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Nonadiabatic Ponderomotive Potentials

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An approximate integral of the Manley-Rowe type is found for a particle moving in a highfrequency field, which may interact resonantly with natural particle oscillations. An effective ponderomotive potential is introduced accordingly and can capture nonadiabatic particle dynamics. We show that nonadiabatic ponderomotive barriers can trap classical particles, produce cooling effect, and generate one-way walls for resonant species. Possible atomic applications are also envisioned.

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I. INTRODUCTION

where v is the particle average velocity, and

$$v/\Delta\omega_{\nu} \ll L,$$
 (3)

Even without a bias, an ac field can exert a significant time-averaged force on a particle [1–3]. This so-called (average) ponderomotive force is comprised of two components: the Miller force due to the dipole interaction of the particle with the ac field and the light pressure due to the particle scattering and absorption of the radiation. Often, the light pressure is negligible compared to the Miller force, and the induced particle dipole moment **p** follows an adiabatic equation of state. The latter means that **p** can be approximately expressed as a local function of the particle location \mathbf{r} , which, in the simplest case, is proportional to the amplitude of the field: $\mathbf{p} = \boldsymbol{\alpha}(\omega; \mathbf{r}) \mathbf{E}_0(\mathbf{r})$. (Here $\boldsymbol{\alpha}$ is the polarizability tensor, and ω is the field frequency; the conventional complex notation is implied.) In this case, the average force on the particle can be approximately described in terms of the ponderomotive, or Miller potential Φ , equal to the average energy of the dipole-field interaction:

$$\Phi = -\frac{1}{4} \left(\mathbf{E}_0^* \cdot \boldsymbol{\alpha} \cdot \mathbf{E}_0 \right). \tag{1}$$

Importantly, Eq. (1) applies both to elementary and compound particles and can capture adiabatic effects connected with particle natural oscillations, such as Larmor rotation in a background dc magnetic field, electron Langmuir oscillations in plasmas, intra-atomic quantum oscillations, and molecular vibrations. Specific properties, which ponderomotive potentials (1) exhibit for particles exhibiting natural oscillations, have been employed in a vast variety of applications in atomic, molecular, and plasma physics, which enjoy experimental verification in a wide range of frequencies and intensities of electromagnetic radiation [2–10].

In the presence of natural oscillations at frequency ω_{ν} comparable with ω , the potential (1) is "seen" by a particle on average over time scales large compared to the field period $2\pi/\omega$, the natural period $2\pi/\omega_{\nu}$, and the beat period $2\pi/|\omega - \omega_{\nu}|$, if any. It is implied then that the drift displacement of the particle on each of these time scales is insignificant as compared to the field scale L, namely,

$$v/\omega \ll L, \qquad v/\omega_{\nu} \ll L,$$
 (2)

where $\Delta \omega_{\nu} = \omega - \omega_{\nu}$ is the beat frequency. If ω_{ν} itself varies in space, the variation of the beat period along the particle trajectory is also required to remain smooth enough:

$$\frac{v}{\Delta\omega_{\nu}} \left| \nabla \ln(\Delta\omega_{\nu}) \right| \ll 1.$$
(4)

Together with the requirement of small amplitude of the particle oscillations as compared to L, Eqs. (2)-(4) represent the validity conditions for Eq. (1).

At resonant interaction ($\omega \approx \omega_{\nu}$), the approximation of a local potential (1) is violated. Near the resonance, the particle polarizability exhibits a singularity $\alpha \propto \Delta \omega_{\nu}^{-1}$ and Φ goes to infinity (assuming negligible dissipation), although the true force on a particle remains finite. Our preliminary studies predict a number of striking fundamental and applied effects in this domain, not captured by the traditional adiabatic model, including quantumlike behavior of classical particles in ponderomotive barriers [11], a possibility of one-way rf walls [12–14], and others [15]. To describe those in detail and predict new, unexplored effects, a generalization of the ponderomotive potential concept is required.

For electrons and ions under rf drive near a cyclotron resonance in a magnetic field, this problem has been addressed previously in a number of works [2, 8, 16– 25, 25–27]. None of those, however, has introduced a non-singular ponderomotive potential in a non-heuristic fashion, except for the essentially perturbative analysis proposed in Refs. [21, 22]. Our recent work though [12] has demonstrated the existence of what can be interpreted as a conservation law for *any* nonadiabatic trajectory. Here we generalize and advance this result to the ponderomotive dynamics of particles of arbitrary nature.

The purpose of this paper is to obtain the general properties of particle dynamics in the resonance domain, when the conditions (2) do hold, whereas those given by Eqs. (3) and (4) may be violated. We derive the effective ponderomotive potential, which is valid for both adiabatic and strongly nonadiabatic interactions and remains non-singular at the resonance $\omega = \omega_{\nu}$. The possi-

bility to introduce such a potential is due to the conservation of an approximate integral of the Manley-Rowe type [28, 29], for which we suggest both a quantum interpretation and a general "classical" derivation directly from the first principles of Hamiltonian mechanics. We show also that the properties of near-resonant ponderomotive barriers are strikingly different from those expected within the traditional adiabatic model framework. In particular, we suggest how such barriers can be employed to produce one-way walls and even cool resonant species. We also discuss the implications of our results with respect to the stability of particle aperiodic bounce oscillations in ponderomotive barriers. As an example, nonlinear dynamics of atomic clusters in an intense ac field is discussed, and the problem of charged particle motion in a magnetic field under resonant drive is revisited from the standpoint of our new approach.

Although the analysis is performed for classical species, we anticipate that our main results apply also to quantum particles, such as atoms and molecules. If so, the proposed methods could supplement the existing techniques of particle manipulation by laser fields [4–6]. The value added could be large then, as these techniques allow present and potential applications in a wide variety of subjects such as light scattering, cloud physics, quantum optics, isotopes separation, and others [7].

The paper is organized as follows. In Sec. II, we develop a general formulation for particle average dynamics under intense high-frequency radiation. In Sec. III, we study particular aspects of ponderomotive dynamics of a single-mode linear oscillator in an ac field resonant with the particle natural oscillations. In Sec. IV, we consider examples of particles (including those with internal degrees of freedom), to which our results apply. In Sec. V, we summarize the main ideas of the paper. Supplementary calculations are given in Appendixes.

II. EFFECTIVE POTENTIAL AND INTEGRALS OF PARTICLE MOTION

A. Example

Consider an example of a ponderomotive force on an oscillator. Namely, consider a charged particle motion in a dc magnetic field \mathbf{B}_0 under the action of the ac field

$$\mathbf{E} = \operatorname{Re}\left[\mathbf{E}_0 \exp(-i\omega t)\right]. \tag{5}$$

Under the conditions (2)-(4) (assuming $\omega_{\nu} \equiv \Omega$), the particle exhibits adiabatic Larmor rotation at frequency $\Omega = eB_0/mc$ superimposed on the induced oscillations at frequency ω . The average effect of the ac field can then be replaced with the particle interaction with the ponderomotive potential (1), which now takes the form

$$\Phi = \sum_{\nu=0,\pm 1} \frac{e^2 |\vec{E}_{\nu}|^2}{4m\omega(\omega + \nu\Omega)}.$$
 (6)

Here $\hat{E}_{\nu} \equiv \boldsymbol{\xi}_{\nu}^* \cdot \mathbf{E}_0$ are the projections of \mathbf{E}_0 on the polarization vectors

$$\boldsymbol{\xi}_{\pm 1} = (\mathbf{x}^0 \pm i\mathbf{y}^0)/\sqrt{2}, \qquad \boldsymbol{\xi}_0 = \mathbf{z}^0, \tag{7}$$

assuming \mathbf{B}_0 is primarily in the z direction (see Ref. [32] and refs. therein). In addition to the average force $-\partial \Phi/\partial z$ seen by the particle in the direction of its onedimensional guiding-center motion along \mathbf{B}_0 , the particle also experiences the diamagnetic acceleration $-\mu B'_0$, where $\mu_0 = m v_{\rm L}^2/2B_0$. (Here $\mathbf{v}_{\rm L} = \mathbf{v}_{\perp} - \mathbf{v}_{\rm ac}$ is the quiver velocity, additional to the velocity of induced highfrequency oscillations $\mathbf{v}_{\rm ac}$.) Like in the case when the ac field is absent, μ_0 is an adiabatic invariant, which is approximately conserved under the conditions (2)-(4) [32]. In this case, the "quasi-energy" of the particle $\mathfrak{E} = \frac{1}{2} m \langle v_z \rangle^2 + \mu B_0 + \Phi$ is also an adiabatic invariant, hence $\Phi + \mu_0 B$ plays a role of an effective potential.

Should (3)-(4) be violated due to the resonant interaction, both μ_0 and \mathfrak{E} will be subjected to substantial variations; hence the potential approximation no longer holds in this case. Remarkably though, the combination of the two, $\mathfrak{E} - (mc\omega/e)\mu_0$, is conserved, as one can deduce from Refs. [12, 15, 17]. This is a sign that a formulation of the average ponderomotive dynamics must be possible in terms of a *generalized* effective potential even at resonant interactions. Such a formulation can be developed for a Hamiltonian oscillator of an arbitrary nature, as we show below in Sec. II. The implications of these results will be considered in Sec. III and Sec. IV.

B. Drift Lagrangian

A particle exhibiting internal oscillations (e.g., a molecule, an atom, etc) is, generally, a system comprised of $N \geq 1$ elementary constituents (e.g., electrons and a nucleus), interacting both with each other and the ac field. Our purpose is to describe the motion of the particle as a whole, treating the individual constituents motion as its internal degrees of freedom. Accordingly, we will assume that the particle cannot dissociate (i.e. the constituents can only exhibit finite oscillations near the common center of mass) and hence represents a well-defined entity described by 3N independent coordinates.

Let us introduce the particle center-of-mass coordinate ρ and the constituents relative displacements $\mathbf{h}_j \ll L$ (j = 1...N), exhibiting finite oscillatory motion with real frequencies. In the absence of the ac field, the particle center of mass will undergo smooth, non-oscillatory behavior, and hence ρ can be chosen as the coordinate of the particle guiding center. An exception would be a case when the particle as a whole interacts with a dc field, which confines its average motion to a subspace of dimensionality n less than that of the real space. For example, an electrostatic potential could attach the drift motion to a surface (n = 2); or, a strong background magnetic field could keep a charged particle moving along a single field line (n = 1). In all such cases we will assume deviations of the particle center of mass from the guiding center subspace as additional oscillatory degrees of freedom, assuming $n \ge 1$. With that, one could define the guiding center coordinate as $\bar{\rho}$ being the projection of ρ on the subspace of the guiding-center motion.

