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Ro-vibrational cooling of diatomic molecules

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Introduction

The photoassociation process (PA) of dimers, e.g. in the MOT, can result in production of translationally cold dimers (e.g. Yb_2 , Cd_2) in the excited ro-vibrational levels of their ground electronic states [1,2]. Here, we present method of ro-vibrational cooling of these molecules employing the $^30_u^+(6^3P_1)\leftrightarrow X^10_g^+(6^1S_0)$ and $c^31_u(5^3P_2)\leftrightarrow X^10_g^+(5^1S_0)$ transitions in Yb_2 and Cd_2, respectively. The method is based on realization of the „internal cooling of molecular degrees of freedom" which was presented in [3].

Idea of vibrational cooling

The method of vibrational cooling is based on an observation, that for certain electronic transitions (e.g. $^30_u^+(6^3P_1)\rightarrow X^10_g^+(6^1S_0)$ in Yb_2 or $c^31_u(5^3P_2)\rightarrow X^10_g^+(5^1S_0)$ in Cd_2) the probability of spontaneous emission process from the upper to the ground electronic state that occurs without changing of the vibrational quantum number υ, is much larger than the probability of similar transition with the change of υ (see Fig.1). The phenomenon is associated with the distribution of Fracnk-Condon factors (FCFs) for the *ground-state←excited-state* transition which depends on potentials parameters of the states involved (e.g. both potentials must have similar equilibrium distances R_e).

If we use laser radiation to excite these molecules, which initially are in the higher vibrational level υ'' of the ground state, to the selected vibrational level υ' of the excited state (with smaller vibrational number $\upsilon' < \upsilon''$), we will observe vibrational cooling after spontaneous emission from the excited state. Usually, due to distribution of FCFs for the *excited-state←ground-state* transition, to provide a high efficiency of the excitation process the molecules should be excited to $\upsilon' = \upsilon'' - 1$ (FCF's for $\upsilon' < \upsilon'' - 1$ are few orders of magnitude smaller).

A cascade of several cycles of excitation-spontanous emission processess can be used to repump the molecules from the initially populated υ'' to $\upsilon'' = 0$ (in each step of the cascade, the vibrational quantum number is reduced by 1). For details see [4].

Ro-vibrational cooling

Under assumption, that the bandwidth of the tunable laser is narrow enough to excite only one selected rotational level in the excited state, the rotational cooling scheme can be proposed. In the method, molecules initially occupy one (or more) ro-vibrational levels in the ground state. In each cooling step, the molecules which populate (υ'',J'') level are pumped to ($\upsilon' = \upsilon'' - 1$,$J' = J'' - 1$) level of the excited state. The idea of vibrational cooling is the same as for "pure" vibrational cooling (see above). The rotational cooling occurs thanks to the selection rule $\Delta J = -1,0,1$ which prevents molecules to de-excite to J'' greater than $J' + 1$. It means that no rotational heating occurs in the excitation-spontaneous emission cycle (see Fig.2).

