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Light-induced atomic desorption dynamics: Theory for a completely illuminated cell

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The light-induced atomic desorption effect, known as LIAD, has been explained so far by a debatable Atutov's theory [S. N. Atutov *et al.*, Phys. Rev. A **60**, 4693 (1999)]. In this Brief Report, we propose an approach in which the difficulties of this theory are removed. The theory yields predictions for the atomic vapor density $n(t)$ in a fully illuminated resonance cell, the time t_{max} that it takes the vapor density to arrive at a maximum value n_{max} and the dependence of n_{max} and $dn/dt_{t=+0}$ on the desorbing light intensity. Some of our results are at variance with outcomes of Atutov's model (such as saturation effect for n_{max} and the dependence of t_{max} on the light intensity) but are in agreement with the experimental data. Additionally, the model presented in this Brief Report satisfactorily explains a reservoir effect recently investigated in experiments where cells equipped with a lockable stem were used.

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I. INTRODUCTION

Experiments prove that even a weak and incoherent light illuminating the films containing adsorbed atoms causes a huge atomic desorption, which is observed as a vast increase in the atomic vapor density in the cell which inner surface is coated with a film made of adsorbing medium. This effect called LIAD (light-induced atomic desorption) has been observed with Na [1], Rb [2], and Cs [3,4] atoms and for many other kinds of coatings such as porous glass [5], paraffin [6,7], sapphire [8], stainless steel [9], porous silica [10,11], and Vycor [12]. Recently LIAD was part of the process used to load magneto-optic trap [13] or achieve Bose-Einstein condensation on a microelectronic chip [14]. For the last decade, the Atutov theory [15] has been used to account for the experimental data concerning LIAD. Problems with this theory are listed in [16]. One more difficulty follows from data presented in [6]. It is shown that after the illumination is stopped and the vapor density already has its equilibrium value n_0 $(dn/dt=0)$, there is a *nonzero* flux of atoms J supplementing the atomic density in the coating. This means that the cornerstone equation of the Atutov theory [Eq. (8) in [15]] governing the vapor density of atoms n in the cell

$$\frac{dn}{dt}=\frac{J}{L}-\gamma(n-n_0) \tag{1}$$

is contradictory in this case (the respective symbols denote: $L=V/S$ is the characteristic cell length, V is the volume of the cell, S is its surface; the last term describes the relaxation of the vapor density to its equilibrium value controlled by the reservoir, and γ^{-1} is the characteristic relaxation time). A model for LIAD dynamics presented in [6] suffers from the same difficulty as the Atutov one and in consequence the two Eqs. 6 in [6] are inconsistent for the case described above. Also the predictions of the model presented in Figs. 9 and 10 of [6] are not satisfactory.

The aim of this Brief Report is to deliver a proper theory for describing LIAD dynamics by taking into consideration the recently achieved experimental results [6,17]. Using our model we also explain the recently investigated reservoir effect [18]. We are going to consider the case of a completely illuminated cell. The case of a partially irradiated coating has already been elaborated in [16].

II. THEORY

As proved in [16], thanks to the very quick diffusion, no diffusion equation regarding the movement of atoms in the coating in the direction normal to the surface needs to be considered. The density of atoms N inside the coating treated as a whole can be determined simply on the basis of the rate equation

$$\frac{dN}{dt}=-\frac{J}{H}. \tag{2}$$

J denotes the flux of atoms $J=\alpha N-\beta n$ through the surface of the coating (α is the desorption rate, β—the adsorption rate). It is assumed that $\alpha=\alpha_0+\alpha_I(\lambda,I)=\alpha_0+k(\lambda)I_L$, where α_0 is the desorption rate in the dark, $k(\lambda)$ is a function of the wavelength only and I_L is the laser light intensity. In equilibrium, before the light is switched on, the flux J is equal to zero, so that $\alpha_0N_0=\beta n_0$, where N_0 is the atomic density in the coating in equilibrium.

