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Light-induced desorption of alkali-metal atoms from paraffin coating

E. B. Alexandrov and M. V. Balabas
S. I. Vavilov State Optical Institute, St. Petersburg, 199034 Russia

D. Budker*
Department of Physics, University of California at Berkeley, Berkeley, California 94720-7300
and Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720

D. English, D. F. Kimball, C.-H. Li, and V. V. Yashchuk
Department of Physics, University of California at Berkeley, Berkeley, California 94720-7300

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Observation of nonthermal, light-induced desorption of Cs and Rb atoms from paraffin coating is reported. Buffer-gas-free paraffin-coated cells containing Cs and Rb were exposed to laser light of various intensities and frequencies, and the change of the vapor densities as a function of time was investigated. Because atomic polarization relaxes very slowly in paraffin-coated cells (spin-relaxation times can be $\gtrsim 1$ s), they are applied in a variety of precision measurements (e.g., magnetometry, discrete symmetry tests, etc.). Light-induced desorption may be useful in such measurements as a method to control atomic density without changing the temperature of the cell.

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I. INTRODUCTION

Coating the walls of an alkali-metal vapor cell with paraffin wax can reduce the spin-relaxation rate due to wall collisions by four orders of magnitude [1–4]. The narrow resonances (~ 1 Hz) that result can be applied in, for example, sensitive magnetometry [3–6], discrete symmetry tests in atomic systems [7–9], the study of light propagation dynamics [10,11], and the generation of atomic spin-squeezed states [12].

Recently, while investigating nonlinear magneto- and electro-optical effects in Rb and Cs [6,8,9,13], we observed a significant increase (roughly by a factor of 2) in both Rb and Cs vapor densities when a paraffin-coated cell was exposed to the ambient room light. Since exposure to the light caused no measurable change in the cell temperature ($<0.1\,^\circ$C), the increase in atomic density did not result from heating of the vapor cell. When the lights were turned off, the atomic density in the cell returned to its original value after a few minutes. These preliminary observations were briefly discussed in Ref. [9].

Nonthermal, light-induced desorption of a number of different atoms has previously been observed from a wide variety of materials. In Ref. [14], desorption of Na, Ni, Zn, and Sn atoms from a sapphire plate was observed when visible laser light of moderate intensity ($1-1000$ mW/cm^2) illuminated the surface. However, no effect was observed for infrared light (wavelength$=1.15\ \mu$m), indicating a threshold in light frequency (more detailed studies of these phenomena are described in Refs. [15,16]). In work more closely related to the present study, there have been several investigations of light-induced atomic desorption (LIAD) of alkali-metal atoms and molecules (Na, Na$_2$, Rb, and Cs) from silane coatings (in particular, polydimethylsiloxane) [17–21]. References [17,20] propose a physical mechanism of LIAD (a charge-transfer reaction) involving the particular chemistry of the silane coating. This mechanism, however, would not account for the observation of LIAD from paraffin, and in fact there have been some statements in the literature that LIAD does not occur from paraffin [20]. References [21,22] emphasize that, in addition to desorption from the coating surface, light-assisted diffusion of atoms inside the silane coating can play an important role in LIAD. Light-induced desorption of Rb atoms from a superfluid ^{4}He film has been observed in Refs. [23,24]. LIAD has proved useful as a relatively low-temperature atomic source [25], which has been employed to load a magneto-optic trap [26]. Very recently, LIAD was part of the process used to achieve Bose-Einstein condensation on a microelectronic chip [27].

In this paper we describe the first detailed investigation of light-induced desorption of Rb and Cs atoms from a paraffin coating. We have studied the time dependence of LIAD in various paraffin-coated, alkali-metal vapor cells as a function of cw desorbing light intensity and frequency. We have also investigated how LIAD from paraffin depends on the amount of time between exposures to cw desorbing light, and have studied LIAD with pulsed desorbing light. From these experiments, we develop a qualitative understanding of the physical processes involved in the effect, and estimate the values of relevant parameters describing LIAD from paraffin coatings.

II. APPARATUS

A. Paraffin-coated cells

Three different types of evacuated (buffer-gas-free), cylindrical paraffin-coated vapor cells (shown in Fig. 1) were used in our present experiments. Cell (a) had a single stem which contained a solid sample of ^{85}Rb. Cell (b) had two

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*Electronic address: budker@socrates.berkeley.edu

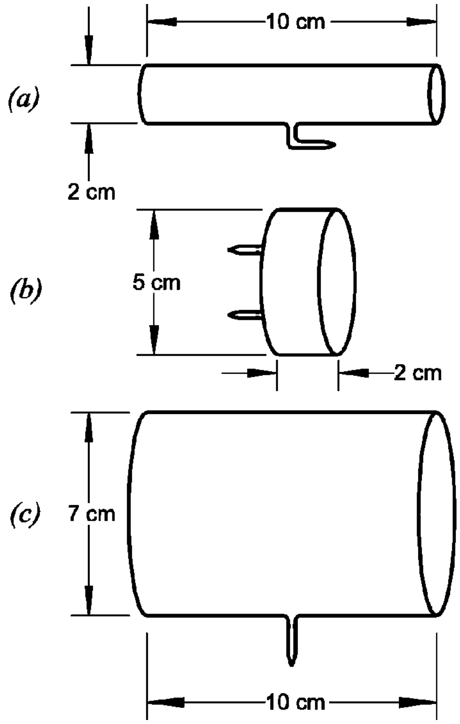

FIG. 1. Geometries of different paraffin-coated vapor cells employed in the present experiments.

stems, one of which contained a sample of ^{85}Rb and the other contained Cs. We used three different cells of type (c), one with two stems, one containing ^{85}Rb and the other stem containing Cs, another with two stems, one containing ^{87}Rb and the other Cs, and a third cell with a single stem containing only Cs. Note that cells of type (a) and type (b) have approximately the same inner surface area and volume.

As has been commonly observed in paraffin-coated alkali vapor cells (see, for example, Refs. [2,28,30,31]), the equilibrium Cs and Rb vapor densities (in the dark) in each of the cells at room temperature (20 °C) were 10–70 % lower (depending on the cell) than the saturated vapor densities of $\approx 3.0\times10^{10}$ cm^{-3} for Cs and $\approx 4.5\times10^{9}$ cm^{-3} for Rb [29]. This suppression of the alkali vapor densities has generally been attributed to the continuous adsorption of atoms into the paraffin coating [2,28,30,31]. As discussed in Sec. IV, we find that in our case, there appears to be some additional suppression of the alkali-metal vapor pressure in the stems, which may occur because the metal samples in the stems become coated with, for example, paraffin. Also, in the case of two-stemmed cells containing Rb and Cs, there may be migration of alkali-metal atoms between the stems leading to the formation of a Rb-Cs amalgam, for which the equilibrium vapor pressure of a given element is given roughly by the fraction of that element making up the amalgam times the element's saturated vapor pressure [32].

The technique for preparing the paraffin-coated cells is discussed in detail in Ref. [33] (a similar procedure is described in Ref. [34]). Because of the possibility that LIAD from paraffin may be dependent on the cell preparation procedure, for completeness we summarize the technique here. To produce a cell, the assembly shown in Fig. 2 is prepared.

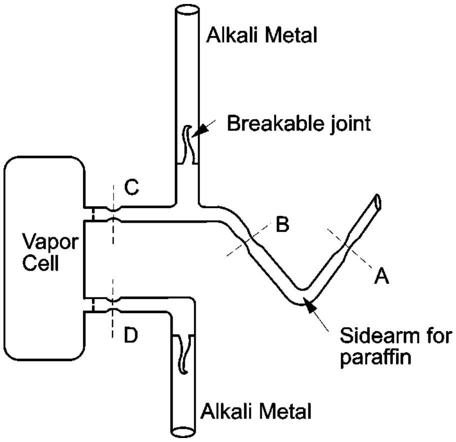

FIG. 2. Diagram of the assembly used to produce a paraffin-coated cell of type (b). *A, B, C,* and *D* denote various points at which the assembly is sealed off during cell preparation, as described in the text. All the cells are prepared in essentially the same manner.