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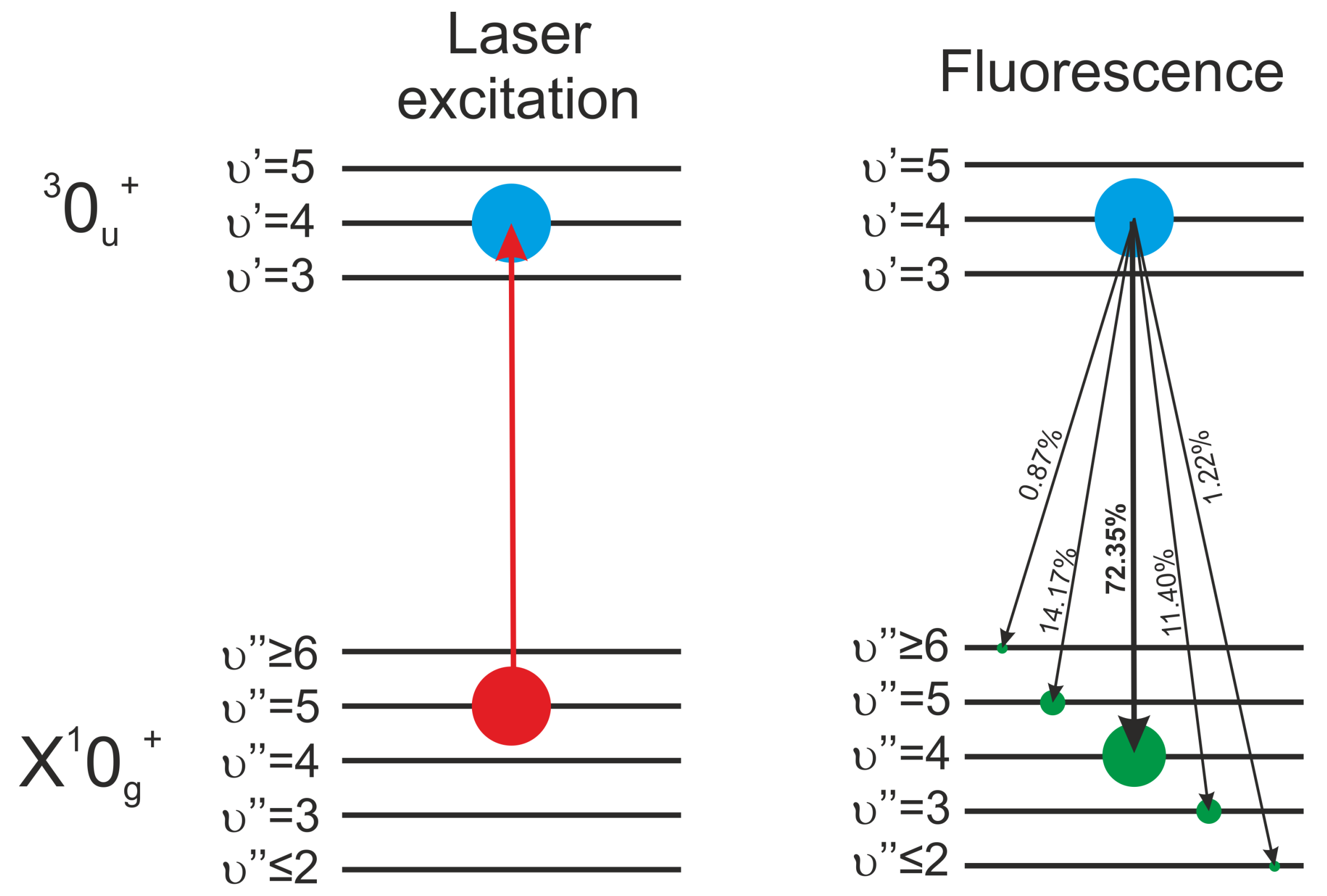

Fig.1. Schematic illustration of one step of "pure" vibrational cooling process in Yb_2 using the $^30_u^+(6^3P_1)\leftrightarrow X^10_g^+(6^1S_0)$ transition. Molecules from the initially populated $X^10_g^+(\upsilon''=5)$ level (red full circle) are excited using laser radiation to the $^30_u^+(\upsilon'=4)$ excited level (blue full circle). After the excitation, 85% (72.35% +11.4%+1.22%) of molecules de-excite to $\upsilon'' \leq 4$ (vibrational cooling), 14.17% of molecules de-exicite to $\upsilon'' = 5$ and can be excited to the $^30_u^+$ state again, 0.87% of molecules de-excite to $\upsilon'' \geq 6$ and are lost from the cooling scheme (green full circles). We assumes that the laser is spectrally wide and can excite molecules from all rotational levels of the $X^10_g^+$ state.

