

## Self-excitation of Rydberg atoms at a metal surface

Bordo, Vladimir

*Published in:*  
Physical Review A (Atomic, Molecular and Optical Physics)

*DOI:*  
[10.1103/PhysRevA.96.023834](https://doi.org/10.1103/PhysRevA.96.023834)

*Publication date:*  
2017

*Document version*  
Final published version

*Citation for pulished version (APA):*  
Bordo, V. (2017). Self-excitation of Rydberg atoms at a metal surface. *Physical Review A (Atomic, Molecular and Optical Physics)*, 96(2), [023834]. <https://doi.org/10.1103/PhysRevA.96.023834>

### Terms of use

This work is brought to you by the University of Southern Denmark through the SDU Research Portal. Unless otherwise specified it has been shared according to the terms for self-archiving. If no other license is stated, these terms apply:

- You may download this work for personal use only.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying this open access version

If you believe that this document breaches copyright please contact us providing details and we will investigate your claim. Please direct all enquiries to [puresupport@bib.sdu.dk](mailto:puresupport@bib.sdu.dk)

**Self-excitation of Rydberg atoms at a metal surface**

V. G. Bordo\*

*NanoSyd, Mads Clausen Institute, Syddansk Universitet, Alsion 2, DK-6400 Sønderborg, Denmark*

(Received 19 May 2017; revised manuscript received 7 July 2017; published 14 August 2017)

The novel effect of self-excitation of an atomic beam propagating above a metal surface is predicted and a theory is developed. Its underlying mechanism is positive feedback provided by the reflective surface for the atomic polarization. Under certain conditions the atomic beam flying in the near field of the metal surface acts as an active device that supports sustained atomic dipole oscillations, which generate, in their turn, an electromagnetic field. This phenomenon does not exploit stimulated emission and therefore does not require population inversion in atoms. An experiment with Rydberg atoms in which this effect should be most pronounced is proposed and the necessary estimates are given.

DOI: [10.1103/PhysRevA.96.023834](https://doi.org/10.1103/PhysRevA.96.023834)**I. INTRODUCTION**

The behavior of atoms and molecules in the vicinity of surfaces or cavities has recently received renewed significant interest due to its importance for control of individual photons and for applications in quantum technologies [1]. This field is a subject of cavity quantum electrodynamics (QED) [2,3] which takes its origin from Purcell's work [4]. Purcell predicted that the spontaneous emission rate of a two-level emitter located in a cavity can be essentially enhanced in comparison with that of an emitter located in free space. This effect can be described in terms of the local density of states, which is determined by the emitter environment [5]. The underlying reason for it is the backaction of the emitted radiation reflected from the cavity walls onto the emitter itself. In the simplest case, where an atom emits radiation in the near field of a reflective surface, the relaxation rate oscillates around its free-space value as a function of the atom-surface distance that was demonstrated in the experiment by Drexhage and coauthors [6]. Despite the quantum-mechanical nature of an atomic emitter, this effect can be well described in the framework of a classical oscillating dipole model as done by Chance, Prock, and Silbey [7]. The emission rate oscillations in such a system reflect the variation of the backaction phase with the distance from the surface. Within some intervals of distances the relaxation rate is enhanced compared with the free-space value, i.e., the backaction provides a negative feedback for the dipole oscillation. On the contrary, within the other intervals the relaxation rate is inhibited, which implies a positive feedback from the reflected field. This problem was discussed originally by Sommerfeld in his 1909 paper, where he calculated the power needed by a dipole antenna above the Earth's surface to radiate radio waves [8,9].

If one turns to the case where a confined ensemble of dipoles oscillates in the vicinity of a surface, the backaction is proportional to the number of dipoles. Then, for a large enough number of emitters, the positive feedback can prevail over the relaxation, which leads to a "negative damping" and self-excitation (self-oscillation) of the ensemble [10]. This effect emerges already in the generalization of the Chance, Prock, and Silbey model to an ensemble of dipoles and can be

rigorously obtained with the use of Green's function [11,12]. A self-oscillating ensemble of dipoles excites, in its turn, the electromagnetic modes of the host cavity without exploiting stimulated emission, which can be used for lasing without inversion in specially designed nanowires [11] or generation of surface plasmon polaritons (SPPs) in metallic nanocavities [12,13].

Despite its fundamental importance and simplicity, the considered effect, to the best of our knowledge, has so far neither been observed nor even been discussed in the literature on cavity QED. In this paper, we consider an atomic beam which propagates at a certain distance above a metal surface and develop the theory of its self-excitation. We analyze the conditions under which the atomic beam becomes self-excited and generates an electromagnetic field. We also propose an experiment with Rydberg atoms in which this phenomenon is expected to be most pronounced due to very large transition dipole moments. This configuration resembles in some respects a Rydberg-atom maser [14–16], however, unlike a maser, the atoms initially have a dipole moment and do not have a population inversion. The effect under consideration is due to the distance-sensitive near-field atom-surface interaction and is principally different from all kinds of lasing without inversion discussed in the literature [17–20].

The paper is organized as follows. Section II introduces the basic equations of the theoretical formalism and their solutions for both transient and steady-state regimes as well as the criterion for the atomic beam self-excitation. In Sec. III the dependence of the gain coefficient on different parameters is investigated. Section IV presents some numerical results and estimates which illustrate the theory. The main results of the paper are summarized in Sec. V.

**II. THEORETICAL FORMALISM**

The setup for the proposed experiment is shown in Fig. 1. We assume that an atomic beam initially excited to the  $nL$  state is polarized by a microwave field resonant to the transition  $nL \rightarrow n'L'$  in the cavity. We assume that the intensity of this field is well below the transition saturation intensity so that the population of the  $n'L'$  state is negligible. The atomic beam then propagates at distance  $d$  above the metal surface and its direction is taken as the  $x$  axis.

