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# Coherent rotational states and their creation and time evolution in molecular and nuclear systems

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We show that molecules and nuclei with well-expressed rotational degrees of freedom can be excited by means of a multistep process, initiated by a short and sufficiently strong electromagnetic pulse, into a coherent superposition of rotational states. Properties of a molecule or a nucleus in such a nonstationary state change (quasi-) periodically in time, due to the fact that the energies of the rotational states satisfy (approximately) the rule $\omega J(J+1)$. Accordingly, their interaction with an electromagnetic field or with other particles changes (quasi-) periodically in time. A gas of molecules in a coherent state exhibits a (quasi-) periodic change of the refractive index, an effect which has been observed for the case of $CS_2$ gas at 325 K. The nuclear case does not lead to direct experimental detection, because of its short coherence period.

## I. INTRODUCTION

In this paper we identify mechanisms by which molecular or nuclear systems can be excited in a coherent superposition of rotational states, whose characteristic properties then show up in the subsequent time evolution of transition probabilities and in the expectation values of dynamical variables. In both cases the superposition of coherent rotational states is obtained by means of a multistep process generated by a strong electromagnetic pulse of short duration in time.

A coherent superposition of states of a pure rotational band exhibits a periodic behavior in time. The periodicity is due to the fact that energy levels of a pure rotational band are given by $\omega J(J+1)$. Actual systems, however, will show a quasiperiodic behavior, since the $\omega J(J+1)$ formula is only approximately satisfied.

All physical properties including the interactions with other systems change periodically in time. These properties include radiation in bursts[1-6] or the periodic change of refractive index[7-10] (when many molecules are simultaneously excited by the same electromagnetic pulse). Analogously, a molecule or a nucleus in a coherent superposition of rotational states can be partly or completely oriented in space.[11] Orientation will also exhibit quasiperiodic behavior.

The periodic properties of coherent rotational states have been discussed in both nuclear and molecular systems.[1-10] While in nuclear systems[1-6] the effect is not directly measurable because of the very short period ($\sim 10^{-19}$ s), it has been measured for a molecular system (where the period is $\sim 10^{-10}$ s).

In the molecular case, the excitation of the molecule provided by the electromagnetic pulse goes via Raman scattering. In Refs. 8 and 9 Lin *et al.* have given the theoretical evaluation of this phenomenon in first-order perturbation theory, using an effective semiclassical interaction Hamiltonian. However, for a strong optical pulse a multistep process occurs with contributions to many orders of perturbation theory. In order to take this into account in Ref. 10 use has been made of the sudden approximation, with the same effective semiclassical Hamiltonian.

Part of this paper consists in describing this phenomenon using, in the sudden approximation, quantum field theoretical methods for the electromagnetic field. It has to be emphasized that the multistep process described by means of the sudden approximation in a low-temperature regime predicts persistence of the effect even close to 0 K, while first-order perturbation theory does not.

The paper makes use of Glauber coherent radiation states for the electromagnetic pulse. For clarity sake we have thought wise to repeat some of the existing results. The paper points out that both the generation and the time evolution of the coherent superposition of rotational states are very similar for molecular and nuclear systems, except for the time scale.

We note that the effect is measurable in molecular systems also because many molecules contribute simultaneously to the same event.

The paper is organized as follows. In Sec. II we give the formal definition and properties of coherent superpositions of rotational states, following the approach of boson operators. We call these states coherent rotational states (CRS). In Sec. III, we present the theory of formation of coherent superpositions of rotational states for realistic systems in the framework of the sudden approximation using pure state or statistical mixture formalisms. Since, for realistic systems, these states are not identical (although they are very similar) to the CRS, we call them realistic coherent rotational states (RCRS). It is shown in Sec. V that indeed the properties of the RCRS are very close to those of the CRS. The subsequent evolution in time of a system in a coherent superposition of rotational states is treated formally in Sec. IV. The electromagnetic decay of such a system is discussed, as well as the change of refractive index of a gas due to the (quasi-) periodic change of properties of the system. Measuring the refractive index is suitable for systems of molecules at room temperature since in that case the electromagnetic decay rate is very small in comparison with the collision rate, while the time of measurement is short in comparison with the average collision time. One says that the gas is "collisionless" in this case.

The applications to the ${}^{238}_{92}\mathrm{U}$ nucleus and the $CS_2$ gas are shown in Sec. V, where we show that the multistep process generates superpositions of about 40 states in the molecular case and about 20 states in the nuclear one. Section VI contains the conclusions. Units $\hbar=c=1$ will be used throughout the paper.

## II. DEFINITION OF COHERENT ROTATIONAL STATES AND THEIR PROPERTIES

In this section we shall present one of the formal definitions of CRS (Ref. 11) to point out the similarities with our realistic coherent states (RCRS). In particular, as we shall see in Sec. V, the coefficients of the states in the two cases are very close, assuring the similarity of physical properties for both states.

Coherent rotational states can be defined in several almost completely analogous ways.[11-13] We choose that definition employing boson creation and annihilation operators[14] because of its similarity with the case of coherent vibrational states[15] and because of the simple analytic properties of expectation values of operators between these CRS. (The fact that in this method the expansion of a state vector on the set of CRS contains both integer and half-integer angular momenta is only an apparent difficulty. In fact, a physically realizable state contains only one type of angular momenta and therefore the coefficients of the expansion pertaining to the other type automatically vanish.)

The method[11,5,6] introduces two pairs of annihilation $(\hat{a}_+,\hat{a}_-)$ and creation $(\hat{a}^\dagger_+,\hat{a}^\dagger_-)$ operators with bosonic commutation relations:

$$[\hat{a}_\pm,\hat{a}^\dagger_\pm]=1,\quad [\hat{a}^\dagger_\pm,\hat{a}^\dagger_\pm]=[\hat{a}_\pm,\hat{a}_\pm]=[\hat{a}_\pm,\hat{a}^\dagger_\mp]=0\ . \tag{2.1}$$

Defining $|\psi_0\rangle$ as the vacuum eigenstate of the angular momentum operators, the operators $\hat{a}^\dagger_\pm$, operating on $|\psi_0\rangle$, create the eigenstate of the angular momentum operator $\hat{J}$ with the expectation value of $\hat{J}^2$ equal to $\frac{3}{4}$ and of $\hat{J}_z$ to $\pm\frac{1}{2}$:

$$\hat{a}^\dagger_\pm|\psi_0\rangle=|\chi^{1/2}_{\pm 1/2}\rangle\ , \tag{2.2a}$$

and

$$\hat{J}^2|\chi^{1/2}_{\pm 1/2}\rangle=\tfrac{3}{4}|\chi^{1/2}_{\pm 1/2}\rangle\ ,$$
$$\hat{J}_z|\chi^{1/2}_{\pm 1/2}\rangle=\pm\tfrac{1}{2}|\chi^{1/2}_{\pm 1/2}\rangle\ .$$

The operators $\hat{a}_\pm$ lower the angular momentum of the state by $\frac{1}{2}$ and the third projection by $\pm\frac{1}{2}$:

$$\begin{aligned}&\hat{a}_\pm|\chi^{1/2}_{\pm 1/2}\rangle=|\psi_0\rangle\ ,\\ &\hat{a}_\mp|\chi^{1/2}_{\pm 1/2}\rangle=0\ ,\\ &\hat{a}_\pm|\psi_0\rangle=0\ .\end{aligned} \tag{2.2b}$$

The angular momentum operators can be expressed as bilinear forms of the operators $(\hat{a}^\dagger_\pm,\hat{a}_\pm)$:[11,5,6]

$$\begin{aligned}&\hat{J}_x=\tfrac{1}{2}(\hat{a}^\dagger_-\hat{a}_+ +\hat{a}^\dagger_+\hat{a}_-)\ ,\\ &\hat{J}_y=(i/2)(\hat{a}^\dagger_-\hat{a}_+ -\hat{a}^\dagger_+\hat{a}_-)\ ,\\ &\hat{J}_z=\tfrac{1}{2}(\hat{a}^\dagger_+ a_+ -\hat{a}^\dagger_-\hat{a}_-)\equiv\tfrac{1}{2}(\hat{N}_+ -\hat{N}_-)\ ,\\ &\hat{J}^2=\tfrac{1}{2}\hat{N}(\tfrac{1}{2}\hat{N}+1)\ ,\\ &\hat{J}_\pm=\hat{a}^\dagger_\pm\hat{a}_\mp\ ,\end{aligned}$$

where $\hat{N}_+=\hat{a}^\dagger_+\hat{a}_+$, $\hat{N}_-=\hat{a}^\dagger_-\hat{a}_-$, and $\hat{N}=\hat{N}_+ +\hat{N}_-$.

The eigenstates of angular momentum $|\phi_{JM}\rangle$ can be expressed as follows:

$$|\phi_{JM}\rangle=\frac{(\hat{a}^\dagger_+)^{J+M}(\hat{a}^\dagger_-)^{J-M}}{[(J+M)!(J-M)!]^{1/2}}|\psi_0\rangle\ . \tag{2.3}$$

CRS $|\psi_\beta\rangle$ are defined (in complete analogy with the vibrational case[15]) as eigenstates of $\hat{a}_\pm$:[11]

$$\hat{a}_\pm|\psi_\beta\rangle=\beta_\pm|\psi_\beta\rangle\ , \tag{2.4}$$

where $\beta_\pm$ can be any complex number. $|\psi_\beta\rangle$ can be normalized: $\langle\psi_\beta|\psi_\beta\rangle=1$. For any pair $(\beta_+,\beta_-)$ of complex numbers one has

$$|\psi_\beta\rangle=\sum_{J=0}^{\infty}\sum_{M=-J}^{J}a^\beta_{JM}|\phi_{JM}\rangle \tag{2.5}$$

with coefficients:

$$a^\beta_{JM}=e^{-(1/2)(|\beta_+|^2+|\beta_-|^2)}\frac{\beta_+^{J+M}\beta_-^{J-M}}{[(J+M)!(J-M)!]^{1/2}}\ . \tag{2.6a}$$

The phases of the coefficients are defined by

$$a^\beta_{JM}=|a^\beta_{JM}|e^{-i\varphi^\beta_{JM}},\quad \varphi^\beta_{JM}=\varphi^\beta_+(J+M)+\varphi^\beta_-(J-M)\ . \tag{2.6b}$$

CRS have the following characteristic properties.

(i) The absolute values of the amplitudes $a^\beta_{JM}$ are peaked around mean values of $(\langle\psi_\beta|\hat{J}^2|\psi_\beta\rangle)^{1/2}$ and $\langle\psi_\beta|\hat{J}_k|\psi_\beta\rangle$.

(ii) The phases $\varphi^\beta_{JM}$ [Eq. (2.6b)] are equidistant for fixed $J$ or $M$. As a consequence, for large $|\beta_+|$ and $|\beta_-|$ the

CRS have large expectation values of the quadrupole moment operator.[1]

(iii) If the vacuum state $|\psi_0\rangle$ represents a rotor, the CRS describe a rotor which is oriented in space for large values of $|\beta_+|$ and $|\beta_-|$.