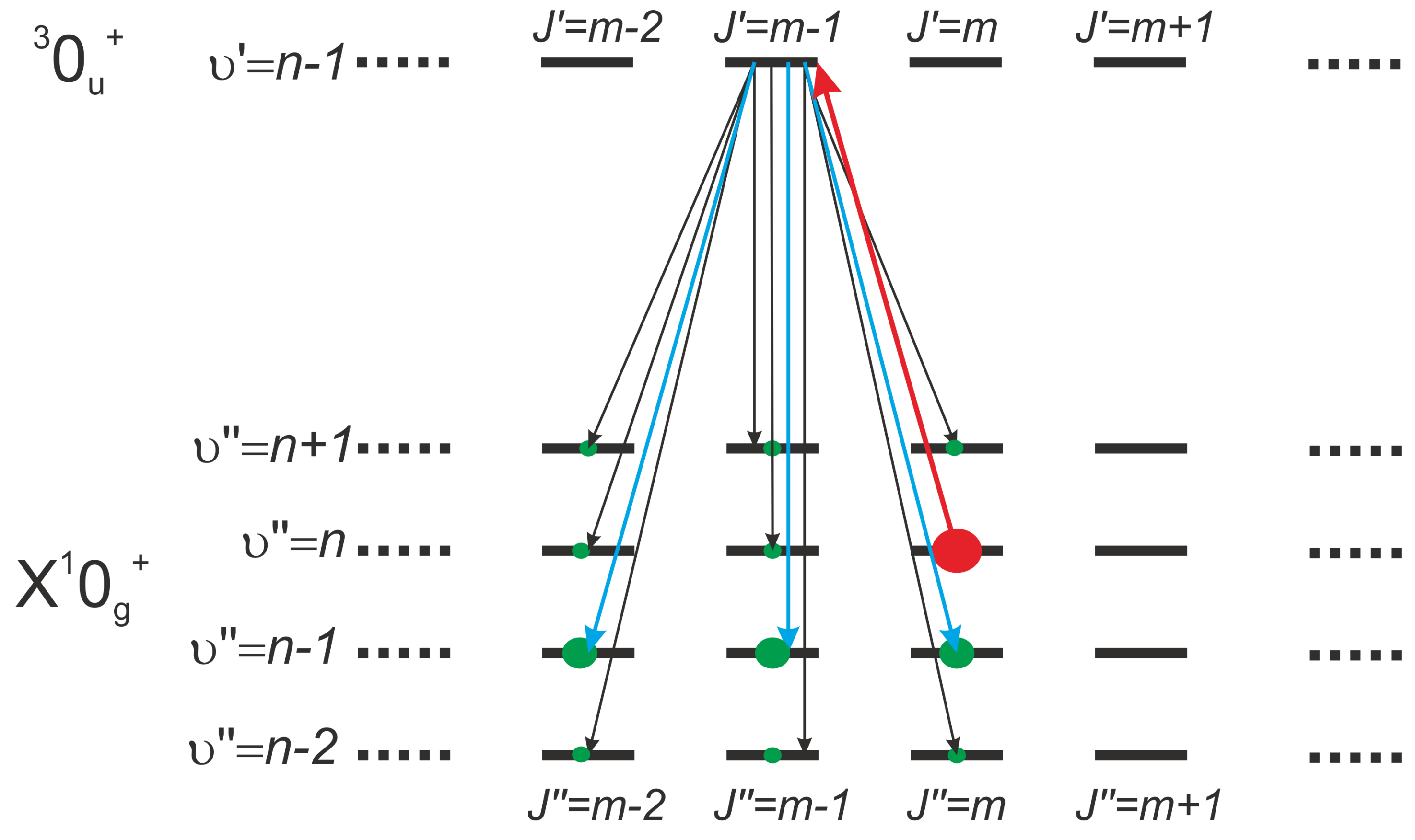

Fig.2. Schematic representation of one step of ro-vibrational cooling process in Yb_2 using the $^30_u^+(6^3P_1)\leftrightarrow X^10_g^+(6^1S_0)$ transition. Initially, a large number of molecules occupies ($\upsilon'' = n$, $J'' = m$) ro-vibrational level in the $X^10_g^+$ state (red full circle). The molecules are excited using laser radiation to ($\upsilon' = n - 1$, $J' = m - 1$) level in the $^30_u^+$ state (red arrow). Due to the fluorescence from the excited level, the molecules are transferred (black arrows) to different ro-vibrational levels in the $X^10_g^+$ state (green full circles). Due to the distribution of FCFs, the majority of molecules is transferred to $\upsilon'' = n-1$ vibrational level (blue arrows). The selection rule $\Delta J = -1,0,1$ ensures rotational cooling and prevents molecule from rotational heating during the process.