\*bordo@mci.sdu.dk

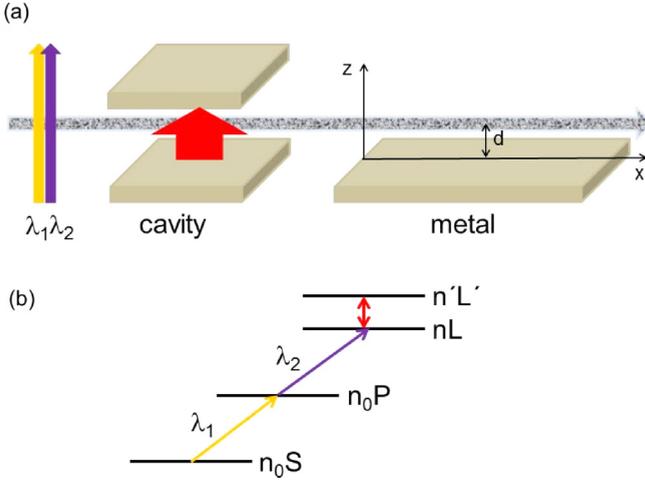


FIG. 1. (a) Proposed experimental setup. The atomic beam propagating along the  $x$  axis is initially excited by two lasers and then by a microwave field in the cavity. The self-excitation process occurs above the metal surface. (b) Atom energy levels scheme, which shows different excitation steps. The Rydberg states between which the transition is self-excited are denoted  $nL$  and  $n'L'$ .

It should be noted that the atom-surface interaction scales as  $1/d^3$  and so does the nonradiative relaxation rate [21,22]. The typical relaxation rate of an electronic excitation of an atom adsorbed at a metal surface ( $d \sim 0.5$  nm) is of the order of  $10^{15} \text{ s}^{-1}$ . In our further discussion we consider the range of distances of the order of the Rydberg transition wavelength,  $d \sim 0.1$  cm, for which the nonradiative relaxation rate is reduced to about  $10^{-4} \text{ s}^{-1}$  and can be neglected in comparison with the radiative relaxation rate. Therefore the atoms are injected into the space above the metal surface with nonzero transition dipole moments if they transverse the distance between the cavity and the surface for times shorter than the radiative relaxation time.

### A. Basic equations

Under the above conditions, the polarized atoms which fly above the metal surface can be considered as the sources of the dipole radiation which, being reflected by the surface, determines the near field of atoms. This field can be written in the form

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}^{(-)}(\mathbf{r}, t)e^{i\omega t} + \mathbf{E}^{(+)}(\mathbf{r}, t)e^{-i\omega t}, \quad (1)$$

$$\mathbf{E}^{(-)}(\mathbf{r}, t) = [\mathbf{E}^{(+)}(\mathbf{r}, t)]^*, \quad (2)$$

where  $\mathbf{E}^{(\pm)}(\mathbf{r}, t)$  are the slowly varying positive- and negative-frequency parts. The amplitude  $\mathbf{E}^{(+)}(\mathbf{r}, t)$  results from the cooperative emission of all atomic dipoles and can be found in terms of the individual dipole amplitudes,  $\mathbf{p}_j^{(+)}(\mathbf{r}_j, t)$ , as [12]

$$\begin{aligned} \mathbf{E}^{(+)}(\mathbf{r}, t) &\approx \sum_j \bar{\mathbf{F}}(\mathbf{r}, \mathbf{r}_j; \omega) \mathbf{p}_j^{(+)}(\mathbf{r}_j, t) \\ &\approx \rho \int \bar{\mathbf{F}}(\mathbf{r}, \mathbf{r}'; \omega) \mathbf{p}^{(+)}(\mathbf{r}', t) dx', \end{aligned} \quad (3)$$

where we have approximated the sum over atoms by the integral over the atomic beam length and introduced the linear number density of atoms,  $\rho$ . Here the quantity  $\rho \mathbf{p}^{(+)}(\mathbf{r}, t)$  is understood as the linear density of the expectation value of the dipole moment distributed over the beam. This approach corresponds to the *polarium* model, which was introduced by Prasad and Glauber [23] to describe a cooperative emission of resonant atoms. The tensor  $\bar{\mathbf{F}}(\mathbf{r}, \mathbf{r}'; \omega)$  is the so-called field susceptibility, which relates the electric field at point  $\mathbf{r}$  generated by a classical dipole, oscillating at frequency  $\omega$ , with the dipole moment itself, located at  $\mathbf{r}'$  [24]. The quantity  $\bar{\mathbf{F}}(\mathbf{r}, \mathbf{r}'; \omega)$  can be decomposed into two parts, one of them originating from the direct dipole field and the other from the field reflected from the metal surface. The first part leads to a contribution to the Lorentz local field in the beam and to the renormalization of the atomic resonance frequencies, which is not important for our discussion. Therefore in what follows we imply that  $\bar{\mathbf{F}}(\mathbf{r}, \mathbf{r}'; \omega)$  is due to the reflected field only.

