becomes. In particular, in a relatively simple case when the second fundamental forms at two points are constant multiple of each other, we have the following volume comparison.

**Lemma 4.4.** *Let  $p, q \in \partial D$  be such that  $A(p) = \beta A(q)$  for some  $\beta > 0$  and  $g(p) = g(q)$ . Then  $|\{H(p, \cdot) = 1\}| = \beta^{d-2} |\{H(q, \cdot) = 1\}|$ . We also have*

$$\int_{\{H(p, \cdot) = 1\}} |\xi|_{g(p)}^{1+2\alpha} d\sigma_{p,H} = \beta^{d-1+2\alpha} \int_{\{H(q, \cdot) = 1\}} |\xi|_{g(q)}^{1+2\alpha} d\sigma_{q,H}.$$

*Proof.* From  $-2$  homogeneity of  $H$ , we have  $H(p, \xi) = H(q, \xi/\beta)$ , and therefore  $\{H(p, \xi) = 1\} = \beta \{H(q, \xi) = 1\}$ , which readily yields the conclusion of the theorem.  $\square$

A better understanding of the localization can be achieved by a more delicate volume comparison of the characteristic variety at different points with the help of Theorems 4.1 and 4.2 and Corollary 4.3. However, it is less easy to give a more explicit comparison of the volumes between  $\{H(p, \cdot) = 1\}$  and  $\{H(q, \cdot) = 1\}$  by their respective second fundamental forms  $A(p)$  and  $A(q)$ . The following lemma provides a detour to control how the (weighted) volume of  $\{H(p, \cdot) = 1\}$  depends on the principal curvatures  $\{\kappa_i(p)\}_{i=1}^{d-2}$ .

**Lemma 4.5.** *Let  $F : \mathbb{R}^{d-2} \rightarrow \mathbb{R}$  be given as*

$$F_\alpha(\{\kappa_i\}_{i=1}^{d-2}) := \int_{\mathbb{S}^{d-2}} \left| \sum_{i=1}^{d-1} \tilde{\kappa}_i \omega_i^2 \right|^{d-1+2\alpha} \sqrt{\sum_{i=1}^{d-1} \tilde{\kappa}_i^2 \omega_i^2} d\omega, \quad (4.9)$$

where

$$\tilde{\kappa}_i := \sum_{j=1}^{d-1} \kappa_j - \kappa_i. \quad (4.10)$$

Then we have the following inequality:

$$F_\alpha(\{\kappa_i(p)\}_{i=1}^{d-2}) \leq \int_{\{H(p, \cdot) = 1\}} |\xi|_{g(p)}^{1+2\alpha} d\sigma_{p,H} \leq 2F_\alpha(\{\kappa_i(p)\}_{i=1}^{d-2}). \quad (4.11)$$

*Proof.* We first simplify the expression of  $H(p, \xi) = 0$  by fixing a point  $p$  and choosing a geodesic normal coordinate with the principal curvatures along the directions  $\xi_i$ . In this case

$$H(p, \xi) = \left( \sum_{i=1}^{d-1} \widetilde{\kappa}_i(p) \xi_i^2 \right)^2 / \left( \sum_{i=1}^{d-1} \xi_i^2 \right)^3.$$

Let us parametrize the surface  $\{H(p, \cdot) = 1\}$  by  $\omega \in \mathbb{S}^{d-2}$  with  $\xi(\omega) := r(\omega)\omega$ , which is legitimate due to the  $-2$  homogeneity of  $H$  with respect to  $\xi$ . With this, we readily see that on  $H = 1$  one has

$$r(\omega) = \sum_{i=1}^{d-1} \widetilde{\kappa}_i(p) \omega_i^2.$$

Hence by virtue of the Sherman-Morrison formula one has

$$\begin{aligned} L_{ij} &:= \frac{\partial \xi}{\partial \omega_{ij}} = r(\omega) \delta_{ij} + 2 \widetilde{\kappa_j(p)} \omega_j \omega_i, \\ (L^{-T})_{ij} &= \frac{1}{r(\omega)} \delta_{ij} - \frac{2}{3r(\omega)} \widetilde{\kappa_i(p)} \omega_i \omega_j, \quad \det(L) = 3(r(\omega))^d, \end{aligned} \quad (4.12)$$

with which, via a change of variable formula, one can further derive that

$$|\xi|_{g(p)}^{1+2\alpha} d\sigma_{p,H} = |r(\omega)|^{d-1+2\alpha} \sqrt{4 \left( \sum_{i=1}^{d-1} \widetilde{\kappa_i(p)}^2 \omega_i^2 \right)^2 - 3 \left( \sum_{i=1}^{d-1} \widetilde{\kappa_i(p)} \omega_i^2 \right) \left( \sum_{i=1}^{d-1} \omega_i^2 \right)} d\omega.$$

Finally by the Cauchy-Schwarz inequality, we therefore have

$$\left| \sum_{i=1}^{d-1} \widetilde{\kappa_i(p)} \omega_i^2 \right|^{d-1+2\alpha} \sqrt{\sum_{i=1}^{d-1} \widetilde{\kappa_i(p)}^2 \omega_i^2} d\omega \leq |\xi|_{g(p)}^{1+2\alpha} d\sigma_{p,H} \leq 2 \left| \sum_{i=1}^{d-1} \widetilde{\kappa_i(p)} \omega_i^2 \right|^{d-1+2\alpha} \sqrt{\sum_{i=1}^{d-1} \widetilde{\kappa_i(p)}^2 \omega_i^2} d\omega,$$

which readily completes the proof.  $\square$

Lemma 4.5 supplies us with a strong tool to obtain the comparison between the ratio of the magnitude of the eigenfunctions via the magnitudes of the principal curvatures at the respective points. For instance, if it happens that  $\min_i |\widetilde{\kappa_i(p)}| \gg \max_i |\widetilde{\kappa_i(q)}|$ , then it is clear that the weighted volume of  $\{H(p, \cdot) = 1\}$  is much bigger than that at  $q$ .