(iv) For a rotor, a CRS is a nonstationary state with periodic time evolution properties:

$$|\psi_\beta(t)\rangle \equiv e^{-i\hat{H}_0 t}|\psi_\beta\rangle = \sum_{J,M} a^\beta_{JM} e^{-i\omega J(J+1)t}|\phi_{JM}\rangle \;, \tag{2.7}$$

where $\hat{H}_0$ is the Hamiltonian describing the rotor (we assume zero ground-state energy for simplicity):

$$\hat{H}_0 = \omega \hat{J}^2 \;. \tag{2.8}$$

The phases $\exp\{i[\varphi^\beta_{JM} + \omega J(J+1)t]\}$ change periodically in time. Therefore, expectation values of operators which do not commute with $\hat{H}_0$ also change periodically in time. As an example, we show in Fig. 1 the expectation value of the quadrupole moment operator as a function of $\omega t$:[6]

$$\langle \psi_\beta(t)|\hat{Q}_{2,0}|\psi_\beta(t)\rangle = \langle \psi_0|\hat{Q}^{\text{int}}_{2,0}|\psi_0\rangle \left[ A + \left[ e^{i6\omega t} \sum_{J,M} e^{i4\omega J t} B_{JM} + \text{c.c.} \right] \right] \tag{2.9}$$

with

$$A = \sum_{J,M} \frac{3e^{-2|\beta|^2}|\beta|^{4J}[2J(J+1)-2M^2-1]}{2(2J+3)(2J-1)(J+M)!(J-M)!} - \tfrac{1}{2} \;,$$

$$B_{JM} = \frac{3e^{i4\varphi^\beta} e^{-2|\beta|^2}|\beta|^{4(J+1)}}{2(2J+3)(J+M)!(J-M)!\sqrt{(2J+5)(2J+1)}} \;,$$

and

$$\hat{Q}_{2,0} = \sum_\nu D^2_{\mu\nu}(\phi,\vartheta,0)\hat{Q}^{\text{int}}_{2\nu} \;,$$

$$\beta_+ = \beta_- = \beta \;,$$

and

$$\varphi^\beta_+ = \varphi^\beta_- = \varphi^\beta \;.$$

The periodic structure of the quadrupole moment is apparent from (2.9). The sum appearing on the right-hand side is periodic with period $\pi/4\omega$ for even $J$, $\pi/2\omega$ for odd $J$. On the other hand, the whole function duplicates itself exactly only after a period $\pi/\omega$ because of the interference between the factor $\exp(i6\omega t)$ and the sum. E.g., for even $J$ we obtain bursts at integral multiples of $\pi/4\omega$, but only every four bursts is the detailed pattern completely reproduced.

Figure 2(a) presents the coefficients $|a^\beta_{J0}|$ of the CRS for different values of $\beta$ and Fig. 2(b) presents the expectation values of the operator $\hat{Q}_{2,0}$ for the corresponding CRS with $\beta = \sqrt{22}$. Since $\langle \psi_\beta|\hat{J}^2|\psi_\beta\rangle = \beta^2(\beta^2+1)$, one notices that the coefficients do not change much with $\beta$.

## III. CREATION OF COHERENT ROTATIONAL STATES

We present in this section a possible mechanism through which a molecule or a nucleus with an almost ideal rotational band can be excited into a coherent superposition of rotational states. We call these states RCRS.

A particle may initially be in the ground state $|\phi_{00}\rangle$, or in one of its excited states $|\phi_{JM}\rangle$, or its state may initially be described by a density operator of statistical nature:

$$\hat{\rho}(t=t_0) = \sum_{J,M} p_{JM}|\phi_{JM}(t_0)\rangle\langle\phi_{JM}(t_0)| \;,$$
$$p_{JM} = \exp(-E_J/kT) \Big/ \left[ \sum_J (2J+1)\exp(-E_J/kT) \right] \;. \tag{3.1}$$

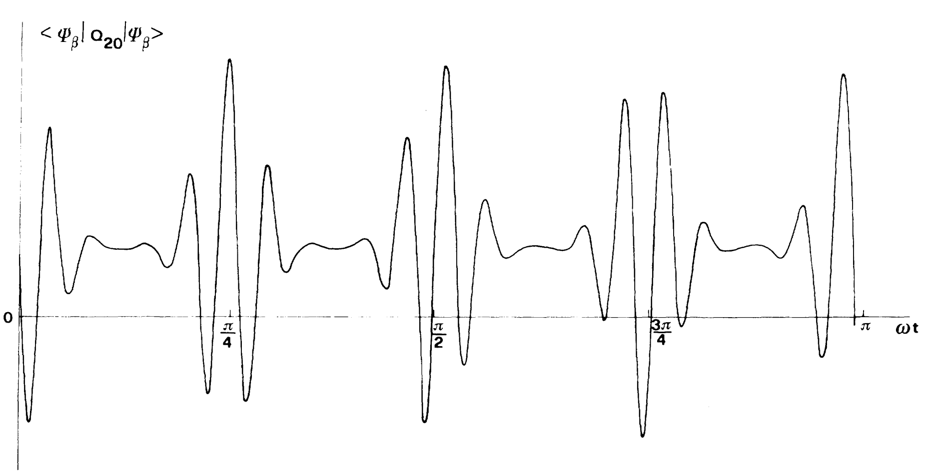

FIG. 1. Time evolution of the expectation value of the quadrupole moment operator $\langle \psi_\beta|\hat{Q}_{2,0}|\psi_\beta\rangle$. Only even integer angular momenta are taken into account. $\beta_+ = \beta_- = \beta = \sqrt{10}$, $\varphi^{\beta_+} = \varphi^{\beta_-} = \varphi^\beta = 0$. $\langle \psi_\beta|\hat{J}^2|\psi_\beta\rangle = \beta^2(\beta^2+1) = 110$.

We show that a short and sufficiently strong electromagnetic pulse can, in a multistep process, excite a molecule or a nucleus into a RCRS. The collision time of the molecules in a gas should be long compared to the period of the coherent state, so that the molecule in the coherent state is not disturbed by collisions. It will be shown (see later Figs. 4 and 5) that the RCRS created from different initial states have similar properties, so that the statistical nature of the initial density operator does not appreciably change the periodic properties of the expectation values of operators.

In this paper a sudden approximation formalism is used since it creates a RCRS with properties close to those of the CRS and since it is rather simple and numerically manageable. Nevertheless, experiments under the conditions for which the sudden approximation is valid, can be performed, as discussed below.

### A. Creation mechanism in the sudden approximation

Let $\hat{H}_0$ describe a system at the $t<t_0$ and let $\hat{H}'(t)$ $(t \geq t_0)$ describe the interaction of the system with an electromagnetic field:

$$\hat{H}=\hat{H}_0+\hat{H}'(t)\ . \tag{3.2}$$

If the system is in a pure state $|\phi_{JM}\rangle$ at $t=t_0$, the state vector at time $t$ has the form:

$$|\psi^{(JM)}(t)\rangle=\hat{U}_0(t-t_0)\hat{U}_1(t,t_0)\,|\phi_{JM}(t_0)\rangle,\quad t\geq t_0 \tag{3.3a}$$

with $\hat{U}_0(t)=e^{-i\hat{H}_0 t}$ and $\hat{U}_1$ determining the evolution of the system under the influence of $\hat{H}'(t)$:

$$\hat{U}_1(t,t_0)=\hat{T}\exp\left[-i\int_{t_0}^{t}\hat{H}'_I(t')dt'\right]\ , \tag{3.4}$$

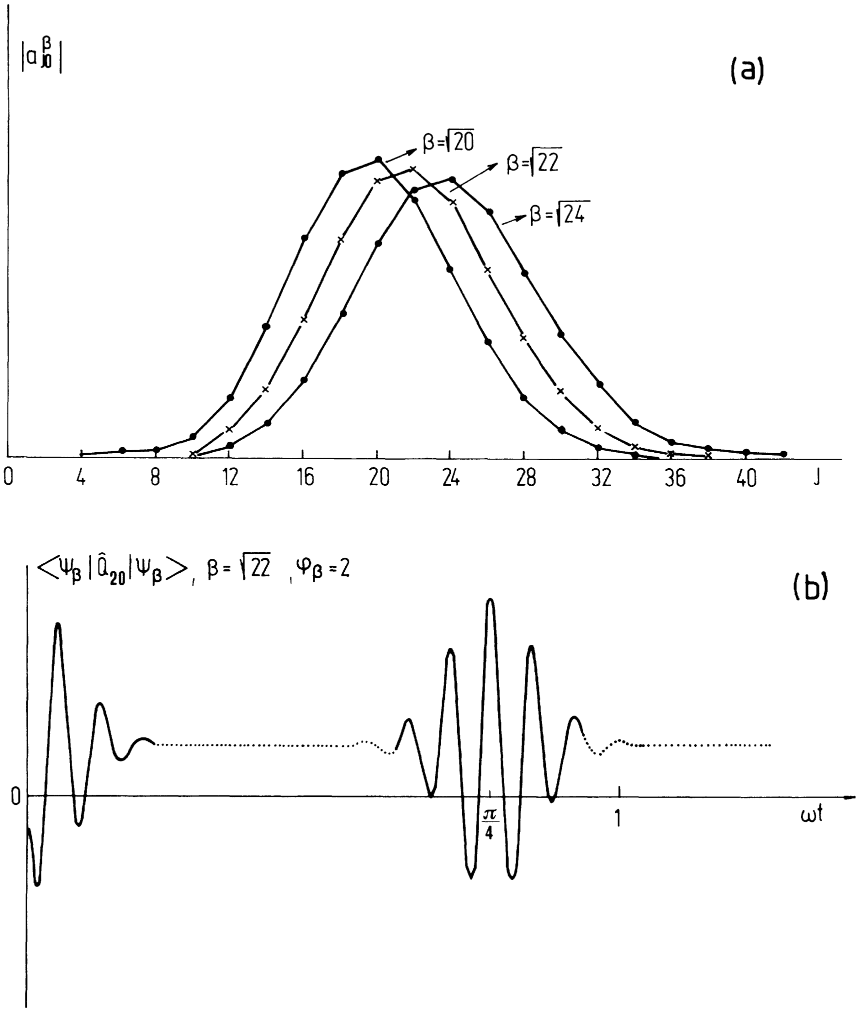

FIG. 2. (a) Coefficients $|a^{\beta}_{J0}|$ determining the coherent rotational states are presented for $\beta_+=\beta_-=\beta=\sqrt{20},\sqrt{22},\sqrt{24}$. (b) The same as in Fig. 1 except that $\beta=\sqrt{22}$. The corresponding coefficients $|a^{\beta}_{J0}|$ are presented in (a). The expectation values of $\hat{Q}_{2,0}$ for $\beta=\sqrt{20}$ and $\beta=\sqrt{24}$ are close to that of $\beta=\sqrt{22}$.

where

$$\hat{H}'_I(t)=e^{i\hat{H}_0 t}\hat{H}'(t)e^{-i\hat{H}_0 t}\ ,$$

and $\hat{T}$ is the ordinary time-ordering operator. The operator $\hat{U}_1(t,t_0)$ [Eq. (3.4)] can be rewritten in a form in which the ordering operator $\hat{T}$ does not appear:[16,17]

$$\hat{U}_1(t,t_0) =\exp\left[-i\int_{t_0}^{t}\hat{H}'_I(t_1)dt_1 -\tfrac{1}{2}\int_{t_0}^{t}dt_2\int_{t_0}^{t_2}dt_1[\hat{H}'_I(t_2),\hat{H}'_I(t_1)]+\cdots\right] \tag{3.5a}$$

where the ellipses represent terms with higher-order commutators.