First, the inner surface of the cell is cleaned with a 10% solution of hydrochloric acid and then repeatedly flushed with distilled water. The assembly is vacuum pumped and baked at 400 °C to outgas the glass (Russian glass type C1-51 is used for the cylindrical parts of the cells and type LK-4 is used for the flat, circular windows). Next, the cell is cooled and a drop of the coating material (polyethylene wax fractionated at 220 °C, a mixture of molecules with chemical formula C_nH_{2n+2} with various n's where $n\sim50$ [33]) is placed in the sidearm. Then the assembly is pumped out (residual pressure$=2\times10^{-5}$ Torr) and sealed off at point *A* in Fig. 2. Next, the assembly is baked in an oven at 230 °C to fill the cell with paraffin vapor. To ensure uniform heating, the air in the oven is circulated to eliminate temperature gradients ($\Delta T<1$ °C over the size of the cell). After 1 h of baking, the cell assembly is cooled back to room temperature. Containers of distilled alkali metals are attached to the vertical arms of the assembly (before the breakable joints), and the vertical arms are vacuum pumped to 10^{-5} Torr. Wax is now removed from the stems of the cell by heating them with a flame. The sidearm where the paraffin was contained is then sealed off at point *B* in Fig. 2. Next, the breakable joints are opened (one at a time) and the alkali metal is deposited on the uncoated glass of the stem. The cell is then sealed off at points *C* and *D* in Fig. 2.

Initially, the alkali-metal vapor density in the cell is extremely small, so that no absorption of resonant light can be observed. Since the alkali vapor pressure in the cell is strongly suppressed compared to the saturated vapor pressure, the rate at which atoms are adsorbed into the paraffin coating must greatly exceed the rate at which atoms are evaporated from the metal samples in the stems. After the cell is baked at 80 °C for ≈ 10 h, a significant alkali vapor density is produced. When this stage of the cell preparation, known as "ripening," is complete, the coating absorbs atoms at a much slower rate, so that the flux of atoms from the stems can support an equilibrium vapor pressure relatively close to the saturated vapor pressure. A similar ripening pro-

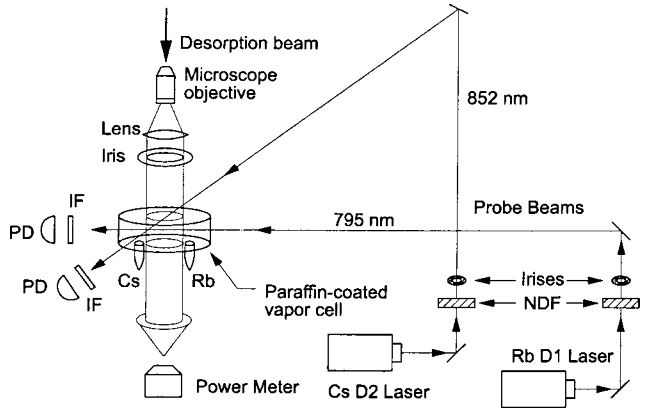

FIG. 3. Setup for light-induced desorption measurements. Neutral density filters (NDF) attenuate the light intensity in the probe beams to $\approx 10\ \mu\text{W/cm}^2$ (well below saturation). Interference filters (IF) at 850 nm and 790 nm (bandwidth = 10 nm) are placed in front of the photodiodes (PD) that monitor transmission. Desorption light is generated by an Ar^+ laser, an Ar^+-pumped dye laser, external-cavity diode lasers, or a pulsed Nd:YAG (yttrium aluminum garnet) laser depending on the experiment. The intensity of the desorbing light was measured to be uniform (to within ~20%) over the illuminated area using a beam profiler.

cess occurs in silane-coated cells [35], and is attributable to a chemical reaction between the alkali atoms and the coating (or impurities in the coating). Additionally, in paraffin-coated cells containing ^{199}Hg [36], measurements of nuclear depolarization rates indicate the presence of paramagnetic impurities on the coating, which may be free radicals or "dangling bonds" [37]. Since there are no chemically active compounds present in the Hg cells to react with the free radicals, they remain indefinitely. Perhaps the ripening process in paraffin-coated cells containing alkali atoms can be attributed to the reaction of alkali atoms with such free radicals in the coating. This would explain why studies of electronic depolarization rates in paraffin-coated alkali vapor cells show no evidence of paramagnetic impurities [2], since they would be eliminated during the ripening process [38].

B. Experimental setup

The setup for the present experiment is shown in Fig. 3. The densities of Rb and Cs are monitored by measuring the transmission of weak light beams resonant with the Rb $D1$ (795 nm) and Cs $D2$ (852 nm) lines. The light is generated by external-cavity diode lasers and the light frequency is scanned over several gigahertz to cover the entire hyperfine structure for each resonance. The transmission spectra are fit to Voigt profiles to extract the densities of Rb and Cs. Alternatively, the probe light is tuned to the center of one of the hyperfine components of the transition, and the time dependence of transmission is measured. In this case, the density is extracted by comparing the transmission of the probe light through the paraffin-coated cell when probe beam is tuned on-resonance to when the probe beam is off-resonance (this ratio $=e^{-\alpha\ell}$, where ℓ is the path length and the absorption coefficient α is proportional to the density). The probe light power and transmission through an uncoated reference cell (in which the density is known) are also measured.

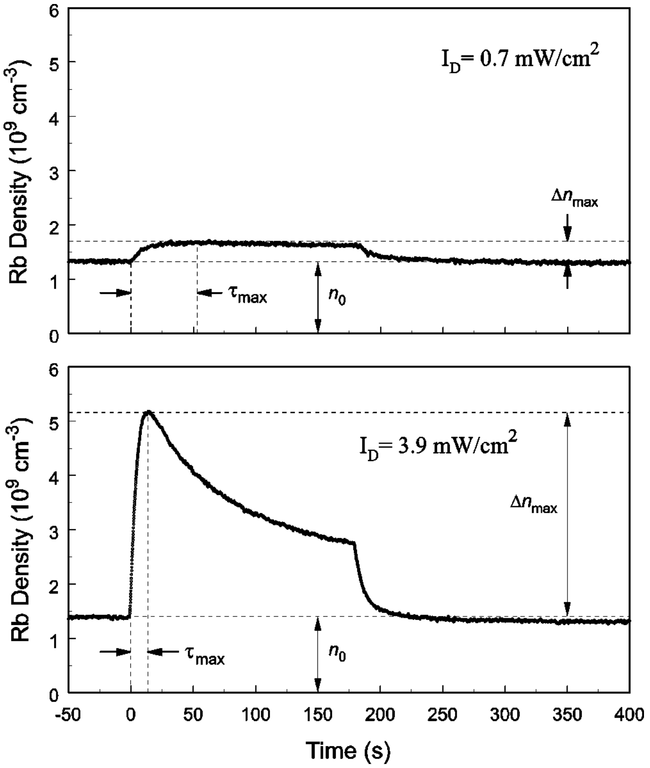

FIG. 4. Rb vapor density in a paraffin-coated cell of type (a) (Fig. 1) as a function of time when the cell is exposed to 514-nm light from an Ar^+ laser. The shutter blocking the desorbing light is opened at time $t=0$ and closed after 180 s. The cell is completely illuminated by the desorbing light for the data shown in these plots. I_D is the desorbing light intensity.

The sequence of events in the experiment is as follows. The desorbing light is initially blocked by a shutter. The shutter is then opened for a period of time after which it is closed. The vapor density in the cell is monitored continuously. The opening and closing of the shutter is very fast ($\lesssim 1$ ms) with respect to the characteristic LIAD dynamics. The stems of the cell (which contain the alkali-metal samples) are not illuminated by the desorbing light.

In many of our experiments we evenly illuminate the entire cell with desorbing light by placing a corner reflector behind the cell (in place of the power meter shown in Fig. 3; the power meter is placed to the side of the cell in this case and the light beam is expanded to cover both the cell and the power meter). We also conducted several experiments in which we cooled the stem of a type (a) cell (Fig. 1) in order to investigate its role in the LIAD dynamics. In these experiments, the stems are placed in an insulated reservoir filled with ethanol chilled to $T \approx -10\,^\circ\text{C}$ by adding dry ice.