**Remark 4.6.** *As we will explore in Appendix A, when  $d = 3$ ,  $\overline{\{H = 1\}} \cap (\partial D \times \{0\}) = \emptyset$  if and only if  $A(p) > c_0 I$  for all  $x \in \partial D$ . In this strictly convex case with  $d = 3$ , we therefore have*

$$\min_{i=1,2} \kappa_i^{3+2\alpha}(p) \leq F_\alpha(\{\kappa_i(p)\}_{i=1}^{1-2}) \leq \max_{i=1,2} \kappa_i^{3+2\alpha}(p), \quad (4.13)$$

and hence

$$\min_{i=1,2} \kappa_i^{3+2\alpha}(p) \leq \int_{\{H(p,\cdot)=1\}} |\xi|_{g(p)}^{1+2\alpha} d\sigma_{p,H} \leq 2 \max_{i=1,2} \kappa_i^{3+2\alpha}(p). \quad (4.14)$$

*This fully captures the desired behaviour that the NP eigenfunctions localize at point at high curvature when  $d = 3$  and when the flow on  $\{H = 1\}$  is non-singular. Further remarks and brief discussions upon certain geometric properties of the Hamiltonian flow is postponed to Appendix A.*

Finally, we discuss the implication of the localization/concentration result of the NP eigenfunctions to the surface plasmon resonances. According to our discussion in Section 1.2, an SPR field  $u$  is the superposition of the plasmon resonant modes of the form

$$u = \sum_i \alpha_i \mathcal{S}_{\partial D}[\phi_i], \quad (4.15)$$

where  $\alpha_i \in \mathbb{C}$  represents a Fourier coefficient and each  $\phi_i$  is an NP eigenfunction, namely  $\mathcal{K}_{\partial D}[\phi_i] = \lambda_i \phi_i$  with  $\lambda_i \in \mathbb{R}$  being an NP eigenvalue. As is widely known in the literature, a main feature of the SPR field is that it exhibits a highly oscillatory behaviour (due to the resonance) and the resonant oscillation is mainly confined in a vicinity of the boundary  $\partial D$ . For a boundary point  $p \in \partial D$ , one handily computes from (1.9) that

$$\frac{\partial}{\partial \nu} (\mathcal{S}_{\partial D}[\phi_i])^\pm(p) = (\pm \frac{1}{2} Id + \mathcal{K}_{\partial D}^*)[\phi_i](p) = (\pm \frac{1}{2} + \lambda_i) \phi_i(p). \quad (4.16)$$

Generically, (4.16) indicates that if  $|\phi_i(p)|$  is large, then  $|\nabla \mathcal{S}_{\partial D}[\phi_i]|$  is also large in a neighbourhood of  $p$ . Hence, by the localization/concentration of the NP eigenfunction  $\phi_i$  established above for a high-curvature point  $p \in \partial D$ , it is unobjectionable to see from (4.16) that the resonant energy of the plasmon resonant mode  $\mathcal{S}_{\partial D}[\phi_i]$  also localizes/concentrates

near the point  $p$ , in the sense that the resonant oscillation near  $p$  is more significant than that near the other boundary point with a relatively smaller magnitude of curvature. According to our earlier analysis following Theorem 4.1, this is particularly the case for the high-mode-number plasmon resonant mode, namely  $\mathcal{S}_{\partial D}[\phi_i]$  with  $i \in \mathbb{N}$  sufficiently large, which corresponds to that  $\lambda_i$  is close to the accumulating point 0. Consequently, one can readily conclude similar localization/concentration results for the SPR field  $u$  in (4.15).

## 5. LOCALIZATION/CONCENTRATION OF PLASMON RESONANCES FOR QUASI-STATIC WAVE SCATTERING

In this section, we consider the scalar wave scattering governed by the Helmholtz system in the quasi-static regime and extend all of the electrostatic results to this quasi-static case. Let  $\varepsilon_0, \mu_0, \varepsilon_1, \mu_1$  be real constants and in particular, assume that  $\varepsilon_0$  and  $\mu_0$  are positive. Let  $D$  be given as that in Section 1, and set

$$\mu_D = \mu_1 \chi(D) + \mu_0 \chi(\mathbb{R}^d \setminus \overline{D}), \quad \varepsilon_D = \varepsilon_1 \chi(D) + \varepsilon_0 \chi(\mathbb{R}^d \setminus \overline{D}).$$

$(\varepsilon_1, \mu_1)$  and  $(\varepsilon_0, \mu_0)$ , respectively, signify the dielectric parameters of the plasmonic particle  $D$  and the background space  $\mathbb{R}^d \setminus \overline{D}$ . Let  $\omega \in \mathbb{R}_+$  denote a frequency of the wave. We further set  $k_0 := \omega \sqrt{\varepsilon_0 \mu_0}$  and  $k_1 := \omega \sqrt{\varepsilon_1 \mu_1}$ , where we would take the branch of the square root with non-negative imaginary part (in the case that  $\varepsilon_1 \mu_1$  is negative). Let  $u_0$  be an entire solution to  $(\Delta + k_0^2)u_0 = 0$  in  $\mathbb{R}^d$ . Consider the following Helmholtz scattering problem for  $u \in H_{loc}^1(\mathbb{R}^d)$  satisfying

$$\begin{cases} \nabla \cdot \left( \frac{1}{\mu_D} \nabla u \right) + \omega^2 \varepsilon_D u = 0 & \text{in } \mathbb{R}^d, \\ \left( \frac{\partial}{\partial |x|} - ik_0 \right) (u - u_0) = o(|x|^{-\frac{d-1}{2}}) & \text{as } |x| \rightarrow \infty, \end{cases} \quad (5.1)$$

where the last limit is known as the Sommerfeld radiation condition that characterises the outgoing nature of the scattered field  $u - u_0$ . The Helmholtz system (5.1) can be used to describe the transverse electromagnetic scattering in two dimensions, and the acoustic wave scattering in three dimensions. Nevertheless, we unify the study for any dimension  $d \geq 2$ . Moreover, we are mainly concerned with the quasi-static case, namely  $\omega \ll 1$ , or equivalently  $k_0 \ll 1$ .

Similar to the electrostatic case, we next introduce the integral formulation of (5.1). To that end, we introduce the associated layer potential operators as follows. Let

$$\Gamma_k(x - y) := C_d (k|x - y|)^{-\frac{d-2}{2}} H_{\frac{d-2}{2}}^{(1)}(k|x - y|), \quad (5.2)$$

be the outgoing fundamental solution to the differential operator  $\Delta + k^2$ , where  $C_d$  is some dimensional constant and  $H_{\frac{d-2}{2}}^{(1)}$  is the Hankel function of the first kind and order  $(d-2)/2$ .