In the presence of the ac field though, $\bar{\rho}$ will generally oscillate at the frequency of the field ω . Hence, more generally, the guiding center coordinates must be introduced as $\mathbf{r} = \bar{\rho} - \mathbf{q}$, where \mathbf{q} is the *n*-dimensional center-ofmass quiver displacement. As, by definition, no natural frequency is associated with the center-of-mass motion in the subspace contemplated, we will assume that *as a whole* a particle experiences adiabatic oscillations $\mathbf{q}^{(a)}$ under the conditions (2). For given fields, $\mathbf{q}^{(a)}(\mathbf{r}, t)$ would be a known function, and therefore \mathbf{r} can be employed as an independent coordinate.

To derive the equation for $\mathbf{r}(t)$, let us consider the particle action

$$S = \int_{t_1}^{t_2} \mathcal{L} \, dt, \tag{8}$$

where \mathcal{L} is the Lagrangian function, and the time scale $\Delta t = t_2 - t_1$ is large compared to ω^{-1} and ω_{ν}^{-1} . The major contribution to the action S, linear in Δt , is the time-averaged part of the Lagrangian, $\langle \mathcal{L} \rangle$, while the contribution of the oscillatory (with characteristic time scale ω^{-1} , ω_{ν}^{-1}) Lagrangian to the integral (8) remains small. Thus, the action S is approximately given by $S = \int_{t_1}^{t_2} \langle \mathcal{L} \rangle dt$, from where it follows that $\mathcal{L}_{d} \equiv \langle \mathcal{L} \rangle$ can be treated as the Lagrangian of the drift motion.

To calculate \mathcal{L}_d consider the Lagrangian \mathcal{L} as a nonlinear operator on the particle 3N-dimensional dipole moment

$$\psi = (e_1 x_1, e_1 y_1, e_1 z_1 \dots e_N x_N, e_N y_N, e_N z_N), \quad (9)$$

where e_j are the electric charges of particle individual constituents, and (x_j, y_j, z_j) are the components of \mathbf{h}_j . Since h_j are assumed small, let us employ a second-order Taylor expansion for ψ to get

$$\mathcal{L} = \mathcal{L}_0 + \mathcal{L}_\psi + \mathcal{L}_{\text{int}}, \qquad (10a)$$

$$\mathcal{L}_{\psi} = \frac{1}{2} \left(\dot{\psi}, M \dot{\psi} \right) - \left(\dot{\psi}, P \psi \right) - \frac{1}{2} \left(\psi, Q \psi \right),$$
(10b)

$$\mathcal{L}_{\text{int}} = (\psi, \mathcal{F}). \tag{10c}$$

Here the Lagrangian $\mathcal{L}_0(\mathbf{r}, \mathbf{v})$, where $\mathbf{v} = \dot{\mathbf{r}}$, describes the particle motion at $\psi_i \equiv 0$; \mathcal{L}_{ψ} describes free oscillations of the dipole moment ψ ; (\cdot, \cdot) stands for a real dot product in 3N-dimensional space; $\mathcal{L}_{int} = \mathbf{p} \cdot \mathbf{E}$ describes the dipole interaction of internal oscillations with the ac field \mathbf{E} ; \mathbf{p} is the total dipole moment of the particle; M, Q, and P are $K \times K$ real matrices (K = 3N) being functions of \mathbf{r} , with M, Q symmetric and P antisymmetric [30, 31]; $\mathcal{F}(\mathbf{r}, t)$ is a K-dimensional force vector

$$\mathcal{F} = (E_x, E_y, E_z \dots E_x, E_y, E_z), \tag{11}$$

consisting of N identical triplets (E_x, E_y, E_z) standing for the electric field components (Appendix A). The guiding center Lagrangian can then be written as

$$\mathcal{L}_{d} = \mathcal{L}_{0} + \langle \mathcal{L}_{\psi} \rangle + \langle \mathcal{L}_{int} \rangle.$$
 (12)

Below we will show how Eq. (12) can be simplified for different cases of interest, and how the average ponderomotive force can be calculated both for adiabatic and nonadiabatic interactions.

C. Adiabatic Interaction

To calculate $\langle \mathcal{L}_{\psi} \rangle$ and $\langle \mathcal{L}_{int} \rangle$, note that the vector ψ can exhibit both *driven* oscillations at frequency ω and *free* oscillations at eigenfrequencies ω_{ν} (Appendix B). Suppose first that all ω_{ν} remain sufficiently far from ω in the sense of Eqs. (3) and (4), so that all eigenmodes ψ_{ν} can be considered evolving adiabatically. In this case, one can write $\psi = \psi^{(0)} + \psi^{(a)}$, where

$$\psi^{(0)} = \operatorname{Re} \sum_{\nu} \psi^{(0)}_{\nu}, \qquad \psi^{(0)}_{\nu} = \chi^{(0)}_{\nu} \exp(-i\omega_{\nu}t), \quad (13)$$

denotes residual free oscillations due to nonzero $\psi_{\nu}(t = -\infty)$ at particle entrance to the ac field, assuming that summation is taken over modes with non-negative ω_{ν} , and $\chi_{\nu}^{(0)} = 0$ for $\omega_{\nu} = 0$; $\psi^{(a)} = \text{Re}[\chi^{(a)} \exp(-i\omega t)]$ is the adiabatic response governed by the equation

$$\mathcal{D}(\omega)\chi^{(a)} = \tilde{\mathcal{F}},\tag{14}$$

with $\chi^{(a)}(t = -\infty) = 0$; $\hat{\alpha} = \mathcal{D}^{-1}$ is the polarizability tensor in K-dimensional space;

$$\mathcal{D}(\omega) = -M\omega^2 + 2iP\omega + Q \tag{15}$$

is a Hermitian response matrix; $\tilde{\mathcal{F}}$ is the complex amplitude of $\mathcal{F} = \operatorname{Re} \left[\tilde{\mathcal{F}} \exp(-i\omega t) \right]$.

The function $\langle \mathcal{L}_{\psi}[\psi] \rangle$ then equals

$$\langle \mathcal{L}_{\psi}[\psi] \rangle = \langle \mathcal{L}_{\psi}[\psi^{(0)}] \rangle + \langle \mathcal{L}_{\psi}[\psi^{(a)}] \rangle, \qquad (16)$$

where the Lagrangian of free oscillations, $\mathcal{L}_{\psi}[\psi^{(0)}] = \sum_{\nu} \mathcal{L}_{\nu}^{(0)}$, can be expressed as follows. Suppose, that the parameters of \mathcal{L}_{ψ} are allowed to vary as the particle moves, and thus the complex amplitudes $\chi_{\nu}^{(0)}$ are generally not constant. Nonetheless, if the variations are slow enough $(v/\omega_{\nu} \ll L)$, then $\dot{\chi}_{\nu}^{(0)} \ll \omega_{\nu} \chi_{\nu}^{(0)}$, and the following approximation can be employed:

$$\mathcal{L}_{\nu}^{(0)} = \frac{\omega_{\nu}}{4} \left(\psi_{\nu}^{(0)*}, \mathcal{D}'(\omega_{\nu})\psi_{\nu}^{(0)} \right) + \frac{i}{8} \left[\left(\dot{\psi}_{\nu}^{(0)*}, \mathcal{D}'(\omega_{\nu})\psi_{\nu}^{(0)} \right) - \left(\psi_{\nu}^{(0)*}, \mathcal{D}'(\omega_{\nu})\dot{\psi}_{\nu}^{(0)} \right) \right].$$
(17)

The first term in Eq. (17) equals $-J_{\nu}\omega_{\nu}$, where J_{ν} is the action of free oscillations at the ν -th mode (Appendix B),

and the second term can be expressed as $J_{\nu}\dot{\varphi}_{\nu}$, where $-\varphi_{\nu}$ is the phase of ψ_{ν} . Hence, one can write

$$\mathcal{L}_{\nu}^{(0)} = \frac{d}{dt} (J_{\nu}\varphi_{\nu}) - \dot{J}_{\nu}\varphi_{\nu} - J_{\nu}\omega_{\nu}.$$
 (18)

The quantity φ_{ν} can be treated as a new variable, with $J_{\nu} = \partial \mathcal{L}_{\rm d} / \partial \dot{\varphi}_{\nu}$ being the associated canonical momentum. Then, the Lagrangian equation for φ_{ν} is $\dot{J}_{\nu} = 0$, meaning that J_{ν} are conserved for all ν , except for those corresponding to zero ω_{ν} . Employing the conservation of J_{ν} and omitting an unimportant full time derivative, from Eq. (18) one gets $\mathcal{L}_{\nu}^{(0)} = -J_{\nu}\omega_{\nu}$. Adding a constant will neither affect the motion equations, and hence

$$\mathcal{L}_{\psi}[\psi^{(0)}] = \sum_{\nu} {}^{\prime} J_{\nu} \Delta \omega_{\nu}, \qquad (19)$$

where the prime denotes summation only modes with positive ω_{ν} .

The Lagrangian of driven oscillations, $\langle L_{\psi}[\psi^{(a)}]\rangle$, can be calculated straightforwardly and yields

$$\langle L_{\psi} \rangle = -\frac{1}{4} \left(\chi^{(a)*}, \mathcal{D}\chi^{(a)} \right), \qquad (20)$$

where $\mathcal{D} \equiv \mathcal{D}(\omega)$. Employing Eq. (14) one gets also

$$\langle L_{\text{int}} \rangle = \frac{1}{2} \left(\chi^{(a)*}, \mathcal{D}\chi^{(a)} \right).$$
 (21)

Therefore,

$$\mathcal{L}_{\rm d} = \mathcal{L}_0 - \Phi + \sum_{\nu} {}^{\prime} J_{\nu} \Delta \omega_{\nu}, \qquad (22)$$

where $\Phi = \langle L_{\psi} \rangle = -2 \langle L_{\text{int}} \rangle$, or

$$\Phi = -\frac{1}{4} \left(\tilde{\mathcal{F}}^*, \hat{\alpha} \tilde{\mathcal{F}} \right).$$
(23)

By definition, $(\tilde{\mathcal{F}}^*, \hat{\alpha}\tilde{\mathcal{F}}) = \mathbf{E}_0^* \cdot \tilde{\mathbf{p}}$, where $\tilde{\mathbf{p}}$ is the complex amplitude of the particle dipole moment in the real threedimensional space: $\tilde{\mathbf{p}} = \boldsymbol{\alpha} \cdot \mathbf{E}_0$, assuming the ac field of the form (5). Therefore, Φ given by Eq. (23) is the same adiabatic ponderomotive potential as the one introduced by Eq. (1).