Result of ro-vibrational cooling for Cd_2 and Yb_2 dimers

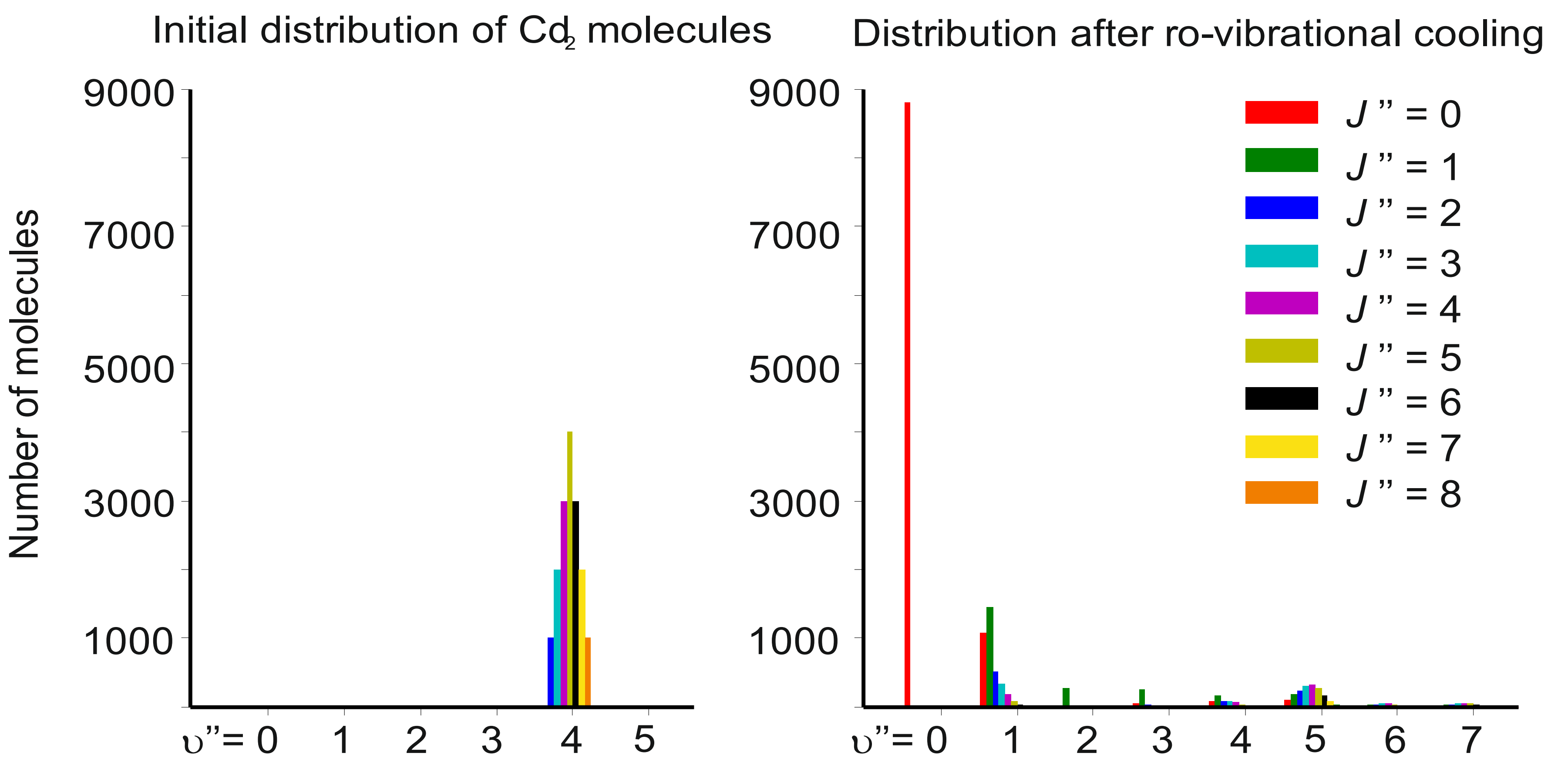

Fig.3. Result of ro-vibrational cooling of group of 16 000 Cd_2 molecules using the $c^31_u\leftarrow X^10_g^+$ transition. Initially, all molecules occupied different rotational levels of $\upsilon''=4$ vibrational level in the $X^10_g^+$ state (1000: $J''=2$, 2000: $J''=3$, 3000: $J''=4$, 4000: $J''=5$, 3000: $J''=6$, 2000: $J''=7$, 1000: $J''=8$). After cooling process, 55% of molecules was transferred to the desired ($\upsilon''=0$, $J''=0$) level. The FCFs were calculated using LEVEL program [5] assuming potential characteristics from [6].