The explicit form of the tensor  $\bar{\mathbf{F}}(\mathbf{r}, \mathbf{r}'; \omega)$  is found in Refs. [24,25], where one should substitute  $y = y' = 0$  and  $z = z' = d$  for the atoms in the beam. Its components can be written in the form

$$F_{\alpha\beta}(x, x'; \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathcal{F}_{\alpha\beta}(\kappa_x; \omega) e^{i\kappa_x(x-x')} d\kappa_x, \quad (4)$$

where, in particular,

$$\mathcal{F}_{yy}(\kappa_x; \omega) = i\tilde{\omega}^2 \int_{-\infty}^{\infty} \frac{d\kappa_y}{W_0} R_s(\kappa) e^{i2W_0d} \quad (5)$$

and

$$\mathcal{F}_{zz}(\kappa_x; \omega) = i \int_{-\infty}^{\infty} \frac{d\kappa_y}{W_0} \kappa^2 R_p(\kappa) e^{i2W_0d}. \quad (6)$$

Here  $\kappa = (\kappa_x^2 + \kappa_y^2)^{1/2}$ ,  $\tilde{\omega} = \omega/c$ , with  $c$  being the speed of light in vacuum,  $W_0 = (\tilde{\omega}^2 - \kappa^2)^{1/2}$ , and  $R_s$  and  $R_p$  are the Fresnel reflection coefficients for  $s$ - and  $p$ -polarized light, respectively.

The atoms in the beam are modeled by a two-level system with two Rydberg states,  $|1\rangle = |nL\rangle$  and  $|2\rangle = |n'L'\rangle$ , the transition frequency  $\omega_0$ , and the transition dipole moment  $\mu_{12}$ . Assuming that the frequency detuning  $\Delta = \omega - \omega_0$  between the electromagnetic field and the Rydberg atom is small [26] and applying the rotating-wave approximation, one comes to the optical Bloch equations [27,28]

$$\begin{aligned} \dot{\mathbf{p}}^{(+)}(x, t) &= -(\gamma_{\perp} - i\Delta) \mathbf{p}^{(+)}(x, t) \\ &\quad - \frac{i}{\hbar} |\mu_{12}|^2 w(x, t) \mathbf{E}^{(+)}(x, t), \end{aligned} \quad (7)$$

$$\begin{aligned} \dot{w}(x, t) &= -\gamma_{\parallel} [w(x, t) - w_0] + \frac{2i}{\hbar} [\mathbf{p}^{(-)}(x, t) \mathbf{E}^{(+)}(x, t) \\ &\quad - \mathbf{p}^{(+)}(x, t) \mathbf{E}^{(-)}(x, t)]. \end{aligned} \quad (8)$$

Here  $w = n_2 - n_1$  is the population inversion and  $w_0 < 0$  is its equilibrium value in the absence of the microwave field, and  $\gamma_{\perp}$  and  $\gamma_{\parallel}$  are the radiative transverse and longitudinal relaxation rates, respectively. A dot above a symbol denotes the full time derivative, i.e.,  $d/dt = \partial/\partial t + v\partial/\partial x$ , with  $v$  being the atom velocity in the beam. Equations (7) and (8) should be complemented by the initial conditions at the moment  $t = 0$

when the atom begins to interact with the metal surface at  $x = 0$ ,  $\mathbf{p}^{(+)}(0,0) = \mathbf{p}_0$ , and  $w(0,0) = w_0$ , where we assume that the microwave field in the cavity is so weak that it does not create a significant population of the upper Rydberg state.

First, we consider the initial stage of the atom evolution when it begins to interact with the surface and its population inversion does not differ significantly from the equilibrium value. Representing the atomic dipole moment in terms of its Fourier transform,

$$\mathbf{p}^{(+)}(x,t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \vec{\mathcal{P}}^{(+)}(\kappa_x,t) e^{i\kappa_x x} d\kappa_x, \quad (9)$$

one finds from Eqs. (3) and (4) the Fourier transform of the near field

$$\vec{\mathcal{E}}^{(+)}(\kappa_x,t) = \rho \vec{\mathcal{F}}(\kappa_x;\omega) \vec{\mathcal{P}}^{(+)}(\kappa_x,t). \quad (10)$$

Introducing the Laplace transform in time,

$$\vec{\pi}^{(+)}(\kappa_x,s) = \int_0^{\infty} \vec{\mathcal{P}}^{(+)}(\kappa_x,t) e^{-st} dt, \quad (11)$$

and taking into account Eq. (10), one obtains from Eq. (7)

$$\pi_{\alpha}^{(+)}(\kappa_x,s) = \frac{2\pi p_0 \delta(\kappa_x)}{s - s_{\alpha}(\kappa_x)}, \quad \alpha = y,z, \quad (12)$$

where

$$s_{\alpha}(\kappa_x) = -\gamma_{\perp} + i\Delta - i\kappa_x v - \frac{i}{\hbar} |\mu_{12}|^2 \rho w_0 \mathcal{F}_{\alpha\alpha}(\kappa_x; \omega) \quad (13)$$

and  $\delta(\kappa_x)$  is the Dirac delta function. We assume here that the initial atomic dipole moment can be oriented along either the  $y$  or the  $z$  axis, which is determined by the direction of the field in the microwave cavity.

Now, performing the inverse Laplace and Fourier transforms, one finds the evolution of the atom dipole moment in the beam above the metal surface:

$$p_{\alpha}^{(+)}(x,t) = p_0 e^{s_{\alpha}(0)t} = p_0 e^{s_{\alpha}(0)x/v}. \quad (14)$$

Accordingly, the field amplitude along the beam is found from Eqs. (10) and (14) as follows:

$$E_{\alpha}(x) = \rho \mathcal{F}_{\alpha\alpha}(0;\omega) p_0 e^{s_{\alpha}(0)x/v}. \quad (15)$$

The reflection coefficient  $R_p(\kappa_y)$ , which determines the integrand of the quantity  $\mathcal{F}_{zz}(0;\omega)$ , has two poles on the real axis at  $\kappa_y = \pm\kappa_0$ , where

$$\kappa_0 \equiv \tilde{\omega} \sqrt{\frac{|\epsilon_m|}{|\epsilon_m| - 1}} > \tilde{\omega}, \quad (16)$$

with  $\epsilon_m < 0$  being the dielectric function of the metal. The contributions of these poles at the points outside the atomic beam are found explicitly as

$$E_{z\pm}^{\text{SPP}} = i\pi\rho p_0 \frac{\kappa_0^2}{\sigma_0} \text{Res}[R_p(\kappa_0)] e^{-\sigma_0(z+d)} e^{\pm i\kappa_0 y} e^{s_{\alpha}(0)x/v}, \quad (17)$$

where  $\sigma_0 = (\kappa_0^2 - \tilde{\omega}^2)^{1/2}$  and  $\text{Res}[R_p(\kappa_0)]$  is the residue of  $R_p(\kappa_y)$  at the pole  $\kappa_0$ . Equation (17) describes two surface electromagnetic waves (surface plasmon polaritons [29]) which propagate along the metal surface in both directions from the atomic beam.