We introduce the following single and double-layer potentials associated with a given wavenumber  $k \in \mathbb{R}_+$ ,

$$\mathcal{S}_{\partial D}^k[\phi](x) := \int_{\partial D} \Gamma_k(x - y) \phi(y) d\sigma(y), \quad x \in \mathbb{R}^d, \quad (5.3)$$

$$\mathcal{D}_{\partial D}^k[\phi](x) := \int_{\partial D} \frac{\partial}{\partial \nu_y} \Gamma_k(x - y) \phi(y) d\sigma(y), \quad x \in \mathbb{R}^d. \quad (5.4)$$

The single-layer potential  $\mathcal{S}_{\partial D}^k$  satisfies the following jump condition on  $\partial D$  (cf. [5, 26]):

$$\frac{\partial}{\partial \nu} \left( \mathcal{S}_{\partial D}^k[\phi] \right)^\pm = \left( \pm \frac{1}{2} Id + \mathcal{K}_{\partial D}^k \right) [\phi], \quad (5.5)$$

where the superscripts  $\pm$  indicate the traces from outside and inside of  $D$ , respectively, and  $\mathcal{K}_{\partial D}^k : L^2(\partial D) \rightarrow L^2(\partial D)$  is the Neumann-Poincaré (NP) operator of wavenumber  $k$  defined by

$$\mathcal{K}_{\partial D}^k[\phi](x) := \int_{\partial D} \partial_{\nu_x} \Gamma_k(x-y) \phi(y) d\sigma(y). \quad (5.6)$$

With this,  $u \in H_{loc}^1(\mathbb{R}^d)$  in (5.1) can be given by

$$u = \begin{cases} u_0 + \mathcal{S}_{\partial D}^{k_0}[\psi] & \text{on } \mathbb{R}^d \setminus \bar{D}, \\ \mathcal{S}_{\partial D}^{k_1}[\phi] & \text{on } D, \end{cases} \quad (5.7)$$

where  $(\phi, \psi) \in L^2(\partial D) \times L^2(\partial D)$  is formally given by (provided that  $k_1^2$  is not a Dirichlet eigenvalue of the Laplacian in  $D$ )

$$\begin{cases} \mathcal{S}_{\partial D}^{k_1}[\phi] - \mathcal{S}_{\partial D}^{k_0}[\psi] = u_0, \\ \frac{1}{\mu_1} \left( -\frac{1}{2} Id + \mathcal{K}_{\partial D}^{k_1} \right) [\phi] - \frac{1}{\mu_0} \left( \frac{1}{2} Id + \mathcal{K}_{\partial D}^{k_0} \right) [\psi] = \frac{1}{\mu_0} \frac{\partial u_0}{\partial \nu}, \end{cases}$$

or that

$$\begin{aligned} & \left\{ \frac{1}{2} \left( \frac{1}{\mu_0} Id + \frac{1}{\mu_1} \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} \mathcal{S}_{\partial D}^{k_0} \right) + \frac{1}{\mu_0} \mathcal{K}_{\partial D}^{k_0} - \frac{1}{\mu_1} \mathcal{K}_{\partial D}^{k_1} \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} \mathcal{S}_{\partial D}^{k_0} \right\} [\psi] \\ &= \frac{1}{\mu_1} \left( -\frac{1}{2} Id + \mathcal{K}_{\partial D}^{k_1} \right) \circ \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} [u_0] - \frac{1}{\mu_0} \frac{\partial u_0}{\partial \nu} \\ &= \left( \frac{1}{\mu_1} - \frac{1}{\mu_0} \right) \frac{\partial u_0}{\partial \nu}. \end{aligned} \quad (5.8)$$

Similar to our treatment in [4] and using (5.8), we can now formally write

$$\begin{aligned} u - u_0 &= \left( \frac{1}{\mu_1} - \frac{1}{\mu_0} \right) \mathcal{S}_{\partial D}^{k_0} \circ \left\{ \frac{1}{2} \left( \frac{1}{\mu_0} Id + \frac{1}{\mu_1} \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} \mathcal{S}_{\partial D}^{k_0} \right) \right. \\ & \quad \left. + \frac{1}{\mu_0} \mathcal{K}_{\partial D}^{k_0} - \frac{1}{\mu_1} \mathcal{K}_{\partial D}^{k_1} \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} \mathcal{S}_{\partial D}^{k_0} \right\}^{-1} \left[ \frac{\partial u_0}{\partial \nu} \right], \end{aligned} \quad (5.9)$$

when the inverses in the equation exist. As in [4], we notice that

$$\mathcal{S}_{\partial D}^k = \mathcal{S}_{\partial D} + \omega^2 \mathcal{S}_{\partial D, -3}^k, \quad \mathcal{K}_{\partial D}^k = \mathcal{K}_{\partial D} + \omega^2 \mathcal{K}_{\partial D, -3}^k \quad \text{and} \quad \Lambda_{k_0} = \Lambda_0 + \omega^2 \Lambda_{k_0, -1}, \quad (5.10)$$

where  $\mathcal{K}_{\partial D, -3}^k, \mathcal{S}_{\partial D, -3}^k, \Lambda_{k_0, -1}$  are uniformly bounded w.r.t.  $\omega$  and are of order  $-3, -3$  and  $-1$  respectively. With this, one quickly observes that the following lemma holds (cf. [4]):

**Lemma 5.1.** *There holds*

$$u - u_0 = \mathcal{S}_{\partial D}^{k_0} \circ \left( \left\{ \lambda(\mu_0^{-1}, \mu_1^{-1}) Id - \mathcal{K}_{\partial D} \right\}^{-1} \circ \Lambda_{k_0}(u_0) + \omega^2 R_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -1}(u_0) \right), \quad (5.11)$$

where  $R_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -1}$  is uniformly bounded with respect to  $\omega \ll 1$  and is of order  $-1$ .

Similar to the static case discussed in Section 1.2, for given  $\mu_0, \varepsilon_0$  and  $\omega \ll 1$ , if the following operator equation

$$\left\{ \frac{1}{2} \left( \frac{1}{\mu_0} Id + \frac{1}{\mu_1} \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} \mathcal{S}_{\partial D}^{k_0} \right) + \frac{1}{\mu_0} \mathcal{K}_{\partial D}^{k_0} - \frac{1}{\mu_1} \mathcal{K}_{\partial D}^{k_1} \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} \mathcal{S}_{\partial D}^{k_0} \right\} \phi = 0 \quad (5.12)$$

has a non-trivial solution  $\phi \in H^{-1/2}(\partial D, d\sigma)$ , then  $(\varepsilon_1, \mu_1)$  is said to be a pair of plasmonic eigenvalue and  $\phi$  is called a perturbed NP eigenfunction. In this case, the plasmon resonant field in  $\mathbb{R}^d \setminus \bar{D}$  is given by  $\mathcal{S}_{\partial D}^{k_0}[\phi]$ . Next, we consider the geometric properties of the perturbed

NP eigenfunctions as well as the associated layer-potentials described above. We quickly realize from (5.10) that (5.12) reads:

$$\left\{ \lambda(\mu_0^{-1}, \mu_1^{-1}) Id - \mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3} \right\} \phi = 0, \quad (5.13)$$

where  $\mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3}$  is uniformly bounded with respect to  $\omega \ll 1$  and is of order  $-3$ . Furthermore, since  $\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3}$  is compact but not-self adjoint, we have a finite dimensional generalized eigenspace whenever the eigenvalue is non zero [15], (we are unsure of what happens when  $\lambda = 0$ , i.e. if the kernel of the operator is finite dimensional and if there is a quasi-nilpotent subspace). We may therefore consider the following generalized plasmon resonance: find  $\phi \in H^{-1/2}(\partial D, d\sigma)$  such that for some  $m \in \mathbb{N}$ ,

$$\left\{ \frac{1}{2} \left( \frac{1}{\mu_0} Id + \frac{1}{\mu_1} \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} \mathcal{S}_{\partial D}^{k_0} \right) + \frac{1}{\mu_0} \mathcal{K}_{\partial D}^{k_0} - \frac{1}{\mu_1} \mathcal{K}_{\partial D}^{k_1} \left( \mathcal{S}_{\partial D}^{k_1} \right)^{-1} \mathcal{S}_{\partial D}^{k_0} \right\}^m \phi = 0. \quad (5.14)$$

We note from our earlier discussion that if  $(\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega)$  is such that (5.14) has a solution, then  $m$  is finite.