If the interaction $\hat{H}'$ has a very short duration $\tau$ in time (that is, $\Delta E\tau \ll 1$, where $\Delta E$ is the energy difference between the highest relevant state excited in this process and the initial state of the system) all the commutators in Eq. (3.5a) are approximately zero. It follows that the operator $\hat{U}_1(t,t_0)$ has, in the sudden approximation, the simple form

$$\hat{U}_1(t,t_0)=e^{\hat{R}(t,t_0)},\quad \hat{R}(t,t_0)=-i\int_{t_0}^{t}\hat{H}'_I(t')dt'\ . \tag{3.5b}$$

Operating on an initial state $|\phi_{JM}\rangle$, the operator $\hat{U}_1(t,t_0)$ populates many excited states of the system as long as the transition probabilities among excited states are high in comparison with the duration $\tau$ of the interaction $\hat{H}'(t)$. One can estimate[17] the probability $P_n$ that an $n$-step process takes place by supposing that the matrix elements for the transitions between any two neighbor states are approximately the same and equal to the matrix element determining the transition from the ground to the first excited rotational state $\langle\phi_{J_0M_0}|\hat{H}'|\phi_{J_1M_1}\rangle$ (for the rigid rotor this is justified). One obtains

$$P_n \propto |\langle\phi_{J_0M_0}|\hat{H}'|\phi_{J_1M_1}\rangle^n\tau^n/n!|^2\ .$$

If $\langle\phi_{J_0M_0}|\hat{H}'|\phi_{J_1M_1}\rangle\tau$ is larger than 1, $P_n$ is appreciably different from zero even for rather high $n$.

If at $t=t_0$ the system is described by a density operator [Eq. (3.1)] then the density operator of the system $\hat{\rho}(t)$ evolves in time as follows:

$$\hat{\rho}(t)=\sum_{J,M}p_{JM}\,|\psi^{(JM)}(t)\rangle\langle\psi^{(JM)}(t)|\ , \tag{3.6}$$

where the time evolution of the states $|\psi^{(JM)}(t)\rangle$ is described by Eq. (3.3a). The statistical nature of the density operator $\hat{\rho}(t)$ is described by coefficients $p_{JM}$ which are independent of time.

If for the interaction $\hat{H}'$ the basis $|\phi_{JM}\rangle$ is complete, we may write

$$|\psi^{(JM)}(t)\rangle=\sum_{J',M'}|\phi_{J'M'}\rangle C^{(JM)}_{J'M'}(t)e^{-iE_{J'}(t-t_0)} \tag{3.3b}$$

with

$$C^{(JM)}_{J'M'}(t)=\langle\phi_{J'M'}(t_0)|\hat{U}_1(t,t_0)|\phi_{JM}(t_0)\rangle\ .$$

In a multistep process many coefficients $C^{(JM)}_{J'M'}$ can become appreciably different from zero so that $|\psi^{(JM)}\rangle$ is a superposition of many rotational states. Figure 3 shows that the coefficients $C^{(JM)}_{J'M'}$ are often similar to the coefficients $a^{\beta}_{J'M'}$ [Eq. (2.6)] determining the CRS. In such cases, the state (3.3b) has properties similar to those of the CRS of Eq. (2.7). To illustrate this, we shall present in Sec. V numerical results for the electromagnetic excitation of a system with rotational degrees of freedom. We will show that a short electromagnetic pulse of a quadrupole type is able to generate the desired RCRS.

### B. Raman excitation of the coherent rotational states in a sudden approximation

Let $\hat{H}_0$ describe a free molecule and the free electromagnetic field:

$$\hat{H}_0=\hat{H}_0^{\,\mathrm{mol}}+\hat{H}_0^{\,\mathrm{em}}\ , \tag{3.7}$$

with $\hat{H}_0^{\mathrm{mol}}=\hat{H}_0^{\mathrm{int}}+\omega\hat{J}^2$ and $\hat{H}_0^{\mathrm{mol}}|\phi_u\rangle=E_u|\phi_u\rangle$. The operator $\hat{H}_0^{\mathrm{int}}$ describes all molecular degrees of freedom except the rotational one. Let the coupling between the rotational and other degrees of freedom be negligibly weak. We express the electromagnetic field $\hat{\mathbf{A}}$ operator in terms of creation $\hat{a}^{\dagger}_{k\sigma}$ and annihilation $\hat{a}_{k\sigma}$ operators:

$$\hat{\mathbf{A}}'(\mathbf{r})=\frac{1}{(2\pi)^{3/2}}\int\frac{d^3\mathbf{k}}{\sqrt{2k}}(\hat{a}^{\dagger}_{k\sigma}e^{-i\mathbf{k}\cdot\mathbf{r}}+\hat{a}_{k\sigma}e^{i\mathbf{k}\cdot\mathbf{r}})\epsilon_{k\sigma}\ , \tag{3.8}$$

Here the symbol of integration means both integration over momenta $\mathbf{k}$ and sum over the polarization vectors $\epsilon_{k\sigma}$.

We suppose a strong electromagnetic field described by the Glauber coherent monochromatic polarized state which does not change appreciably during the interval $\tau$

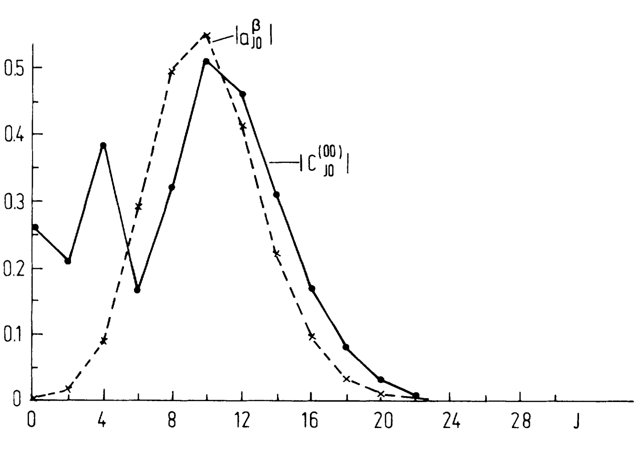

FIG. 3. Coefficients $|C^{(00)}_{J0}|$ of Eq. (3.17a) obtained in a heavy ion collision of ${}^{40}_{20}\mathrm{Ca}$ and ${}^{238}_{92}\mathrm{U}$ (see Table I) for the coherent superposition of rotational states of ${}^{238}_{92}\mathrm{U}$ are presented together with the corresponding coefficients $|a^{\beta}_{JM}|$ ($\beta=\sqrt{10}$).

when the interaction is switched on (in the calculations which follow we actually made use of box quantization for the electromagnetic field so that $|\alpha_{k\sigma}\rangle$ are strictly normalized states. In this spirit we also have $\hat{a}_{k\sigma}|\alpha_{k0\sigma0}\rangle=\delta_{kk0}\delta_{\sigma\sigma0}\alpha_{k\sigma}|\alpha_{k0\sigma0}\rangle$):

$$|\alpha_{k_0\sigma_0}\rangle\equiv\exp(-\tfrac{1}{2}|\alpha_{k_0\sigma_0}|^2)\exp(\alpha_{k_0\sigma_0}\hat{a}^{\dagger}_{k_0\sigma_0})|0\rangle\ , \tag{3.9}$$

where $|0\rangle$ is the vacuum photon state and

$$\hat{a}_{k_0\sigma_0}|\alpha_{k_0\sigma_0}\rangle=\alpha_{k_0\sigma_0}|\alpha_{k_0\sigma_0}\rangle\ .$$

The interaction between the molecule and the electromagnetic field can be written as

$$\hat{H}'(t)=-\sum_{i=1}^{Z}\frac{e}{2m_i}[\hat{\mathbf{A}}(\mathbf{r}_i)\cdot\hat{\mathbf{p}}_i+\hat{\mathbf{p}}_i\cdot\hat{\mathbf{A}}(\mathbf{r}_i)]\theta(t,t_0,\tau)\ , \tag{3.10}$$

with

$$\frac{i\hat{\mathbf{p}}_i}{m_i}=[\mathbf{r}_i,\hat{H}_0^{\mathrm{mol}}]$$

and

$$\theta(t,t_0,\tau)=\begin{cases}1 & \text{if}\ \ t_0\le t\le t_0+\tau\ ,\\ 0 & \text{otherwise}\ .\end{cases}$$

$Z$ is the total number of electrons in the molecule. We have neglected the term $\hat{\mathbf{A}}^2(e^2/2m_i)$ which gives zero contribution to the coefficients $C^{(JM)}_{J'M'}$ of Eq. (3.3a) in second order but may contribute to higher orders. Expanding $e^{\pm i\mathbf{k}\cdot\mathbf{r}}$ to first order in Eq. (3.8) (since $\mathbf{k}\cdot\mathbf{r}\sim10^{-3}$ for a molecule and a monochromatic pulse with $\lambda\sim1$ $\mu$m), one obtains

$$\hat{H}'(t)=\frac{i}{(2\pi)^{3/2}}\int\frac{d^3k}{\sqrt{2k}}\epsilon_{k\sigma}\times[\hat{\boldsymbol{\mu}}_e,\hat{H}_0^{\mathrm{mol}}](\hat{a}^{\dagger}_{k\sigma}+\hat{a}_{k\sigma})\theta(t,t_0,\tau)\ , \tag{3.11}$$

where $\hat{\boldsymbol{\mu}}_e=\sum_i e_i\hat{\mathbf{r}}_i$ is the electric dipole operator. We have omitted higher terms in Eq. (3.11) by supposing that contributions of the magnetic dipole, electric quadrupole and all higher-order operators are negligible in comparison with the electric dipole operator.

We assume in addition that the duration $\tau$ of the interaction between the electromagnetic field and the molecule is so short that the sudden approximation (3.5b) can be used (for the estimates see Sec. V).

Since $|\phi_{JM}\rangle$ are pure rotational states and $\hat{R}(t,t_0)$ is proportional to an electric dipole operator, then $\langle\alpha_{k_0\sigma_0}|\langle\phi_{JM}|\hat{R}^{2n+1}|\phi_{J'M'}\rangle|\alpha_{k_0\sigma_0}\rangle=0$ ($n$ is an integer) because all states of the rotational band have the same parity. On the other hand, we have

$$\langle\alpha_{k_0\sigma_0}|\langle\phi_{JM}|\hat{R}^2|\phi_{J'M'}\rangle|\alpha_{k_0\sigma_0}\rangle=\sum_{\phi_u}\prod_{k_u\sigma_u}\int\frac{d^2\alpha_{k_u\sigma_u}}{\pi}\langle\alpha_{k_0\sigma_0}|\langle\phi_{JM}|\hat{R}|\phi_u\rangle|\alpha_{k_u\sigma_u}\rangle\langle\alpha_{k_u\sigma_u}|\langle\phi_u|\hat{R}|\phi_{J'M'}\rangle|\alpha_{k_0\sigma_0}\rangle\neq0\ . \tag{3.12}$$

Here $|\phi_u\rangle$ represents all molecular excited states and $|\alpha_{k_u\sigma_u}\rangle$ is an (over) complete basis of coherent photon states. All molecular states with a parity opposite that of $|\phi_{JM}\rangle$ contribute to the sum.