## B. Criterion of self-excitation

As follows from Eq. (14), the quantity

$$\text{Im}[s_{\alpha}(0)] - \Delta = -\frac{1}{\hbar} |\mu_{12}|^2 \rho w_0 \text{Re}[\mathcal{F}_{\alpha\alpha}(0;\omega)] \quad (18)$$

determines the transition frequency shift which an atom acquires when flying in the beam above the surface. Besides that, the quantity

$$\begin{aligned} g_{\alpha} &\equiv \frac{1}{v} \text{Re}[s_{\alpha}(0)] \\ &= \frac{1}{v} \left\{ \frac{1}{\hbar} |\mu_{12}|^2 \rho w_0 \text{Im}[\mathcal{F}_{\alpha\alpha}(0;\omega)] - \gamma_{\perp} \right\} \end{aligned} \quad (19)$$

describes the variation of the dipole moment amplitude when an atom propagates along the surface. In particular, for distances  $d$  from the surface for which  $\text{Im}[\mathcal{F}_{\alpha\alpha}(0;\omega)]$  is negative (as well as  $w_0$ ) the dipole moment is amplified due to the interaction with the metal surface. If, besides that,

$$\eta_{\alpha} \equiv \frac{|\mu_{12}|^2}{\hbar \gamma_{\perp}} \rho w_0 \text{Im}[\mathcal{F}_{\alpha\alpha}(0;\omega)] > 1, \quad (20)$$

the quantity

$$G_{\alpha} = 2g_{\alpha} = \frac{2\gamma_{\perp}}{v} (\eta_{\alpha} - 1) \quad (21)$$

gives the net intensity gain coefficient. Condition (20) therefore specifies the criterion of generation of the microwave field by the atomic beam. A part of the generated field propagates in both directions from the beam along the metal surface as sustained SPPs.

## C. Steady-state regime

If the generated field amplitude becomes so high that it populates remarkably the upper Rydberg state, one should take into account the evolution of the population inversion given by Eq. (8). In the steady-state regime, which can be reached for a large enough propagation length over the metal, the inversion acquires a constant value,

$$w_{\text{ss}} = \frac{w_0}{1 + (E_{\text{ss}}^{(+)} / E_s)^2}, \quad (22)$$

where

$$E_s = \frac{\hbar(\gamma_{\perp}\gamma_{\parallel})^{1/2}}{2|\mu_{12}|} \quad (23)$$

is the saturation field,  $E_{\text{ss}}^{(+)}$  is the steady-state amplitude of the field, and we have assumed  $\Delta = 0$ . The value of  $E_{\text{ss}}^{(+)}$  is found, in its turn, from the condition

$$\frac{1}{\hbar} |\mu_{12}|^2 \rho w_{\text{ss}} \text{Im}[\mathcal{F}_{\alpha\alpha}(0;\omega)] = \gamma_{\perp} \quad (24)$$

or, equivalently,

$$\left( \frac{E_{\text{ss}}^{(+)}}{E_s} \right)^2 = \eta_{\alpha} - 1. \quad (25)$$

In the steady-state regime one can take  $\dot{\mathbf{p}}^{(+)} = 0$  in Eq. (7) and obtain

$$\mathbf{p}_{\text{ss}}^{(+)} = -\frac{i}{\hbar} \frac{|\mu_{12}|^2}{\gamma_{\perp} - i\Delta} w_{\text{ss}} \mathbf{E}_{\text{ss}}^{(+)}. \quad (26)$$

This quantity does not depend on the microwave-field amplitude in the cavity.

It is noteworthy that a system which is slightly above the threshold is equivalent to the van der Pol oscillator (see the Appendix).

### III. GAIN COEFFICIENT

In this section we explore the dependence of the gain coefficient on different parameters which specify the atomic species and the transition between the Rydberg states. For approximate calculations it is sufficient to neglect the fine structure of the atomic levels and to consider the transition dipole matrix element between state  $|nLM\rangle$  and state  $|n'L'M'\rangle$ . This quantity can be represented as the product of the radial matrix element,  $\langle nL|r|n'L'\rangle$ , which does not depend on the magnetic quantum numbers or the matrix element between the angular parts of the wave functions, which we denote  $D_{L'M'}^{LM}$  [30]. In the quasiclassical approximation the dipole radial matrix element for close states such that  $\Delta n = |n - n'| \ll n, n'$  one finds [31]

$$\langle nL|er|n'L - 1\rangle \approx \frac{n^*n'^*}{\Delta n^*} F_0(\Delta n^*) e a_0, \quad (27)$$

where  $n^* = n - \delta_L$  and  $n'^* = n' - \delta_{L-1}$ , with  $\delta_L$  being the quantum defect,  $\Delta n^* = |n^* - n'^*|$ , the function  $F_0(\Delta n^*)$  is tabulated in Ref. [31],  $e$  is the electron charge,  $a_0$  is the Bohr radius, and we have used the condition  $L/n^* \ll 1$ .