The following lemma characterizes the plasmon resonance when  $\omega \ll 1$ .

**Lemma 5.2.** *Under Assumption (A), suppose  $\omega \ll 1$ , a solution  $((\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega), m, \phi_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, m})$  satisfying the generalized plasmon resonance equation (5.14) with unit  $L^2$ -norm possesses the following property for all  $s \in \mathbb{R}$ :*

$$\begin{cases} \| |D|^s \phi_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, m} - |D|^s \phi_i \|_{C^0(\partial D)} &= \mathcal{O}_{i,s}(\omega^2), \\ \lambda(\mu_0^{-1}, \mu_1^{-1}) - \lambda_i &= \mathcal{O}_i(\omega^2), \end{cases}$$

for some eigenpair  $(\lambda_i, \phi_i)$  of the Neumann-Poincaré operator  $\mathcal{K}_{\partial D}^*$  with zero wavenumber, and  $m \leq m_i$ , where  $\|\phi\|_{L^2(D)} = 1$ , and  $m$  and  $m_i$  signify the algebraic multiplicities of  $\lambda$  and  $\lambda_i$ , respectively. Here the constant in  $\mathcal{O}_{i,s}$  depends on both  $i$  and  $s$ , and that in  $\mathcal{O}_i$  depends only on  $i$ .

*Proof.* Since the family  $\{\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3}\}_{\omega \geq 0}$  is collectively compact, it readily follows from Osborn's Theorem [37] and the equivalence of  $\|\cdot\|_{H^{-1/2}(\partial D, d\sigma)}$  and  $\|\cdot\|_{L^2_{S_{\partial D}}(\partial D)}$  and that a solution  $((\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega), m, \phi_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, m})$  satisfying (5.14) will satisfy:

$$\begin{cases} \|\phi_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, m} - \phi_i\|_{H^{-1/2}(\partial D, d\sigma)} &= \mathcal{O}_i(\omega^2), \\ \lambda(\mu_0^{-1}, \mu_1^{-1}) - \lambda_i &= \mathcal{O}_i(\omega^2), \end{cases}$$

for some eigenpair  $(\lambda_i, \phi_i)$  of the Neumann-Poincaré operator  $\mathcal{K}_{\partial D}^*$ .

It remains to obtain the  $\| |D|^s(\cdot) \|_{C^0(\partial D)}$  bounds instead of  $H^{-1/2}(\partial D, d\sigma)$  bounds. For this purpose, let us look into the generalized eigenspace  $E_{\lambda(\mu_0^{-1}, \mu_1^{-1})}$  of  $\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3}$  and pick  $\widetilde{\phi}_{i,j} \in E_{\lambda(\mu_0^{-1}, \mu_1^{-1})}$  be with unit  $H^{-1/2}$  norm such that  $\widetilde{\phi}_{i,m} = \phi_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, m}$ . Then there exists  $\{\varepsilon_{j,j-1}\}_{j=2}^m$  with  $|\varepsilon_{j,j-1}| = \mathcal{O}_i(\omega^2)$  such that

$$\begin{cases} (\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3} - \lambda(\mu_0^{-1}, \mu_1^{-1})) \widetilde{\phi}_{i,j} = \varepsilon_{j,j-1} \widetilde{\phi}_{i,j-1} \text{ for } j = 2, \dots, m, \\ (\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3} - \lambda(\mu_0^{-1}, \mu_1^{-1})) \widetilde{\phi}_{i,1} = 0, \end{cases}$$

which can always be done by rescaling the basis giving the Jordan block representation with a scaling factor of  $1/\varepsilon_{j,j-1}$ . Then Osborn's Theorem and the equivalence of norms yield

$$\|\widetilde{\phi}_{i,j} - \phi_{i,j}\|_{H^{-1/2}(\partial D, d\sigma)} = \mathcal{O}_i(\omega^2)$$



for some  $\phi_{i,j} \in \mathcal{C}^\infty(\partial D)$  sitting in the eigenspace of  $\mathcal{K}_{\partial D}^*$ . Taking the difference between the system in the generalized eigenspace and the original eigenvalue equations:

$$\left\{ \begin{array}{l} (\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3}) (\phi_{i,j} - \widetilde{\phi}_{i,j}) - \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3} \phi_{i,j} \\ = \lambda(\mu_0^{-1}, \mu_1^{-1})(\phi_{i,j} - \widetilde{\phi}_{i,j}) + (\lambda_i - \lambda(\mu_0^{-1}, \mu_1^{-1}))\phi_{i,j} - \varepsilon_{j,j-1} \widetilde{\phi}_{i,j-1} \text{ for } j = 2, \dots, m, \\ (\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3}) (\phi_{i,1} - \widetilde{\phi}_{i,1}) - \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3} \phi_{i,1} \\ = \lambda(\mu_0^{-1}, \mu_1^{-1})(\phi_{i,1} - \widetilde{\phi}_{i,1}) + (\lambda_i - \lambda(\mu_0^{-1}, \mu_1^{-1}))\phi_{i,1}. \end{array} \right.$$

Now, under Assumption (A),  $\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3} : H^s(\partial D) \rightarrow H^{s+1}(\partial D)$ , and therefore we have from the above system that

$$\|\widetilde{\phi}_{i,j} - \phi_{i,j}\|_{H^{1/2}(\partial D, d\sigma)} = \mathcal{O}_i(\omega^2),$$

for all  $j = 1, \dots, m$  with a different constant. One now gazes at the above system. Together with a bootstrapping argument and the fact that Assumption (A) gives  $\phi_{i,j} \in \mathcal{C}^\infty(\partial D)$ , we arrive at, for all  $l \in \mathbb{R}$ ,

$$\|\widetilde{\phi}_{i,j} - \phi_{i,j}\|_{H^l(\partial D, d\sigma)} = \mathcal{O}_{i,l}(\omega^2).$$

Our conclusion follows after applying the Sobelov embedding theorem to bound the  $\| |D|^s(\cdot) \|_{\mathcal{C}^0(\partial D)}$  semi-norm by the  $H^{s+l}(\partial D, d\sigma)$  norm for large enough  $l$ .