In the optical region, the wavelength of the electromagnetic field is much smaller than the characteristic wavelength $\lambda_{\mathrm{rot}}$ of the rotational band and only high-energy states $|\phi_u\rangle$ contribute appreciably to the matrix element $\langle\alpha_{k_0\sigma_0}|\langle\phi_{JM}|\hat{R}^2|\phi_{J'M'}\rangle|\alpha_{k_0\sigma_0}\rangle$. Since, according to Eq. (3.11), $\langle\alpha_{k_0\sigma_0}|\langle\phi_{u'}|\hat{R}|\phi_u\rangle|\alpha_{k_0\sigma_0}\rangle$ is proportional to $(E_{u'}-E_u)\tau$, then $\langle\alpha_{k_0\sigma_0}|\langle\phi_{JM}|\hat{R}|\phi_u\rangle|\alpha_{k_0\sigma_0}\rangle$ is much larger than $\langle\alpha_{k_0\sigma_0}|\langle\phi_{u'}|\hat{R}|\phi_u\rangle|\alpha_{k_0\sigma_0}\rangle$ if $|\phi_u\rangle$ and $|\phi_{u'}\rangle$ are from high excited states.

One can then approximately express coefficients $C^{(JM)}_{J'M'}$ from Eq. (3.3a) in a rather simple form by performing the diagonalization of the operator $\hat{R}^2$ within the (truncated) subspace $\{|\phi_{JM}\rangle\}$:

$$\langle\alpha_{k_0\sigma_0}(t_0)|\langle\phi_{JM}(t_0)|\hat{V}^{\dagger}\hat{R}^2(t,t_0)\hat{V}|\phi_{J'M'}(t_0)\rangle|\alpha_{k_0\sigma_0}(t_0)\rangle=\delta_{JJ'}\delta_{MM'}\lambda_{JM} \tag{3.13a}$$

with $\hat{V}^{\dagger}\hat{V}=I$, and eigenvalues $\lambda_{JM}$. The matrix elements (3.12) are given by Eq. (A6) of Appendix A.

Due to the properties of the operator $\hat{R}$ discussed above, we can write

$$\langle\alpha_{k_0\sigma_0}|\langle\phi_{JM}|e^{\hat{R}}|\phi_{J'M'}\rangle|\alpha_{k_0\sigma_0}\rangle=\tfrac{1}{2}\langle\alpha_{k_0\sigma_0}|\langle\phi_{JM}|(e^{(R^2)^{1/2}}+e^{-(R^2)^{1/2}})|\phi_{J'M'}\rangle|\alpha_{k_0\sigma_0}\rangle\ .$$

Restricting ourselves to the truncated space $\{\phi_{JM}\}$, one finally obtains

$$C^{(JM)}_{J'M'}(t)=\tfrac{1}{2}\sum_{J''M''}\langle\phi_{J'M'}|\hat{V}|\phi_{J''M''}\rangle(e^{(\lambda_{J''M''})^{1/2}}+e^{-(\lambda_{J''M''})^{1/2}})\langle\phi_{J''M''}|\hat{V}^{\dagger}|\phi_{JM}\rangle\ . \tag{3.14}$$

In Sec. V we shall present some numerical results for a gas of molecules both for temperatures close or far from 0 K to show how closely the coefficients $C^{(JM)}_{J'M'}$ resemble the coefficients $a^{\beta}_{J'M'}$ given by Eq. (2.6a). We shall see that about 40 states contribute to the RCRS.

One can formally rewrite the matrix element (3.12), describing a two-step process as a one-step process. For the special case of Raman scattering, for $t>t_0+\tau$, it follows that

$$\langle \alpha k_0\sigma_0 | \langle \phi_{JM} | \hat{R}^{\,2} | \phi_{J'M'} \rangle | \alpha_{k_0\sigma_0} \rangle$$
$$= -\langle \phi_{JM} | (\Delta\alpha \cos^2\vartheta + \alpha_\perp) | \phi_{J'M'} \rangle \overline{\xi^2} \tau \ . \qquad (3.15a)$$

The derivation is performed in Appendix A. $\overline{\xi^2}$ is the time average of the square of the electric field:

$$\overline{\xi^2} = \frac{1}{\tau} \int_{t_0}^{t_0+\tau} [\langle \alpha_{k_0\sigma_0}(t) | \hat{\mathbf{E}} | \alpha_{k_0\sigma_0}(t) \rangle]^2 dt \ . \qquad (3.15b)$$

The two constants $\alpha_{\|}$ and $\alpha_\perp$ ($\Delta\alpha \equiv \alpha_{\|} - \alpha_\perp$) determine the expectation values of the polarization tensor operator with respect to the molecular ground state (in our approximation) in the diagonal representation; $\vartheta$ is the angle between the polarization vector of the electric field and the symmetry axis of the rotational ellipsoid. The polarization tensor operator is defined as follows:

$$\hat{\alpha}_{ij} = \tau \sum_u \hat{\mu}_{ei} | \phi_u(t_0) \rangle \langle \phi_u(t_0) | \hat{\mu}_{ej} \ . \qquad (3.15c)$$

The summation is limited to highly excited states with $E_u - E_J \sim k_0$, where $E_J$ is the energy of rotational state $|\phi_{JM}\rangle$. The expression is valid for a strong monochromatic polarized electromagnetic field in the sudden approximation:

$$k_0\tau \gg 1, \ (E_J - E_{J'})\tau \ll 1 \ .$$

Coefficients $C_{J'M'}^{(JM)}$ [Eq. (3.14)] can then be obtained by diagonalizing the matrix:

$$-\langle \phi_{JM}(t_0) | \hat{V}^\dagger (\Delta\alpha \cos^2\vartheta + \alpha_\perp) \overline{\xi^2} \tau \hat{V} | \phi_{J'M'}(t_0) \rangle$$
$$= \lambda_{JM} \delta_{JJ'} \delta_{MM'} \ . \qquad (3.13b)$$

We shall make use of this expression in Sec. V to point out the similarity between nuclear and molecular cases.

### C. Comments on formation of the RCRS in a nuclear system

Since the formation of nuclear RCRS has already been described in Refs. 1–6, we shall give here only a short review and present the final results.

A short electromagnetic pulse of a quadrupole type, with which two colliding heavy ions interact, can, through a multistep process, excite one or both nuclei into a RCRS.[1–6] The energy of the projectile has to be kept below the Coulomb barrier, so that $\hat{H}'$ of Eq. (3.2) is of a pure electromagnetic type. Under special conditions (see Table I of Sec. V), the relative motion of the two nuclei can be described by a classical trajectory and also the sudden approximation, presented in Sec. III A can be used. The duration of the interaction is however very short in this case ($\lesssim 10^{-21}$ s) so that one can approximate $\hat{H}'_I(t')$ with $H'(t')$ in (3.5b).

If one of the nuclei has well expressed rotational degrees of freedom, the interaction Hamiltonian $\hat{H}'(t)$ [Eq. (3.2)] is of quadrupole type:

$$\hat{H}'(t) = \sum_m \frac{4\pi Z_t e}{5(r(t))^3} Y_{2m}^*(\mathbf{r}(t)) \hat{Q}_{2,m} \ , \qquad (3.16)$$

where $\mathbf{r}(t)$ is the position vector of the projectile referred to a coordinate system with its origin in the target[17] and $\hat{Q}_{2,m}$ denotes the electric quadrupole moment operator. In this case one diagonalizes directly the operator $\hat{R}$, obtaining

$$\langle \phi_{JM} | \hat{V}^\dagger \hat{R} \hat{V} | \phi_{J'M'} \rangle = \delta_{JJ'} \delta_{MM'} \lambda_{JM} \qquad (3.17a)$$

with $\hat{V}^\dagger \hat{V} = \hat{I}$ and

$$C_{J'M'}^{(JM)} \cong \sum_{J''M''} \langle \phi_{J'M'} | \hat{V} | \phi_{J''M''} \rangle e^{\lambda_{J''M''}} \langle \phi_{J''M''} | \hat{V}^\dagger | \phi_{JM} \rangle \ .$$
$$(3.17b)$$

When the backscattering of the projectile is considered, the operator $\hat{R}(t_0, t_0+\tau)$ obtains the simple form:[17]

$$\hat{R} = -i\tfrac{8}{3}\sqrt{\pi/5}\, q Y_{20}(\vartheta) \qquad (3.18)$$

with

$$q = \frac{Z_t e Q_0}{4 v b^2} \ .$$

We shall present numerical examples in Sec. V.

## IV. TIME EVOLUTION OF COHERENT ROTATIONAL STATES

Since the time evolution of the RCRS exhibits periodic properties, (spontaneous) $\gamma$ decay of the RCRS exhibits radiation in bursts.[1–6] For a molecular gas the periodic properties of the RCRS can also be measured through the refractive index of the gas.[7–10] If a gas is at room temperature, the electromagnetic decay rate is much smaller than the collision rate. Since the system has to be observed at times shorter than the collision time (so that collisions do not disturb the measurement process), the electromagnetic decay of the molecules in the RCRS is observable only at low temperatures.

We shall present in this section the theory of the periodic change of a refractive index of a gas of molecules in RCRS as well as the theory of electromagnetic decay.

### A. Refractive index of a molecular gas in a RCRS measured with a short pulse

At time ($t > t_0 + \tau$) molecules of a gas are in a RCRS. The density operator is in this case [Eqs. (3.6)]:

$$\hat{\rho}_0^{\mathrm{mol}}(t) = \sum_{J,M} p_{JM} | \psi_\beta^{(JM)}(t) \rangle \langle \psi_\beta^{(JM)}(t) | \ .$$

Weights $p_{JM}$ are due to scattering of molecules before $t_0$, when a short electromagnetic pulse appeared, leaving each molecule in the RCRS. If the temperature of the gas is zero, all $p_{JM}$ except one are 0. We shall test the time behavior of the molecules in the RCRS by using additional polarized monochromatic electromagnetic pulses of short duration $\tau$ at delayed times $t_{01}$. At time $t_{01}$, with $t_{01} > t_0 + \tau$, we let a pulse go through the gas and we predict the change of the polarization of the pulse as a function of $t_{01}$.

Let, at time $t > t_0 + \tau$, the molecule (evolving with $H_0^{\mathrm{mol}}$) and the free electromagnetic field (evolving with

$H_0^{\mathrm{em}}$) be described by the density matrix $\hat\rho_0(t)$:

$$\hat\rho_0(t)=|\alpha_{k_0\sigma_0}(t)\rangle\hat\rho_0^{\mathrm{mol}}(t)\langle\alpha_{k_0\sigma_0}(t)|\ ,\quad t>t_0+\tau\ . \tag{4.1}$$

The free monochromatic field has a polarization $\epsilon_{k_0\sigma_0}$. At time $t_{01}$ the interaction $\hat H'$ between a molecule and the electromagnetic field takes place:

$$\hat H=\hat H_0+\hat H',\quad t_{01}\le t<t_{01}+\tau\ , \tag{4.2}$$

with $\hat H_0=\hat H_0^{\mathrm{mol}}+\hat H_0^{\mathrm{em}}$. The interaction $H'$ is (approximately) described by Eq. (3.11). After the interaction takes place, the density operator for the system can be written as follows:

$$\hat\rho(t)=\hat U_0(t-t_{01})\hat U_1(t,t_{01})|\alpha_{k_0\sigma_0}(t_{01})\rangle\hat\rho_0^{\mathrm{mol}}(t_{01})\langle\alpha_{k_0\sigma_0}(t_{01})|\hat U_1^{\dagger}(t,t_{01})\hat U_0^{\dagger}(t-t_{01})\ . \tag{4.3}$$

The sudden approximation is now used. (See Sec. III).