The quantities  $\text{Im}[\mathcal{F}_{\alpha\alpha}(0; \omega)]$ , which enter the expression for the gain coefficient, Eq. (19), can be evaluated as follows. Assuming the Drude model for the dielectric function of the metal,

$$\epsilon_m = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}, \quad (28)$$

with  $\omega_p$  being the plasma frequency and  $\gamma$  being the relaxation rate, one finds that for the microwave frequency range, where  $\omega \ll \omega_p$ ,  $|\epsilon_m| \gg 1$ . In particular, for niobium, which is usually used as a superconducting material for microwave cavities in maser experiments at cryogenic temperatures,  $\omega_p = 5.8$  eV and  $\gamma = 150$  cm<sup>-1</sup> [32]. Then, for example, for  $\lambda = 0.3$  cm,  $\epsilon_m = (-1 + 43.5 \cdot i) \times 10^5$  and, to a good approximation,  $R_p \simeq 1$  and  $R_s \simeq -1$ . In this case the quantities  $\text{Im}[\mathcal{F}_{\alpha\alpha}(0; \omega)]$  can be represented as

$$\text{Im}[\mathcal{F}_{\alpha\alpha}(0; \omega)] = 2\tilde{\omega}^2 f_\alpha(\delta), \quad (29)$$

where

$$f_y(\delta) = \int_0^\delta \frac{\cos 2x}{\sqrt{\delta^2 - x^2}} dx, \quad (30)$$

$$f_z(\delta) = \frac{1}{\delta^2} \int_0^\delta \sqrt{\delta^2 - x^2} \cos 2x dx, \quad (31)$$

and  $\delta = \tilde{\omega}d = 2\pi d/\lambda$ . Plots of the functions  $f_\alpha(\delta)$  are shown in Fig. 2.

The quantity  $\tilde{\omega} = 2\pi/\lambda$  can be expressed in terms of the quantum numbers of the Rydberg states as

$$\tilde{\omega} = 2\pi R \left( \frac{1}{n^{*2}} - \frac{1}{n'^{*2}} \right) \approx 4\pi R \frac{\Delta n^*}{n^3}, \quad (32)$$

where  $R$  is the Rydberg constant.

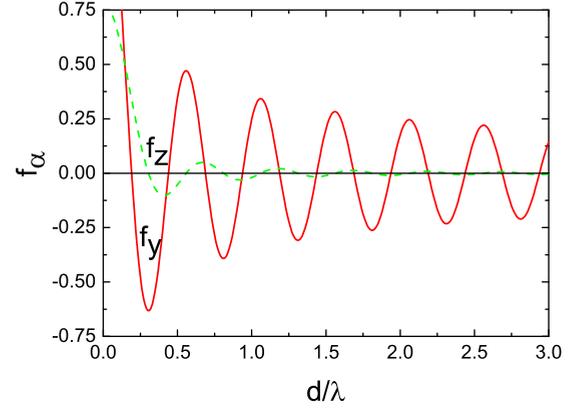


FIG. 2. Plots of the functions  $f_y$  (solid red line) and  $f_z$  (dashed green line) versus  $d/\lambda$ .

The quantities  $\eta_\alpha$ , Eq. (20), which determine the threshold condition, can be found assuming, for a purely radiative relaxation,

$$\gamma_\perp = \frac{1}{2}\gamma_\parallel = \frac{2\tilde{\omega}^3}{3\hbar} |\mu_{12}|^2 g_L, \quad (33)$$

where  $g_L$  is the statistical weight of the lower Rydberg state. Taking into account Eq. (32) one finds

$$\eta_\alpha \approx \frac{3}{4\pi g_L R} \frac{n^3}{\Delta n^*} \rho w_0 f_\alpha(\delta), \quad (34)$$

where we have neglected the quantum defects in comparison with the principal quantum number in the numerator.

Finally, for a system well above the threshold, i.e., for  $\eta_\alpha \gg 1$ , summing up the above results one obtains for the intensity gain coefficient

$$G_\alpha \approx \frac{64\pi^2}{n^2} \frac{\rho w_0}{\hbar v} R^2 e^2 a_0^2 F_0^2(\Delta n^*) |D_{L'M'}^{LM}|^2 f_\alpha(\delta). \quad (35)$$

The saturation field, Eq. (23), can be written in terms of the same parameters as

$$E_s = \frac{(4\pi)^3 \sqrt{2} (\Delta n^*)^2}{3} \frac{R^3 e a_0 F_0(\Delta n^*)}{n^7} |D_{L'M'}^{LM}|. \quad (36)$$

### IV. NUMERICAL RESULTS AND DISCUSSION

We illustrate the above theory with some numerical estimates. We assume that the atomic beam is initially excited by two laser beams to either  $nS$  or  $nD$  Rydberg states. We consider different microwave transitions from these initial states to close overlying Rydberg states with  $n' - n = 0, \pm 1$ , and  $\pm 2$  for sodium, rubidium, and cesium atomic species.

Table I summarizes the values of the parameters which determine the gain coefficient, Eq. (35), for specific Rydberg transitions. The calculations have been carried out using the available data on the quantum defects for Na [33,34], Rb [35,36] and Cs [37] atoms. The corresponding transition wavelength  $\lambda$  can be calculated from Eq. (32). Then, using the plots shown in Fig. 2, one can calculate the gain coefficient for a given distance  $d$  and for a given field orientation in the cavity. The maximum gain is expected at  $d \approx 0.3\lambda$  for the  $y$  orientation of the field ( $f_y = -0.63$ ) and at  $d \approx 0.4\lambda$

TABLE I. Parameters which determine the gain coefficient, Eq. (35), for different atomic species and Rydberg transitions.

