

Vibrational cooling of cesium molecules using noncoherent broadband light

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We demonstrate selective vibrational population transfer in cold cesium dimers using a simple approach based on the use of a shaped incoherent broadband diode laser near threshold. Optical pumping into a single vibrational level is accomplished with an incoherent light source by eliminating transitions from the targeted vibrational level. The broadband spectrum of the laser is wide enough to electronically excite several vibrational states of the molecule simultaneously. This method is relatively inexpensive, simple, and flexible to allow for development of new applications, in particular, the preparation of optically closed molecular system, opening the way to direct laser cooling of molecules.

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Population transfer in quantum systems is essential for a variety of fields, such as precision spectroscopy, quantum computing, control of molecular dynamics, and chemical reactions [1–3]. In the domain of cold molecules, recent activities related with controlling both the internal and external degrees of freedom of a molecule are expected to lead significant advances in collision dynamics of chemical reactions, molecular spectroscopy, molecular clocks, fundamental test in physics, controlled photochemistry studies, and quantum computation [4–8]. Moreover, several theoretical approaches have been proposed to control the internal degrees of freedom of a molecule [7,9–12].

Preparation of molecules in their ground vibrational state using optical pumping has been recently demonstrated in our laboratory with the use of a shaped femtosecond mode-locked laser [13]. This work has been extended to different target vibrational levels, with the use of a liquid-crystal spatial light modulator (SLM) [14]. In both experiments, the population transfer is achieved by performing amplitude shaping of the pulse in the frequency domain. These results can find further exciting applications as far as some limitations can be confronted, namely, the high complexity and cost of the setup as well as the limited extinction ratio of the SLM [15,16].

In this Rapid Communication we describe a pair of experiments demonstrating the optical pumping of diatomic molecules to a desired target vibrational state of the electronic ground state using an incoherent broadband diode laser (BDL) operating near threshold. Cs₂ dimers are created in a mixture of vibrational levels of the ground molecular state through photoassociation of laser cooled Cs atoms. In the first experiment, filters diminish the light intensity corresponding to transitions originating from the target vibrational state, while addressing the other vibrational levels. The result is vibrational cooling of a fraction of the population to the $v=0$ state. The second experiment uses an optical setup with

a reflection grating to remove the light corresponding to transitions from the $v=1$ state, resulting in optical pumping into that state.

The experiment is conducted in a Cs vapor loaded magneto-optical trap (MOT), formed with $\sim 10^7$ atoms at a temperature of ~ 100 μ K and at a peak density of 10^{11} cm⁻³. In the cold atomic sample, molecules are formed via photoassociation (PA), a process in which two colliding atoms resonantly absorb a photon and form an excited molecule, which then decays either into a bound ground state molecule or back into two free atoms. In Fig. 1(a) we show cesium potential curves involved in photoassociation, in the optical pumping, and in the resonantly enhanced multiphoton ionization (REMPI) detection. PA is performed at $\sim 11\,730.1$ cm⁻¹, with radiation available by an Ar⁺ pumped cw Ti:sapphire laser (intensity 300 W/cm²), which permits the preparation of Cs₂ in a collection of $v_X=1-10$ vibrational levels of the $X\ ^1\Sigma_g^+$ ground electronic state (hereafter referred as X). Once formed, the molecules undergo ballistic expansion as they fall freely under the influence of gravity and are available for manipulation and detection for a period of ~ 10 ms. In this lap of time, an optical pumping scheme is applied to accumulate molecules into a chosen vibrational level of the ground state. It involves several absorption-spontaneous emission cycles with the $B\ ^1\Pi_u$ excited state, hereafter referred as B .

Molecules are detected by converting them into molecular ions via a pulsed two-photon REMPI scheme through the $C\ ^1\Pi_u$ excited state, hereafter referred as C . The laser radiation, in the $\sim 16\,000$ cm⁻¹ range, is provided by a pulsed dye laser (spectral bandwidth 0.1 cm⁻¹) pumped by the second harmonic of a pulsed Nd:YAG laser (repetition rate 10 Hz and duration 7 ns). Cs₂⁺ ions are detected using a pair of microchannel plates in a time-of-flight mass spectrometer. By scanning the ionization wavelength, transitions from v_X ground vibrational levels to various v_C levels of the C state are monitored and assigned according to Ref. [17]. Any redistribution of population among the vibrational levels of the ground state introduced by additional light sources results in a clear change in the relative intensities of the lines present in the REMPI spectrum [13,14]. Thus, molecular population