The change of polarization of the electromagnetic field can be tested by measuring the intensity of photons with polarization changed over to $\epsilon_{k_0\sigma_1}$:

$$\mathrm{Tr}[\hat\rho(t)\hat a_{k_0\sigma_1}^{\dagger}\hat a_{k_0\sigma_1}]\ , \tag{4.4}$$

with $\epsilon_{k_0\sigma_0}\cdot\epsilon_{k_0\sigma_1}=0$. Equation (4.4) in the sudden approximation takes the form

$$\mathrm{Tr}[\hat\rho(t)\hat a_{k_0\sigma_1}\hat a_{k_0\sigma_1}]=\frac{k_0}{2}\tau\frac{1}{(2\pi)^3}\mathrm{Tr}[\hat\rho_0^{\mathrm{mol}}(t_{01})(\Delta\alpha\cos^2\vartheta+\alpha_\perp)]\ . \tag{4.5a}$$

The derivation is presented in Appendix B. Since $\hat Q_{2,0}=\hat Q_{2,0}^{\mathrm{int}}Y_{2,0}(\vartheta)$ for a pure rotational band with $K=0$, and $\cos^2\vartheta=\frac{2}{3}\sqrt{4\pi/5}Y_{20}+\frac{1}{3}\sqrt{4\pi}Y_{00}$, Eq. (4.5a) can be written as follows:

$$\mathrm{Tr}[\hat\rho(t)\hat a_{k_0\sigma_1}^{\dagger}\hat a_{k_0\sigma_1}]=\frac{k_0}{2}\tau\frac{1}{(2\pi)^3}\left[\frac{2}{3}\sqrt{4\pi/5}\Delta\alpha\frac{\mathrm{Tr}[\hat\rho_0^{\mathrm{mol}}(t_{01})\hat Q_{2,0}]}{\langle\psi_0|\hat Q_{2,0}^{\mathrm{int}}|\psi_0\rangle}+\frac{1}{3}(\Delta\alpha-\alpha_\perp)\right]\ . \tag{4.5b}$$

The time dependence of Eq. (4.5b) is determined by the expectation value of the quadrupole moment operator $\mathrm{Tr}[\hat\rho_0^{\mathrm{mol}}(t_{01})\hat Q_{2,0}]$. Such an expectation value was already graphically presented in Sec. II for the case when $\hat\rho_0^{\mathrm{mol}}(t_{01})\equiv|\psi_\beta\rangle\langle\psi_\beta|$. The discussion there given on the coherent periodicity is therefore also valid here. We shall present some more examples in Sec. V.

### B. Spontaneous electromagnetic decay of a system in a coherent rotational state

Let $\hat{\mathscr{H}}^{p}$, $\hat{\mathscr{H}}^{\mathrm{em}}$, and $\hat{\mathscr{H}}^{\mathrm{int}}$ be the Hamiltonian densities of a free molecule or nucleus, a free electromagnetic field, and an interaction between the molecule or nucleus and the electromagnetic field, respectively. We shall treat a system at zero temperature so that the electromagnetic decay can be followed at times long enough to notice the periodicity of the RCRS and short in comparison with the electromagnetic decay time so that first-order perturbation theory can be used. According to energy conservation one has

$$\frac{d}{dt}\left\langle\psi(t)\left|\int\mathscr{H}dV\right|\psi(t)\right\rangle=\frac{d}{dt}\left\langle\psi(t)\left|\int\hat{\mathscr{H}}^{p}dV+\int\hat{\mathscr{H}}^{\mathrm{em}}dV+\int\hat{\mathscr{H}}^{\mathrm{int}}dV\right|\psi(t)\right\rangle=0\ . \tag{4.6a}$$

Here $|\psi(t)\rangle$ is the wave function of the system, described by the total Hamiltonian density:

$$\hat{\mathscr{H}}=\hat{\mathscr{H}}^{p}+\hat{\mathscr{H}}^{\mathrm{em}}+\hat{\mathscr{H}}^{\mathrm{int}}\ . \tag{4.6b}$$

Since $\hat{\mathscr{H}}^{p}$ and $\hat{\mathscr{H}}^{\mathrm{int}}$ are different from zero only within a small sphere of molecular or nuclear radius $R$, the radiation power can be defined as

$$\begin{aligned}P_\gamma(t)&\equiv-\frac{d}{dt}\left\langle\psi(t)\left|\int_{\text{within }R}\hat{\mathscr{H}}dV\right|\psi(t)\right\rangle\\&=\frac{d}{dt}\left\langle\psi(t)\left|\int_{\text{outside }R}\hat{\mathscr{H}}^{\mathrm{em}}\right|\psi(t)\right\rangle\\&\cong\frac{d}{dt}\left\langle\psi(t)\left|\int_{\text{over all space}}\hat{\mathscr{H}}^{\mathrm{em}}dV\right|\psi(t)\right\rangle\\&=\frac{d}{dt}\left\langle\psi(t)\left|\int d^3k\,k\hat a_{k\sigma}^{\dagger}\hat a_{k\sigma}\right|\psi(t)\right\rangle\ .\end{aligned} \tag{4.7}$$

We follow the derivations of Refs. 1 and 4.

Let us suppose that our system obeys the rule $\omega J(J+1)$. Then in $\hat H^{\mathrm{int}}=\int\hat{\mathscr{H}}^{\mathrm{int}}dV$ only a quadrupole interaction appears:

$$\hat H^{\mathrm{int}}=\frac{1}{6(2\pi)^{3/2}}\int d^3\mathbf{k}\sqrt{k/2}\hat a_{k\sigma}^{\dagger}[\hat Q_{k\sigma},\hat H^{\mathrm{mol}}]+\mathrm{H.c.}\ , \tag{4.8}$$

where again the integral contains also a sum over polarizations. Dipole terms and higher than quadrupole terms have been neglected. The quadrupole moment operator $\hat Q_{k\sigma}$ is defined by

$$\hat Q_{k\sigma}=\sum_i 3e_i\frac{(\mathbf{k}\cdot\mathbf{r}_i)(\epsilon_{k\sigma}\cdot\mathbf{r}_i)}{k}\ .$$

Let the system be in the RCRS [Eq. (3.3a)] at time $t=0$: $|\psi(0)\rangle \equiv |\psi^{(J_0M_0)}(0)\rangle$ and let $|\psi_k^{(JM)}(t)\rangle$ denote one-photon states:

$$|\psi_k^{(JM)}(t)\rangle = \hat{a}^{\dagger}_{k\sigma} e^{-ikt} |\psi^{(JM)}(t)\rangle \ .$$

Looking for solutions of the equation

$$\hat{H}\,|\psi(t)\rangle = i\frac{d}{dt}\,|\psi(t)\rangle \ , \tag{4.9}$$

we expand the state vector on states with $0,1,\ldots,$ photons:

$$|\psi(t)\rangle = \sum_{J,M} a^{(JM)}(t)\,|\psi^{(JM)}(t)\rangle + \sum_{J,M} \int d^3k\, a_k^{(JM)}(t)\,|\psi_k^{(JM)}(t)\rangle + \cdots \ .$$

For the radiation power [Eq. (4.7)] to second-order perturbation theory one obtains

$$P_\gamma(t) \cong \frac{d}{dt} \sum_{J,M} \int d^3k\, k\, |a_k^{(JM)}(t)|^2 \ . \tag{4.10}$$

After some algebra,[1,4] one finally gets

$$P_\gamma(t) = \frac{1}{36\pi}\frac{d}{dt} \sum_{J,M''} \sum_{\substack{I,I',L,L',\\ M,M',N,N'}} [\Delta^5_{II'}\Delta_{LL'}\theta(\Delta_{II'}) + \Delta^5_{LL'}\Delta_{II'}\theta(\Delta_{LL'})] \overline{Q^{(JM'')}_{II'MM'} Q^{(JM'')*}_{L'LN'N}}\, e^{i(\Delta_{LL'}-\Delta_{II'})t/2} \frac{\sin(\Delta_{II'}-\Delta_{LL'})t/2}{\Delta_{II'}-\Delta_{LL'}} \ . \tag{4.11}$$

Here

$$\Delta_{JJ'} = E_J - E_{J'} \ ,$$

$$Q^{(JM'')}_{II'MM'} = C^{(JM'')*}_{IM} C^{(JM'')}_{I'M'} \langle \phi_{IM} \,|\, \hat{Q}_{k\sigma} \,|\, \phi_{I'M'} \rangle \ ;$$

the expression $\overline{QQ^*}$ means that the average over directions $\mathbf{k}/k$ and polarizations of the photon has been performed. $\theta(x)$ is 1 for $x>0$ and 0 otherwise. The expression (4.11) is obtained under the restriction that $t \gg 1/\Delta_{JJ'}$ for any $J \neq J'$. In this way we loose only the description of the first few revolutions, which is short in comparison with the time interval which we are interested in.

In Sec. V we shall present the radiation power for the CRS and for the RCRS generated in the collision of ${}^{40}_{20}\mathrm{Ca}$ against ${}^{238}_{92}\mathrm{U}$.

## V. NUMERICAL EXAMPLES

When either a molecule or a nucleus with well-expressed rotational degrees of freedom is excited into a RCRS, they both exhibit properties similar to those of the CRS. Only the energy scale, and consequently the time scale, is different, Also, the multistep processes exciting the particles in coherent states are similar in both cases. We shall here illustrate the derivations, presented in the preceding sections, by numerical examples.

### A. Formation of the coherent rotational state of particles with well-expressed rotational degrees of freedom

We shall present in this section the numerical evaluation of the coefficients as given by (3.14) and (3.17b), respectively.

These coefficients, both in the molecular and in the nuclear case, are obtained from the diagonalization of the same matrix

$$\langle \phi_{J'M'} \,|\, \sqrt{4\pi/5}\,Y_{20} \,|\, \phi_{JM} \rangle = \delta_{MM'}[(2J+1)(2J'+1)]^{1/2} \begin{pmatrix} J & 2 & J' \\ M & 0 & -M \end{pmatrix} \begin{pmatrix} J & 2 & J' \\ 0 & 0 & 0 \end{pmatrix} (-1)^{2J+M} \ , \tag{5.1}$$

where $J' = J, J\pm 2$.

$$\langle \phi_{J'M'} \,|\, \hat{V}^{\dagger}\sqrt{4\pi/5}\,\hat{Y}_{20} V \,|\, \phi_{JM} \rangle = \tilde{\lambda}_{JM}\delta_{JJ'}\delta_{M'M} \ . \tag{5.2}$$

For the eigenvalue $\lambda_{JM}$ one gets

$$\lambda_{JM}, = \begin{cases} -\frac{4}{3} iq\tilde{\lambda}_{JM}, & \text{nuclear case} \\ -\Delta\alpha\tau\overline{\xi}^2 \left[ \left[ \dfrac{1}{3} + \dfrac{\alpha_\perp}{\Delta\alpha} \right] + \dfrac{2}{3}\tilde{\lambda}_{JM} \right] , & \text{molecular case} \ . \end{cases}$$

In the nuclear case we consider the heavy-ion collision between the projectile ${}^{40}_{20}\mathrm{Ca}$ and the target ${}^{238}_{92}\mathrm{U}$ in the sudden approximation, assuming classical trajectories of ions with relative kinetic energy below the Coulomb barrier. The nucleus ${}^{40}_{20}\mathrm{Ca}$ remains in the ground state while ${}^{238}_{92}\mathrm{U}$ is excited into a RCRS. In Table I, we represent the quantities needed in numerical calculations of the coefficients $C^{(JM)}_{J'M'}$ [Eq. (3.17b)] together with the conditions needed for the approximations used.