The Euler equation yielded by the Lagrangian (22),

$$\frac{d}{dt} \left(\frac{\partial \mathcal{L}_{\mathrm{d}}}{\partial \mathbf{v}} \right) = \frac{\partial \mathcal{L}_{\mathrm{d}}}{\partial \mathbf{r}},\tag{24}$$

would be the equation of adiabatic drift motion. Together with $J_{\nu} = \text{const}$, an integral of such motion (assuming $\partial \mathcal{L}_d / \partial t \equiv 0$) is the Hamiltonian $\mathcal{H}_d = \mathbf{v} \cdot \partial \mathcal{L}_d / \partial \mathbf{v} - \mathcal{L}_d$, or

$$\mathcal{H}_{d} = \mathcal{E}_{d} + \Phi - \sum_{\nu} {}^{\prime} \frac{\Delta \omega_{\nu}}{\omega_{\nu}} \mathcal{E}_{\nu}, \qquad (25)$$

where \mathcal{E}_d stands for the particle kinetic energy (plus the energy of interaction with low-frequency background fields, if any), and $\mathcal{E}_{\nu} = J_{\nu}\omega_{\nu}$ is the energy of *free* oscillations stored in a ν -th mode. In the simplest case when interactions with low-frequency fields (if any) are inessential, Eq. (24) takes the form

$$m\frac{d\mathbf{v}}{dt} = -\nabla\Phi - \sum_{\nu}{}^{\prime}J_{\nu}\nabla\omega_{\nu},\qquad(26)$$

so that

$$\Phi_{\rm eff} = \Phi - \sum_{\nu} {}^{\prime} J_{\nu} \, \Delta \omega_{\nu} \tag{27}$$

plays a role of the effective potential. [The terms $J_{\nu}\omega_{\nu}(\mathbf{r})$ can be omitted for constant $\omega_{\nu}(\mathbf{r})$ at adiabatic interaction, but otherwise result in essential forces analogous, say, to the diamagnetic force on a charged particle in inhomogeneous magnetic field (Sec. IV B).]

D. Near-resonant Interaction

Rewrite Eq. (23) in the eigenmode representation:

$$\Phi = -\frac{1}{4} \sum_{\nu} \frac{|\tilde{\mathcal{F}}_{\nu}|^2}{\mathcal{D}_{\nu}(\omega)},\tag{28}$$

where $\tilde{\mathcal{F}}_{\nu}$ are projections of $\tilde{\mathcal{F}}$ on the eigenvectors χ_{ν} , and \mathcal{D}_{ν} are the eigenvalues of $\mathcal{D} = \hat{\alpha}^{-1}$. By definition, $\mathcal{D}_{\nu}(\omega) \to 0$ as $\Delta \omega_{\nu} \to 0$. Hence, keeping only the leading term with respect to $\Delta \omega_{\nu}^{-1}$, the adiabatic Miller potential equals

$$\Phi = -\frac{|\tilde{\mathcal{F}}_{\nu}|^2}{4\mathcal{D}_{\nu}'(\omega_{\nu})\Delta\omega_{\nu}} = -\frac{|\tilde{E}_{\nu}|^2}{4\Delta\omega_{\nu}} \left[(\alpha_{\nu}^{-1})'(\omega_{\nu}) \right]^{-1}, \quad (29)$$

where we also introduced the corresponding resonant eigenvalue α_{ν} of the particle polarizability $\boldsymbol{\alpha}$, and the projections $\tilde{E}_{\nu} \equiv \boldsymbol{\xi}_{\nu}^{*} \cdot \mathbf{E}_{0}$ of the ac field amplitude on the eigenvectors $\boldsymbol{\xi}_{\nu}$ of the tensor $\boldsymbol{\alpha}$. Note now that at $\Delta \omega_{\nu} \to 0$ one has $|\tilde{\mathcal{F}}_{\nu}|^{2}/4\mathcal{D}_{\nu}(\omega_{\nu}) \approx -J_{\nu}^{(a)}\Delta\omega_{\nu}$, where $J_{\nu}^{(a)}$ is the action corresponding to oscillations at the ν -th mode with the amplitude $\chi_{\nu} = \chi_{\nu}^{(a)}$ (Appendix B). Since $J_{\nu}^{(a)} = \mathcal{E}_{\nu}^{(a)}/\omega_{\nu}$, where $\mathcal{E}_{\nu}^{(a)}$ is, respectively, the energy of near-resonant adiabatic oscillations, one can write

$$\Phi = \frac{\Delta\omega_{\nu}}{\omega_{\nu}} \, \mathcal{E}_{\nu}^{(a)}.\tag{30}$$

For a stable particle $\mathcal{E}_{\nu}^{(a)} > 0$. Hence, close to the resonance, the adiabatic potential is attractive for $\Delta \omega_{\nu} < 0$ and repulsive for $\Delta \omega_{\nu} > 0$. However, since $\mathcal{E}_{\nu}^{(a)}$ itself goes to infinity at $\omega = \omega_{\nu}$, $\Phi(\omega)$ exhibits a singularity at the resonance, as follows from Eq. (29). As the true force exerted by the ac field on a particle is finite, the fact that $\Phi(\omega)$ appears to be a singular function is a clear sign of the adiabatic approximation failure in the resonance region. Let us then derive a more precise expression for \mathcal{L}_{d} to obtain the true, non-singular average force applied to a particle near the resonance.

Consider the general case when for some (possibly more than one) modes the adiabaticity conditions (3), (4) may be violated. To do so, divide ψ into the nonresonant (certainly adiabatic) part $\psi_{\rm nr}$ consisting of modes with $\Delta \omega_{\nu}/\omega \gtrsim 1$ and the remaining part $\psi_{\rm r}$ consisting of those with $\Delta \omega_{\nu}/\omega \ll 1$. Among the latter we might find both adiabatic and nonadiabatic modes, which we will treat equivalently, hence no precise discrimination between the two types of oscillations is required. The drift Lagrangian of the particle can then be written as

$$\mathcal{L}_{\rm d} = \mathcal{L}_0 + \mathcal{L}_{\rm nr} + \mathcal{L}_{\rm r}, \qquad (31)$$

$$\mathcal{L}_{\rm nr} = -\phi_{\rm nr} + \sum_{\nu} {}^{\prime} J_{\nu} \Delta \omega_{\nu}, \qquad (32)$$

where ϕ_{nr} is the part of the adiabatic potential Φ corresponding to the interaction with nonresonant modes, over which also the summation is taken in Eq. (32).

To calculate $\mathcal{L}_{\rm r}$, consider the particle near-resonant response in the form $\psi_{\rm r} = {\rm Re} \left[\chi \exp(-i\omega t) \right]$, where χ is a slow function as compared to the exponent. We can write then $\mathcal{L}_{\rm r} = \mathcal{L}_{\rm r}^{(1)} + \mathcal{L}_{\rm r}^{(2)}$, where

$$\mathcal{L}_{\mathbf{r}}^{(1)} = \frac{1}{2} \operatorname{Re}\left(\chi^*, \mathcal{D}\chi^{(a)}\right) - \frac{1}{4} \left(\chi^*, \mathcal{D}\chi\right), \qquad (33a)$$

$$\mathcal{L}_{\mathrm{r}}^{(2)} = \frac{i}{8} \left[\left(\dot{\chi}^*, \mathcal{D}' \chi \right) - \left(\chi^*, \mathcal{D}' \dot{\chi} \right) \right].$$
(33b)

Here $\mathcal{D} \equiv \mathcal{D}(\omega)$; the adiabatic response $\chi^{(a)}$ is governed by Eq. (14), with only resonant component of $\tilde{\mathcal{F}}$ taken into account, and $\chi^{(a)}(t = -\infty) = 0$. The Lagrangian $\mathcal{L}_{\rm r}^{(1)}$ can be expressed as $\mathcal{L}_{\rm r}^{(1)} = -\phi_{\rm r} + \delta\phi_{\rm r}$, where $\delta\phi_{\rm r}$ is the correction to the adiabatic potential $\phi_{\rm r}$:

$$\delta\phi_{\mathbf{r}} = -\frac{1}{4} \left(\delta\chi^*, \mathcal{D}\,\delta\chi \right) = \sum_{\nu} {}^{\prime} J_{\nu} \Delta\omega_{\nu}. \tag{34}$$

Here the summation is taken over near-resonant modes, $\delta \chi \equiv \chi - \chi^{(a)}$, and

$$J_{\nu} = -\frac{1}{4} \left(\delta \chi_{\nu}^{*}, \mathcal{D}'(\omega_{\nu}) \, \delta \chi_{\nu} \right) \tag{35}$$

is the action of free oscillations at a ν -th mode, $J_{\nu} = \mathcal{E}_{\nu}/\omega_{\nu}$, where \mathcal{E}_{ν} is the energy of these oscillations (Appendix B).

The drift Lagrangian can then be put in the form

$$\mathcal{L}_{\rm d} = \mathcal{L}_0 - \Phi_{\rm eff} + \mathcal{L}_{\rm r}^{(2)}, \qquad (36)$$

with Φ_{eff} given by Eq. (27). The difference from the adiabatic case is that now J_{ν} are not necessarily constant, and Φ_{eff} must generally be considered as a function of χ :

$$\Phi_{\text{eff}} = -\frac{1}{4} \left(\chi^{(a)*}, \mathcal{D}(\omega)\chi^{(a)} \right) + \frac{1}{4} \sum_{\nu} \Delta \omega_{\nu} \left(\delta \chi^{*}_{\nu}, \mathcal{D}'(\omega_{\nu}) \delta \chi_{\nu} \right), \quad (37)$$

where the first term again equals the adiabatic potential Φ . Note that this expression is not obtained by a perturbation method, and the two terms in Eq. (37) are allowed to be of the same order. In particular, near each resonance $\omega = \omega_{\nu}$, the terms quadratic in $\chi_{\nu}^{(a)} \propto \Delta \omega_{\nu}^{-1}$ cancel out, whereas linear terms enter the expression being multiplied by $\Delta \omega_{\nu}$. It means that, unlike $\Phi(\omega)$, the effective potential (37) is a non-singular function.