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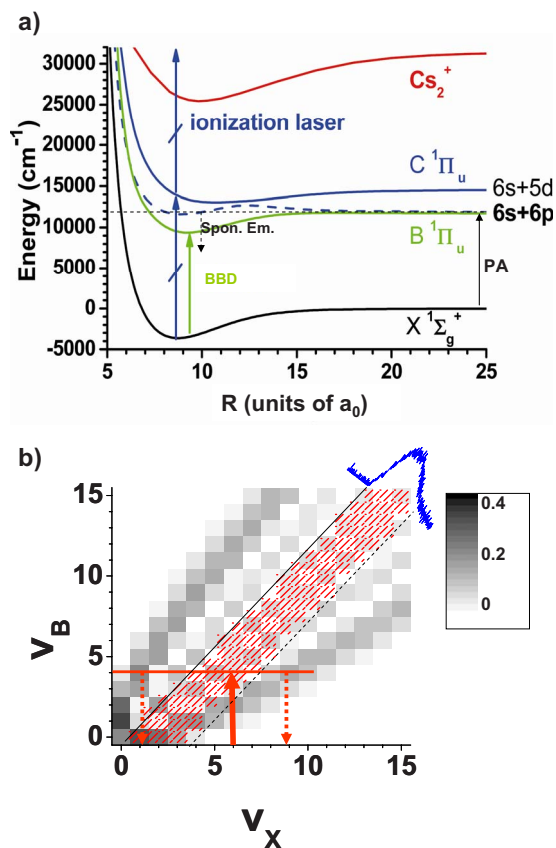


FIG. 1. (Color online) (a) Cs dimer potential curves relevant to the experiment. The PA connects the $6s+6s$ limit of the $X\ ^1\Sigma_g^+$ state to the $6s+6p$ limit. Part of the created molecules decays spontaneously to the $X\ ^1\Sigma_g^+$ state. The ionization takes place through the $C\ ^1\Pi_u$ intermediate state. The BDL performs optical pumping between the $X\ ^1\Sigma_g^+$ and the $B\ ^1\Pi_u$ states. (b) Franck-Condon factors plotted in grayscale. The hatched squares indicate transitions that are excited from the optical pumping radiation shown in the upper part, the arrows describe the possible evolution of population excited from the $v_X=6$ level.

transfer can be monitored by comparing spectra obtained with or without the optical pumping. We note that the present detection scheme does not allow rotational transitions to be resolved since this would require a spectral resolution of $\sim 0.01\text{ cm}^{-1}$.

The principle of the optical pumping technique is explained in Fig. 1(b). Franck-Condon factors between ground and excited vibrational levels, which represent the relative strength of transitions, form a so-called “Condon parabola.” As shown, excitation from a particular X level, for example, from $v_X=6$ to $v_B=4$, results in an important number of molecules decaying to $v_X=1$ and also in some molecules decaying to higher vibrational levels like level $v_X=9$. While the former process decreases the vibrational number, the latter increases it. The broadband nature of the optical pumping radiation ensures re-excitation of these molecules while its shaping is necessary to remove all frequencies that would excite the target state (in this case $v_X=0$) and to make it a dark state of the system. Molecular population is thus accumulated in the target state after a series of excitation—spontaneous emission cycles.

In the initial demonstration of the vibrational cooling of molecules [13], as well as in following work [14], the broadband radiation was provided by a femtosecond laser (Tsunami, mode-locked Ti:sapphire laser, 10 nm bandwidth, and 80 MHz repetition rate). In the current experiment, the broadband radiation is provided by a Laser 2000 LD1227 HPD 1110 diode laser operating near threshold in the 780 nm range and focused to a waist of $\sim 800\ \mu\text{m}$ on the MOT position. This represents not only a substantial simplification of the required laser apparatus but also a conceptual change in the sense that we demonstrate that the coherent nature of the laser light is not necessary and the only requirement is to provide enough energy density at the correct frequencies. In fact we exploit all the BDL bandwidth, that is 2 nm for the laser part of the spectrum, but it increases to $\sim 10\text{ nm}$ when considering also the luminescent part. At this level, our method does not require any temporal control of the BDL radiation that is provided in cw mode.

We modified in two different ways the spectrum of the diode radiation. In the first case shown in Fig. 2(a), where the goal is to transfer the molecular population into the $v_X=0$ level, we need to remove only the blue part of the spectrum so that transitions from $v_X=0$ level to any vibrational level of the B state are no longer excited [13]. In this way the molecular population is accumulated in the $v_X=0$ level that now becomes a dark state of the system. This requires the removal of wave numbers larger than $13\ 030\text{ cm}^{-1}$. The stiffness of the shaping is not important as long as all other vibrational levels of the X state are excited by the laser light. The desired shaping was obtained with the use of three Semrock RazorEdge 785 long-pass filters placed on top of each other in order to achieve an extinction ratio of $\sim 99\%$. The filters were mounted at an angle with respect to the incident diode radiation to allow fine tuning of the cut-off frequency to the desired value. In Fig. 2(b) we show the diode spectra, before and after shaping, monitored by an Ando AQ6317B optical spectrum analyzer. The BDL was at all times operated below lasing threshold, with a bias current of 308 mA, producing an output power of 26.5 mW. We note that operating the diode above threshold does not modify the results. After the shaping, the remaining collimated beam power was 17.1 mW.