Rydberg transition	$\Delta n^*$	$F_0^2(\Delta n^*)$
Na		
$nS \rightarrow nP$	0.49	0.306
$nS \rightarrow (n+1)P$	1.49	0.043
$nD \rightarrow (n+1)F$	1.01	0.106
$nD \rightarrow (n+2)P$	1.16	0.022
Rb		
$nS \rightarrow nP$	0.49	0.306
$nS \rightarrow (n+1)P$	1.49	0.043
$nD \rightarrow nF$	1.33	0.007
$nD \rightarrow (n+2)P$	0.71	0.311
Cs		
$nS \rightarrow nP$	0.49	0.306
$nS \rightarrow (n+1)P$	1.49	0.043
$nD \rightarrow (n-2)F$	0.44	0.280
$nD \rightarrow (n-1)F$	1.44	0.032
$nD \rightarrow (n+2)P$	0.91	0.183

for the  $z$  orientation of the field ( $f_z = -0.10$ ). For estimates one can take  $|D_{L'M'}^{LM}| \sim 1$ . Besides the parameters specified in Table I, the gain coefficient is determined by the beam average velocity. For a given Rydberg transition and for the same number density and temperature of the atomic source for different atomic beams, the gain scales with the atomic mass,  $m_A$ , as  $m_A^{1/2}$ . In particular, the gain coefficients for Na, Rb, and Cs beams correlate as  $G_{\text{Na}}:G_{\text{Rb}}:G_{\text{Cs}} = 1:1.9:2.4$ .

As an example, let us consider sodium atoms with  $w_0 = -0.5$  which are injected into the space above a metal surface having a dipole moment at the  $nS \rightarrow nP$  transition ( $\lambda = 9.1 \times 10^{-6} n^3$  cm) oriented along the  $y$  axis. The generation condition, Eq. (20), is realized for the atom densities  $\rho > \rho_{\text{th}} = 1.5 \times 10^6/n^3$  cm $^{-3}$ . Assuming  $v = 8.9 \times 10^4$  cm/s [15] and  $\rho \gg \rho_{\text{th}}$  one obtains the gain coefficient as  $G_y \approx 5.3 \times 10^{-2} \rho/n^2$ . In particular, for  $n \sim 30$  this gives  $\rho_{\text{th}} \sim 60$  cm $^{-3}$ , which corresponds to the mean distance of 0.02 cm ( $\sim 0.07\lambda$ ) between the atoms in the beam, and  $G_y \approx 6 \times 10^{-5} \rho$ . The maximum gain is expected at the distance  $d \approx 0.08$  cm.

Figures 3–5 illustrate the dependence of the gain coefficient on the beam-surface distance for the  $y$  orientation of the microwave field and for different atomic transitions and atomic species.

The atomic beam approaches the steady-state regime when the generated field amplitude becomes comparable to the saturation field, Eq. (36). For the Rydberg transition in sodium atoms considered above with  $n \sim 30$ ,  $E_s \sim 6 \times 10^{-9}$  V/cm. The distance  $x_s$  from the metal edge  $x = 0$  at which such an amplitude can be reached depends on the initial value of the field,  $E_\alpha(0)$  [Eq. (15)], as follows:

$$x_s = \frac{2}{G_\alpha} \ln \frac{E_s}{E_\alpha(0)}. \quad (37)$$

For the example considered above and  $\rho = 10^5$  atoms/cm,  $E(0)/E_s = 10^{-2}$ , one obtains  $x_s \approx 1.5$  cm. This estimate corresponds to the atom injection rate of  $8.9 \times 10^9$  atoms/s.

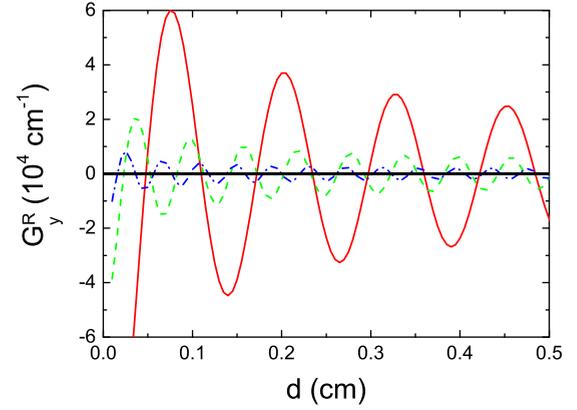


FIG. 3. Dependence of the reduced gain coefficient,  $G_y^R = G_y/|D_{L'M'}^{LM}|^2$ , on the distance  $d$  for  $\rho = 10^9$  atoms/cm,  $v = 8.9 \times 10^4$  cm/s,  $w_0 = -0.5$  and for different transitions in sodium:  $30S \rightarrow 30P$  (solid red line),  $30D \rightarrow 31F$  (dashed green line), and  $30S \rightarrow 31P$  (dash-dotted blue line). Positive values of  $G_y^R$  correspond to gain.

The total power radiated by the atomic beam in the steady-state regime can be estimated using the expression for the dipole radiation [38],

$$W = \frac{4\omega^4}{3c^3} |N \mathbf{p}_{\text{ss}}^{(+)}|^2, \quad (38)$$

where  $N = \rho L$  is the number of self-excited atoms in the beam, with  $L$  being the length of the beam above the metal surface and  $\mathbf{p}_{\text{ss}}^{(+)}$  the steady-state value of the dipole moment, Eq. (26). For  $\Delta \approx 0$  and  $\eta_\alpha \gg 1$  one finds using Eq. (33)

$$|\mathbf{p}_{\text{ss}}^{(+)}| \approx \frac{3}{2g_L} \frac{w_0}{\tilde{\omega}^3 \sqrt{\eta_\alpha}} E_s. \quad (39)$$

Taking for the estimate  $\rho \sim 10^9$  atoms/cm (atom injection rate,  $\sim 10^{14}$  atoms/s) and  $L \sim 1$  cm, one obtains  $W \sim 2 \times 10^{-11}$  W.