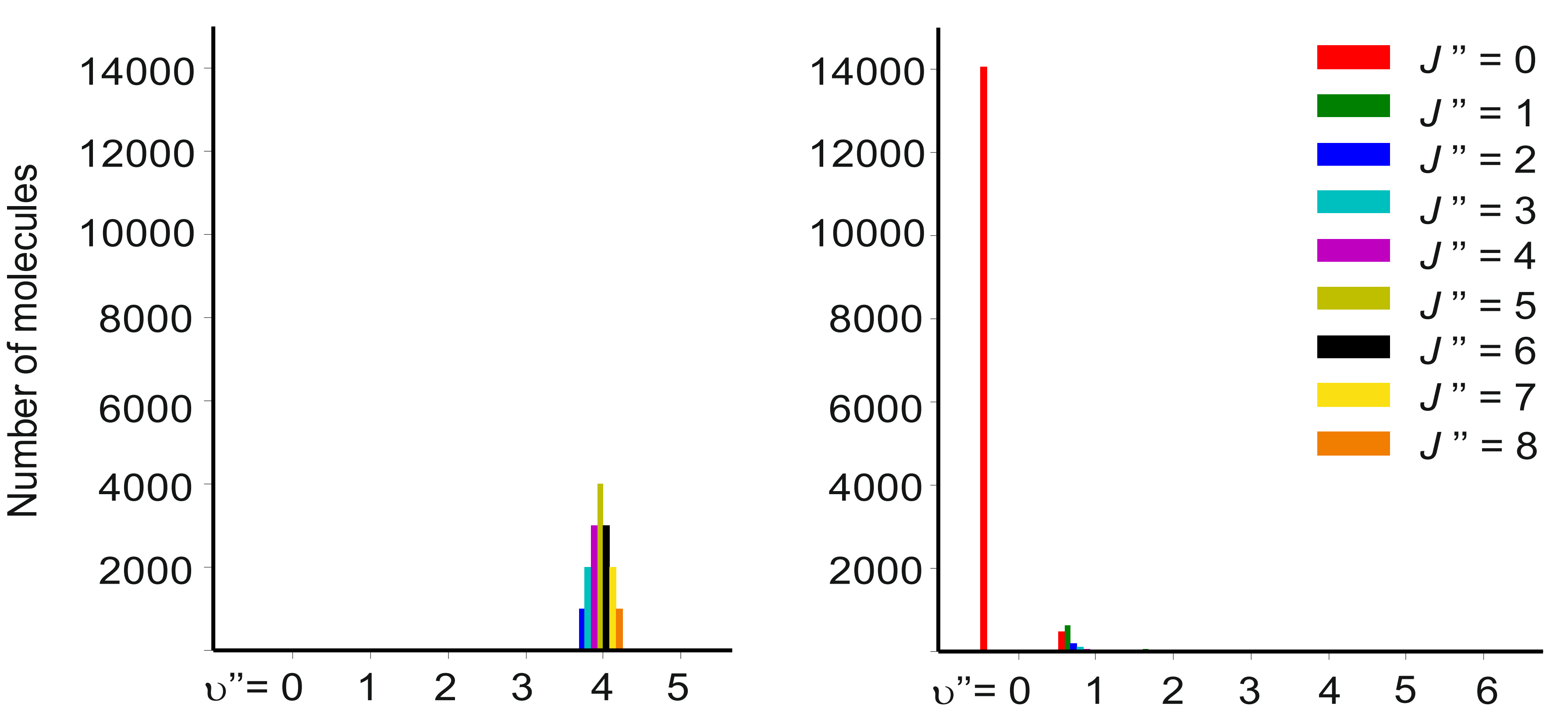

Fig.4. Result of ro-vibrational cooling of group of 16 000 Yb_2 molecules using the $^30_u^+\leftarrow X^10_g^+$ transition. Initially, all molecules occupied different rotational levels of $\upsilon''=4$ vibrational level in the $X^10_g^+$ state (1000: $J''=2$, 2000: $J''=3$, 3000: $J''=4$, 4000: $J''=5$, 3000: $J''=6$, 2000: $J''=7$, 1000: $J''=8$). After cooling process, 88% of molecules was trensferred to the desired ($\upsilon''=0$, $J''=0$) level. The FCFs were calculated using LEVEL program [5] assuming potential characteristics from [7].

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