The quantity χ (as well as χ^*) should now be treated as an independent variable, for which the Lagrangian equation can be derived as follows. Consider the variation of \mathcal{L}_d [Eq. (36)] with respect to χ^*_{ν} :

$$\frac{\delta \mathcal{L}_{\rm d}}{\delta \chi_{\nu}^*} = \frac{\delta}{\delta \chi_{\nu}^*} \left\{ -\frac{1}{4} \Delta \omega_{\nu} \left(\delta \chi_{\nu}^*, \mathcal{D}' \, \delta \chi_{\nu} \right) + \frac{i}{8} \left[\left(\dot{\chi}_{\nu}^*, \mathcal{D}' \chi_{\nu} \right) - \left(\chi_{\nu}^*, \mathcal{D}' \dot{\chi}_{\nu} \right) \right] \right\}, \quad (38)$$

where we used $\delta \mathcal{L}_0 / \delta \chi_{\nu}^* \equiv 0$, $\delta \Phi / \delta \chi_{\nu}^* \equiv 0$, $\mathcal{D}'(\omega_{\nu}) \approx \mathcal{D}'(\omega) \equiv \mathcal{D}'$, and the $\delta \chi_{\nu}$ and $\delta \psi$, and the terms corresponding to the ν -th mode contribute to Eq. (38). Since both $\chi_{\nu}^{(a)}$ and $\chi_{\nu}^{(a)*}$ are independent of χ_{ν}^* , Eq. (38) can be rewritten as follows:

$$\frac{\delta \mathcal{L}_{\rm d}}{\delta \chi_{\nu}^*} = -\frac{i}{4} \left[\mathcal{D}' \dot{\chi}_{\nu} + \frac{1}{2} \dot{\mathcal{D}}' \chi_{\nu} - i \Delta \omega_{\nu} \, \mathcal{D}' \delta \chi_{\nu} \right] + \frac{d}{dt} \left[\frac{i}{8} \frac{\delta}{\delta \chi_{\nu}^*} \left(\chi_{\nu}^*, \mathcal{D}' \chi_{\nu} \right) \right]. \quad (39)$$

The second term represents a full time derivative and hence can be neglected. Then to obtain the Euler equation $\delta \mathcal{L}_{\rm d} / \delta \chi^*_{\nu} = 0$, one must require that the first term in Eq. (39) equals zero, which yields

$$\dot{\chi}_{\nu} + \tau_{\nu}^{-1} \chi_{\nu} = i \Delta \omega_{\nu} \, \delta \chi_{\nu}, \tag{40}$$

where $\tau_{\nu}^{-1} = \frac{1}{2} (\mathcal{D}')^{-1} \dot{\mathcal{D}}'$. While Eq. (40) is, strictly speaking, derived for modes with $\Delta \omega_{\nu} \ll \omega$, it can formally be applied to any mode with nonzero ω_{ν} . Even in this case, Eq. (40) properly describes adiabatic evolution of free oscillations and predicts that at large $\Delta \omega_{\nu}$ the amplitude of induced oscillations approaches $\chi_{\nu}^{(a)}$.

The guiding-center equation of motion readily follows [see Eq. (24)] from the drift Lagrangian, which we can finally put in the following form:

$$\mathcal{L}_{\rm d} = \mathcal{L}_{\rm d} + \mathcal{L}_{\chi},\tag{41a}$$

$$\bar{\mathcal{L}}_{d} = \mathcal{L}_{0}(\mathbf{r}, \mathbf{v}) - \Phi_{\text{eff}}(\mathbf{r}, \chi), \qquad (41b)$$

$$\mathcal{L}_{\chi} = \frac{\imath}{8} \sum_{\nu} \left[\left(\dot{\chi}_{\nu}^{*}, \mathcal{D}'(\omega_{\nu})\chi_{\nu} \right) - \left(\chi_{\nu}^{*}, \mathcal{D}'(\omega_{\nu})\dot{\chi}_{\nu} \right) \right].$$
(41c)

While J_{ν} corresponding to nonresonant modes are integrals of such motion (assuming $\partial \mathcal{L}_{\rm d}/\partial t \equiv 0$), those of resonant modes are not conserved, and hence a particle can generally exhibit stochastic behavior. Surprisingly though, one more independent integral can be identified in this case. Employing Eq. (24) together with Eq. (40), one can prove by direct calculation that the full time derivative of $\mathcal{H}_{\rm d} = \mathbf{v} \cdot \partial \bar{\mathcal{L}}_{\rm d} / \partial \mathbf{v} - \bar{\mathcal{L}}_{\rm d}$ equals zero. The approximate integral

$$\mathcal{H}_{\rm d} = \mathcal{E}_{\rm d} + \Phi_{\rm eff} = \text{const},\tag{42}$$

coincides with that given by Eq. (25) and is conserved under the limitations (2) only, regardless of conditions (3) and (4).

Like in the adiabatic case, at nonadiabatic interaction the function Φ_{eff} can also be considered as the effective potential seen by the particle. Directly from Eq. (42) it follows that

$$m\frac{dv}{dt} = -\frac{d\Phi_{\text{eff}}}{ds},\tag{43}$$

assuming that $\mathcal{E}_{d} = \frac{1}{2}mv^{2}$, and ds = v dt is a length element along the particle trajectory. In the simplest case when no background fields are present in the system, it is also convenient to employ the actual Lagrangian equation (24), which now takes an "intuitive" form

$$m \frac{d\mathbf{v}}{dt} = -\nabla \Phi_{\text{eff}}.$$
(44)

Note that the gradient in Eq. (44) applies to both terms of Eq. (37), assuming $\delta \chi_{\nu} = \chi_{\nu} - \chi_{\nu}^{(a)}(\mathbf{r})$, where χ_{ν} is an independent variable.

Despite of deceptive similarity in form with a truly conservative force, near-resonant ponderomotive acceleration is not reversible. Only *n* of the particle 3N degrees of freedom are governed by Eq. (44). The remaining 3N - n degrees of freedom, described by Eq. (40), are "frozen" at adiabatic interaction, but otherwise are indispensable and can be viewed as hidden variables of the particle guiding center, should the latter be considered as a "black box". Also associated with the particle average motion are a complex phase arg χ and the natural "location uncertainty" $\lambda = v/\min |\Delta \omega_{\nu}|$, which makes the guiding center similar to a quantum object. As shown in Ref. [11], the quantum analogy can be elaborated upon further and can even be made quantitative.

E. Hamiltonian Interpretation and Quantum Analogy

The conservation law (42) can also be explained alternatively, by employing basic principles of Hamiltonian dynamics. To show that, note that J_{ν} are the canonical momenta corresponding to angle variables $\varphi_{\nu}, \dot{\varphi}_{\nu} \equiv \omega_{\nu}$. For clarity, label these modes (with nonzero ω_{ν}) with indexes $\nu = 2 \dots q$ and introduce the quantity

$$J_1 = -\frac{1}{\omega} \left(\mathcal{E}_{\mathrm{d}} + \Phi + \sum_{\nu=2}^q J_\nu \omega_\nu \right), \qquad (45)$$

which is the action variable corresponding to a generalized coordinate φ_1 oscillating at frequency $\omega_1 = \omega$ (see, e.g., Ref. [32]). At adiabatic interaction, when all ω_{ν} are well separated, each of J_{ν} will represent an invariant, which results in conservation of \mathcal{H}_d [Eq. (25)] being a combination of J_{ν} . On the other hand, at nonadiabatic interaction, when, say, $\omega_1 \dots \omega_s$ are close to each other, $J_{\nu \leq s}$ will not be conserved individually. In this case though, the *sum* of the resonant modes actions $I_s = \sum_{\nu=1}^s J_{\nu}$ will be an adiabatic invariant (Appendix C). Since the action of each nonresonant mode $J_{\nu>s}$ remains constant in any case, adding $J_{\nu>s}$ to I_s will not violate the conservation law. Hence, the integral can also be expressed as $\sum_{\nu=1}^q J_{\nu} = \text{const, or}$

$$-\frac{1}{\omega}\left(\mathcal{E}_{d} + \Phi + \sum_{\nu}' J_{\nu}\omega_{\nu}\right) + \sum_{\nu}' J_{\nu} = \text{const}, \quad (46)$$

where now *any* of the modes may be resonant with the ac field. The obtained equation is clearly equivalent to the above-predicted conservation of \mathcal{H}_d given by Eq. (25), both in adiabatic and nonadiabatic regimes.

Note that the conservation law (46) follows also from a quantum-mechanical argument, if one recalls that J_{ν} are proportional to the number of quanta N_{ν} in corresponding modes, $N_{\nu} = J_{\nu}/\hbar$. Consider the total energy of the system \mathcal{E}_{Σ} , which includes the energy of the "dressed" particle average motion $\mathcal{E}_{d} + \Phi$ [33–37], the energy of internal oscillations $\sum_{\nu} N_{\nu} \hbar \omega_{\nu}$, and the energy of the ac field $N_{f}\hbar\omega$. As \mathcal{E}_{Σ} is conserved, one can write that

$$\mathcal{E}_{\rm d} + \Phi - \sum_{\nu} N_{\nu} \hbar \Delta \omega_{\nu} + N_0 \hbar \omega = \text{const}, \qquad (47)$$

where the first three terms constitute \mathcal{H}_{d} , and $N_{0} = N_{f} + \sum_{\nu} N_{\nu}$ is the total number of quanta. Since N_{0} is conserved as the particle absorbs or emits photons (which is what constitutes nonadiabaticity in classical terms), then Eq. (47) yields $\mathcal{H}_{d} = \text{const}$, in agreement with the above results.

One can see then that the conservation law for \mathcal{H}_d is of the same type as Manley-Rowe relations, which are similar classical manifestations of inherently quantum interactions between resonant oscillators [28]. (One can show also that such relations originate from specific Noether symmetry of resonant interactions [17, 29].) Like those, the new integral allows to conclude upon global stability of particle oscillations and obtain other results of interest. In Sec. III, we will consider some applications of this conservation law on the simplest example of a particle with a single natural mode ν . More complicated dynamics of particles with richer eigenspectrum can be approached analogously.

III. SINGLE-MODE OSCILLATOR IN RESONANT FIELD

For a particle with a single natural mode ν , the conservation law (42) takes the form

$$\mathcal{E}_{\rm d} + \Phi - \frac{\Delta \omega_{\nu}}{\omega_{\nu}} \, \mathcal{E}_{\nu} = \mathcal{H}_{\rm d},\tag{48}$$

with \mathcal{H}_d being a constant determined by initial conditions. At adiabatic interaction, when all of the conditions (2)-(4) are satisfied, $J_{\nu} = \mathcal{E}_{\nu}/\omega_{\nu}$ represents an invariant, so that (48) yields an expression for the drift velocity at each **r**. This fact allows to integrate the particle equation of motion and actually find the dependence $\mathbf{r}(t)$ (at least, in quadratures and for one-dimensional drift). At nonadiabatic interaction though, J_{ν} varies in time, and particle sees the effective potential

$$\Phi_{\rm eff} = \frac{\Delta\omega_{\nu}}{\omega_{\nu}} \Big[\mathcal{E}_{\nu}^{(a)} - \mathcal{E}_{\nu} \Big], \tag{49}$$

with $\mathcal{E}_{\nu}(t)$ essentially being an unknown function [cf. Eqs. (27) and (30)]. Hence construction of the analytic solution is generally impossible in this case. Nonetheless, applying the $\mathcal{H}_{\rm d}$ conservation together with Eq. (49) renders important information on the properties of particle dynamics in the resonance domain. Some of these properties are described below.