III. RESULTS

A. Time dependence of alkali vapor density

Figure 4 shows characteristic time dependences of the Rb vapor density in a cell of type (a) (Fig. 1) when the cell is exposed to 514-nm desorbing light. For these measurements, the corner reflector was used to uniformly illuminate the cell (see Sec. II B). The entire cell was at room temperature ($\approx 20\,^{\circ}\mathrm{C}$). As discussed in Sec. III B, depending on the light intensity and duration of exposure to desorbing light, it is necessary to wait for 45 min or longer between exposures in order to obtain repeatable measurements.

We identify several properties (illustrated in Fig. 4) of the time dependence of the vapor density $n(t)$ that can be used to characterize the LIAD dynamics: the initial density before the desorbing light is unblocked (n_0), the maximum increase in density ($\Delta n_{\max}$) when the cell is exposed to desorbing light, and the time ($\tau_{\max}$) for the density to reach its maximum value. In addition, we define the relative rate of increase of the vapor density in the cell at $t=+0$,

$$\gamma_0 = \frac{1}{n_0}\left(\frac{dn}{dt}\right)_{t=+0}. \tag{1}$$

We choose this particular set of parameters because they can be determined directly from the data in a model-independent way.

One of the features of the Rb-density time dependence not described by the chosen parameters is the decrease in the Rb density after it has reached its maximum value. The decrease in density while the cell is exposed to desorbing light indicates that LIAD depletes the paraffin of adsorbed atoms. If the flux of atoms desorbed from the paraffin were constant, the density of atoms in the cell would increase until reaching a new equilibrium value, then remain constant. Since the density falls after reaching a maximum, the flux of atoms desorbed from the paraffin must decrease as the cell continues to be exposed to desorbing light. Furthermore, there must be a sink for the desorbed atoms, which, presumably, is the stem of the cell, where the metal sample attempts to maintain a constant density through evaporation and condensation of Rb.

Another observation is that, for the case of higher light intensity (the lower plot of Fig. 4), the Rb density in the cell after the desorbing light is blocked reaches a value slightly lower than the initial density n_0. This, too, we believe is a result of LIAD depleting the paraffin of adsorbed atoms (see Sec. IV).

B. Repeated exposures to desorbing light

When a paraffin-coated cell is exposed to desorbing light, some of the alkali-metal atoms that have been adsorbed into the coating are released. As we have seen in Sec. III A, after a period of exposure to the desorbing light, the flux of atoms from the coating decreases due to depletion. If the desorbing light is blocked, then subsequently unblocked after a brief time (~ 10 min), we find that $\Delta n_{\max}$ is smaller than for the first exposure (see Fig. 5). However, for a cell at room temperature, if one waits ~ 45 min between exposures, repeatable results for $\Delta n_{\max}$ are obtained.

To study the effect of repeated exposures to desorbing light, we measured LIAD in the type (a) cell containing ^{85}Rb, and employed 514-nm desorbing light with intensity ≈ 5 mW/cm^2. The corner reflector is used to illuminate the entire cell.

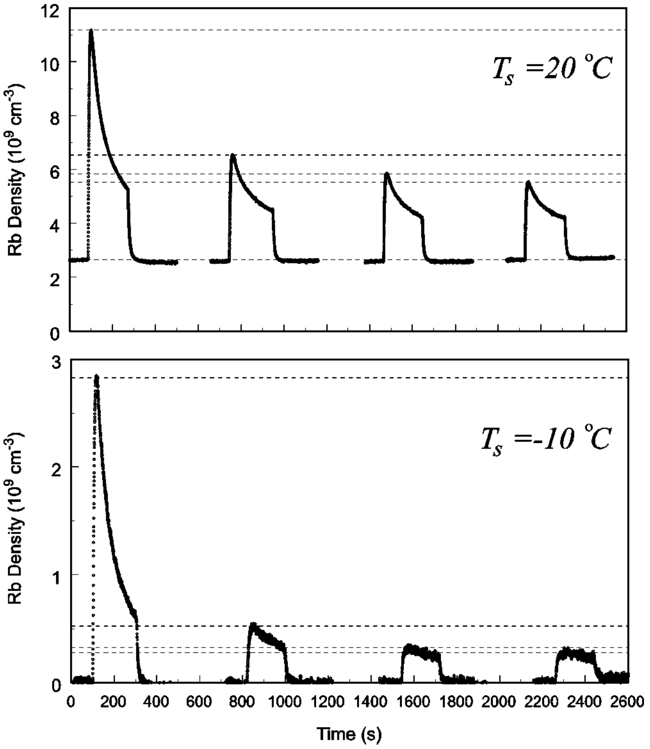

FIG. 5. Rb vapor density in a paraffin-coated cell of type (a) (see Fig. 1) as a function of time when the cell is repeatedly exposed to 514-nm light with intensity ≈ 5 mW/cm^2. The cell is completely illuminated by the desorbing light for these files. The upper plot shows data taken while the entire cell was at room temperature ($T_s \approx 20\,^{\circ}\mathrm{C}$), the lower plot shows data taken when stem of the cell was immersed in dry-ice-chilled ethanol ($T_s \approx -10\,^{\circ}\mathrm{C}$). Note that the vertical scale of the lower plot is four times smaller.

The first set of data (upper plot in Fig. 5) is obtained with the cell and stem at room temperature. The dashed horizontal lines designate the values of $n_0 + \Delta n_{\max}$ for each of the exposures, and the lowest dashed horizontal line designates n_0 prior to the first exposure. Note that n_0 for each of the exposures is approximately the same, so the vapor density in the cell returns to its initial value in between exposures. However, $\Delta n_{\max}$ becomes successively smaller for each exposure, suggesting that there are fewer atoms available to be desorbed from the coating. It is also interesting to note that LIAD can cause the alkali-metal density to greatly exceed the saturated vapor density at room temperature ($\approx 5 \times 10^9$ atoms/cm^3). This shows that the light does not merely prevent adsorption of atoms into the coating, but rather causes atoms inside the coating to be released into the cell.

The lower plot of Fig. 5 shows the Rb vapor density during repeated exposures to desorbing light while the stem is kept at a temperature of $T_s \approx -10\,^{\circ}\mathrm{C}$. The dry-ice-chilled ethanol was added to the insulated reservoir in which the stem was placed ≈ 5 min prior to the first exposure to desorbing light. In this case n_0 is much smaller, since the Rb vapor density in the stem is ≈ 50 times smaller than at room temperature. The average temperature of the outer surface of the rest of the cell remained constant within ~2 °C (measured with a thermocouple). It should be noted that the slight change of coating temperature could affect the mobility of atoms trapped inside the paraffin.

The observation of LIAD when the stem of the cell is cooled confirms that the effect of the light is not to merely prevent the adsorption of atoms into the coating, since such an effect would cause no significant change in the vapor density in the cell in this case. Therefore we conclude that the primary mechanism causing the increase in the vapor density is indeed the desorption of atoms from the paraffin coating.

Another important feature of the data shown in Fig. 5 is that for the room-temperature case, $n_0+\Delta n_{\max}$ always exceeds the vapor density in the cell just prior to blocking the desorbing light for the previous exposure, while in the case of the cold stem, $\Delta n_{\max}$ is always about the same as the vapor density in the cell just prior to blocking the desorbing light for the previous exposure. This is clear evidence that in the room-temperature case, the coating is replenished by absorbing atoms evaporated from the metal samples in the stem.

Note that $\Delta n_{\max}$ for each of the exposures for $T_s \approx -10\,^\circ\mathrm{C}$ is considerably less than that obtained when the stem was at room temperature. This difference is primarily due to the fact that the light-induced atomic flux $\Phi(t)$ from the coating is smaller when $T_s \approx -10\,^\circ\mathrm{C}$ [39]. We can estimate the atomic flux from the coating at $t=0$ using the relation [see Eq. (1)]

$$\Phi(0)=n_0 V \gamma_0 = V\left(\frac{dn}{dt}\right)_{t=+0}, \tag{2}$$

where V is the volume of the cell. This relation holds since just prior to $t=0$, the system is in equilibrium, so that $dn/dt=0$. Thus the sudden change in dn/dt can be attributed entirely to the light-induced flux. We find that for the initial exposure taken with the stem held at room temperature,

$$\Phi(0)\approx 47\times 10^9 \ \text{atoms/s}, \tag{3}$$

while for the cooled stem,

$$\Phi(0)\approx 9\times 10^9 \ \text{atoms/s}. \tag{4}$$

This shows that the reduction in the number of atoms available to be desorbed by light happens before the cell is illuminated.