Table II contains the analogous quantities for the molecular case. We treat the gas $CS_2$, excited into RCRS by a strong electromagnetic pulse of short duration $\tau$, again in the sudden approximation.

TABLE I. Conditions which the projectile $^{40}_{20}$Ca and the target $^{238}_{92}$U satisfy for the calculations presented in this paper. Here $b$ is the distance of closest approach; $\bar{\lambda}$ is the wavelength of the relative motion; $L$, $E$, and $v$ are the total angular momentum, the total energy, and the relative velocity, respectively; $\Delta L$ and $\Delta E$ are the angular momentum and energy transfer from the relative motion to the intrinsic motion of the target $^{238}_{92}$U, $Z_p=20$, $Z_t=92$, $eQ_0=\sqrt{16\pi/5}\langle \psi_0 | \hat{Q}^{\text{int}}_{2,0} | \psi_0 \rangle$ (quadrupole moment of the uranium nucleus), and $\omega$ is the rotational frequency.

| Quantity | Value | Condition |
|---|---|---|
| $b$ | 25 fm | |
| $b/(2\bar{\lambda})$ | 168 | $\gg 1$ |
| $L$ | 340 | $\gg 1$ |
| $\Delta L/L$ | 0.03 | $\ll 1$ |
| $v/c$ | 0.08 | $\ll 1$ |
| $b/v$ | $10^{-21}$ s | |
| $E$ | 106 MeV | |
| $\Delta E/E$ | 0.01 | $\ll 1$ |
| $q=Z_p e^2 Q_0/(4vb^2)$ | 7 | |
| $Z_t Z_p e^2/[r_0(A_t^{1/3}+A_p^{1/3})]=E_c$ | 230 MeV | |
| $E/E_c$ | 0.46 | $<1$ |
| $(b/v)(2\pi/\omega)$ | 0.002 | $\ll 1$ |
| $4\pi/\omega$ (coherence period) | $6.4\times 10^{-20}$ s | |

In Fig. 3 the coefficients $|C_{J0}^{(00)}|$ are presented, determining the RCRS for the nucleus $^{238}_{92}$U, if before the collision it was in the ground state. Coefficients $|C_{J0}^{(00)}|$ are compared with coefficients $a_{JM}^{\beta}$ for $\beta=\sqrt{10}$, which determine the CRS with $\langle \psi_\beta | \hat{J}^2 | \psi_\beta \rangle = \beta^2(\beta^2+1)$. One can notice the similarity between coefficients $|C_{J0}^{(00)}|$ and $|a_{J0}^{\beta}|$. In Table III the phase differences $|\varphi_J-\varphi_{J-2}|$ of the coefficients $C_{J0}^{(00)}$ are presented. They must be compared with $\varphi_{\beta+}=\varphi_{\beta-}=0.4$ of the CRS.

TABLE II. Quantities which appear in the calculations presented in this paper for excitation of molecules $CS_2$ by means of a strong laser pulse of short duration. $2\pi/\omega$ is the molecular rotational period, $\Delta\alpha=\alpha_{\parallel}-\alpha_{\perp}$, with $\alpha_{\parallel}$ and $\alpha_{\perp}$ being the two polarization constants, $k_0$ is the wave vector of the (excitation) electromagnetic field, $\tau$ is the duration of the pulse, and $\xi$ is the intensity of the electric field.

| Quantity | Value | Condition |
|---|---|---|
| $\omega$ | $1.35\times 10^{-5}$ eV | |
| Coherence period $\pi/4\omega$ | $38\times 10^{-12}$ s | |
| $\Delta\alpha$ | $75\times 10^{-25}$ cm$^3$ | |
| $\lambda_0$ | $1.06\times 10^{-4}$ cm | |
| $k_0$ | 1.17 eV | |
| $\omega/k_0$ | $1.15\times 10^{-5}$ | $\ll 1$ |
| $\tau$ | $1\times 10^{-12}$ s | |
| $\omega\tau$ | $2.07\times 10^{-2}$ | $\ll 1$ |
| $k_0\tau$ | $1.8\times 10^{3}$ | $\gg 1$ |
| $\frac{1}{3}\Delta\alpha\tau\xi^2$ | 15.6 | |

Figure 4 shows the coefficients $|C_{J0}^{(26,0)}|$, $|C_{J0}^{(28,0)}|$, and $|C_{J0}^{(30,0)}|$ determining the coherent state of $^{238}_{92}$U after collision with $^{40}_{20}$Ca takes place under the same conditions as in Fig. 3, except that initially $^{238}_{92}$U was in the state $|\phi_{26,0}\rangle$, $|\phi_{28,0}\rangle$, and $|\phi_{30,0}\rangle$, respectively. The states, described by the coefficients from Fig. 4, have the expectation value of the angular momentum operator equal to 20, 22, and 24, respectively. It can be noticed that the coefficients do not change appreciably if the coherent state is generated in a neighboring excited state. The reader should notice here that the parameter $q$ is rather large.

Figure 5 shows the corresponding coefficients $|C_{J0}^{(20,0)}|$, $|C_{J0}^{(22,0)}|$, and $|C_{J0}^{(24,0)}|$ obtained when Raman scattering is used to excite molecules into the coherent superposition of rotational states [Eq. (3.14)] for $\Delta\alpha\tau\xi^2/3=15.6$. Initially the molecule was in the state $|\phi_{20,0}\rangle$, $|\phi_{22,0}\rangle$, and $|\phi_{24,0}\rangle$, respectively. The similarity with the coefficients of the CRS and the coefficients of the nuclear case can be noticed. Again, the corresponding states have the expectation value of the angular momentum operator equal to 20, 22, and 24, respectively.

### B. Time evolution of systems in the coherent superpositions of rotational states

In Figs. 6(a) and 6(b) we present the $\gamma$ decay of the $^{238}_{92}$U nucleus. The corresponding coefficients are presented in Fig. 3. While marked periodic bursts appear in Fig. 6(a), where the ideal $\omega J(J+1)$ spectrum is assumed, the realistic nucleus exhibits a quasiperiodic behavior in time as shown in Fig. 6(b). But bursts are still present.

Figure 7(a) presents the $\gamma$ decay of a CRS, described by $\beta_+=\beta_-=\sqrt{22}$. Figure 7(b) shows the $\gamma$ decay of a coherent superposition of rotational states described by the coefficients $C_{J0}^{(22,0)}$ presented in Fig. 5. In both cases the $\omega J(J+1)$ rule is supposed determining the periodic nature of bursts. The width of the profile is determined by the number of states $|\phi_{JM}\rangle$ contributing appreciably to the coherent state.

The statistical nature of the initial state of a particle (nonzero temperature) can be treated through Eq. (3.6). To study the influence of the statistical nature of the initial state on CRS, we simulated Eq. (3.6) by

$$\hat{\rho}(t)=\sum_{\beta} e^{-E_\beta/kT} |\psi_\beta\rangle\langle\psi_\beta| \Big/ \sum_{\beta} e^{-E_\beta/kT} ,$$

with

$$E_\beta=\langle \psi_\beta | \hat{H}_0^{\text{mol}} | \psi_\beta \rangle .$$

Figure 8 shows the expectation values of the quadrupole moment operator taking into account the statistical nature of the initial states. One can notice that the periodic time dependence remains well expressed.

In Sec. IV we derived Eq. (4.5b), which measures at time $t=t_{01}$ the change of polarization of the electromagnetic field passing through the gas of molecules which were excited into a RCRS at a previous time $t=t_0$. We saw that the time evolution of this physical quantity is completely determined by the time dependence of the

TABLE III. Phase differences for the coefficients $C_{J0}^{(00)} = | C_{J0}^{(00)} | e^{i\varphi_J}$, obtained in the calculation of a heavy ion collision of $^{238}_{92}U$ and $^{40}_{20}Ca$ under conditions presented on Table I.

| | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $J$ | 0 | | 2 | | 4 | | 6 | | 8 | | 10 | |
| $\lvert \varphi_J - \varphi_{J-2} \rvert$ | | 3.4047 | | 3.7205 | | 2.3929 | | 2.7370 | | 1.8534 | | 1.6731 |
| $J$ | 12 | | 14 | | 16 | | 18 | | 20 | | 22 | |
| $\lvert \varphi_J - \varphi_{J-2} \rvert$ | | 1.5134 | | 1.6090 | | 1.5985 | | 1.5919 | | 1.5540 | | |

quadrupole moment operator. Therefore Fig. 8 also determines the time dependence of the refractive index in the gas.

This effect has been observed by Heritage, Gustafson, and Lin[7] for the $CS_2$ gas at 325 K. For this gas, whose molecules are linear and symmetrical, the statistics of the identical sulfur nuclei allows the existence of only even rotational states ($J$ is even). At 325 K about 20 rotational states ($0 \leq J \leq 40$) are relevant in the formation of the RCRS, while collision times are a few hundred picoseconds. Since the duration of their laser pulse was around 5 ps, the gas was in good approximation "collisionless" with regard to the phenomenon under study.

The experimental results,[7] are shown in Fig. 9, where bursts show up at 38 and 76 ps, after the initial excitation, in agreement with the discussion of the coherent behavior of the quadrupole moment (2.9) given in Sec. II. The structure shown in Fig. 9 is not perfectly periodic because of the interference provoked by the coupling to vibrational levels and, to some extent, by molecular collisions.

In Refs. 8 and 9 Lin *et al.* have given a theoretical evaluation of the effect using perturbation theory to the first order in the effective dipole-electric field semiclassical Hamiltonian $\widetilde{H}' = -\frac{1}{2}\xi^2(t)(\Delta\alpha \cos^2\vartheta + \alpha_\perp)$. Even though the interaction lasts a short time, due to the intensity of the optical pulse a many-step process actually occurs with contributions to all orders of perturbation theory and this has been taken into consideration in this paper through the use of the sudden approximation with quantum field theoretic Hamiltonian (3.11).

The difference between the two approaches is dramatic as far as the gas temperature dependence of the final result is concerned. In particular, when the temperature of the gas reaches zero, the effect of the periodic change of refractive index disappears completely in the approach by Lin *et al.*[8,9] Instead, in a multistep process many rotational states are populated, leaving the effect well expressed even when the temperature goes to zero.

## VI. CONCLUSIONS

We have shown in this paper that a short and sufficiently strong electromagnetic pulse of a quadrupole type generates, in a multistep process, a coherent superposition of rotational states for a system with well-expressed rotational degrees of freedom. This process occurs in a very similar way both in molecular and in nuclear cases in spite of the fact that the relative mechanisms seem completely different in the two cases. We used as a generating mechanism in the nuclear case the collision between two ions below the Coulomb barrier, and in the molecular case the Raman scattering of a laser pulse on a molecule.