The proof is complete.  $\square$

We aim to know whether the generalized plasmon resonance (cf. (5.14)) always exists when  $\omega \ll 1$ . The following lemma addresses this issue.

**Lemma 5.3.** *Given any non-zero  $\lambda_i \in \sigma(\mathcal{K}_{\partial D}^*)$ , the spectrum of  $\mathcal{K}_{\partial D}^*$ , for any  $(\tilde{\mu}_0, \tilde{\mu}_1) \in D_i := \{(\mu_0, \mu_1) \in \mathbb{C}^2 \setminus \{(0,0)\} : \lambda(\tilde{\mu}_0^{-1}, \tilde{\mu}_1^{-1}) = \lambda_i, \mu_0 - \mu_1 \neq 0\}$  (which is non-empty), there exists  $0 < \omega_i \ll 1$  such that for all  $\omega < \omega_i$ , the set*

$$\left\{ (\mu_0, \mu_1, \varepsilon_0, \varepsilon_1) \in \mathbb{C}^2 \setminus \{\mu_0 - \mu_1 = 0\} \times (\mathbb{C} \setminus \mathbb{R}^+)^2; \right.$$

*there exists  $m \in \mathbb{N}, \phi \in H^{-1/2}(\partial D, \sigma)$  such that  $((\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega), \phi, m)$  satisfies (5.14)  $\left. \right\}$*

*forms a complex co-dimension 1 surface in a neighborhood of  $(\tilde{\mu}_0, \tilde{\mu}_1)$ .*

*Proof.* Given a non-zero  $\lambda_i \in \sigma(\mathcal{K}_{\partial D}^*)$ , we consider a function  $F_i$  defined over  $\partial D$  and  $\lambda_i \in \sigma(\mathcal{K}_{\partial D}^*)$ . In particular, by Osborn's Theorem [37] and the smooth dependence of  $\mathcal{E}$  on  $(\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega)$ , there exists  $0 < \tilde{\omega}_i \ll 1$  (depending on  $i$ ) such that we have a (non-unique) smooth choice of function:

$$\begin{aligned} F_{i,\delta} : \mathbb{C}^2 \setminus \{\mu_0 - \mu_1 = 0\} \times (\mathbb{C} \setminus \mathbb{R}^+)^2 \times (0, \tilde{\omega}_i) &\rightarrow \mathbb{C}, \\ F_i(\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega) &= \tilde{\lambda}_i(\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega) - \lambda(\mu_0^{-1}, \mu_1^{-2}), \end{aligned}$$

where

$$\tilde{\lambda}_i(\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega) \in \sigma(\mathcal{K}_{\partial D}^* + \omega^2 \mathcal{E}_{\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega, \partial D, -3})$$

is such that

$$\lim_{\varepsilon \rightarrow 0} \tilde{\lambda}_i(\mu_0, \mu_1, \varepsilon_0, \varepsilon_1, \omega) = \lambda_i.$$

We now note that, for any  $(\tilde{\mu}_0, \tilde{\mu}_1) \in D_i$  in this set,

$$F_i(\tilde{\mu}_0, \tilde{\mu}_1, \varepsilon_0, \varepsilon_1, 0) = 0,$$

for all  $\varepsilon_0, \varepsilon_1$  in the domain of the function. Moreover, we can directly verify that

$$\partial_\omega F_i(\tilde{\mu}_0, \tilde{\mu}_1, \varepsilon_0, \varepsilon_1, 0) = 0, \quad \partial_{\varepsilon_0, \varepsilon_1} F_i(\tilde{\mu}_0, \tilde{\mu}_1, \varepsilon_0, \varepsilon_1, 0) = 0,$$

whereas

$$\partial_{\mu_0, \mu_1} F_i(\tilde{\mu}_0, \tilde{\mu}_1, \varepsilon_0, \varepsilon_1, 0) = \partial_{\mu_0, \mu_1} \lambda(\tilde{\mu}_0^{-1}, \tilde{\mu}_1^{-1}) = \left( -\frac{\tilde{\mu}_1}{2(\tilde{\mu}_1 - \tilde{\mu}_0)^2}, \frac{\tilde{\mu}_0}{2(\tilde{\mu}_1 - \tilde{\mu}_0)^2} \right).$$

Hence, we have

$$\partial_{\mu_0} F_i(\tilde{\mu}_0, \tilde{\mu}_1, \varepsilon_0, \varepsilon_1, 0) \neq 0 \quad \text{or} \quad \partial_{\mu_1} F_i(\tilde{\mu}_0, \tilde{\mu}_1, \varepsilon_0, \varepsilon_1, 0) \neq 0.$$

Therefore, applying the inverse function theorem in a neighborhood of any chosen point in  $D_i \times (\mathbb{C} \setminus \mathbb{R}^+)^2 \times \{0\}$ , we obtain either a unique smooth function  $l_0 : B_\delta(\tilde{\mu}_0) \rightarrow l_0(B_\delta(\tilde{\mu}_0))$  fulfilling

$$F_i(\mu_0, l_{0,1}(\mu_0), l_{0,2}(\mu_0), l_{0,3}(\mu_0), l_{0,4}(\mu_0)) = 0,$$

or a unique smooth function  $l_1 : B_\delta(\tilde{\mu}_1) \rightarrow l_1(B_\delta(\tilde{\mu}_1))$  fulfilling

$$F_i(l_{1,1}(\mu_1), \mu_1, l_{1,2}(\mu_1), l_{1,3}(\mu_1), l_{1,4}(\mu_1)) = 0.$$

If we obtain  $l_0$ , let us take  $\omega_i \leq \tilde{\omega}_i$  such that  $\omega_i \in l_{0,3}(B_\delta(\tilde{\mu}_0))$ . Otherwise, we take  $\omega_i \leq \tilde{\omega}_i$  such that  $\omega_i \in l_{1,3}(B_\delta(\tilde{\mu}_1))$ . The conclusion stated in the lemma readily follows.  $\square$

By Lemma 5.3, we easily see that there are infinitely many choices of  $(\varepsilon_1, \mu_1)$  such that the (generalized) plasmon resonance occurs around  $\lambda_i$ . Combining with a similar perturbation argument as in the proof of Lemma 5.3, our conclusions of the plasmon resonance in the electrostatic case transfers to the Helmholtz transmission problem to show concentration of plasmon resonances at high-curvature points. For instance, we have the following result.