A. Bounce Oscillations in Nonadiabatic Barriers

The conservation of \mathcal{H}_d allows to determine the stability conditions for particle bounce oscillations in ponderomotive barriers. Suppose, for example, that $\Delta \omega_{\nu} < 0$, in which case $\Phi < 0$ [cf. Eq. (30)]. Conservation of \mathcal{H}_d requires then that $\mathcal{E}_{d} \leq \mathcal{H}_{d} + |\Phi|$ for all t. Hence a particle cannot leave the field if $\mathcal{H}_{d} < 0$, as otherwise it must arrive in the region $\Phi(z) = 0$ having $\mathcal{E}_{d} \leq \mathcal{H}_{d} < 0$. Thus, at negative \mathcal{H}_d , even aperiodic bounce oscillations remain stable and a particle remains trapped by an attractive ponderomotive potential. (Instability of such oscillations can only result from dissipative effects like spontaneous emission [33, 38], not considered here.) Unstable oscillations develop otherwise and result in particle escaping from the interaction region. Examples of trapped and untrapped particle trajectories are depicted in Fig. 1, which exhibits the agreement between the sign of \mathcal{H}_d and particle confinement.

B. Ponderomotive Cooling

Breaking the adiabaticity allows irreversible energy exchange between particles and the ac field. If the radiation is redshifted from the resonance frequency ω_{ν} , thermal particles lose their drift energy as they scatter off a nonadiabatic ponderomotive barrier, regardless of their actual trajectories. Should particle natural oscillations thermalize between consecutive interactions with the field, the effect can be employed for cooling particles.

The idea can be explained as follows. When a free particle scatters off a ponderomotive barrier $[\Phi(\pm\infty) = 0]$, the conservation of \mathcal{H}_d requires that the overall changes of the guiding center kinetic energy \mathcal{E}_d and the internal oscillations energy \mathcal{E}_{ν} are bound to each other:

$$\Delta \mathcal{E}_{\rm d} = \Delta \left[\left(\frac{\omega}{\omega_{\nu}} - 1 \right) \mathcal{E}_{\nu} \right]. \tag{50}$$

If ω_{ν} is constant, Eq. (50) reads $\Delta \mathcal{E}_{\rm d} = (\omega/\omega_{\nu} - 1) \Delta \mathcal{E}_{\nu}$, so for given $\Delta \omega_{\nu}$, the sign of $\Delta \mathcal{E}_{\rm d}$ is determined by the sign of $\Delta \mathcal{E}_{\nu}$. Let us show that $\Delta \mathcal{E}_{\nu} > 0$ for moderate $\eta = T_0/\Phi_{\rm max}$, where T_0 is the initial temperature. As follows from Eq. (40), $\Delta \mathcal{E}_{\nu} \sim \Lambda \Phi_{\rm max}$, where [39]

$$\Lambda = |1 - \omega_{\nu}/\omega|^{-1} \gg 1.$$
(51)

If $\Lambda \gg \eta$, then $\Delta \mathcal{E}_{\nu} \gg T_0$, hence $\Delta \mathcal{E}_{\nu} \approx \mathcal{E}_{\nu}(+\infty) > 0$ for all particles, regardless of the initial value of \mathcal{E}_{ν} . As a result, if $\omega < \omega_{\nu}$, one has $\Delta \mathcal{E}_{d} < 0$, so that *all* thermal particles are decelerated [Fig. 3(a)].

Suppose now that each particle encounters the field repeatedly, and the time between consecutive encounters exceeds the relaxation time of particle natural oscillations. At each impact, the particle will lose about $|\omega/\omega_{\nu} - 1| \Delta \mathcal{E}_{\nu} \sim |\Phi|_{\text{max}}$ of its drift energy \mathcal{E}_{d} , and yet get to the next encounter with negligible \mathcal{E}_{ν} , as compared to $\Lambda |\Phi|_{\text{max}}$. Then, after about η interactions, each particle will be cooled down to $\mathcal{E}_{d} \sim |\Phi|_{\text{max}} \ll T_{0}$, assuming $\eta \gg 1$. At lower energies though, new effects come into play, and further cooling slows down significantly, as we explain below.

C. Dynamic Trapping

What impedes cooling below the limit $\mathcal{E}_{d} \sim |\Phi|_{max}$ is the dynamic trapping of particles by the ac field. It is possible that, due to nonadiabatic deceleration in an attractive ponderomotive potential, a particle can lose all of its kinetic energy \mathcal{E}_d even before leaving the interaction region. In this case, an initially free particle will be bounced back by the decelerating slope of the wave barrier, and hence will remain inside the potential well at least for one bounce oscillation (Fig. 2). More oscillations may also follow after that; yet, because of the phase space conservation requirement, particles may not stay trapped forever. Rather, only dynamic (i.e. temporary) trapping is possible in this case. On the other hand, if the number of bounce oscillations within a potential well is large, the post-trapping dynamics of a particle correlates little with its pre-trapping dynamics. Hence the direction, to which the particle is released, is almost uncorrelated with the initial velocity, and a particle can randomly escape either forward or backward with respect to the direction



FIG. 1: Longitudinal velocity v_z vs guiding center location z for a charged particle in the ac field with the spatial profile $\mathbf{E}_0(z) = \mathbf{x}^0 \bar{E}_0 \exp(-z^2/2L^2)$ imposed over uniform dc magnetic field $\mathbf{B}_0 = \mathbf{z}^0 B_0$. Here v_z is measured in units $\hat{v} = (e|\bar{E}_0|/m\omega)\sqrt{\Lambda} \sim (|\Phi|_{\max}/m)^{1/2}$; z is measured in units $\hat{z} = c/\omega$; $e|\bar{E}_0|/mc\omega = 0.001$; $\Lambda = |1 - \Omega/\omega|^{-1} = 100$; $\Omega = eB_0/mc$; $L = 0.4\hat{z}$; $\mathcal{E}_{\nu}(t=0) = 0$. (a) $v_0 = 0.47\hat{v}$, that is, $\mathcal{H}_d < 0$ (b) $v_0 = 0.53\hat{v}$, that is, $\mathcal{H}_d > 0$; $v_0 \equiv v_z(t=0)$. The bifurcation from stable to unstable bounce oscillations is analytically predicted at $|v_0| = \frac{1}{2}\hat{v}$, that is, $\mathcal{H}_d = 0$.



FIG. 2: Longitudinal velocity v_z vs z for a particle being trapped and released by an attractive ponderomotive potential in a dc magnetic field (same notation and parameters as in Fig. 1; $L = \lambda(\hat{v})/\hat{\epsilon}$; $\hat{\epsilon} = 3$): $v_z = 0.30 \hat{v}$ (black) and $v_z = 0.31 \hat{v}$ (gray).

of its original drift (Fig. 2). The overall scattering is then stochastic, and may lead to both transmission or reflection of incident particles.

To derive the condition, under which these effects become possible, suppose for simplicity that $\omega_{\nu} = \text{const.}$ Also introduce the particle drift displacement on the beat period $\lambda(v) = v/|\Delta\omega_{\nu}|$, the characteristic velocity change of the particle as it encounters the ac field $\hat{v} \sim (|\Phi|_{\max}/m)^{1/2}$, and the dimensionless parameter $\hat{\epsilon} = \lambda(\hat{v})/L$. Slow particles with initial velocity $v_0 \ll \hat{v}$ are accelerated ponderomotively up to the velocity of the order of \hat{v} inside the barrier. If \hat{v} itself is large enough $(\hat{\epsilon} \gtrsim 1)$, nonadiabatic effects have to reveal for all, even initially slow particles, some of which may then experience trapping. On the contrary, at $\hat{\epsilon} \ll 1$, slow particles remain adiabatic and hence cannot be trapped. As for fast particles ($v_0 \gg \hat{v}$), in both cases they have enough energy to overcome the ponderomotive deceleration and avoid trapping. Therefore, if $\hat{\epsilon} \gtrsim 1$, at sufficiently small v_0 a particle may be trapped within a potential well, but if $\hat{\epsilon} \lesssim 1$, trapping is impossible regardless of v_0 .

To illustrate these conditions, consider a Gaussian field $\mathbf{E}_0(z) = \mathbf{x}^0 \bar{E}_0 \exp(-z^2/2L^2)$ applied to a particle traveling in a uniform magnetic field $\mathbf{B}_0 = \mathbf{z}^0 B_0$ (see also Sec. IV B). The energy change of a particle as it goes through the ponderomotive barrier can be estimated under the assumption $v_z \approx v_0 = \text{const.}$ In this case, as follows from Eq. (40), $\Delta \mathcal{E}_{\nu} = -2\pi |\Phi|_{\text{max}} f(\epsilon_0^{-2})$, where $|\Phi|_{\text{max}} = \frac{1}{8} m \hat{v}^2$, $\hat{v} = (e|\bar{E}_0|/m\omega) \sqrt{\Lambda}$, $f(x) = xe^{-x}$, and $\epsilon_0 = \lambda(v_0)/L$. Assuming that $v \approx \text{const}$ remains a good approximation also for $u = v_0/\hat{v} \sim 1$, we can expect then that a particle having

$$\frac{u^2}{2} < \frac{\pi}{4} f\left[(\hat{\epsilon}u)^{-2}\right]$$
(52)

must be released from the interaction region with negative \mathcal{E}_d , which is impossible. Therefore, for u satisfying the condition (52), the approximation $v_z \approx \text{const}$ is strongly violated. It is a sign that a turning point appears on the particle trajectory, which means that the particle gets trapped in the potential well. Trapping remains possible for $\hat{\epsilon}$, at which Eq. (52) has a real solution for u. In compliance with the general condition derived above, such solution exists only for $\hat{\epsilon} > 1.08$, as can be shown numerically. These analytic predictions are confirmed with high accuracy in our numerical calculations, as depicted in Fig. 3.

It is clear now why the cooling mechanism described in Sec. III B cannot be efficient at $\mathcal{E}_{d} \ll \Phi_{\max}$, that is, at $v_0 \ll \hat{v}$. For a barrier with $\hat{\epsilon} \ll 1$ [Fig. 3(a)], the condition $v_0 \ll \hat{v}$ guarantees adiabatic dynamics. Correspondingly, the energy exchange between particles and the ac field will be exponentially small with respect to ϵ_0 , hence substantial cooling will be possible only on exponentially large time scales. On the other hand, at $\hat{\epsilon} \gtrsim 1$ [Fig. 3(b)], slow particles will get trapped by the ac field, and the characteristic trapping time increases with decrease of v_0 . Indeed, a released particle must have kinetic energy \mathcal{E}_d satisfying $0 < \mathcal{E}_d < \mathcal{H}_d = \frac{1}{2} m v_0^2$. As v_0 approaches zero, this interval shrinks, and it becomes less probable for a particle to escape the interaction region. Therefore, at $\mathcal{E}_d \ll \Phi_{\text{max}}$ all particles will eventually get trapped by the ac field, hence further cooling, as described in Sec. III B, will become impossible. On the other hand, specific properties of nonadiabatic ponderomotive barriers allow other cooling mechanisms operating also at $v_0 \ll \hat{v}$ via employing one-way walls, which we will discuss in Sec. III D.

