Proc. Estonian Acad. Sci. Phys. Math., 1993, 42, 3, 264—266
https://doi.org/10.3176/phys.math.1993.3.09

OPTICAL ORIENTATION OF THE NUCLEAR SPIN IN THE MOLECULE

Olev SILD [a] and Mihhail BENDER [b]

[a] Eesti Teaduste Akadeemia Füüsika Instituut (Institute of Physics, Estonian Academy of Sciences), Riia 142, EE-2400 Tartu, Eesti (Estonia)

[b] Tartu Ülikool (Tartu University), Tähe 4, EE-2400 Tartu, Eesti (Estonia)

Presented by V. Hizhnyakov

Received March 12, 1993; accepted March 31, 1993

TUUMASPINNI OPTILINE ORIENTEERIMINE MOLEKULIS. Olev SILD, Mihhail BENDER

ОПТИЧЕСКАЯ ОРИЕНТАЦИЯ ЯДЕРНОГО СПИНА В МОЛЕКУЛЕ. Олев СИЛЬД, Михаил БЕНДЕР

Key words: strong light, rotating molecule hyperfine interaction, nuclear polarization.

In [1, 2], the rotation of a linear molecule in a linearly polarized strong light field resonant with the electronic $\Sigma \leftrightarrow \Sigma$ or $\Sigma \leftrightarrow \Pi$ transition has been studied theoretically. As shown, in a strong field limit the molecular axis will be oriented with respect to the polarization vector. At the same time, the linearly polarized coherent light field induces the dynamic dipole moment in the molecule [3].

One can expect that the circularly polarized light field resonant with electronic transition will induce the electronic angular momentum in the molecule. In its turn the hyperfine interaction between the induced angular momentum and the nuclear spin can orient the latter.

An analogous mechanism of the optical orientation of the nuclear spin in atoms has been studied in [4–6]. In atoms the monochromatic light field interacts with a certain electronic transition of the atom considered as a two-level system, which results in the saturation of this transition. In case of a strong field limit the saturation means the maximum orbital angular momentum and the maximum nuclear polarization.

In a rotating molecule, the strong monochromatic light field is simultaneously interacting with a number of electron-rotational transitions. As a result, the eigenstates of the light field—molecule system are superpositions of a number of electron-rotational eigenstates of the free molecule and phonon states. These superpositions depend on the strength of the light field. In these eigenstates, the electronic angular momentum of the molecule depends on the strength of the circularly polarized light field, too. Such dependence will also determine that of nuclear polarization on the light field strength. The aim of the present note is to report on this dependence.

Consider a linear molecule in a monochromatic circularly polarized light field of the frequency ω near the frequency ω_0 of the electronic $\Sigma \leftrightarrow \Pi$ transition. The rotation Hamiltonians of the molecule are

$$H(\Sigma)=B^0(\vec{J}-\vec{L})^2, \quad H(\Pi)=B^1(\vec{J}-\vec{L})^2,$$

where $\vec{J}$ and $\vec{L}$ are the full and the electronic angular momentum operator; B^0 and B^1 are the rotational constants in the ground (Σ) and the excited (Π) electronic state.

The light field is modelled through the optical mode polarized circularly in the xy-plane. Its electric field vector is

$$\vec{E}(\vec{r},t)=i\sqrt{\hbar\omega/4}\,[(\vec{e}_x\pm i\vec{e}_y)\hat{a}e^{ikz}-(\vec{e}_x\mp i\vec{e}_y)\hat{a}^+e^{-ikz}],$$

where $\vec{e}_{x,y}$ are the polarization unit vectors; $\hat{a}$ and $\hat{a}^+$ are the annihilation and the creation operator of photons; k is the plane wave vector; $+$ and $-$ mean the left-hand and right-hand circular polarization.

Here the dipole interaction $V=-\vec{d}\cdot\vec{E}$ serves as the interaction of the molecule with the light field. By using the rotating-wave approximation the problem of eigenvalues and eigenstates of the light field — molecule system is solved by the dressed states method analogously to [1, 2].

The eigenstates $|C_k\rangle$ are the following superpositions:

$$|C_k\rangle=\sum_{JMN} C_k(\Sigma JMN)\,|\Sigma\rangle\,|JM\rangle\,|N\rangle+ \\ +\sum_{J'M'N} C_k(\Pi J'M'N)\,|\Pi\rangle\,|J'M'\rangle\,|N\rangle,$$

where $|\Sigma\rangle$ and $|\Pi\rangle$ are electronic eigenstates; $|JM\rangle$ and $|J'M'\rangle$ are rotational eigenstates in the electronic ground and the excited state, respectively; J and M are the quantum numbers of the angular momentum and its projection; $|N\rangle$ is the eigenstate of the optical mode with N photons. The coefficients C_k of the superpositions depend on the strength of the light field.