We have also taken data (not shown) where we vary the length of time (from a few minutes to over an hour) for which the stem is chilled before the cell is exposed to desorbing light. After the initial reduction in $\Phi(0)$ [compared to when the stem is at room temperature, see Eqs. (3) and (4)] over the first few minutes, the initial flux becomes relatively independent (to within about 10%) of the length of time for which the stem has been chilled. Thus a certain fraction of the atoms inside the paraffin remain available to be desorbed, i.e., they are not depleted when the influx of new atoms into the coating ceases. As discussed in Secs. III F and IV, perhaps a certain fraction of the adsorbed atoms are "mobile" within the paraffin only in the presence of light (i.e., some of the atoms are more tightly bound inside the paraffin than others). Such an effect may also explain why in the lower plot of Fig. 5, $\Delta n_{\max}$ levels off at a nonzero value.

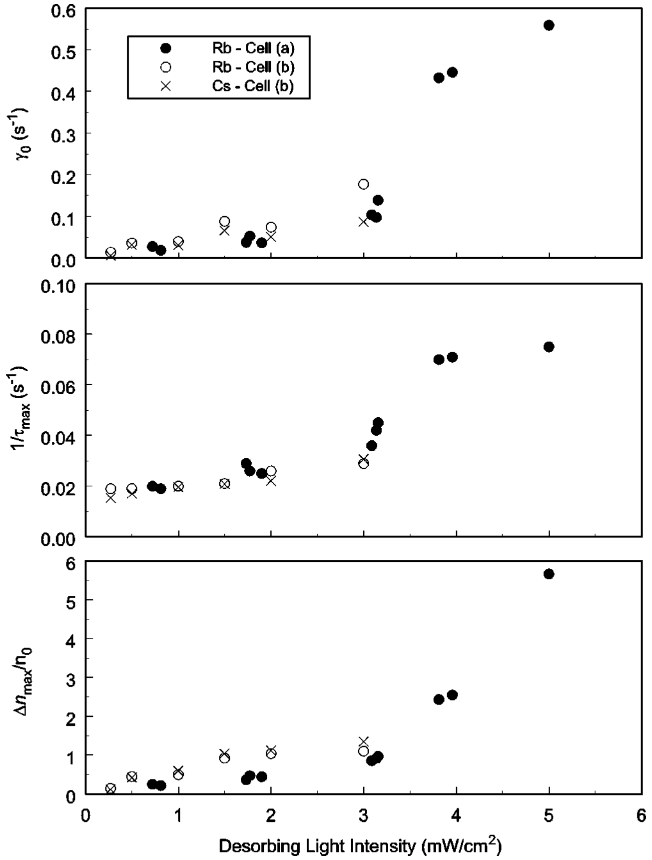

FIG. 6. Results of measurements of γ_0, $\tau_{\max}{}^{-1}$, and $\Delta n_{\max}/n_0$ as a function of 514-nm desorbing light intensity for alkali-metal vapor contained in type (a) and type (b) cells (see Fig. 1). Cells are kept at room temperature ($\approx 20\,^\circ\mathrm{C}$). The type (a) cell contains only Rb, while the type (b) cell contains both Rb and Cs. Uncertainty in the determination of γ_0, $\tau_{\max}{}^{-1}$, or $\Delta n_{\max}/n_0$ for a given measurement is about the size of the data point on the plot. An estimate of the reproducibility of the data can be inferred from the spread of points at a given desorbing light intensity. Measurements for Rb and Cs in the type (b) cell were performed simultaneously. For these measurements, the initial density n_0 for Cs was $\approx 13\times 10^9$ atoms/cm^3 and it was $\approx 2\times 10^9$ atoms/cm^3 for Rb.

C. LIAD as a function of desorbing light intensity

Figure 6 shows γ_0, $\tau_{\max}{}^{-1}$, and $\Delta n_{\max}/n_0$ (defined in Sec. III A, see Fig. 4) as a function of desorbing light intensity. The measurements were taken for two different types of cells [types (a) and (b), Fig. 1], and Fig. 6 compiles data taken on three different days. Recall that the type (a) cell contains only ^{85}Rb, while the type (b) cell contains Cs in one stem and ^{85}Rb in another stem. The desorbing light is generated by an Ar$^+$-laser at 514 nm, and the beam is expanded so that each cell is entirely illuminated. Each cell is at room temperature ($\approx 20\,^\circ\mathrm{C}$). There is a delay of at least 45 min between each exposure to desorbing light, during which the cells are kept in the dark, allowing the paraffin coating to be replenished. In order to further minimize the possibility of systematic errors due to depletion of the coating, data for different light intensities is acquired in a nonsequential manner.

The measurements of LIAD in the type (b) cell for Rb and Cs were taken simultaneously using the setup shown in Fig. 3. Despite their substantially different geometries and the fact that the type (b) cell contains both Rb and Cs, the data for the two different types of cells is relatively similar. Moreover, the values of γ_0, $\tau_{\max}{}^{-1}$, and $\Delta n_{\max}/n_0$ are very similar for both Rb and Cs (Fig. 6). This suggests that LIAD is generally defined by the properties of the coating.

The close correspondence of $\Delta n_{\max}/n_0$ at each light power for Rb and Cs suggests that the difference between $\Delta n_{\max}$ for Rb and Cs is related to the difference in n_0 for the two species.

Furthermore, we find that for the range of light intensities for which we have investigated LIAD of both Cs and Rb ($\lesssim 3$ mW/cm^2; Fig. 6) the ratio of the initial light-induced fluxes for the two species approximately equals the ratio of their initial densities:

$$\frac{\Phi(0)_{\mathrm{Rb}}}{\Phi(0)_{\mathrm{Cs}}} \approx \frac{n_0(\mathrm{Rb})}{n_0(\mathrm{Cs})}. \tag{5}$$

If one supposes that the light-induced desorption rate for both species is the same (as our data suggests), then Eq. (5) implies that the majority of the initial number of atoms available for desorption from the coating at room temperature is proportional to n_0. However, as previously discussed in Sec. III B, a certain fraction of atoms remain in the coating even when $n_0 \approx 0$ for a few hours (observed with the stem chilled).

Note that $\gamma_0 \rightarrow 0$ as the desorbing light intensity approaches zero. However, $\tau_{\max}{}^{-1} \rightarrow 0.02\ \mathrm{s}^{-1}$ as the light intensity approaches zero, suggesting that for sufficiently low light intensities ($\lesssim 3$ mW/cm^2), other factors (such as the exchange of atoms between the volume of the cell and the stems and/or light-independent atomic adsorption by the coating) determine $\tau_{\max}$.

At present, we do not have a conclusive explanation for the nontrivial dependence of γ_0, $\tau_{\max}{}^{-1}$, and $\Delta n_{\max}$ on light power, but we suspect that it may be related to the different ways in which atoms can be bound inside the paraffin coating and/or the onset of the regime where light-assisted diffusion becomes important (discussed in Secs. III B and IV).

D. LIAD as a function of desorbing light frequency

As has been observed in studies of LIAD from other materials [14–18,20,23], we found that γ_0 and $\Delta n_{\max}$ for both Rb and Cs increase with desorbing photon energy (Fig. 7). The data in Fig. 7 were taken at low light powers to minimize the depletion of adsorbed atoms from the coating during the measurements, and there was at least 45 min delay between each exposure to desorbing light.