D. One-way Walls

Internal oscillations at $\omega_{\nu} \approx \omega$ essentially decrease the potential seen by the particle by the factor γ : $\Phi_{\text{eff}} = \gamma \Phi$, where $\gamma = 1 - \mathcal{E}_{\nu}/\mathcal{E}_{\nu}^{(a)} < 1$ [cf. Eq. (49)]. Remarkably, $\gamma = \gamma(\mathcal{E}_{\nu})$ can be made different for different particle trajectories even at the same **r**. In particular, it means that particles incident on a nonadiabatic ponderomotive barrier from opposite directions can be arranged to see different Φ_{eff} . As a result, a barrier can become asymmetric and operate as a one-way wall.

Various techniques to produce one-way walls on the base of this principle (or what effectively amounts to it) have been proposed recently in rf frequency range for electrons and ions [12–14] and optical frequency range for atoms [40, 41]. To illustrate how the idea can be employed on practice in the simplest case $\Delta \omega_{\nu} = \text{const}$, let us consider the field configuration depicted in Fig. 4(a), assuming that it is encountered by particles with initially zero \mathcal{E}_{ν} [13]. (As explained in Sec. III B, the presence of nonzero $\mathcal{E}_{\nu} \sim \mathcal{E}_{d}$ can be neglected.) Assume also that $L_1 \gg \lambda \gg L_2$ and, at first, consider a particle incident on the ponderomotive barrier from the right. At its entrance to the ac field, such a particle will not have enough time to establish adiabatic oscillations. As it passes the vanishingly narrow right slope of the barrier, it will still have $\chi \approx 0$, or $\delta \chi_{\nu} \approx -\chi_{\nu}^{(a)}$. Hence, at z = 0 the particle will see

$$\Phi_{\text{eff}}(z=0) \approx \Phi_{\text{eff}}(z=L_2 \to 0) = 0, \qquad (53)$$

in compliance with the fact that Φ_{eff} must be a continuous function. As z is changing from 0 to $-\infty$, the effective potential can only decrease, as both J_{ν} and $\Delta \omega_{\nu}$ remain constant on the adiabatic left slope, whereas Φ gradually changes from its maximum value to zero. A particle incident from the right then sees an *attractive* potential $\Phi_{\text{eff}} \approx \Phi - \Phi_{\text{max}} < 0$ [Fig. 4(b)] and eventually gains energy

$$\Delta \mathcal{E}_{d} = \Phi_{\max}, \qquad \Delta \mathcal{E}_{\nu} = \Lambda \Phi_{\max}. \tag{54}$$

A particle incident from the left, however, sees an adiabatic *repulsive* potential $\Phi_{\text{eff}} \approx \Phi > 0$. Assuming that Φ_{max} is large enough, this particle will never get to the nonadiabatic region $0 < z < L_2$; it will rather be reflected by the adiabatic slope and leave the interaction region with zero $\Delta \mathcal{E}_d$ and $\Delta \mathcal{E}_{\nu}$. It is then clear that the contemplated ponderomotive barrier is asymmetric and acts essentially like a Maxwell demon, except that it increases the energy of transiting particles, as required by laws of thermodynamics [12, 42].

One-way walls of the described type might find numerous applications employing selective manipulations with plasma particles. For example, in Refs. [12–14], it was proposed how the contemplated Maxwell demon effect (MDE) can be used for driving electric current in magnetized plasmas. (The flexibility in rearranging phase space makes these techniques at least as efficient, and in some regimes more efficient, as the conventional current drive techniques [43].) Mechanisms of cooling particles below $\Phi_{\rm max}$ by what effectively amounts to ponderomotive one-way walls have also been proposed recently [40, 44]. In addition, one can also imagine how the sensitivity of MDE with respect to particle resonance properties could help in separating plasma constituents (including isotope separation), and how asymmetry of nonadiabatic barriers in general could be employed for enhancing plasma confinement of mirror traps and other applications [15].

If successful also on neutral particles, MDE could supplement the existing techniques of manipulating atoms by means of laser fields [4-6], which broke important ground in atomic physics. Similar capabilities apply also for other small neutral objects ranging from molecules to micron-sized particles and permit one to selectively and stably trap particles, levitate them against gravity, channel particles along laser beams and use them as sensitive probes for measuring optical, electric, magnetic, viscous drag, and gravity forces [7, 45–48]. These lightpressure techniques allow present and potential applications in a wide variety of subjects such as light scattering, cloud physics, quantum optics, and high-resolution spectroscopy [7]. If these techniques could be additive with those proposed in this paper, the value added could be large.

IV. CLASSICAL PARTICLES EXHIBITING NATURAL OSCILLATIONS

Let us now consider the actual examples of particles (including those with internal degrees of freedom), to which the above results can be applied. The purpose of this section is to derive, within the framework of our new approach, the expressions for the Miller potentials Φ for two types of such particles, to obtain expressions for the corresponding effective potentials Φ_{eff} , and to show how particular results of our previous works [12, 32] follow from the general consideration proposed above.



FIG. 3: Change of the drift energy $\Delta \mathcal{E}_{d}$ of a particle traveling in a dc magnetic field as it scatters off a nonadiabatic attractive ponderomotive barrier: numerical result (dots) and analytic prediction (solid). Same notation and parameters as in Fig. 2, energy is measured in units $m\hat{v}^2$: (a) $\hat{\epsilon} = 0.5$ (no trapping); (b) $\hat{\epsilon} = 3.3$ (shaded is the region of particle trapping as established numerically). For reference to compare with the analytically predicted trapping condition (52), shown also is a graph $\frac{1}{2} m v_0^2$ (dashed).



FIG. 4: (a) high-frequency field profile producing a one-way wall: $L_1 \gg \lambda \gg L_2$, $\lambda = v_0/|\Delta \omega_{\nu}|$; (b) effective potentials for particles incident from the left (positive repulsive potential; particles are reflected) and from the right (negative attractive potential; particles are transmitted).

A. Atomic Cluster

Consider first an atomic cluster, i.e. a compound particle containing electron gas, which can oscillate in the attractive Coulomb field of the ion core. Since under an ac drive the electron cloud oscillates as a whole (assuming cold electrons), it can be treated as a single constituent of the "macro-particle". Hence, counting also the ion core as one, we have N = 2. Since the guiding center motion is three-dimensional (n = 3), there can be K - n = 3independent modes (K = 3N), each corresponding to some non-negative frequency ω_{ν} , plus K - 3 = 3 modes with zero eigenfrequencies, corresponding to the centerof-mass oscillations.

To obtain the eigenfrequencies ω_{ν} , it is sufficient to find poles of the particle polarizability $\boldsymbol{\alpha}$. This can be done as follows. Assume that collisional heating of the cluster is insignificant during the time when the ac field is on, and the size of the cluster is small compared to the radiation wavelength. Then an adequate model for a cluster would be a polarizable sphere characterized by a real dielectric constant ϵ and a fixed radius R [49, 50]. In this case $\boldsymbol{\alpha}$ is a diagonal tensor, $\boldsymbol{\alpha} = \alpha \mathbf{I}$, with all three eigenvalues α_{ν} equal to

$$\alpha = R^3 \frac{\epsilon - 1}{\epsilon + 2},\tag{55}$$

so that ω_{ν} must satisfy the dispersion equation $\epsilon(\omega_{\nu}) = -2$. The dielectric constant ϵ equals that of a plasma, $\epsilon(\omega) = 1 - \omega_p^2 / \omega^2$, where ω_p is the plasma frequency of the electron gas inside the cluster [49]. Hence, the (Fröhlich) resonance takes place at $\omega_{\nu} = \omega_p / \sqrt{3}$, whereas the coefficient is modified as the shape of a cluster deviates from spherical [51, 52].

The Miller potential (1) then takes the form

$$\Phi = \frac{1}{4} |E_0|^2 R^3 \frac{\omega_p^2}{3\omega^2 - \omega_p^2},\tag{56}$$

and the effective ponderomotive potential equals

$$\Phi_{\rm eff} = \Phi + \left(1 - \sqrt{3} \,\frac{\omega}{\omega_p}\right) \mathcal{E}_p,\tag{57}$$

where \mathcal{E}_p is the energy of free electron (Langmuir) oscillations inside the cluster. At adiabatic interaction, \mathcal{E}_p is conserved (since $J_{\nu} = \text{const}$, and ω_p is constant by definition), and thus $\Phi_{\text{eff}} = \Phi + \text{const.}$ Hence, the quasienergy $\mathfrak{E} = \mathcal{E}_d + \Phi$, where $\mathcal{E}_d = \frac{1}{2} m v^2$, will represent an integral of particle motion in this case. A more general integral, which is conserved even when the cluster interacts resonantly with the ac drive, would be the quantity $\mathcal{H}_d = \mathcal{E}_d + \Phi_{\text{eff}}$. Even when the conservation of \mathcal{E}_d is violated, the conservation of \mathcal{H}_d still bounds the change of the particle drift energy to the change of its "internal" energy at $t \to \infty$:

$$\Delta \mathcal{E}_{\rm d} = \left(\sqrt{3}\,\frac{\omega}{\omega_p} - 1\right) \Delta \mathcal{E}_p,\tag{58}$$

as follows from Eq. (50).

B. Particle in a Magnetic Field

Return now to an elementary charged particle (electron or ion) in a high-frequency field (5) in the presence of a dc magnetic field $\mathbf{B}_0 = \nabla \times \mathbf{A}_0$. Employ a linear approximation for the vector potential $\mathbf{A}_0(\mathbf{r}) = \frac{1}{2} B_0(z) (\mathbf{z}^0 \times \mathbf{r})$ with respect to the particle displacement $\mathbf{z}^0 \times \mathbf{r}$ from the location of the guiding center, assuming that B_0 is a slow function of z. Assuming also that the gyrofrequency $\Omega = eB_0/mc$ is comparable or larger than ω , we must treat the guiding center motion as one-dimensional (n = 1). Hence there can be at most two "internal" eigenmodes with distinct non-negative frequencies ω_{ν} (plus, the same number of modes with $-\omega_{\nu}$). To find those, one can either start with the actual expression for \mathcal{L}_{ψ} (see Ref. [15]), or, more easily, employ the already known expression for the polarizability tensor [32]

$$\boldsymbol{\alpha} = -\frac{e^2}{m\omega^2} \begin{pmatrix} \frac{1}{1-b^2} & \frac{ib}{1-b^2} & 0\\ \frac{-ib}{1-b^2} & \frac{1}{1-b^2} & 0\\ 0 & 0 & 1 \end{pmatrix}, \quad (59)$$

where $b = \Omega/\omega$. From Eq. (59), it is seen that $\boldsymbol{\alpha}$ exhibits singularities at the cyclotron resonance $\omega = \pm \Omega$, which gives us one of non-negative eigenfrequencies ω_{ν} . The remaining ω_{ν} are apparently zero, which can also be proved directly by solving the full characteristic equation $\mathcal{D}\psi = 0$, as shown in Ref. [15].