As proved in [6], after irradiation, when the light is switched off and n has reached its equilibrium value n_0, a flux of atoms ("dark flux") defined as

$$J_D=\alpha_0N-\beta n_0, \tag{3}$$

fills the coating. J_D does *not* contribute to the change in the vapor density when the light is switched off. We can expect that during illumination only the part of the flux J *exceeding* the dark flux J_D influences the change in the vapor density n in the cell. It means that the rate equation for n, instead of Eq. (1), should be

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$$\frac{dn}{dt} = \frac{J - J_D}{L} - \gamma(n - n_0), \tag{4}$$

Contrary to Eq. (1), Eq. (4) is self-consistent in the case when the vapor is in equilibrium after the light is switched off (i.e., when $n = n_0$, $dn/dt = 0$, and $\alpha = \alpha_0$).

Solving Eq. (4) we get

$$n(t) = \frac{H}{\beta}[A_1(s_1 + a)\exp(s_1 t) + A_2(s_2 + a)\exp(s_2 t) + aA_3], \tag{5}$$

where

$$s_{1,2} = \frac{-(a+g) \pm \sqrt{(a-g)^2 + 4ab - \frac{4b^2 L n_0}{HN_0}}}{2}, \tag{6}$$

$$A_3 = \frac{g n_0 b L}{Ha(g-b) + \frac{b^2 L n_0}{N_0}}, \tag{7}$$

$$A_{1,2} = \mp \frac{s_{2,1}(N_0 - A_3) + aN_0 - \frac{bLn_0}{H}}{s_1 - s_2}. \tag{8}$$

and $a = \alpha/H$, $b = \beta/L$, $g = \beta/L + \gamma$.

When the light is switched off, $\alpha_I = 0$, Eq. (4) reduces to

$$\frac{dn}{dt} = -g(n - n_0), \tag{9}$$

so that $n(t)$ decays as $\exp(-gt)$. To fit $n(t)$ given by Eq. (5) to experimental data, first we determine g from the exponential decay when the light is switched off and next the remaining parameters a, b, and HN_0 are obtained from the best fit of the curve Eq. (5) to the LIAD signal (H and N_0 always occur together as the product HN_0, so that HN_0 is treated as a single parameter). Fits of the function Eq. (5) to the experimental results provided in [6] are presented in Fig. 1.

Having Eq. (5), from the condition $dn/dt = 0$ we find the expression for the time t_{max} for the vapor density n to reach its maximum value

$$t_{max} = \frac{\ln(-d_2/d_1)}{s_1 - s_2}, \tag{10}$$

where: $d_{1,2} = A_{1,2}s_{1,2}(s_{1,2} + a)$. For established parameters b, g, and HN_0 Eq. (10) represents a function of a. Remembering that α_0 is much smaller than $\alpha_I = k(\lambda)I_L$, the dependence on $a \approx k(\lambda)I_L/H$ may be regarded as the dependence on the light intensity I_L. The plot of t_{max}^{-1} as a function of a is given in Fig. 2. The theoretical curve is fitted to the experimentally achieved dependence of t_{max}^{-1} on the light intensity presented in [6]. Agreement is satisfactory up to the light intensity equal to 3 mW/cm^2. (The data for light intensity above 3 mW/cm^2 are quite extraordinary and seem to reveal a heating process that occurred during irradiation.) Let us remark that the Atutov approach [15] yields t_{max} as being independent of the light intensity, which makes our theory essentially different from the Atutov one.