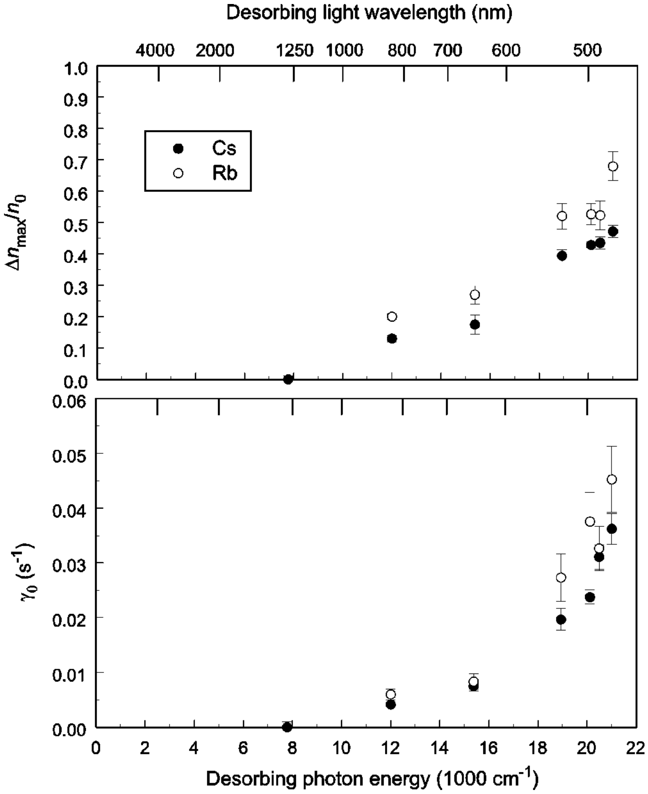

FIG. 7. $\Delta n_{\max}/n_0$ and γ_0 as a function of desorbing photon energy for Cs and Rb. The data were taken at the same light intensity (0.56 mW/cm^2) and cross-sectional beam area (≈ 18 cm^2), except for the data at 650 nm (15 400 cm^{-1}), which was taken at 0.28 mW/cm^2 and rescaled (Fig. 6 gives the intensity dependences). The data at the four highest photon energies were obtained with the Ar$^+$ laser, while data at 650 nm, 833 nm (12 000 cm^{-1}), and 1285 nm (780 cm^{-1}) were obtained with external-cavity diode lasers. All data were taken with the type (b) cell.

Also, as is the case for LIAD from sapphire surfaces [14,15] and from silane coatings [20], there is a threshold in photon energy for the desorbing light—we do not observe LIAD using light at 1285 nm, while LIAD is observed at the other wavelengths we employed.

E. Comparison of LIAD in different cells

Figure 8 shows the time dependences of Cs vapor density in four available paraffin-coated cells [three type (c) cells, and one type (b) cell] when they were exposed to a beam of 514-nm desorbing light with intensity ≈ 100 mW/cm^2 generated by an Ar$^+$ laser. The light beam had a cross-sectional area of ≈ 8 cm^2, so it did not fully illuminate any of the cells. The measurements were all performed with the same setup by merely swapping the cells. Some relevant parameters are listed in Table I.

Even though the same technique is used for preparation of each of the cells, there is a wide variation of the initial vapor density n_0 among the cells. The cell with the largest initial density is the one containing only Cs, which suggests that there may be additional suppression of the alkali-metal vapor

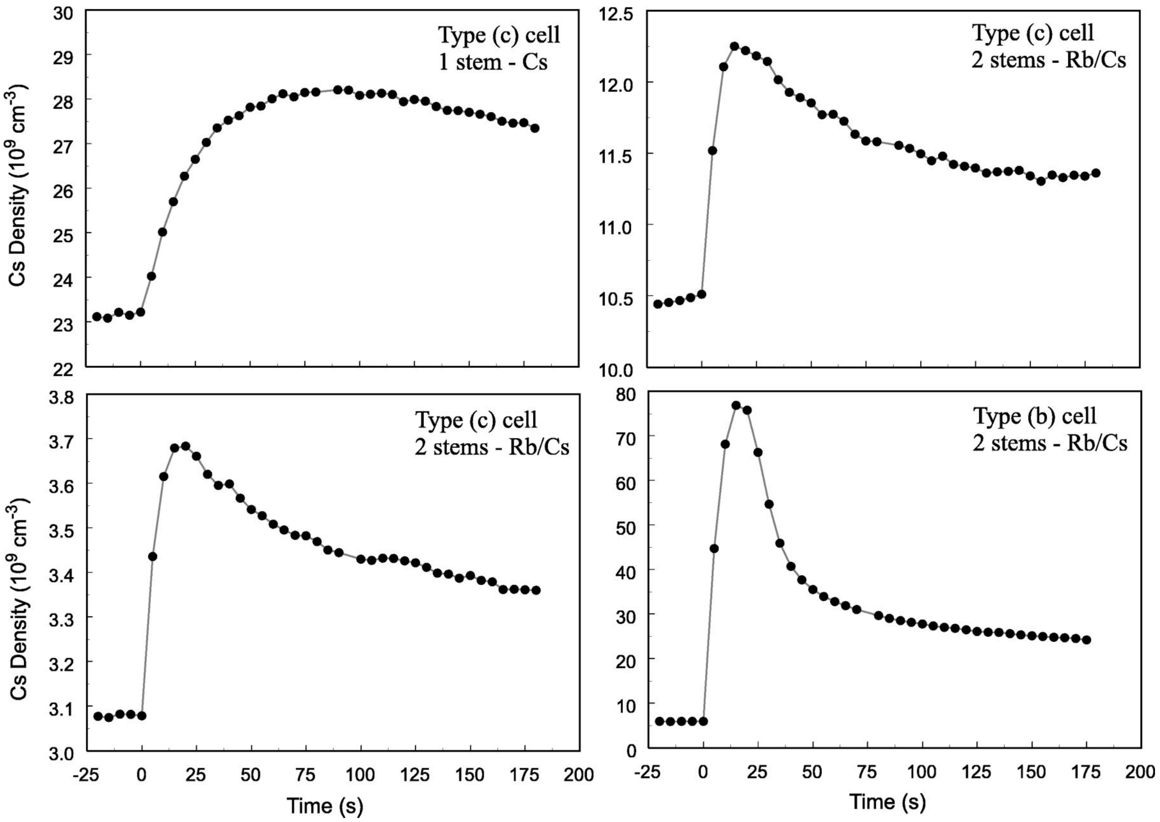

FIG. 8. Cs vapor density as a function of time in four different cells. Each cell is exposed to a beam of 514-nm desorbing light with intensity ≈ 100 mW/cm^2 and cross-sectional area ≈ 8 cm^2. The desorbing light is turned on at $t=0$ in each case. Note that the vertical scale on each of the plots is different.

densities in cells containing both Rb and Cs. As mentioned earlier, a possible explanation for this would be migration of atoms between the stems, leading to the formation of a Rb-Cs amalgam.

In spite of the great variation of n_0 between the cells, the ratio $\Delta n_{\max}/n_0$ for each of the type (c) cells is quite similar. We have already seen that the same result holds for Rb and Cs contained in the type (b) cell at significantly lower light intensity (Fig. 6). The observed difference between $\Delta n_{\max}/n_0$ for the type (c) cells and that obtained for the type (b) cell suggests that, for a given cell temperature, desorbing light beam area, intensity, and frequency, $\Delta n_{\max}/n_0$ is primarily a function of the cell geometry (its volume, surface area, number of stems, etc.).

The decline in density after $\tau_{\max}$ differs for each of the cells. For the type (c) cells, those with two stems have faster rates of decline than the cell with only one stem. Since the stems act as sinks for the Cs atoms when the density in the volume of the cell exceeds that in the stems, the difference can be explained by the differences in the cell volumes and numbers of stems.

The variation in $\Phi(0)$ between the cells could be due to, for example, differences in the coating thicknesses or the histories of the cells [at the time of the measurement the type (b) cell was about 1 yr old and the type (c) cells were 6 months old, and the type (b) cell had electric fields of up to 5 kV/cm applied to it during the investigation of nonlinear electro-optic effects [9,13]].