To calculate the Miller potential, it is convenient to introduce the complex amplitudes of the ac field $\tilde{E}_{\nu} \equiv \boldsymbol{\xi}_{\nu}^* \cdot \mathbf{E}_0$ in the new basis formed by the eigenvectors $\boldsymbol{\xi}_{\nu}$ of the polarizability tensor:

$$\boldsymbol{\xi}_{\pm 1} = \frac{\mathbf{x}^0 \pm i\mathbf{y}^0}{\sqrt{2}}, \qquad \alpha_{\pm 1} = -\frac{e^2}{m\omega(\omega \pm \Omega)}, \qquad (60a)$$

$$\boldsymbol{\xi}_0 = \mathbf{z}^0, \qquad \qquad \alpha_0 = -\frac{e^2}{m\omega^2}, \qquad (60b)$$

where α_{ν} are the corresponding eigenvalues of $\boldsymbol{\alpha}$. The adiabatic ponderomotive potential $\Phi = -\frac{1}{4} \sum_{\nu} \alpha_{\nu} |\tilde{E}_{\nu}|^2$ [Eq. (23)] then is given by (6), and the effective potential can be expressed as $\Phi_{\text{eff}} = \Phi - J(\omega - \Omega)$, where J is the

action variable (35) corresponding to free natural oscillations at frequency Ω : $J = mv_{\rm L}^2/2\Omega$. (Here $\mathbf{v}_{\rm L} = \mathbf{v}_{\perp} - \mathbf{v}_{\rm ac}$ is the quiver velocity, additional to the velocity of induced high-frequency oscillations $\mathbf{v}_{\rm ac}$.) Then, introducing the particle magnetic moment $\mu = mv_{\rm L}^2/2B_0$, one gets

$$\Phi_{\rm eff} = \Phi + \mu (B_0 - B_{\rm res}), \tag{61}$$

where $B_{\rm res} = mc\omega/e$ is the magnetic field strength, at which a particle would be in exact cyclotron resonance with the ac drive at frequency ω . In the adiabatic limit, when $\mu = (e/mc)J$ is conserved, one has $\Phi_{\rm eff} = \Phi + \mu B_0 + {\rm const.}$ Hence, the quasi-energy $\mathfrak{E} = \mathcal{E}_{\rm d} + \mu B_0 + \Phi$, where $\mathcal{E}_{\rm d} = \frac{1}{2} m \langle v_z \rangle^2$, will represent an integral of the particle motion in this case (cf. Ref. [32]). A more general integral, which is conserved even when a particle interacts resonantly with the ac drive, would be the quantity $\mathcal{H}_{\rm d} = \mathcal{E}_{\rm d} + \Phi_{\rm eff}$. Even when the μ conservation is violated, the conservation of $\mathcal{H}_{\rm d}$ still bounds the change of the particle drift energy to the change of the magnetic moment at $t \to \infty$:

$$\Delta \mathcal{E}_{\rm d} = \Delta [\mu (B_{\rm res} - B_0)], \tag{62}$$

as follows from Eq. (50), in full agreement with the result we obtained in Ref. [12] by straightforward averaging of particle the motion equations.

V. CONCLUSIONS

In this paper, we generalize the ponderomotive formulation to particles (including those with internal degrees of freedom) moving under the action of a high-frequency field, which may interact resonantly with natural particle oscillations. The effective ponderomotive potential is derived for both adiabatic and strongly nonadiabatic interactions and remains non-singular even at resonant drive. The possibility to introduce such a potential is due to the conservation of an approximate integral of the Manley-Rowe type, for which we suggest a natural quantum interpretation. We show that the properties of near-resonant wave barriers are strikingly different from those expected from the traditional adiabatic model. On one hand, nonadiabatic ponderomotive potentials can repel or attract particles in measurably predictable ways, in which sense they can operates just like normal potentials. On the other hand though, nonadiabatic potentials are more flexible as tools for controlling particle motion. They are not limited by the requirement of conservativeness, hence allowing more freedom in manipulating particles, which can be either charged plasma particles, or neutral particles, such as atomic clusters and others. We show that, as a result, nonadiabatic ponderomotive barriers can be arranged into stable traps for classical particles, produce cooling effect, and generate one-way walls for resonant species.

Although the analysis is performed for classical species, we anticipate that our main results can be extrapolated also on quantum particles, such as atoms and molecules. If so, the proposed methods could supplement the existing techniques of particle manipulation by laser fields. The value added could be large then, as these techniques allow present and potential applications in a wide variety of subjects such as quantum optics, isotopes separation, high-resolution spectroscopy, and others.

VI. ACKNOWLEDGEMENTS

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APPENDIX A: COMPOUND PARTICLE LAGRANGIAN

Consider the Lagrangian of a compound particle consisting of $N \ge 1$ individual elementary particles:

$$\mathcal{L} = \sum_{j} \left[\frac{1}{2} m_{j} v_{j}^{2} - e_{j} \phi(\mathbf{r}_{j}) + \frac{e_{j}}{c} \mathbf{v}_{j} \cdot \mathbf{A}(\mathbf{r}_{j}) - \frac{1}{2} \sum_{i \neq j} U_{ij} \left(|\mathbf{r}_{j} - \mathbf{r}_{i}| \right) \right]. \quad (A1)$$

Here m_j , e_j , \mathbf{r}_j , and \mathbf{v}_j are, respectively, the mass, the charge, the location, and the velocity of a *j*-th particle; $\phi = \phi_{\rm ac} + \phi_{\rm bg}$ and $\mathbf{A} = \mathbf{A}_{\rm ac} + \mathbf{A}_{\rm bg}$ are the scalar and the vector potentials, which determine the ac and the low-frequency background fields correspondingly; *c* is the speed of light; $U_{ij}(|\mathbf{r}_j - \mathbf{r}_i|)$ is the energy of interaction between particles labeled with indexes *j* and *i*.

Assuming that $|\mathbf{r}_j - \mathbf{r}| \ll L$, where L is the minimum scale of the fields, and \mathbf{r} is the guiding-center location, one can rewrite Eq. (A1) as

$$\mathcal{L} = \mathcal{L}_0 + \mathcal{L}_{\sim} + \sum_j \mathcal{L}_j.$$
 (A2)

Here \mathcal{L}_0 is the Lagrangian of the guiding center motion as it would be without excitation of particle internal degrees of freedom:

$$\mathcal{L}_0 = \frac{1}{2} m v^2 - e \,\phi_{\rm bg}(\mathbf{r}) + \frac{e}{c} \,\mathbf{v} \cdot \mathbf{A}_{\rm bg}(\mathbf{r}), \tag{A3}$$

where $m = \sum_{j} m_{j}$, $e = \sum_{j} e_{j}$, and $\mathbf{v} = \dot{\mathbf{r}}$. [Note though that, in the presence of a dc magnetic field \mathbf{B}_{bg} when the particle gyrofrequency $\Omega = eB_{bg}/mc$ is of the order of ω , the proposed model captures the guiding center motion only along a single field line. In this case, the component of \mathbf{v} transverse to $\mathbf{B}_{bg} = \nabla \times \mathbf{A}_{bg}$ should be taken equal to zero by definition (see Sec. II B and IV B for details)]. The term

$$\mathcal{L}_{\sim} = \sum_{j} m_{j} \mathbf{v}_{j} \cdot \mathbf{v} - e \,\phi_{\rm ac}(\mathbf{r}) + \frac{e}{c} \,\mathbf{v} \cdot \mathbf{A}_{\rm ac}(\mathbf{r}) \qquad (A4)$$

is approximately a full time derivative, and can be omitted as insignificant. The Lagrangians \mathcal{L}_j are those of elementary particles relative motion. Assuming that U_{ij} give rise to linear oscillations, we model them with quadratic functions of $\mathbf{r}_j - \mathbf{r}_i$, and thus, when elaborated to the second order in $\mathbf{h}_j = \mathbf{r}_j - \mathbf{r}$, \mathcal{L}_j can be put in the form

$$\mathcal{L}_j = \mathcal{L}_j^{(0)} + \mathcal{L}_j^{(\text{int})}, \tag{A5a}$$

$$\mathcal{L}_{j}^{(0)} = \frac{1}{2} m_{j} \dot{h}_{j}^{2} + \frac{e_{j}}{c} \dot{\mathbf{h}}_{j} \cdot \left[(\mathbf{h}_{j} \cdot \nabla) \mathbf{A}_{\mathrm{bg}}(\mathbf{r}) \right] - \frac{1}{2} \sum_{i \neq j} \mathbf{h}_{i} \cdot \mathbf{U}_{ij} \cdot \mathbf{h}_{j}, \qquad (A5b)$$

$$\mathcal{L}_j^{(\text{int})} = e_j \mathbf{h}_j \cdot \mathbf{E}', \tag{A5c}$$

where \mathbf{E}' is the external field in the guiding center rest frame:

$$\mathbf{E}' = \mathbf{E} + \frac{1}{c} \mathbf{v} \times \mathbf{B}.$$
 (A6)

The field \mathbf{E}' consists of both the ac component $\mathbf{E}'_{\rm ac}$ and the low-frequency background component $\mathbf{E}'_{\rm bg}$. For an ac field, the amplitudes of the electric and the magnetic fields are connected with each other: $E_{\rm ac} \sim (L\omega/c)B_{\rm ac}$, as follows from the Faraday's law. Since $v/c \ll 1$, one can usually neglect the term $\frac{1}{c}\mathbf{v} \times \mathbf{B}_{\rm ac}$ in comparison with $\mathbf{E}_{\rm ac}$, and thus, $\mathbf{E}'_{\rm ac} \approx \mathbf{E}_{\rm ac}$. In turn, $\mathbf{E}'_{\rm bg} = \mathbf{E}_{\rm bg}$ both in the absence of $\mathbf{B}_{\rm bg}$ and in the opposite case when $\mathbf{B}_{\rm bg}$ is sufficiently strong, so that one must consider \mathbf{v} as parallel to \mathbf{B} (see above). We will therefore assume for simplicity that $\mathbf{E}' = \mathbf{E}$, while a more general case can also be contemplated.

With the above formulas, the Lagrangian \mathcal{L} can be finally put in the form (10a), with

$$\mathcal{L}_{\psi} = \sum_{j} \left\{ \frac{m_{j}}{2e_{j}^{2}} \dot{\mathbf{p}}_{j}^{2} + \frac{1}{ce_{j}} \dot{\mathbf{p}}_{j} \cdot \left[(\mathbf{p}_{j} \cdot \nabla) \mathbf{A}_{\text{bg}}(\mathbf{r}) \right] - \sum_{i \neq j} \frac{1}{2e_{i}e_{j}} \mathbf{p}_{i} \cdot \mathbf{U}_{ij} \cdot \mathbf{p}_{j} \right\}, \quad (A7a)$$

$$\mathcal{L}_{\text{int}} = \sum_{j} \mathbf{p}_{j} \cdot \mathbf{E}(\mathbf{r}), \qquad (A7b)$$

where $\mathbf{p}_j = e_j \mathbf{r}_j$ stand for the dipole moments of the particle individual constituents, and $\mathbf{p} = \sum_j \mathbf{p}_j$ is the total dipole moment of the particle.