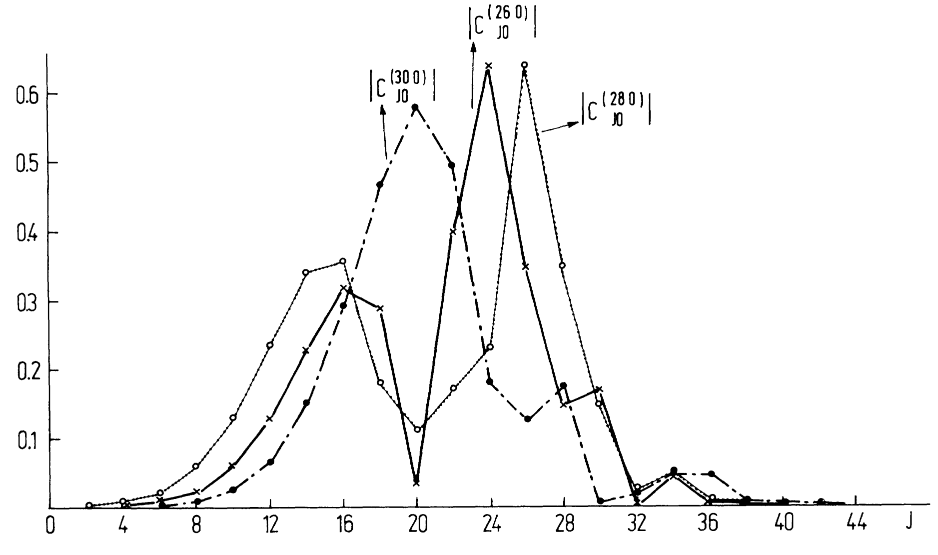

FIG. 4. Coefficients $| C_{J0}^{(26,0)} |$, $| C_{J0}^{(28,0)} |$, and $| C_{J0}^{(30,0)} |$ for $^{238}_{92}U$ are presented for the same conditions as in Fig. 3, except that initially the nucleus is in $| \phi_{26,0} \rangle$, $| \phi_{28,0} \rangle$, and $| \phi_{30,0} \rangle$, respectively.

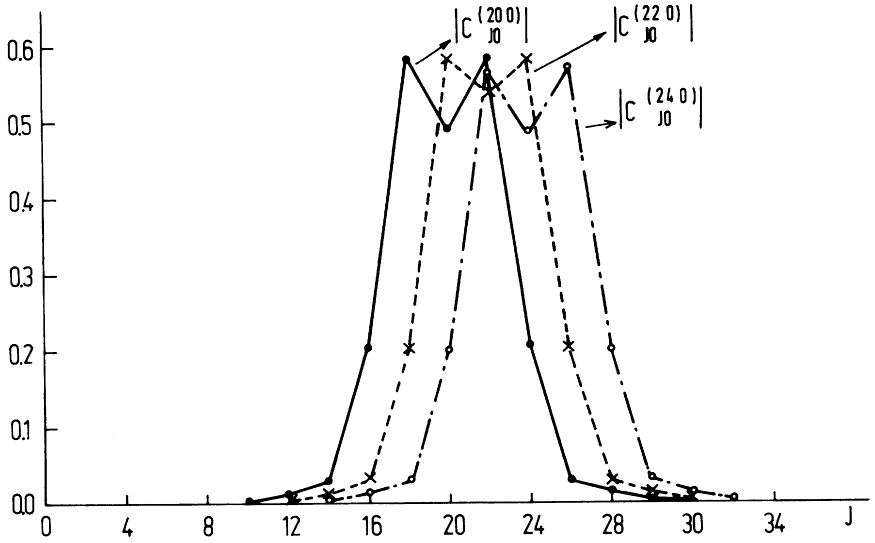

FIG. 5. Coefficients $|C_{J0}^{(20,0)}|$, $|C_{J0}^{(22,0)}|$, and $|C_{J0}^{(24,0)}|$ for the Raman scattering of a laser pulse on $CS_2$. The parameters used in the calculations are shown in Table II.

Such systems show a periodic behavior in time for their dynamical observables. The origin of the periodicity lies in the fact that each of the coherent states evolves in time in a different way destroying the initially arranged phases. Since the energies of the rotational band obey the $\omega J(J+1)$ rule, the phases recover periodically. Molecules and nuclei differ only in the time scale. While in the nuclear case the scale is $10^{-19}$ s and therefore the effect is not directly measurable, the scale of $10^{-10}$ s makes it feasible in the molecular case. An experiment of this kind has been, in fact, performed[7] showing periodic changes of

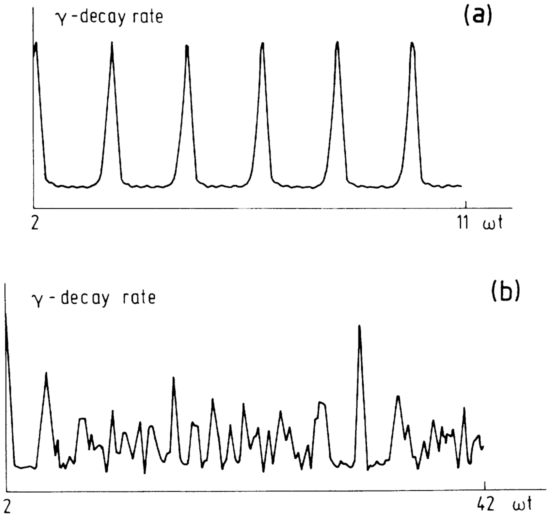

FIG. 6. (a) $\gamma$-decay rate for an even-even nucleus with the ideal rotational band. $\omega$ is taken to be 0.007 MeV (which reproduces rather well the energies of ${}^{238}_{92}U$). The nucleus was initially in the ground state (see Fig. 3 and Table I). (b) $\gamma$-decay rate for ${}^{238}_{92}U$, obtained using actual experimental energies.

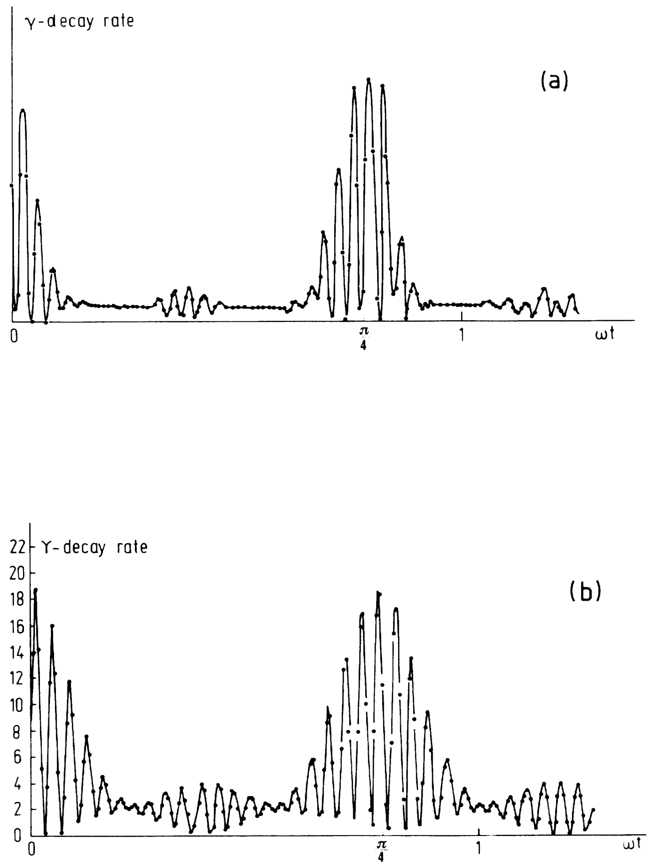

FIG. 7. (a) Electromagnetic decay rate for a CRS with $\beta_+ = \beta_- = \sqrt{22}$. The $\omega J(J+1)$ rule is assumed. (b) Electromagnetic decay rate for a CRS described by coefficients $C_{J0}^{(22,0)}$ from Fig. 5. The $\omega J(J+1)$ rule is assumed.

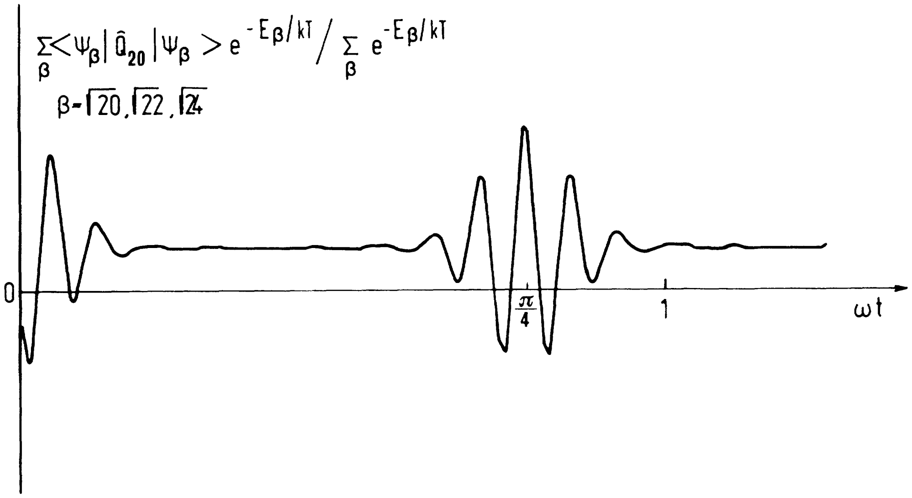

FIG. 8. The time evolution of the expectation value of the quadrupole moment operator. The statistical nature of the initial density matrix operator is simulated by $\sum_\beta \langle \psi_\beta | \hat{Q}_{2,0} | \psi_\beta \rangle e^{-E_\beta/kT} / \sum_\beta e^{-E_\beta/kT}$. We choose $kT=\frac{1}{40}$ eV and $\beta=\sqrt{20},\sqrt{22},\sqrt{24}$.

the refractive index in the gas $CS_2$. In both the nuclear and the molecular case, the statistical nature of the initial density matrix does not spoil the effect, if the generating mechanism is strong enough.

Since molecules and nuclei never obey perfectly the $\omega J(J+1)$ rule and since other degrees of freedom (vibrational, internal) are coupled to the rotational one, they express quasiperiodic rather than periodic structure in time evolution. Additionally, the statistical nature of collisions in a gas eventually spoils this effect.

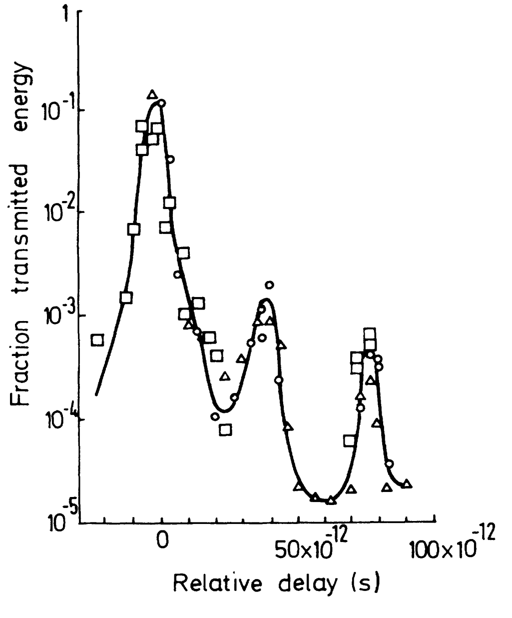

FIG. 9. The measured time dependence of the fraction of the energy transmitted through the analyzing polarizer Ref. 7. Time $t=0$ is chosen when the first pulse excites molecules of $CS_2$.