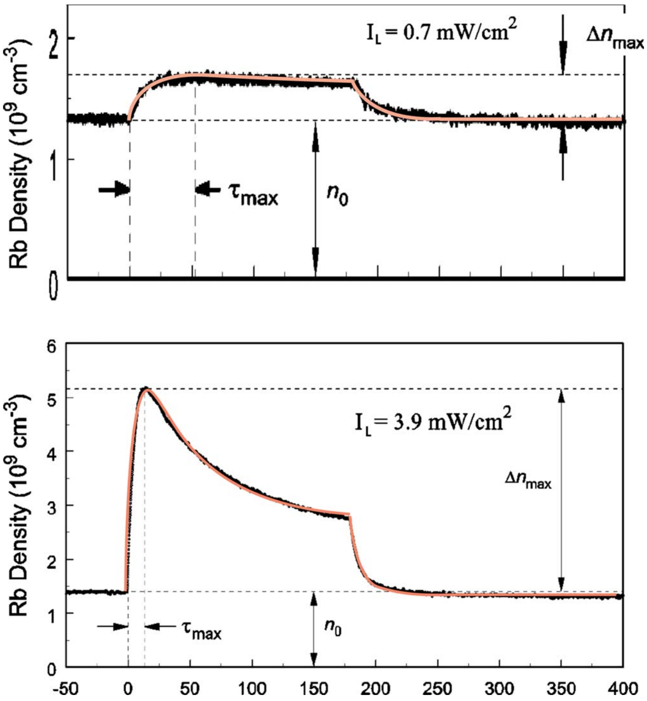

FIG. 1. (Color online) Fits of the theoretical curve Eq. (5) to the experimental outcomes taken from [6]. From the cell parameters we get $L \approx 0.5$ cm. The remaining parameters for $I_L = 0.7$ mW/m^2 are: $g \approx 0.066$ s^{-1}, $a \approx 0.0034$ s^{-1}, $b \approx 0.010$ s^{-1}, and $HN_0 \approx 6.2 \cdot 10^9$ cm^{-2}. For $I_L = 3.9$ mW/m^2 the parameters are: $g \approx 0.12$ s^{-1}, $a \approx 0.055$ s^{-1}, $b \approx 0.064$ s^{-1}, and $HN_0 \approx 7.4 \cdot 10^9$ cm^{-2}.

Now, inserting Eq. (10) into Eq. (5) we can obtain an analytical function showing the dependence of n_{max} on the light intensity (i.e., on $a = k(\lambda)I_L/H$)

$$n_{max} = \frac{H}{\beta}\left[A_1(s_1 + a)\left(-\frac{d_2}{d_1}\right)^{s_1/(s_1 - s_2)} + A_2(s_2 + a)\left(-\frac{d_2}{d_1}\right)^{s_2/(s_1 - s_2)} + aA_3\right]. \tag{11}$$

What is important is that, according to Eq. (11), n_{max} does

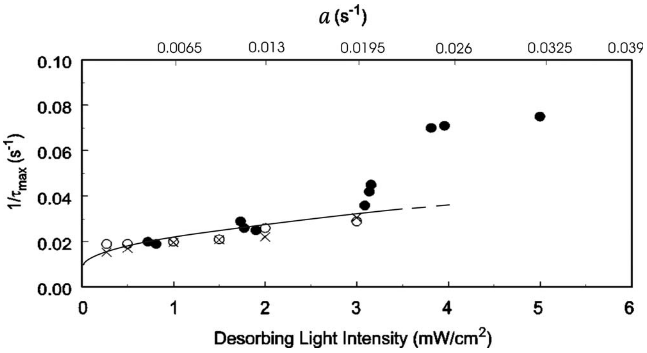

FIG. 2. Results of the measurements of t_{max}^{-1} as a function of the light intensity (taken from [6]) and the theoretical prediction Eq. (10) for t_{max}^{-1} as a function of a (the solid line). Good agreement is achieved up to $I_L = 3$ mW/cm^2. One can suspect that heating effects for the higher light intensity changed conditions of the experiment.

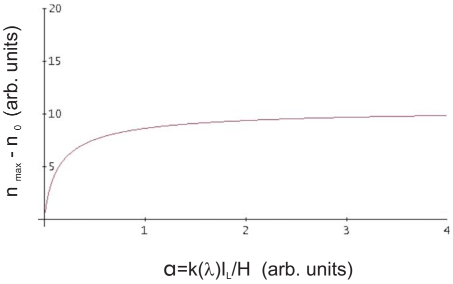

FIG. 3. (Color online) $n_{max}-n_0$ plotted according to the function given in Eq. (11).

not increase infinitely with increasing light intensity (see Fig. 3) but is *bounded* by the value

$$\lim_{a\to\infty}(n_{max}-n_0)=2HN_0. \tag{12}$$

This "saturation" effect we expect from LIAD dynamics is missing in the Atutov theory, which predicts an unbounded increase in $n_{max}-n_0$ proportional to $\sqrt{I_L}$.