Figure 2(c) shows the REMPI spectra obtained respectively in presence and in absence of the optical pumping process. In the vertical axis we show the absolute number of Cs_2^+ ions. In presence of the BDL, lines corresponding to the $v_X=0 \rightarrow v_C=0, 1$ are greatly enhanced compared to the case without the BDL present which is the signature of a vibrational cooling into $v_X=0$.

The intensity used in this experiment was 1.2 W/cm^2 . Population transfer efficiency saturates for higher intensity values, while it drops to half this value at an intensity of 0.3 W/cm^2 . Taking into account the efficiency of the REMPI detection, the total number of molecules in the $v_X=0$ level is estimated to be of the order of 10^3 . This number could seem small compared to the initial number of cold atoms ($\sim 10^7$). But, this relatively large inefficiency lies in the initial PA process and not in the optical pumping one which we estimated to be $\sim 80\%$. This value has been obtained using simulations but is consistent with experimental

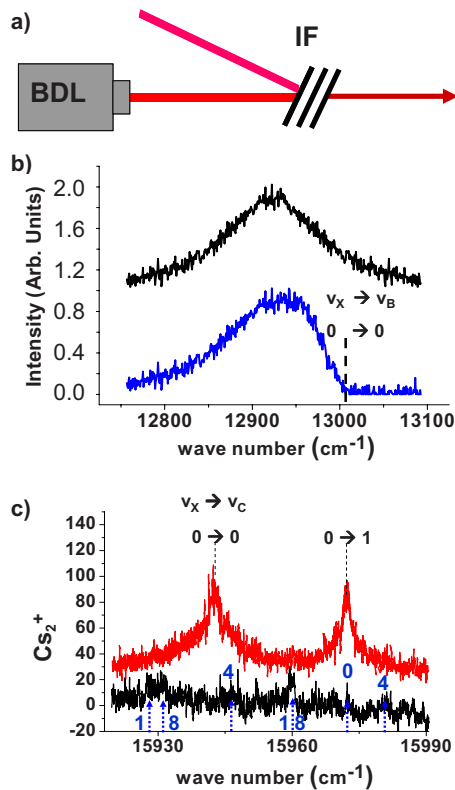


FIG. 2. (Color online) (a) Schematic of the BDL shaping realized with three interference filters (IFs) in order to transfer the population to the $v_x=0$ level. The filters are placed at an angle of $\sim 30^\circ$ in order to finely adjust the cut-off frequency to the desired value, resulting in a less sharp “cut” than for 0° . (b) Spectrum of the diode laser before (upper line) and after passing through the interference filters (lower line). All the BDL frequencies are suppressed above the $v_x=0 \rightarrow v_B=0$ transition frequency. (c) REMPI spectra of the $X^1\Sigma_g^+ \rightarrow C^1\Pi_u$ transitions [13,17]. The spectrum (black lower trace) without the BDL radiation contains labels of some detected vibrational levels v_x . The spectrum (red upper trace, offset of 20 ions for clarity) with the BDL radiation shows lines corresponding to the $v_x=0 \rightarrow v_c=0,1$ transitions.

observation of the REMPI signals. Simulations show that the efficiency of the cooling process could be improved if either more complicated shaping, or more broadband sources are used. These issues and ways to extend this technique to include rotational cooling have been discussed in Refs. [14,18].

A different, but still very simple setup allows us to extend this method to perform molecular transfer to levels other than the $v_x=0$. Again, the idea is to remove all frequency components that can excite molecules from the target level while still depopulating all other vibrational levels by optical pumping. We demonstrated, as an example, the molecular population transfer toward the $v_x=1$ level. Interference filters cannot be used since the transitions from the $v_x=0$ level, which have now to be addressed, are at a higher frequency than the transitions from the $v_x=1$ level that we need to block. In preceding experiments [14,18], the necessary shaping was achieved with the use of a Liquid Crystal SLM, which allowed a great versatility but at the price of high cost, experimental complexity, and nonperfect (97%) extinction