The total power of the SPP generated in the steady-state regime can be estimated under the condition  $\omega \ll \omega_p$ . One can show that the main contribution to the SPP intensity

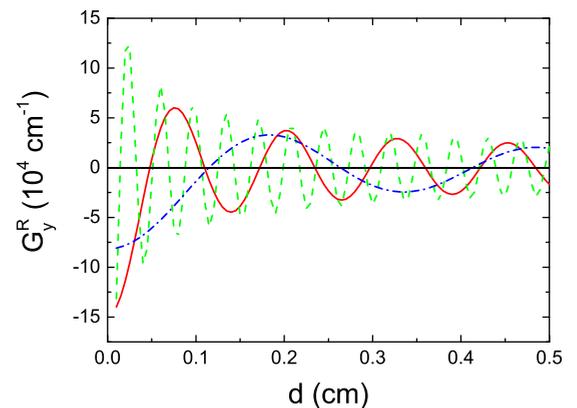


FIG. 4. Same as Fig. 3, but for the transitions  $30S \rightarrow 30P$  (solid red line),  $20S \rightarrow 20P$  (dashed green line), and  $40S \rightarrow 40P$  (dash-dotted blue line).

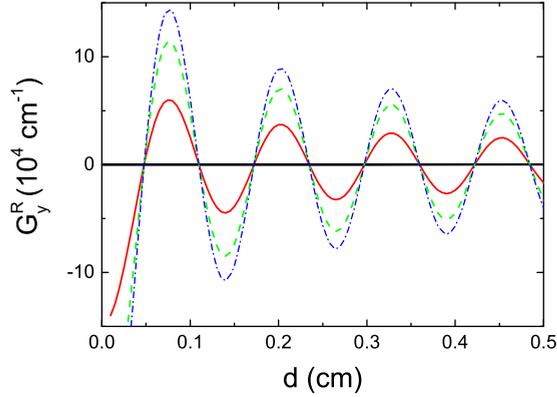


FIG. 5. Same as Fig. 3, but for the transition  $30S \rightarrow 30P$  in sodium (solid red line), rubidium (dashed green line), and cesium (dash-dotted blue line).

originates from the  $z$  component of the electric field in vacuum. Its penetration depth into the vacuum is found as  $1/\sigma_0 \approx (2\pi/\lambda)(\omega_p/\omega)$  and the SPP power is obtained as

$$W_{\text{spp}} \approx \frac{\pi}{4} \frac{\rho^2 |\mathbf{p}_{\text{ss}}^{(+)}|^2 L \omega^4}{c^2 \omega_p} e^{-2\sigma_0 d}. \quad (40)$$

Substitution of the same parameters as above gives  $W_{\text{spp}} \sim 2 \times 10^{-16}$  W, which is hardly detectable in experiments. This result is a consequence of the smallness of the factor  $\omega/\omega_p \approx 8.6 \times 10^{-5}$  which enters Eq. (40). However, this effect can, in principle, be observable for much shorter wavelengths.

## V. CONCLUSION

In conclusion, we have developed the theory of self-excitation of an atomic beam propagating above a metal surface. The underlying mechanism stems from the positive feedback which a reflective surface provides for the dipole polarization of atoms. This phenomenon does not exploit stimulated emission and therefore does not require population inversion in atoms. We have also proposed an experiment with Rydberg atoms in which this effect should be significant. The self-excitation process can be detected either directly by recording the microwave signal or indirectly by monitoring the atomic populations by means of field ionization [15]. This effect can open up new opportunities for studying the cooperative behavior of atoms near surfaces.

## APPENDIX: ANALOGY WITH THE VAN DER POL OSCILLATOR

The system under consideration has a close analogy with a system of oscillating classical dipoles which interact with each

other through their radiation fields reflected from a surface,  $E_R$  [11]. Introducing a saturation in this interaction one can write the equation of their motion as

$$\ddot{p} + \gamma \dot{p} + \omega_0^2 p = \frac{e^2}{m} \frac{E_R}{1 + \kappa |E_R|^2}, \quad (A1)$$

where  $p$  is the dipole moment of the system,  $\gamma$  is the damping constant,  $\omega_0$  is the frequency of a single oscillator, and  $\kappa$  is the parameter which is related to the saturation field. Taking into account that the reflected field is proportional to the dipole moment of the system, i.e.,  $E_R = Fp$ , and assuming a weak saturation ( $\kappa |E_R|^2 \ll 1$ ), one arrives at the equation

$$\ddot{p} + \gamma \dot{p} + \omega_0^2 p = \frac{e^2}{m} F p (1 - \kappa |F p|^2). \quad (A2)$$

In the absence of saturation the quantity  $\text{Re}F$  determines the frequency of oscillations,  $\bar{\omega}_0$ , renormalized due to the reflected field. Therefore in what follows we assume that such a renormalization is done and take  $F = -iG$ , with  $G$  being a real positive quantity that corresponds to self-excitation. The evolution of the dipole moment is described by the time dependence  $p(t) = q(t)e^{-i\bar{\omega}_0 t}$ , with  $q(t)$  being a function which varies on a time scale much larger than  $\bar{\omega}_0^{-1}$ . Then one can approximate the quantity  $p$  in the factor in front of the parenthesis on the right-hand side of Eq. (A2) as  $p \approx \dot{p}/(-i\bar{\omega}_0)$ . Thus one obtains the following equation:

$$\ddot{p} - (\alpha - \beta |p|^2) \dot{p} + \bar{\omega}_0^2 p = 0, \quad (A3)$$

where

$$\alpha = \frac{e^2 G}{m \bar{\omega}_0} - \gamma \quad (A4)$$

and

$$\beta = \frac{e^2 G^3 \kappa}{m \bar{\omega}_0}. \quad (A5)$$

For a self-excited system the constant  $\alpha$  is positive. In such a case for real solutions Eq. (A3) is identical to the equation for the van der Pol oscillator [10]. Its steady-state amplitude is given by

$$p_{\text{ss}} = \sqrt{\frac{\alpha}{\beta}}. \quad (A6)$$

For the system under consideration  $\alpha \ll \bar{\omega}_0$ , which implies that any small oscillations build up to amplitude  $p_{\text{ss}}$  and no chaotic behavior is possible.