The fit of Eq. (11) to the experimental data taken from [15] is shown in Fig. 4. As can be seen, a good agreement with the experimental results is achieved. Let us note that for the typical (relatively low) light intensities used in experiments, the dependence of Δn_{max} on I_L is very similar to a function proportional to $\sqrt{I_L}$, which makes it easy to be falsely interpreted, as is done in [15].

III. RESERVOIR EFFECT

An important verification of our theory is the possibility to explain a reservoir effect described recently in [18]. Using lockable-stem cells the authors were able to reduce (by closing the reservoir) the relaxation of the alkali-atom density to the stem. The configuration with a completely closed stem is equivalent, in our model, to putting $\gamma=0$ (no relaxation to the reservoir). In consequence, the relaxation after the light is switched off, Eq. (9), is determined by the factor $g=\beta/L$ instead of $g=\beta/L+\gamma$, as it is for the open stem configuration. Our theory then predicts that the relaxation time, denoted in [18] as τ_3, should be increased for the cell with the closed stem, which is confirmed by the experimental data (see Table II in [18]).

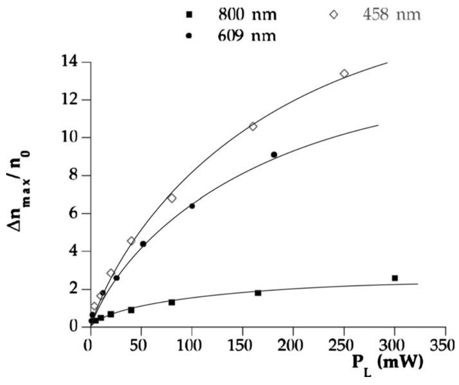

FIG. 4. $(n_{max}-n_0)/n_0\equiv\Delta n_{max}/n_0$ fitted according to Eq. (11) to the experimental results presented in [15].

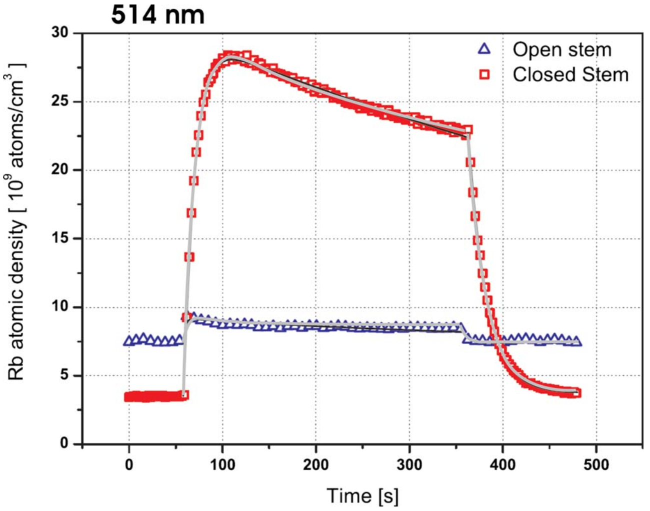

FIG. 5. (Color online) Change in the atomic density with the LIAD effect in two configurations: open and closed stem in a Rb paraffin-coated lockable-stem cell. The solid line represents a fit of the function (5). $I_L=50$ mW/cm^2 in both cases.