**Theorem 5.4.** *Given any  $x \in \partial D$ , let us consider  $\{\chi_{x,\delta}\}_{\delta>0}$  being a family of smooth nonnegative bump functions compactly supported in  $B_\delta(x)$  with  $\int_{\partial D} \chi_{p,\delta} d\sigma = 1$ . Under Assumption (A), given  $r \leq s$ ,  $\alpha \in \mathbb{R}$  and  $p, q \in \partial D$ , we have a choice of  $\delta(h)$  and  $\omega(h)$  both depending on  $r, s, p, q$  and  $\alpha$  such that for any  $\omega < \omega(h)$ , there exists*

$$((\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega), m_i, \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i})$$

solving (5.14), and as  $h \rightarrow +0$ , we have  $\delta(h) \rightarrow 0$ ,  $\omega(h) \rightarrow 0$  and

$$\frac{\sum_{r h \leq \lambda_i^2 \leq s h} c_i \int_{\partial D} \chi_{p,\delta(h)}(x) \|D\|^\alpha |\phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i}(x)|^2 d\sigma(x)}{\sum_{r h \leq \lambda_i^2 \leq s h} c_i \int_{\partial D} \chi_{q,\delta(h)}(x) \|D\|^\alpha |\phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i}(x)|^2 d\sigma(x)} = \frac{\int_{\{H(p,\cdot)=1\}} |\xi|_{g(p)}^{1+2\alpha} d\sigma_{p,H}}{\int_{\{H(q,\cdot)=1\}} |\xi|_{g(q)}^{1+2\alpha} d\sigma_{q,H}} + o_{r,s,p,q,\alpha}(1), \quad (5.15)$$

where  $c_i := |\phi_i|_{H^{-\frac{1}{2}}(\partial D, d\sigma)}^{-2}$ . Here, the little- $o$  depends on  $r, s, p, q$  and  $\alpha$ .

*Proof.* From Theorem 4.1, we have a choice of  $\delta(h)$  depending on  $r, s, p, q$  and  $\alpha$  such that, for any given  $\varepsilon > 0$ , there exists  $h_0$  depending on  $r, s, p, q, \alpha$  such that for all  $h < h_0$ ,

$$\left| \frac{\sum_{r h \leq \lambda_i^2 \leq s h} c_i \int_{\partial D} \chi_{p,\delta(h)}(x) \|D\|^\alpha |\phi_i(x)|^2 d\sigma}{\sum_{r h \leq \lambda_i^2 \leq s h} c_i \int_{\partial D} \chi_{q,\delta(h)}(x) \|D\|^\alpha |\phi_i(x)|^2 d\sigma} - \frac{\int_{\{H(p,\cdot)=1\}} |\xi|_{g(p)}^{1+2\alpha} d\sigma_{p,H}}{\int_{\{H(q,\cdot)=1\}} |\xi|_{g(q)}^{1+2\alpha} d\sigma_{q,H}} \right| \leq \varepsilon. \quad (5.16)$$

Now for each  $h < h_0$ , from Lemma 5.3, there exists  $\tilde{\omega}(h) := \min_{\{i \in \mathbb{N} : r h \leq \lambda_i^2 \leq s h\}} \{\omega_i\}$  such that for all  $\omega < \omega(h)$ , there exists

$$((\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega), m_i, \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i})$$

solving (5.14). By Lemma 5.2, upon a rescaling of  $\phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i}$  while still denoting it as  $\phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i}$ , we have

$$\| \|D\|^\alpha \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i} - \|D\|^\alpha \phi_i \|_{C^0(\partial D)} \leq C_{i,\alpha} \omega^2.$$

In particular, we can make a smaller choice of  $\omega(h) < \tilde{\omega}(h)$  depending on  $r, s, p, q, \alpha$  such that for all  $\omega < \omega(h)$ , we have

$$\begin{aligned} & \left| \| |D|^\alpha \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i} \|^2 - \| |D|^\alpha \phi_i \|^2 \right|_{C^0(\partial D)} \\ & \leq 10^{-2} \varepsilon / \sum_{rh \leq \lambda_i^2 \leq sh} c_i / \min \left\{ 1, \min_{y=p,q} \left\{ \left( \sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{y, \delta(h)}(x) \| |D|^\alpha \phi_i(x) \|^2 d\sigma \right)^{-2} \right\} \right\}. \end{aligned}$$

Therefore, with this choice of  $\omega(h)$ , we have, for all  $\omega < \omega(h)$

$$\begin{aligned} & \left| \frac{\sum_{rh \leq \lambda_i^2 \leq sh} c_i \int_{\partial D} \chi_{p, \delta(h)}(x) \| |D|^\alpha \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i}(x) \|^2 d\sigma(x)}{\sum_{rh \leq \lambda_i^2 \leq sh} c_i \int_{\partial D} \chi_{q, \delta(h)}(x) \| |D|^\alpha \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i}(x) \|^2 d\sigma(x)} \right. \\ & \quad \left. - \frac{\sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{p, \delta(h)}(x) \| |D|^\alpha \phi_i(x) \|^2 d\sigma}{\sum_{r \leq \lambda_i^2(h) \leq s} c_i \int_{\partial D} \chi_{q, \delta(h)}(x) \| |D|^\alpha \phi_i(x) \|^2 d\sigma} \right| \leq \varepsilon. \end{aligned} \quad (5.17)$$

Combining (5.17) with (5.16) readily yields our conclusion.

The proof is complete.  $\square$

Likewise we obtain the following result.

**Theorem 5.5.** *Under Assumption (A), given  $r, s$ , there exists  $S(h) \subset J(h) := \{i \in \mathbb{N} : rh \leq \lambda_i^2 \leq sh\}$  and  $\omega(h)$  such that, for all  $\varphi \in C^\infty(\partial D)$ , we have for any  $\omega < \omega(h)$ , there exists*

$$((\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega), m_i, \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i})$$

solving (5.14), such that as  $h \rightarrow +0$ , we have  $\omega(h) \rightarrow 0$  and

$$\begin{aligned} & \max_{i \in S(h)} \left| \int_{\partial D} \varphi(x) \left( c_i \| |D|^{-\frac{1}{2}} \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i}(x) \|^2 \right. \right. \\ & \quad \left. \left. - \int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(x) g_i(\mu) d\nu(\mu) \right) d\sigma(x) \right| = o_{r,s}(1). \end{aligned} \quad (5.18)$$

Here,  $S(h)$ ,  $\{g_i : M_{X_H, \text{erg}}(\{H=1\}) \rightarrow \mathbb{C}\}_{i \in \mathbb{N}}$  and  $\mu(p)$  are described as in Theorem 4.2. In particular, we remind that

$$\frac{\int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(p) d\nu(\mu)}{\int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(q) d\nu(\mu)} = \frac{\int_{\{H(p, \cdot)=1\}} d\sigma_{p,H}}{\int_{\{H(q, \cdot)=1\}} d\sigma_{q,H}} \quad \text{a.e. } (d\sigma \otimes d\sigma)(p, q).$$