- [1] S. Haroche, *Rev. Mod. Phys.* **85**, 1083 (2013).  
 [2] P. R. Berman (ed.), *Cavity Quantum Electrodynamics* (Academic Press, New York, 1994).  
 [3] H. Walther, B. T. H. Varcoe, B.-G. Englert, and T. Becker, *Rep. Prog. Phys.* **69**, 1325 (2006).  
 [4] E. M. Purcell, *Phys. Rev.* **69**, 681 (1946).

- [5] W. L. Barnes, *J. Mod. Opt.* **45**, 661 (1998).  
 [6] K. H. Drexhage, H. Kuhn, and F. P. Schäfer, *Ber. Bunsenges. Phys. Chem.* **72**, 329 (1968).  
 [7] R. R. Chance, A. Prock, and R. Silbey, *J. Chem. Phys.* **60**, 2744 (1974).  
 [8] A. Sommerfeld, *Ann. Phys.* **333**, 665 (1909).

- [9] A. Sommerfeld, *Partial Differential Equations in Physics*, Lectures on Theoretical Physics Vol. VI (Academic Press, New York, 1964), Secs. 32, 33.
- [10] A. Jenkins, *Phys. Rep.* **525**, 167 (2013).
- [11] V. G. Bordo, *Phys. Rev. A* **88**, 013803 (2013).
- [12] V. G. Bordo, *Phys. Rev. B* **93**, 155421 (2016).
- [13] V. G. Bordo, *Appl. Phys. Lett.* **110**, 033110 (2017).
- [14] J. M. Raimond, P. Goy, M. Gross, C. Fabre, and S. Haroche, *Phys. Rev. Lett.* **49**, 1924 (1982).
- [15] L. Moi, P. Goy, M. Gross, J. M. Raimond, C. Fabre, and S. Haroche, *Phys. Rev. A* **27**, 2043 (1983).
- [16] P. Goy, L. Moi, M. Gross, J. M. Raimond, C. Fabre, and S. Haroche, *Phys. Rev. A* **27**, 2065 (1983).
- [17] N. Lu, *Phys. Lett. A* **143**, 457 (1990).
- [18] O. Kocharovskaya and P. Mandel, *Phys. Rev. A* **42**, 523 (1990).
- [19] J. Mompert and R. Corbalán, *J. Opt. B: Quantum Semiclass. Opt.* **2**, R7 (2000).
- [20] A. A. Svidzinsky, L. Yuan, and M. O. Scully, *Phys. Rev. X* **3**, 041001 (2013).
- [21] See, e.g., V. G. Bordo and H.-G. Rubahn, *Optics and Spectroscopy at Surfaces and Interfaces* (Wiley-VCH, Weinheim, 2005).
- [22] I. A. Larkin, M. I. Stockman, M. Achermann, and V. I. Klimov, *Phys. Rev. B* **69**, 121403(R) (2004).
- [23] S. Prasad and R. J. Glauber, *Phys. Rev. A* **61**, 063814 (2000).
- [24] J. M. Wylie and J. E. Sipe, *Phys. Rev. A* **30**, 1185 (1984).
- [25] Strictly speaking, the field polarizability given in Ref. [24] is derived for a dipole above an infinite vacuum-metal interface and does not take into account the edge effects due to the finite dimensions of the interface. Such effects occur, however, within distances of the order of the wavelength from the edges.
- [26] In the problem under consideration the quantity  $\Delta$  has a sense of a frequency shift of the atomic transition which originates from the atom-atom interactions. It is reasonable to assume that this shift is much less than the transition frequency  $\omega_0$ .
- [27] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, Mineola, NY, 1987).
- [28] H. Haken, *Light* (North-Holland, Amsterdam, 1985), Vol. 2.
- [29] H. Raether, *Surface Plasmons on Smooth and Rough Surfaces and on Gratings* (Springer, Berlin, 1988).
- [30] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Pergamon, Oxford, UK, 1977).
- [31] N. B. Delone, S. P. Goreslavsky, and V. P. Krainov, *J. Phys. B: At. Mol. Opt. Phys.* **27**, 4403 (1994).
- [32] A. V. Pronin, M. Dressel, A. Pimenov, A. Loidl, I. V. Roshchin, and L. H. Greene, *Phys. Rev. B* **57**, 14416 (1998).
- [33] T. F. Gallagher, R. M. Hill, and S. A. Edelstein, *Phys. Rev. A* **13**, 1448 (1976).
- [34] C. Fabre, S. Haroche, and P. Goy, *Phys. Rev. A* **18**, 229 (1978).
- [35] C.-J. Lorenzen and K. Niemax, *Phys. Scripta* **27**, 300 (1983).
- [36] W. Li, I. Mourachko, M. W. Noel, and T. F. Gallagher, *Phys. Rev. A* **67**, 052502 (2003).
- [37] P. Goy, J. M. Raimond, G. Vitrant, and S. Haroche, *Phys. Rev. A* **26**, 2733 (1982).
- [38] L. D. Landau and E. M. Lifshitz, *The Classical Theory of Fields* (Pergamon, Oxford, UK, 1971).