APPENDIX B: PROPERTIES OF GENERALIZED LINEAR OSCILLATORS

The Lagrangian

$$\mathcal{L}_{\psi} = \frac{1}{2} \left(\dot{\psi}, M \dot{\psi} \right) - \left(\dot{\psi}, P \psi \right) - \frac{1}{2} \left(\psi, Q \psi \right)$$
(B1)

describes free linear oscillations in coordinates ψ and yields a Euler equation $\hat{\mathcal{D}}\psi = 0$, where

$$\hat{\mathcal{D}} = M \, \frac{d^2}{dt^2} - 2P \, \frac{d}{dt} + Q. \tag{B2}$$

The eigenmodes $\psi_{\nu} = \chi_{\nu} \exp(-i\omega_{\nu}t)$ are then defined by the characteristic equation $\mathcal{D}(\omega_{\nu})\chi_{\nu} = 0$, where

$$\mathcal{D}(\omega) = -M\omega^2 + 2R\omega + Q, \tag{B3}$$

and R = iP. We will assume that all ω_{ν} are real, for which we will require that M, Q, and P are $K \times K$ real matrices, with M, Q symmetric and non-negatively defined, P antisymmetric, and R imaginary Hermitian correspondingly [30, 31].

Under these conditions, the eigenmodes are orthogonal with weight function. To show that, note first that for any $\psi_{\nu} = \chi_{\nu} \exp(-i\omega_{\nu}t)$ the function $\psi_{-\nu} = \chi_{\nu}^* \exp(i\omega_{\nu}t)$ will also be an eigenmode [31]. We thus can represent a *real* function ψ in the form $\psi = \sum_{\nu} \chi_{\nu} \exp(-i\omega_{\nu}t)$, where summation is taken over all ν , both positive and negative. Consider then a real expression $(\dot{\psi}, P\psi)$, which can be put in two alternative complex forms: first, as

$$(\dot{\psi}, P\psi) = \sum_{\nu,\mu} \omega_{\mu}(\chi^*_{\mu}, R\chi_{\nu}) \exp[i(\omega_{\mu} - \omega_{\nu})t], \quad (B4)$$

and second, after taking a complex conjugate and exchanging the dummy indexes, as

$$(\dot{\psi}, P\psi) = \sum_{\nu,\mu} \omega_{\nu}(\chi_{\mu}^*, R\chi_{\nu}) \exp[i(\omega_{\mu} - \omega_{\nu})t], \quad (B5)$$

where we employed the fact that $R^{\dagger} = R$. By subtracting Eq. (B4) from Eq. (B5) and noting that the difference between the two must remain zero at all t, one gets $(\omega_{\mu} - \omega_{\nu})(\chi_{\mu}^{*}, R\chi_{\nu}) = 0$, meaning that $(\chi_{\mu}^{*}, R\chi_{\nu}) =$ $\delta_{\mu\nu}R_{\nu}$. Using this result together with the characteristic equation and $M^{\dagger} = M$, one can show also that $(\chi_{\mu}^{*}, M\chi_{\nu}) = \delta_{\mu\nu}M_{\nu}$, and $(\chi_{\mu}^{*}, Q\chi_{\nu}) = \delta_{\mu\nu}Q_{\nu}$.

Since of the 2K vectors ψ_{ν} the first K modes are complex conjugate to the remaining ones, it is convenient to consider ψ in the form

$$\psi = \operatorname{Re} \sum_{\nu} \chi_{\nu} \exp(-i\omega_{\nu} t).$$
(B6)

The Lagrangian function (B1) can then be expressed as $\mathcal{L}_{\psi} = \sum_{\nu} \mathcal{L}_{\nu}$, where summation is taken over half of modes, i.e. those with non-negative ω_{ν} , and

$$\mathcal{L}_{\nu} = \frac{1}{4} \left(\dot{\psi}_{\nu}^{*}, M \dot{\psi}_{\nu} \right) - \frac{1}{4} \left(\dot{\psi}_{\nu}^{*}, P \psi_{\nu} \right) + \frac{1}{4} \left(\psi_{\nu}^{*}, P \dot{\psi}_{\nu} \right) - \frac{1}{4} \left(\psi_{\nu}^{*}, Q \psi_{\nu} \right).$$
(B7)

Consider also the energy $\mathcal{H}_{\psi} = \mathcal{P}_{\psi}\dot{\psi} - \mathcal{L}_{\psi}$, where $\mathcal{P}_{\psi} = \partial \mathcal{L}_{\psi} / \partial \dot{\psi}$ is the canonical momentum associated with ψ :

$$\mathcal{H}_{\psi} = \frac{1}{2} \left(\dot{\psi}, M \dot{\psi} \right) + \frac{1}{2} \left(\psi, Q \psi \right). \tag{B8}$$

Like the Lagrangian, \mathcal{H}_{ψ} can also be written as a sum over individual modes with non-negative ω_{ν} : $\mathcal{H}_{\psi} = \sum_{\nu} \mathcal{H}_{\nu}$, where

$$\mathcal{H}_{\nu} = \frac{1}{4} \left(\dot{\psi}_{\nu}^{*}, M \dot{\psi}_{\nu} \right) + \frac{1}{4} \left(\psi_{\nu}^{*}, Q \psi_{\nu} \right). \tag{B9}$$

In turn, \mathcal{H}_{ν} can be expressed in terms of the ν -th action variable

$$J_{\nu} = \frac{1}{2\pi} \oint \mathcal{P}_{\nu} \, d\psi_{\nu}, \qquad (B10)$$

which remains an adiabatic invariant as the parameters of the system experience slow variations [53]. Since

$$J_{\nu} = -\frac{1}{4} \left(\chi_{\nu}^*, \mathcal{D}'(\omega_{\nu}) \chi_{\nu} \right), \tag{B11}$$

where $\mathcal{D}'(\omega)$ is the derivative of (B3), the energy of a ν th mode equals $\mathcal{H}_{\nu} = J_{\nu}\omega_{\nu}$; hence the associated phase variable φ_{ν} oscillates in time at frequency $\partial \mathcal{H}_{\nu}/\partial J_{\nu} = \omega_{\nu}$.

APPENDIX C: ADIABATIC INVARIANTS FOR RESONANT OSCILLATORS

Consider a system governed by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_0(\mathbf{J}, t) + \epsilon \mathcal{H}_{\sim}(\mathbf{J}, \boldsymbol{\varphi}, t), \tag{C1}$$

where $\mathbf{J} = (J_1 \dots J_s)$ denote action variables, $\boldsymbol{\varphi} =$ $(\varphi_1 \dots \varphi_s)$ denote angle variables, t is time, $\epsilon \ll 1$ is a small parameter, and \mathcal{H}_{\sim} is the perturbation Hamiltonian. If all frequencies $\omega_i = \partial \mathcal{H}_0 / \partial J_i$ are aliquant and ϵ is sufficiently small, the system exhibits regular dynamics, with the image point moving in phase space along an s-dimensional invariant torus provided by the conservation of all J_i [54]. Suppose though that the first p frequencies ω_i are close to each other $(|\omega_i - \omega_j| \ll \omega_i)$, in which case even a small perturbation can destroy the invariant torus and resonantly drive the system away from its unperturbed trajectory. The overall deviation will then depend on the tensor $g_{ij} = \partial \omega_i / \partial J_j$, and in general can be arbitrarily large. Let us show that even in this case the system will nonetheless conserve an adiabatic invariant being a combination of $J_1 \ldots J_p$.

Consider a canonical transformation $(\mathbf{J}, \boldsymbol{\varphi}) \rightarrow (\mathbf{I}, \boldsymbol{\theta})$, where

$$\theta_{i < p} = \varphi_i - \bar{\varphi}, \qquad \theta_p = \bar{\varphi}, \qquad \theta_{i > p} = \varphi_i \qquad (C2)$$

are the new angle variables, with $\bar{\varphi}$ being the average over φ_i :

$$\bar{\varphi} = \frac{1}{p} \sum_{i=1}^{p} \varphi_i, \tag{C3}$$

and where the new action variables are derived as $I_i = \partial F / \partial \varphi_i$ from the generating function

$$F(\boldsymbol{\varphi}, \mathbf{I}) = \sum_{i=1}^{p-1} (\varphi_i - \bar{\varphi}) I_i + \bar{\varphi} I_p + \sum_{i=p+1}^{s} \varphi_i I_i, \quad (C4)$$

so that

$$J_{i < p} = I_i + \frac{I_p}{p} - \frac{1}{p} \sum_{j=1}^{p-1} I_j, \qquad (C5a)$$

$$J_p = \frac{I_p}{p} - \frac{1}{p} \sum_{j=1}^{p-1} I_j,$$
 (C5b)

$$J_{i>p} = I_i. \tag{C5c}$$

In terms of the new variables, the Hamiltonian of the system is given by

$$\mathcal{H}' = \mathcal{H}_0(\mathbf{I}, t) + \epsilon \mathcal{H}_{\sim}(\mathbf{I}, \boldsymbol{\theta}, t), \qquad (C6)$$

where the frequencies $\dot{\theta}_i = \omega'_i, \, \omega'_i = \partial \mathcal{H}' / \partial I_i$, equal

$$\omega'_{i < p} = \omega_i - \bar{\omega}, \qquad \omega'_p = \bar{\omega}, \qquad \omega'_{i > p} = \omega_i, \qquad (C7)$$

and where

$$\bar{\omega} = \frac{1}{p} \sum_{i=1}^{p} \omega_i \tag{C8}$$

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is the average frequency of resonant oscillators. Since $\omega'_{i < p}$ are small, the oscillations in variables (θ_i, I_i) (i < p) may be resonant, and thus $I_{i < p}$ may be subjected to substantial variations. On the contrary, the frequency ω'_p is large compared to $\omega'_{i < p}$. Since by the initial assumption it also remains far from $\omega_{i > p}$, the corresponding action I_p is approximately conserved throughout the interaction. The value of the new adiabatic invariant can be found by taking a sum of Eqs. (C5) over $i = 1 \dots p$, which yields

$$I_p = \sum_{i=1}^p J_i.$$
 (C9)

A similar argument can also be applied to the remaining $q \equiv p-1$ resonant oscillators, with frequencies inside the characteristic interval $\Delta \omega$ around the new average frequency $\bar{\omega}' = \sum_{i=1}^{q} \omega'_i/q$. If $\epsilon \ll \Delta \omega/\bar{\omega}$, the new action $I'_q = \sum_{i=1}^{q} I_i$ will also be an adiabatic invariant. Extending the technique even further, one can say then that at least $k + 1 \leq p$ independent integrals exist if, roughly, $\epsilon \ll (\Delta \omega/\bar{\omega})^k$.

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