To check whether our model correctly explains the difference between the LIAD signal for the open and closed-stem configurations we refer to the experimental data provided in Figs. 6 and 7 in [18]. Since the light intensity is the same for the open and closed-stem configurations, the proper fits to the LIAD signal should be obtained for the same parameter a. Similarly, the parameter $b=\beta/L$, representing the rate of the atomic adsorption to the coating, must remain unchanged. We also have no choice as concerns the parameter g, because it is determined from the exponential decay when the light is switched off. As discussed above, our theory predicts that for the cell with a *perfectly closed* stem, the best fit of the theoretical curve to LIAD signal should be obtained for $g=b$. In effect there remains only *one* parameter, HN_0, that can be modified while switching between the two LIAD

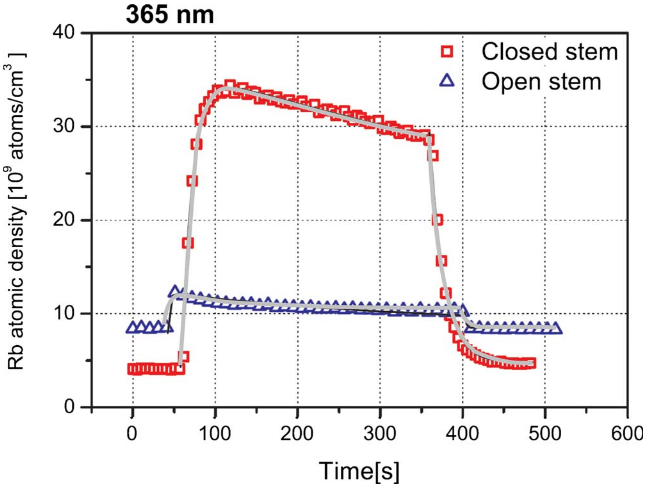

FIG. 6. (Color online) Density change with LIAD in a Rb paraffin-coated lockable-stem cell. Two cases are shown: open-stem and closed-stem configurations. The solid line represents a fit of the function (5). $I_L=5$ mW/cm^2 in both cases.

TABLE I. LIAD characterization parameters extracted from the model fit to the LIAD experimental data. The parameters are presented for the Rb cells in open (Rb-o) and closed (Rb-c) stem configurations. The desorption light used is characterized by wavelength (λ) and intensity (I).

Cell	λ (nm)	I (mW/cm^2)	g (s^{-1})	a (s^{-1})	b (s^{-1})	HN_0 (cm^{-2})
Rb-o	514	50	0.17	0.039	0.045	6.97×10^9
Rb-c	514	50	0.051	0.039	0.045	25.3×10^9
Rb-o	365	5	0.17	0.02	0.051	21.0×10^9
Rb-c	365	5	0.057	0.02	0.051	51.7×10^9

signals for the open and closed-stem configurations. The data from Fig. 4 in Ref. [18] prove that N_0 indeed must vary for cells with open and closed stems. After the stem is locked (with no illumination), there is observed a decrease in the vapor density in the cell, which means that the alkali atoms are adsorbed by the cell coating. Thus, for successive LIAD experiments, the initial atomic density N_0 in the coating is increased in the case of the closed stem.

Fits of the function (5) to the experimental data are presented in Figs. 5 and 6 and the best fit parameters are listed in Table I. The data shown in Table I confirm all the expected facets of a proper model. It should be noticed that, in any case of the closed-stem configuration, g is not precisely equal to b, but is slightly bigger. This reflects the fact that the stem opening is never completely closed by a moving glass bullet, so that γ is never reduced precisely to zero.

IV. CONCLUSIONS

We have developed a self-consistent theory that is devoid of all the conceptual difficulties one encounters in the Atutov approach. Satisfactory fits of the theoretical curves to the experimental results, the saturation effect as concerns increase in n_{max} with I_L and successful explanation of the reservoir effect may be regarded as an important verification of our theory.

Both the theory [16] for a locally illuminated cell and the one presented here take into account that the process of diffusion in the adsorbing films proceeds very quickly in the direction normal to the surface. Therefore no diffusion equation needs to be considered for this direction. Persistence of LIAD effect described in [16] is ensured by the lateral atomic diffusion and for a completely illuminated cell follows from the adsorption process that prevents the coating from being depleted from the atoms within a very short time.

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