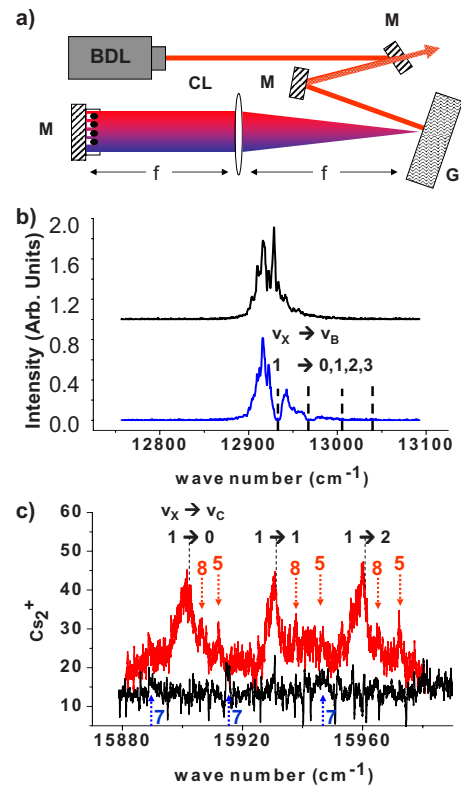


FIG. 3. (Color online) (a) The BDL shaping realized with the folded 4-f line setup, consisted of three mirrors (*m*) a grating (*g*) a CL and a mask. (b) Spectrum of the diode source with a bias current of 660 mA before (upper line) and after passing through the 4f line (lower line). At this bias current the diode is just above the lasing threshold, as the dominant frequency at $\sim 12\,930$ cm⁻¹ becomes more apparent. The frequencies of the $v_x=1 \rightarrow v_B=0,1$ transitions are removed. (c) The REMPI spectrum of the $X^1\Sigma_g^+ \rightarrow C^1\Pi_u$ transitions with (upper trace, offset of 10 ions for clarity) and (lower trace) without optical pumping with label to v_x levels and to $v_x \rightarrow v_c$ transitions.

ratio for the removed frequency components. This last point was considerably limiting the population transfer efficiency [14]. In the present experiment we can recover the selective molecular transfer to the $v_x=1$ level by removing from the BDL radiation the undesired frequency components with a mechanical mask placed in the Fourier plane of a 4f dispersion line [see Fig. 3(a)].

The BDL radiation is diffracted by a 1800 lines/mm optical grating. A cylindrical lens (CL) is placed at a distance equal to the effective focal length of the grating (~ 500 mm in this case) is used to collimate the radiation and to focus each of its frequency components in the Fourier plane where a mirror is placed. In the reflected beam all frequency components are recombined again by the grating while a slight misalignment in the vertical direction allows the radiation to exit the system by “missing” one of the guiding mirrors. In the Fourier plane we remove some frequency components by blocking them with a dark mask. In this case it is composed by four 1.5-mm-wide needles but other types of (photo)mask can be manufactured. The optical throughput of this second setup is much lower than in the filters case because of the limited reflectivity of the grating in the first diffraction order.

Thus, in order to have enough laser power after the shaping system, we operate the BDL with 30% higher bias current (~ 660 mA), i.e., above the lasing threshold, to have nearly 0.5 mW of shaped laser power on the molecules. The BDL spectrum is shown in Fig. 3(b) before and after the shaping. The gaps in the spectrum are ~ 4 cm^{-1} wide and correspond to the strongest transitions originating from the $v_X=1$ level. The efficiency of the technique is not sensitive to the removal of more frequencies than necessary as long as no other dark states than the target one are created. The resulting REMPI spectra, with and without BDL radiation, are shown in Fig. 3(c). Once more we see a clear effect of the optical pumping since the spectral features corresponding to the $v_X=1$ to the $v_C=0, 1$, and 2 transitions emerge in the presence of the shaped diode radiation. We can notice that some molecules appear to be pumped into the $v_X=5$ and $v_X=8$ levels, this is due to the imperfect shaping of our mask.

In conclusion, we have demonstrated the selective vibrational population transfer, i.e., vibrational cooling, in cesium dimers with the use of a collimated broadband diode laser. The fact that cooling was performed with the diode laser operating below threshold shows that coherence related effects are not relevant in this process. This is, to our knowledge, the first demonstration of a “Kastler’s type” [19] of optical pumping in molecules. The simple and inexpensive experimental setup for the population transfer can make this technique very attractive with respect to the previous demonstration, which involved a femtosecond laser and a spatial light modulator, and suitable for more general experiments

involving thermal samples of molecules. The use of a photomask eliminates the problem of the nonperfect extinction ratio that existed in the SLM setup. The extension of this method to cool the rotational degrees of freedom is possible as long as the required resolution to the shaping can be achieved. Additional and more important advances that can be foreseen for such a method is the use of a shaped BDL as a repumper light in a scheme for a direct laser cooling of molecules [20]. The relatively low cost and the simplicity of such a setup allows for the use of more than one diode laser if needed for instance each of them addressing transitions between different vibrational levels or even different electronic states. As the coherence of the light is not concerned, other light sources other than a BDL could be used provided that they can be collimated enough to be shaped and focused on the molecular sample.

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