TABLE I. Initial density n_0 of Cs, maximum change in density over the initial density $\Delta n_{\max}/n_0$, γ_0, $\tau_{\max}{}^{-1}$, and initial flux $\Phi(0)$ for four different paraffin-coated cells illuminated by light at 514 nm with intensity ≈ 100 mW/cm^2. The cross-sectional area of the light beam is ≈ 8 cm^2. Also listed are the volumes V of the cells and the ratio of the cells' inner surface area to the illuminated area $A_i \approx 16$ cm^2 (twice the cross-sectional area of the beam since the light illuminates the front and back faces).

Cell type	Stems	n_0 (cm^{-3})	$\Delta n_{\max}/n_0$	γ_0 (s^{-1})	$1/\tau_{\max}$ (s^{-1})	$\Phi(0)$ (atoms/s)	V(cm^3)	A/A_i
(c)	1, Cs	23.1×10^9	0.22	0.008	0.011	7×10^{10}	360	17.5
(c)	2, ^{87}Rb and Cs	10.5×10^9	0.17	0.019	0.062	7×10^{10}	370	18.1
(c)	2, ^{85}Rb and Cs	3.7×10^9	0.20	0.019	0.059	3×10^{10}	370	18.1
(b)	2, ^{85}Rb and Cs	5.9×10^9	12.2	1.34	0.059	24×10^{10}	30	3.9

Finally, it is interesting that ${\tau_{\max}}^{-1}$ is similar for all cells with two stems containing Cs and Rb, while the one-stemmed cell with only Cs reaches the maximum density much more slowly.

F. LIAD with pulsed desorbing light

The light-induced desorption of atoms from paraffin coating by 532-nm pulsed light from a Nd:YAG (yttrium aluminum garnet) laser were investigated with the type (b) cell containing ^{85}Rb and Cs. The Nd:YAG laser operated at a repetition rate of 10 Hz, the light pulses directed at the cell had energy ≈ 2 mJ and duration ≈ 10 ns. The cross-sectional area of the beam was $\approx 2.3\ \mathrm{cm}^2$. We found that $\Delta n_{\max}/n_0 \approx 0.3$ for both Rb and Cs. This result can be compared to the cw case with equivalent average power (20 mW), beam area, and wavelength, for which we have determined $\Delta n_{\max}/n_0 \approx 3.0$ from measurements. The order-of-magnitude difference between $\Delta n_{\max}/n_0$ for the cw and pulsed cases demonstrates that the desorption of atoms from the paraffin depends not only on the total light power but also on the length of time for which the light interacts with the coating. This suggests that diffusion of atoms inside the coating plays an important role in LIAD. These observations support the view that a certain fraction of the adsorbed atoms are mobile inside the paraffin only when the coating is exposed to light, as suggested in Sec. III B.

IV. INTERPRETATION

A. Overview of observations

Based on the experimental observations discussed in the preceding section, in this section we develop a simple picture of the physical processes involved in the light-induced desorption of alkali-metal atoms from paraffin coatings.

We have found that the increase in the alkali-metal vapor densities when paraffin-coated cells are exposed to light is primarily due to light-induced desorption of atoms from the coating (see Secs. III A and III B). For example, the light-induced increase in vapor density when the stem was immersed in dry-ice-chilled ethanol can only be explained by a flux of atoms from the coating (Fig. 5). Therefore, we suppose that there is a population of alkali atoms N_c within the coating (near the surface), which are released at a rate γ into the volume of the cell when it is exposed to light.

As can be seen from the data in Secs. III C and III D, γ depends on the light intensity (Fig. 6) and frequency (Fig. 7). There are several proposals to explain the dependence of LIAD on the frequency of the desorbing light. References [15,16] suggest that the frequency dependence is related to the absorbtion spectrum of adsorbed atoms. It is possible that if an atom in the coating absorbs a photon, its mobility is increased via superelastic collisions [40]. Another idea is that the alkali-metal atoms form quasimolecular bonds with the paraffin, and the photon energy must exceed some threshold binding energy. In spite of the fact that we have observed a threshold for the necessary photon energy for LIAD, it is still difficult to distinguish between these various possibilities with our present data.

The decrease in $\Delta n_{\max}$ after repeated exposures to desorbing light (Sec. III B, Fig. 5) shows that the coating is depleted of atoms by the light. When the stem of the cell is at room temperature, the coating is replenished after about 45 min. However, when the stem is immersed in dry-ice-chilled ethanol, we observe no recovery of the coating. Thus we conclude that the coating is replenished by atoms evaporated from the stems. Hence, the coating must adsorb atoms: i.e., when an atom collides with the wall, there is some probability ρ for an atom to become trapped inside the paraffin.

Furthermore, the vapor density (in the dark) in the cell volume does not change appreciably between exposures to desorbing light (Fig. 5), even though N_c has changed considerably (as evidenced by the successively smaller values of $\Delta n_{\max}$ for repeated exposures to desorbing light of the same frequency and intensity). This indicates that the number of alkali atoms in the coating N_c only weakly affects the flux of atoms into the coating in the absence of light. This agrees with previous observations that the coating continuously adsorbs atoms at a relatively constant rate [2,28,30,31]. Therefore, we assume that the adsorption probability ρ is light independent. The slight depression of the equilibrium vapor density after the desorbing light has been turned off, evident in the lower plot of Fig. 4, can be explained by the existence of a small rate of light-independent desorption of atoms from the coating γ_d (leading to some small flux of atoms from the coating, $\gamma_d N_c$, even in the absence of light).

How can it be that the coating always adsorbs atoms at a relatively constant rate, yet it seems to "fill up" ~ 45 min after being depleted? There must be a sink for atoms that have been adsorbed into the coating. Perhaps atoms stick irreversibly to the glass surface, or they diffuse inside the coating until they encounter an "impurity" in the paraffin (for example, dangling bonds buried beneath the surface of the coating). In either case, the criterion for replenishment of the coating is that the flux of atoms adsorbed into the coating ($\rho n \bar{v} A/4$) must be balanced by the flux of atoms to the sink, where n is the vapor density of atoms in the volume of the cell, $\bar{v} \approx 2 \times 10^4$ cm/s is the average atomic velocity in the volume of the cell, and A is the inner surface area of the cell. Then the overall system (stem, cell volume, coating) is in equilibrium. This explains the observation that the flux from the coating always seems to be proportional to the equilibrium density n_0 of the atomic species in the cell in the absence of light (Sec. III C). We assume that the flux of atoms into the sink is given by ΓN_c, so when this flux balances $\rho n_0 \bar{v} A/4$, the system is in equilibrium and we find that $N_c \propto n_0$.

The idea that there is a mechanism causing "free" (mobile) atoms diffusing in the coating to be irreversibly lost is supported by the fact that the initial light-induced flux from the coating is suppressed by a factor of ~ 5 when the stem has been immersed in dry-ice-chilled ethanol for a few minutes before being exposed to light (Sec. III B). This indicates that N_c decreases when the flow of atoms into the coating is stopped. Light-independent desorption from the coating also contributes to this depletion. But, as we discussed in Sec. III B, neither of these processes is capable of fully depleting the coating of atoms which can be desorbed by light.

Finally, we find from the data taken with the cell's stem immersed in dry-ice-chilled ethanol that there appears to be a certain fraction of atoms inside the coating that are neither noticeably depleted by LIAD nor by other mechanisms. Therefore, we also suppose there is a light-dependent flux of atoms Φ_c from this seemingly inexhaustable source. This flux could result from light-induced diffusion of atoms which are deep inside the coating. Φ_c is independent of N_c, and, for most of our data (the exception being the data shown in the lower plot of Fig. 5), the contribution of Φ_c to LIAD is small. In principle, if the "free" alkali-metal atoms are trapped in potential wells with random depths ΔE, which in disordered systems are often distributed according to an exponential law, such behavior (where some of the trapped atoms are desorbed quickly while others are desorbed very slowly) may be expected. In such cases it is necessary to invoke Lévy statistics (see, for example, Ref. [41–43]) to describe the system dynamics. In general, this analysis leads to a continuous distribution of rates, but for simplicity we assume two discrete subsamples of atoms.