The problem of the eigenvalues reduces to the diagonalization of the five-diagonal energy matrix. In case of a strong field limit the lower part of the energy spectrum is available analytically:

$$E_{nm}=-5B/12-\Gamma-(4\pi\alpha)^2\frac{\Gamma}{2}+(2\pi)^{3/2}\alpha\left(B\frac{\Gamma}{2}\right)^{1/2}(n+1)+Bm^2/2,$$

where $B=B^0=B^1$; $\Gamma=d\sqrt{N\hbar\omega/2}$; d is the value of the $\Sigma \leftrightarrow \Pi$ transition dipole moment; $\alpha=[\Gamma(1/4)]^{-2}$; $\Gamma(1/4)$ is the gamma function.

The formula determines the spectrum of the two-dimensional libration of the molecular axis with respect to the orientation of the plane wave vector, $n=0, 1, 2, \dots$ being the quantum number of the librational vibration, $m=n, n-2, \dots, -n$ is the quantum number of the vibrational momentum.

In a strong field, due to the orientation of the molecular axis and the mixing of the Σ and the Π state, the electronic angular momentum is induced in the molecule. Further the hyperfine interaction between the angular momentum and the nuclear spin is considered assuming the inversion symmetry of the molecule to have the nucleus with the spin I in the centre of the molecule (e. g. the molecule $C^{13}O^{16}_2$ having $I=1/2$ of C^{13}). The nuclear eigenstates are $|IM_I\rangle$, where M_I is the quantum number of the nuclear spin projection.

Now, the eigenstates of the light field—molecule—nuclear spin system are the following superpositions:

$$|C_k\rangle = \sum_{JMNIM_I} C_k(\Sigma JMNIM_I)\,|\Sigma\rangle\,|JM\rangle\,|N\rangle\,|IM_I\rangle +$$

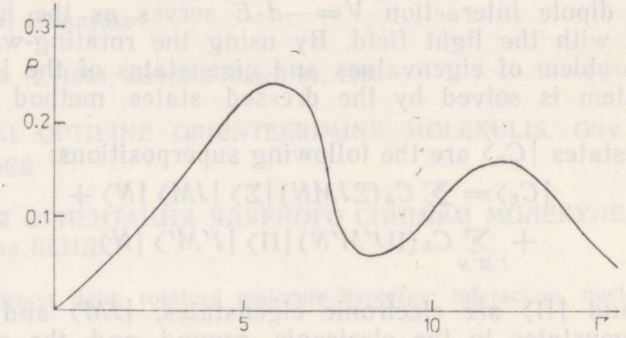
$$+ \sum_{J'M'NIM_I} C_k(\Pi J'M'NIM_I)\,|\Pi\rangle\,|J'M'\rangle\,|N\rangle\,|IM_I\rangle,$$

where the coefficients C_k depend on the strength of the light field. The problem of eigenvalues and eigenstates of the system, consisting of the dipole interaction as well as hyperfine interaction, reduces to the diagonalization of the nine-diagonal energy matrix.

Knowing the eigenstates $|C_k\rangle$, one can calculate the nuclear polarization

$$P = \langle C_k | M_I | C_k \rangle$$

as a function of the strength of the light field—molecule interaction determined by the parameter Γ.



Dependence of nuclear polarization on the strength of the light field.

The Figure presents this function in the ground state of the light field—molecule system for the constant of the hyperfine interaction of the value 10^9 Hz ($I=1/2$). Unlike in the case of the atom it appears that the maximum nuclear polarization $P=0.24$ occurs at the light field—molecule interaction parameter $\Gamma \approx 6B$. For example, at $B=1$ cm^{-1} and $d=1D$, $\Gamma \approx 6B$ corresponds to the electric field strength $\sim 10^5$ V/cm. In the region $\Gamma > 6B$, the nuclear polarization will (after some oscillation) decrease with the growth of Γ. This is conditioned by the increase of the electronic angular momentum on the growing of Γ and by the simultaneous decrease of the difference between the probabilities of occupancy of the states $M_I = 1/2$ and $M_I = -1/2$.

REFERENCES

1. Sild, O., Rozman, M., Hizhnyakov, V. Opt. Commun., 1985, 53, 3, 173—178.
2. Сильд О., Розман М. Изв. АН ЭССР. Физ. Матем., 1989, 38, 1, 12—18.
3. Сильд О. Препринт F-57, Тарту, 1990.
4. Зарецкий Д. Ф., Козлинский А. В. Ядерная физика, 1980, 31, 4, 891—898.
5. Зарецкий Д. Ф., Козлинский А. В. Ядерная физика, 1985, 42, 2, 317—323.
6. Гангрский Ю. П., Градечны Ч., Зарецкий Д. Ф., Изосимов И. Н., Козлинский А. В., Марков Б. Н. ЖЭТФ, 1990, 98, 5, 1531—1538.