If the Hamiltonian flow given by  $X_H$  is uniquely ergodic on  $\{H=1\}$ , then

$$\max_{i \in S(h)} \left| \int_{\partial D} \varphi(x) \left( c_i \| |D|^{-\frac{1}{2}} \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i}(x) \|^2 - \frac{\int_{\{H(x, \cdot)=1\}} d\sigma_{x,H}}{\sigma_H(\{H=1\})} \right) d\sigma(x) \right| = o_{r,s}(1).$$

*Proof.* Let  $r, s$  be given. Consider  $\varphi \in C^\infty(\partial D)$ . Given  $\varepsilon > 0$ , by Theorem 4.2 and considering  $h_0$  small enough such that for all  $h < h_0$ , we have

$$\max_{i \in S(h)} \left| \int_{\partial D} \varphi(x) \left( c_i \| |D|^{-\frac{1}{2}} \phi_i(x) \|^2 - \int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(x) g_i(\mu) d\nu(\mu) \right) d\sigma(x) \right| \leq \varepsilon.$$

Now, for each  $h < h_0$ , from Lemma 5.3, there exists  $\tilde{\omega}(h) = \min \{ \min_{i \in S(h)} \omega_i, 1 \}$  such that for all  $\omega < \tilde{\omega}(h)$ , there exists

$$((\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega), m_i, \phi_{\mu_{0,i}, \mu_{1,i}, \varepsilon_{0,i}, \varepsilon_{1,i}, \omega, m_i})$$

solving (5.14). By Lemma 5.2, again upon a rescaling of  $\phi_{\mu_0,i,\mu_1,i,\varepsilon_0,i,\varepsilon_1,i,\omega,m_i}$  while still denoting it as  $\phi_{\mu_0,i,\mu_1,i,\varepsilon_0,i,\varepsilon_1,i,\omega,m_i}$ , we have

$$\begin{aligned} & \max_{i \in S(h)} c_i \left| \int_{\partial D} \varphi(x) \left( \|D\|^{-\frac{1}{2}} \phi_i(x)^2 - \|D\|^{-\frac{1}{2}} \phi_{\mu_0,i,\mu_1,i,\varepsilon_0,i,\varepsilon_1,i,\omega,m_i}(x)^2 \right) d\sigma(x) \right| \\ & \leq C_{S(h)} \|\varphi\|_{C^0(\partial D)} \omega^2. \end{aligned} \quad (5.19)$$

We may now choose

$$\omega(h) \leq \min \{ \varepsilon, \tilde{\omega}(h), \tilde{\omega}(h)/C_{S(h)} \}.$$

Then for all  $\omega < \omega(h)$ , we finally have from (5.19) and Corollary 4.3 that

$$\begin{aligned} & \max_{i \in S(h)} \left| \int_{\partial D} \varphi(x) \left( c_i \|D\|^{-\frac{1}{2}} \phi_{\mu_0,i,\mu_1,i,\varepsilon_0,i,\varepsilon_1,i,\omega,m_i}(x)^2 \right. \right. \\ & \quad \left. \left. - \int_{M_{X_H, \text{erg}}(\{H=1\})} \mu(x) g_i(\mu) d\nu(\mu) \right) d\sigma(x) \right| \\ & \leq (1 + \|\varphi\|_{C^0(\partial D)}) \varepsilon. \end{aligned}$$

The proof is complete.  $\square$

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#### APPENDIX A. FURTHER REMARKS UPON SOME GEOMETRIC BEHAVIORS OF THE HAMILTONIAN FLOW

In this appendix, we would like to briefly explore some geometric behaviours of the Hamiltonian flow, which should help our understanding of  $M_{X_H, \text{erg}}(\{H=1\})$  and translate to the understanding of the NP eigenfunctions. We also want to understand and explore Assumption (A), which is equivalent to  $\overline{\{H=1\}} \cap (\partial D \times \{0\}) = \emptyset$ , which we always assume in our work (until now). First, we have the following elementary property of  $X_H$ :

**Lemma A.1.** *If  $\xi \neq 0$ , then  $X_H \neq 0$ .*

*Proof.* Consider  $S := \log H$ . Since  $X_H$  preserves  $H$ , let us consider its action only on  $\{H=c\}$  with  $c \neq 0$ . By choosing a local coordinate, one can directly compute that

$$\partial_\xi H = H \partial_\xi S = 2c \left( 2 \frac{(d-1)H(x)g^{-1}(x) - g^{-1}(x)A(x)g^{-1}(x)\xi}{|\langle (d-1)H(x)g^{-1}(x) - g^{-1}(x)A(x)g^{-1}(x)\xi, \xi \rangle|} - 3 \frac{g^{-1}\xi}{\langle g^{-1}(x)\xi, \xi \rangle} \right).$$

Therefore we immediately have that  $\langle \partial_\xi H, \xi \rangle = 2(\pm 2 - 3)c$ , which ensures that  $\partial_\xi H \neq 0$ , and hence our conclusion holds.  $\square$

**A.1. The non-singular case when  $\overline{\{H=1\}} \cap (\partial D \times \{0\}) = \emptyset$ .** Let us assume  $\overline{\{H=1\}}$  does not contain  $(x, 0) \in \partial D \times \{0\} \leftrightarrow T^*(\partial D)$ . We would like to look into the local property of the flow. As an example, let us consider  $d=3$ . In fact, the condition  $\overline{\{H=1\}} \cap (\partial D \times \{0\}) = \emptyset$  readily implies that the Gaussian curvature  $\kappa(x) := \kappa_1(x)\kappa_2(x) \neq 0$ . By the compactness of the surface, we have  $\kappa(x) > 0$  for some  $x \in \partial D$ . Then by continuity we have  $\kappa(x) > c$  for all  $x \in \partial D$  for some  $c > 0$ . An application of the Gauss-Bonnet theorem readily yields the Euler characteristic of the surface  $\chi(\partial D) > 0$ , and hence  $\partial D$  is diffeomorphic to a sphere. Moreover, there exists  $c_0$  such that the matrix  $A(x) > c_0 Id$  for all  $x \in \partial D$ , i.e., the domain  $D$  is strictly convex. The following figure shows a typical example of  $\{H(x, \cdot) = 1\}$ .