B. Model

The basic set of ideas discussed above can be collected in a simple model based on rate equations which describe the time-dependent number of atoms in three regions: the stem, the volume of the cell, and the paraffin coating. We assume that the vapor density in the stem of the cell, n_s, is constant, and that there is an exchange rate between the stem and the volume of the cell described by a constant ξ (which has units of cm^3/s). The flux of atoms adsorbed into the coating is given by $\rho n(t)\overline{v}A/4$. The number of "free" atoms in the coating is $N_c(t)$, and the atoms in the coating are irreversibly lost at a rate of Γ to the glass or impurities. There is also a relatively small, light-independent flux of atoms from the coating $\gamma_d N_c(t)$.

Our data have indicated that the "free" atoms diffuse through the coating, and in fact the diffusion rates in the presence of light and in the absence of light are probably different. A more accurate description of LIAD should be possible by solving the diffusion equation for the alkali-metal atoms in the coating, as is done in Ref. [21].

Using these approximations, the rate equations in the absence of light are given by

$$V\frac{d}{dt}n(t) = -\rho\frac{\overline{v}A}{4}\,n(t) + \xi[n_s - n(t)] + \gamma_d N_c(t),$$

$$\frac{d}{dt}N_c(t) = +\rho\frac{\overline{v}A}{4}\,n(t) - \Gamma N_c(t) - \gamma_d N_c(t). \tag{6}$$

When the desorbing light is turned on, there is an additional flux of atoms from the coating into the volume of the cell, $\Phi(t) = \gamma N_c(t) + \Phi_c$, where Φ_c is a term describing a time-independent light-induced flux (relatively small compared to $\gamma N_c(t)$ for the data taken with the cell and stem at room temperature). So with the desorbing light on, the rate equations are

$$V\frac{d}{dt}n(t) = -\frac{\rho\overline{v}A}{4}\,n(t) + \xi[n_s - n(t)] + (\gamma+\gamma_d)N_c(t) + \Phi_c,$$

$$\frac{d}{dt}N_c(t) = \frac{\rho\overline{v}A}{4}\,n(t) - \Gamma N_c(t) - (\gamma+\gamma_d)N_c(t). \tag{7}$$

According to Eq. (6), in equilibrium prior to exposure to the desorbing light, the initial density in the cell $n_0 \equiv n(0)$ is given by

$$n_0 = \frac{(\Gamma+\gamma_d)\xi}{(\rho\overline{v}A\Gamma)/4 + (\Gamma+\gamma_d)\xi}\,n_s, \tag{8}$$

and the initial number of atoms in the coating near the surface is

$$N_c(0) = \frac{\rho\overline{v}A}{4(\Gamma+\gamma_d)}\,n_0. \tag{9}$$

From Eq. (7), we find that the initial light-induced flux from the coating $\Phi(0)$ [Eq. (2)] is given by

$$\Phi(0) = \gamma N_c(0) + \Phi_c = \frac{\gamma}{\Gamma+\gamma_d}\,\frac{\rho\overline{v}A}{4}\,n_0 + \Phi_c, \tag{10}$$

and the relative rate of increase of the vapor density in the cell at $t=0$ [Eq. (1)] is given by

$$\gamma_0 = \frac{\gamma}{\Gamma+\gamma_d}\,\frac{\rho\overline{v}A}{4V} + \frac{\Phi_c}{n_0 V}. \tag{11}$$

Equation (10) shows a linear dependence of $\Phi(0)$ on n_0, which [recalling that $\Phi_c \ll \Phi(0)$], is consistent with observations described in Sec. III C [Eq. (5)]. Also, we see that γ_0 is inversely proportional to the volume, which accounts for some of the difference between γ_0 for the type (b) and type (c) cells (Sec. III E). The rest of the difference is most likely attributable to the difference between the ratio of the total surface area to the illuminated surface area A/A_i (see Table I) [44].

Figures 9 and 10 show the model's predictions for the density of atoms in the vapor cell and the number of atoms in the coating as a function of time when the cell is repeatedly exposed to desorbing light. The data from Sec. III B (Fig. 5) is also shown for comparison. The parameters listed in the captions of Figs. 9 and 10 are chosen to obtain best agreement with the data. We have also used the model to fit other relevant data [45], and the average values of some of the fitted parameters for the type (a) and (b) cells are listed in Table II. In spite of the simplicity of the model and differences between the coatings and geometries of the cells, the values of light-independent parameters $(\Gamma, \gamma_d, \rho, \xi)$ are in general agreement for the two cells.

We find a roughly linear dependence of γ on light intensity (Fig. 11). Knowledge of the light-induced desorption rate γ allows one to estimate a cross section σ for LIAD:

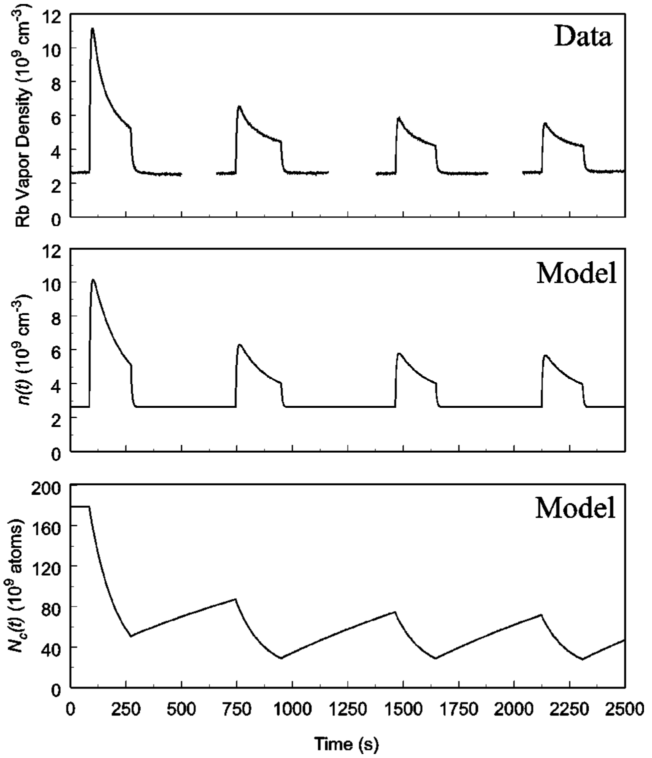

FIG. 9. Upper plot shows Rb vapor density in a paraffin-coated cell of type (a) (see Fig. 1) as a function of time when the cell is repeatedly exposed to 514-nm light with intensity ≈ 5 mW/cm^2, with the stem of the cell at room temperature—this is the same data shown in the upper plot of Fig. 5. The middle plot shows the predictions of the model for the time-dependent vapor density in the cell $n(t)$ for the following parameters (see text for definitions): $\gamma \approx 0.01$ s^{-1}, $\Gamma \approx 5\times10^{-4}$ s^{-1}, $\gamma_d \approx 2\times10^{-4}$ s^{-1}, $\rho \approx 0.14\times10^{-6}$, $\xi \approx 0.15$ cm^3/s, and $\Phi_c \approx 10^9$ atoms/s. The lower plot shows the number of atoms in the coating $N_c(t)$ as a function of time.

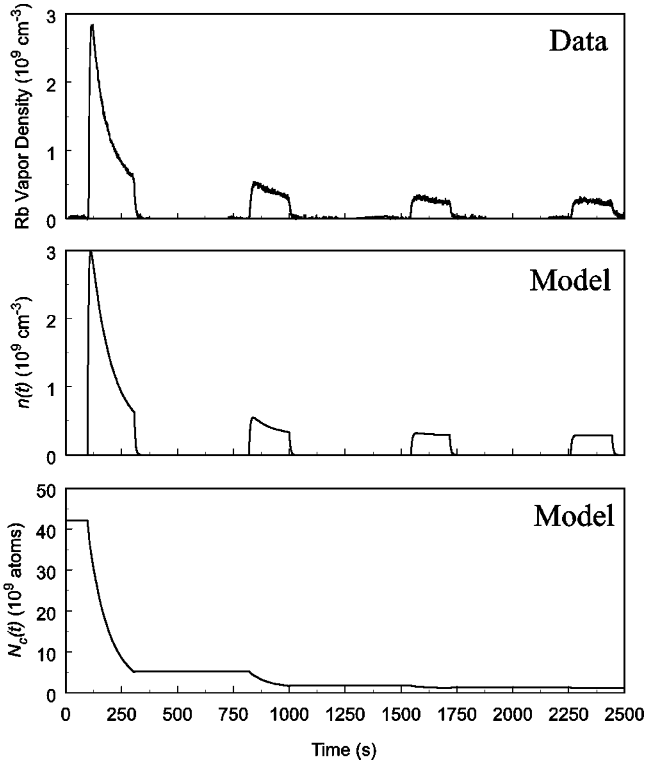

FIG. 10. Upper plot shows Rb vapor density in a paraffin-coated cell of type (a) as a function of time when the cell is repeatedly exposed to 514-nm light with intensity ≈ 5 mW/cm^2, with the stem of the cell immersed in dry-ice-chilled ethanol—this is the same data shown in the lower plot of Fig. 5. The middle plot shows the predictions of the model for the time-dependent vapor density in the cell $n(t)$ for the same parameters as in Fig. 9. The lower plot shows the number of atoms in the coating $N_c(t)$ as a function of time.

$$\sigma \approx \frac{\gamma\hbar\omega}{I}, \tag{12}$$

where I is the desorbing light intensity and ω is the desorbing light angular frequency. We find that $\sigma \sim 10^{-18}$ cm^2, similar to cross sections for LIAD from other materials (for example, for Na atoms on sapphire it was also found that $\sigma \sim 10^{-18}$ cm^2 [14]).

We can also estimate the vapor density in the stems n_s from our fits [Eq. (8)], which we find to be $\approx$25–50 % less than the saturated vapor density [29], varying from cell to cell. This suppression may be due to the metal stems becoming coated with paraffin or the products of chemical reactions occurring during the ripening process.

From the prediction of the model for $N_c(t)$ (shown in the lower plot of Fig. 9) it is clear why the coating takes around 45 min to recover after being exposed to light. The vapor density in the cell recovers relatively quickly to its initial value n_0. From then on, according to Eq. (6), the density in the coating is governed by the differential equation

$$\frac{d}{dt}N_c(t) = \rho\frac{\overline{v}A}{4}n_0 - (\Gamma+\gamma_d)N_c(t), \tag{13}$$

whose solution is

$$N_c(t) = N_c(0) - \Delta N_c e^{-(\Gamma+\gamma_d)(t-t_0)}, \tag{14}$$

where ΔN_c describes the initial depletion of the coating and t_0 is the time at which the desorbing light is blocked. From our data $(\Gamma+\gamma_d)^{-1} \approx 24$ min, which gives the scale for the coating recovery time. For $N_c(t)$ shown in Fig. 9, $N_c(0) \approx 1.8\times10^{11}$ atoms and $\Delta N_c \approx 1.4\times10^{11}$ atoms after the first exposure. In this case, after 48 min of recovery in the dark, N_c would be $\approx$90% of its initial value.

The probability for adsorption of atoms by the coating per wall collision [46], $\rho = 0.15(4)\times10^{-6}$ [average of data from

TABLE II. Average values of various light-independent parameters from the model extracted from fits to the data from Secs. III B, III C, and III D.

Parameter	Type (a) cell	Type (b) cell
Γ	$5(2)\times10^{-4}$ s^{-1}	$7(4)\times10^{-4}$ s^{-1}
γ_d	$2(1)\times10^{-4}$ s^{-1}	$2(1)\times10^{-4}$ s^{-1}
ρ	$1.7(1)\times10^{-7}$	$1.4(6)\times10^{-7}$
ξ	0.151(5) cm^3/s	0.045(1) cm^3/s

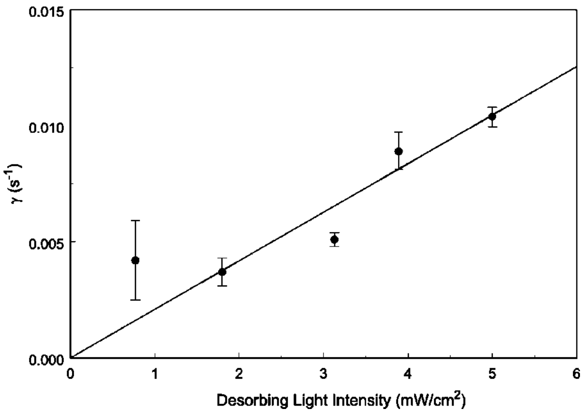

FIG. 11. Fitted value of γ as a function of 514-nm desorbing light intensity for Rb vapor contained in a type (a) cell. The cell and stem are at room temperature ($\approx$20 °C).

both type (a) and (b) cells], is in good agreement with the results of Refs. [2,28,30,31], in which ρ was determined by completely different means.

If one assumes that the alkali-metal atoms within the coating are trapped inside potential wells created by the paraffin molecules, one can perform a crude estimate of the average depth ΔE of the potential wells from the fitted value of $\gamma_d \approx 2\times10^{-4}$ s^{-1}. Supposing that $\gamma_d \approx \omega_v e^{-\Delta E/kT}$, where $\omega_v \sim 10^{11}$ s^{-1} is a rough estimate of the vibrational frequency for the alkali-metal–paraffin quasimolecule (see, for example, Refs. [2,47]), we find that $\Delta E \sim 1$ eV. It is interesting that this is approximately equal to the measured threshold in desorbing photon energy (Fig. 7).

Finally, we note that the initial number of atoms in the coating, $N_c(0) \sim 2\times10^{11}$, is very small compared to the number of atoms absorbed by the coating during the cell ripening process (discussed in Sec. II A). During the ripening process, since the vapor density in the volume of the cell is negligible, we can assume that the coating is absorbing atoms as fast as the stems release them: $\xi n_s \sim 10^{11}$ atoms/s (for a cell temperature of 80 °C, where ξ is obtained from the results of our fitting). The cell is generally kept at this temperature for 10 h, which means that the coating absorbs $\sim 4\times10^{15}$ atoms during the ripening process. The number of atoms N_c available for desorption is very small compared to this number, which suggests that the atoms which "ripen" the coating are absorbed irreversibly. This would agree with the hypothesis that ripening consists of a chemical reaction between the alkali atoms and impurities in the coating (or perhaps gaseous impurities). This large amount of atoms inside the coating can also provide the "inexhaustable" source for Φ_c.

V. CONCLUSION

In conclusion, we have made an observation of nonthermal, light-induced desorption of Cs and Rb from paraffin coatings. We have measured the properties of the effect as a function of desorbing light frequency and intensity, observed the effect in several different cells (with the same type of paraffin coating), and compared the effect for cw and pulsed desorbing light.

In combination with previous observations [14–26], our work confirms that this effect is rather general in nature and not restricted to a specific type of wall coating. Light-induced atomic desorption from paraffin is of particular interest because of the very long spin-relaxation times obtainable in paraffin-coated cells, which makes them useful for a wide variety of applications [1–13].

There are a number of interesting topics to be addressed in future studies, such as the effect of nonresonant desorbing light on relaxation of atomic polarization in wall collisions, and light-induced desorption of other alkali atoms from paraffin, in particular, potassium, which has a low vapor pressure at room temperature and is widely used in magnetometry [3,4].

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[38] Another possible explanation for the ripening is that there is some residual gas in the cell with which the alkali-metal atoms react.
[39] This difference cannot be explained by the change in the alkali-metal vapor pressure in the stem, because at room temperature n_0 is only about 10% smaller than the vapor density in the stem n_s (see Sec. IV). Therefore, for a given Δn, the net flux of atoms into the stem is approximately the same regardless of n_s (we find that this effect can only explain a difference in $\Delta n_{\max}$ between the two cases of $\approx 10^9$atoms/cm^3).
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[44] The present model is only valid for the case of a cell that is totally illuminated by the desorbing light.
[45] Here we compare the fitted parameters for the data from Secs. III B, III C, and III D. For this data, the entire cell is illuminated and light intensities were relatively low ($\lesssim 6$ mW/cm^2).
[46] In Refs. [2,28,30,31], it was found that ρ was temperature dependent, so our result is only valid for when the coating is at room temperature.
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