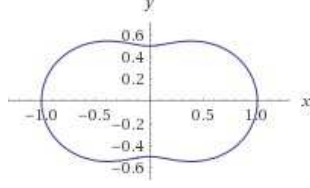


FIGURE 1. Level curve  $\overline{\{H(a, \xi) = 1\}}$  for a fixed  $a \in \partial D$  when  $A(a) = \text{diag}(1, 0.5)$ , where  $\xi = (x, y)$ .

In this case, locally around a point  $(x, \xi) \in \{H = 1\}$ , the flow  $X_H$  given by  $\partial_t(x(t), p(t)) = X_H(x(t), p(t))$  projects to the  $x$ -coordinate to give  $\partial_t x(t) = \partial_\xi H$ , which is the normal of the level set  $\{H(x, \cdot) = 1\}$ .

In general when  $d > 2$ , suppose  $X_H$  generates a Hamiltonian circle action (i.e.  $\mathbb{T}^1$  action) over  $T^*(\partial D)$ . Then by Lemma A.1, the circle action has no critical point on  $\{H = 1\} = \overline{\{H = 1\}}$ , and that 1 is a regular value of  $H$ . We may therefore perform a symplectic reduction to obtain  $M := \{H = 1\}/\mathbb{T}^1$ . From the compactness of  $\{H = 1\}$ ,  $M$  is now a compact symplectic manifold of dimension  $2d - 4$ .  $M$  also provides a parametrization of the set of periodic orbits (equivalently, of the ergodic measures in this case). In this case, we can appeal to results of classical symplectic geometry to classify the global structure of the flow.

We would like to remark that the non-singular case is rather restrictive, e.g. in  $d = 3$ , any  $\partial\Omega$  not diffeomorphic to  $\mathbb{S}^2$  would admit a flow  $X_H$  on  $\{H = 1\}$  with singularities.

**A.2. The singular case when  $\overline{\{H = 1\}} \cap (\partial D \times \{0\}) \neq \emptyset$ .** In general, we would also like to consider the dynamics where  $\overline{\{H = 1\}}$  may contain points  $(x, 0) \in \partial D \times \{0\}$ . The critical set may now be highly singular and degenerate. To appropriately (and mildly) resolve the singularity of  $H$  and  $X_H$  around  $(x, 0)$ , we consider

$$\tilde{H}(x, \xi) := \arctan(H(x, \xi)) = \arctan(\exp(S(x, \xi))).$$

One directly verifies that  $\tilde{H}$  removes the  $(-2)$ -order singularity of  $H$  (which is smooth away from zero) at  $\xi = 0$  in the following sense: that  $\tilde{H}$  is now furthermore bounded, and directionally differentiable at 0, all the while generating a rescaled flow of  $X_H$  away from the singularities. In particular, one quickly checks that

$$\partial \tilde{H} = \frac{H}{1 + H^2} \partial S = 0 \quad \Leftrightarrow \quad \xi = 0.$$

Therefore we have the following lemma.

**Lemma A.2.**  $X_{\tilde{H}} = 0$  if and only if  $\xi = 0$ .

The set of critical points  $\{X_{\tilde{H}} = 0\} = \partial D \times \{0\}$  are still highly degenerate and very singular. As an example, let us take  $d = 3$  for illustration purpose. When  $d = 3$ , one of the cases that  $\overline{\{H = 1\}} \cap (\partial D \times \{0\}) \neq \emptyset$  is when  $\overline{\{H = 1\}}$  contains a point  $(x, 0)$  with its mean curvature  $H(x) = 0$ . Near such point  $x \in \partial D$ , write  $\lambda(x) := \kappa_1(x) = -\kappa_2(x)$  and  $\xi = r\omega = r(\cos(\theta), \sin(\theta))$ , then  $\overline{\{H(x, r\omega) = 1\}}$  can be parametrized by

$$r^2(\theta) = \lambda^2(x) \cos^2(2\theta).$$

The following figure shows  $\overline{\{H(p, \cdot) = 1\}}$  in this degenerate and singular case.

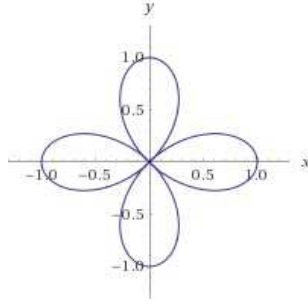


FIGURE 2. The closure of the level curve  $\overline{\{H(a, \xi) = 1\}}$  for a fixed  $a \in \partial\Omega$  when  $A(a) = \text{diag}(1, -1)$ , where  $\xi = (x, y)$ .

Locally around a point  $(x, \xi) \in \overline{\{H = 1\}}$  away from  $(x, 0)$  where  $H(x) = 0$ , the flow  $X_{\tilde{H}}$  is again given by  $\partial_t(x(t), p(t)) = X_{\tilde{H}}(x(t), p(t))$ . It projects to the  $x$ -coordinate to give  $\partial_t x(t) = \partial_\xi \tilde{H}$ , which is the normal of the level set  $\{H(x, \cdot) = 1\}$ . However, when  $(x, \xi) \in \overline{\{H = 1\}}$  is close to  $(x, 0)$  where  $H(x) = 0$ , we can see from the above figure that the normal of the level set  $\overline{\{H(x, \cdot) = 1\}}$  is behaving pathologically, creating a pathological behaviour of the flow around that point.

**Remark A.3.** *We expect further study of the local and global structures of the flow  $X_H$  (e.g. its dynamical property or its symplectic geometric property) in the singular case to be possible via the study of  $X_{\tilde{H}}$  near the critical set. For instance, we suspect a generalized version of Duistermaat-Heckman formula [39] to hold via a stationary phase approximation (only along the co-normal directions in a tabulated neighbour of  $\partial D \times \{0\}$ ) of*

$$\int_{T^*(\partial D)} g(x, \xi) \exp(-i/h \tilde{H}(x, \xi))(d\sigma \otimes d\sigma)(x, \xi)$$

for any  $g \in C_c^\infty(T^*(\partial D))$  as an expansion of  $h$  to provide further topological information of the global structure of flow. The exploration of these properties will be the subject of a forthcoming work.

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DEPARTMENT OF MATHEMATICS, ETH ZÜRICH, RÄMISTRASSE 101, CH-8092, SWITZERLAND  
E-mail address: [habib.ammari@math.ethz.ch](mailto:habib.ammari@math.ethz.ch)

DEPARTMENT OF MATHEMATICS, UNIVERSITY OF CALIFORNIA, RIVERSIDE, USA  
E-mail address: [yattinc@ucr.edu](mailto:yattinc@ucr.edu)

DEPARTMENT OF MATHEMATICS, CITY UNIVERSITY OF HONG KONG, HONG KONG SAR, CHINA  
E-mail address: [hongyu.liuip@gmail.com](mailto:hongyu.liuip@gmail.com), [hongyliu@cityu.edu.hk](mailto:hongyliu@cityu.edu.hk)