## ACKNOWLEDGMENTS

We would like to thank M. Čopič for having pointed out to us some of the literature for the molecular case. One of the authors (N. M.-B.) would like to thank J. Brickmann for the much needed information that a gas of molecules of very low temperature can be prepared so that the effect of the periodic change of refractive index can be measured even at very low temperature. In such a case a multistep process, exciting each molecule into a coherent superposition of many rotational states, is needed in order that the periodic time behavior of the refractive index is measurable. The same author would like to thank W. G. Harter and J. Heller for information about the work presented in Ref. 18 which also studies the time evolution of the coherent states obtained as superpositions of states $|\psi_{JMK}\rangle$ with fixed $J$ and $M$ and different $K$: $\hat{H}\,|\psi_{JMK}\rangle = E_{JK}\,|\psi_{JMK}\rangle$, with $\hat{H}=A\hat{J}^2+B\hat{J}_z^2$. Coherent states of this type are very useful, e.g., to study the spectra of systems with octahedral structure. One of the authors (L.F.) was supported in part by the University of Trieste and by the National Institute for Nuclear Physics (INFN), Section of Trieste.

## APPENDIX A

In this Appendix we shall prove Eq. (3.15a). Starting from the matrix element (3.12) and using (3.5b) and (3.11) one gets

$$\langle\alpha_{k_0\sigma_0}(t_0)|\langle\phi_{JM}(t_0)|\hat R^2|\phi_{J'M'}(t_0)\rangle|\alpha_{k_0\sigma_0}(t_0)\rangle$$

$$=\sum_u (E_u-E_J)(E_{J'}-E_u)\frac{1}{2(2\pi)^3k_0}$$

$$\times\int_{t_0}^{t_0+\tau}dt'\int_{t_0}^{t_0+\tau}dt''\,e^{-i(E_u-E_J)(t'-t_0)}e^{-i(E_{J'}-E_u)(t''-t_0)}$$

$$\times\langle\phi_{JM}(t_0)|(\epsilon_{k_0\sigma_0}\cdot\hat{\boldsymbol{\mu}}_e)|\phi_u(t_0)\rangle\langle\phi_u(t_0)|(\epsilon_{k_0\sigma_0}\cdot\hat{\boldsymbol{\mu}}_e)|\phi_{J'M'}(t_0)\rangle$$

$$\times\int\frac{d^2\alpha_u}{\pi}|\langle\alpha_{k_0\sigma_0}(t_0)|\alpha_u\rangle|^2$$

$$\times(\alpha^*_{k_0\sigma_0}e^{ik_0(t'-t_0)}+\alpha_u e^{-ik_0(t'-t_0)})(\alpha^*_u e^{ik_0(t''-t_0)}+\alpha_{k_0\sigma_0}e^{-ik_0(t''-t_0)})\ . \quad \text{(A1)}$$

The wavelength $\lambda_0=2\pi/k_0$ of the monochromatic polarized field should be short, so that $k_0\tau\gg1$ (in our case $k_0\tau\sim10^4$). Consequently, after integration over $t'$ and $t''$, one can drop all the terms proportional to $(\sin k_0\tau)/k_0\tau$. Keeping only those terms for which $E_u-E_J\sim k_0$, one finally gets

$$\langle\alpha_{k_0\sigma_0}(t_0)|\langle\phi_{JM}(t_0)|\hat R^2|\phi_{J'M'}(t_0)\rangle|\alpha_{k_0\sigma_0}(t_0)\rangle$$

$$=-\frac{\tau^2k_0|\alpha_{k_0\sigma_0}|^2}{(2\pi)^3}\left\langle\phi_{JM}\left|(\epsilon_{k_0\sigma_0}\cdot\hat{\boldsymbol{\mu}}_e)\sum_u\right|\phi_u\right\rangle\langle\phi_u|(\epsilon_{k_0\sigma_0}\cdot\hat{\boldsymbol{\mu}}_e)|\phi_{J'M'}\rangle\ . \quad \text{(A2)}$$

Let us now introduce the electric field vector operator:

$$\hat{\mathbf{E}}=\frac{i}{(2\pi)^{3/2}}\int d^3k\sqrt{k/2}(\hat a_{k\sigma}e^{i\mathbf{k}\cdot\mathbf{r}}-\hat a^\dagger_{k\sigma}e^{-i\mathbf{k}\cdot\mathbf{r}})\epsilon_{\mathbf{k}\sigma}\ . \quad \text{(A3)}$$

Following the same line of thought as above, in the dipole approximation $\exp(\pm i\mathbf{k}\cdot\mathbf{r})\simeq1$ one gets

$$\overline{\xi^2}\equiv\frac{1}{\tau}\int_{t_0}^{t_0+\tau}[\langle\alpha_{k_0\sigma_0}(t)|\hat{\mathbf{E}}|\alpha_{k_0\sigma_0}(t)\rangle]^2dt$$

$$=\frac{k_0}{(2\pi)^3}|\alpha_{k_0\sigma_0}|^2\ , \quad \text{(A4)}$$

where by $|\alpha_{k_0\sigma_0}(t)\rangle$ we mean $\exp[-i\hat H_0^{\rm em}(t-t_0)]|\alpha_{k_0\sigma_0}(t_0)\rangle$. On the other hand, the matrix element appearing in (A2) can be simplified using the notion of polarizability. In general, we define the polarizability tensor from (3.15c). In the diagonal representation one splits $\boldsymbol{\mu}$ into $\boldsymbol{\mu}_\parallel+\boldsymbol{\mu}_\perp$ where $\boldsymbol{\mu}_\parallel=(\boldsymbol{\mu}\cdot\mathbf{a})\cdot\mathbf{a}$, $\mathbf{a}$ being the symmetry axis of the molecule. One finally gets

$$\tau\left\langle\phi_{JM}\left|(\epsilon_{k_0\sigma_0}\cdot\boldsymbol{\mu}_e)\sum_u\right|\phi_u\right\rangle\langle\phi_u|(\epsilon_{k_0\sigma_0}\cdot\boldsymbol{\mu}_e)|\phi_{J'M'}\rangle$$

$$=\alpha_\parallel\langle\phi_{JM}|\cos^2\vartheta|\phi_{J'M'}\rangle+\alpha_\perp\langle\phi_{JM}|\sin^2\vartheta|\phi_{J'M'}\rangle\ , \quad \text{(A5)}$$

where $\vartheta$ is the angle between the polarization vector of the electric field $\epsilon_{k_0\sigma_0}$ and the symmetry axis $\mathbf{a}$; $\alpha_\parallel$ and $\alpha_\perp$ are the expectation values of the polarization tensor operator in the diagonal representation. In order to get (A5), we have used the fact that the parities of $|\phi_{JM}\rangle$ and $|\phi_{J'M'}\rangle$ are the same so that the mixed term $\sin\vartheta\cdot\cos\vartheta$ does not contribute.

Introducing the anisotropy $\Delta\alpha\equiv\alpha_\parallel-\alpha_\perp$, one finally gets (3.15a):

$$\langle\alpha_{k_0\sigma_0}|\langle\phi_{JM}|\hat R^2|\phi_{J'M'}\rangle|\alpha_{k_0\sigma_0}\rangle$$

$$=-\langle\phi_{JM}|(\Delta\alpha\cos^2\vartheta+\alpha_\perp)|\phi_{J'M'}\rangle\overline{\xi^2}\tau\ . \quad \text{(A6)}$$

## APPENDIX B

In this Appendix we derive Eq. (4.5a), which describes the change of polarization of an electromagnetic field when interacting for a short time $\tau$ with a molecule of a gas.

Let $\hat\rho_0$ describe the state of the system of a free molecule and a free monochromatic polarized electromagnetic field:

$$\hat\rho_0(t)=|\alpha_{k_0\sigma_0}(t)\rangle\hat\rho_0^{\rm mol}(t)\langle\alpha_{k_0\sigma_0}(t)|\ . \quad \text{(B1)}$$

Let at time $t_{01}$ the interaction $\hat H'$ [Eq. (3.11)] of a short duration $\tau$ take place. The density matrix operator $\hat\rho$ of the system at $t\geq t_{01}$ is then

$$\hat\rho(t)=\hat U_0(t-t_{01})\hat U_1(t,t_{01})|\alpha_{k_0\sigma_0}(t_{01})\rangle\hat\rho_0^{\rm mol}(t_{01})\langle\alpha_{k_0\sigma_0}(t_{01})|\hat U_1^\dagger(t,t_{01})\hat U_0^\dagger(t-t_{01})\ . \quad \text{(B2)}$$

Here, $\hat{U}_0(t)=e^{-i\hat{H}_0^{\text{em}}t}e^{-i\hat{H}_0^{\text{mol}}t}$ and $\hat{U}_1(t,t_{01})=e^{\hat{R}(t,t_{01})}$. We check the change of the polarization of the electromagnetic field by counting the number of photons which changed polarization from $\epsilon_{k_0\sigma_0}$ to $\epsilon_{k_0\sigma_1}$:

$$\text{Tr}[\hat{\rho}(t)\hat{a}^{\dagger}_{k_0\sigma_1}\hat{a}_{k_0\sigma_1}] \tag{B3}$$

with $\boldsymbol{\epsilon}_{k_0\sigma_0}\cdot\boldsymbol{\epsilon}_{k_0\sigma_1}=0$. One readily obtains

$$\text{Tr}[\hat{\rho}(t)\hat{a}^{\dagger}_{k_0\sigma_1}\hat{a}_{k_0\sigma_1}]=\text{Tr}[\hat{\rho}_0^{\text{mol}}(t_{01})\langle\chi(t)\,|\,\chi(t)\rangle]\ , \tag{B4}$$

where $|\chi(t)\rangle$ is a photon state defined by

$$|\chi(t)\rangle=\hat{a}_{k_0\sigma_1}e^{\hat{R}(t,t_{01})}\,|\,\alpha_{k_0\sigma_0}(t_{01})\rangle\ .$$

Using the identity

$$[\hat{a},e^{\hat{R}}]=e^{\hat{R}}[\hat{a},\hat{R}]$$

which holds since $[\hat{a},[\hat{a},\hat{R}]]=[\hat{R},[\hat{a},\hat{R}]]=0$, one gets

$$|\chi(t)\rangle=e^{\hat{R}}\int_{t_{01}}^{t_{01}+\tau}dt'\,e^{ik_0(t'-t_{01})}e^{i\hat{H}_0^{\text{mol}}(t'-t_{01})}\hat{O}_{k_0\sigma_1}e^{-i\hat{H}_0^{\text{mol}}(t'-t_{01})}\,|\,\alpha_{k_0\sigma_0}(t_{01})\rangle\ ,$$

$$\begin{aligned}\langle\chi(t)\,|\,\chi(t)\rangle=&\left[\int_{t_{01}}^{t_{01}+\tau}dt'\,e^{ik_0(t'-t_{01}}e^{i\hat{H}_0^{\text{mol}}(t'-t_{01})}\hat{O}_{k_0\sigma_1}e^{-i\hat{H}_0^{\text{mol}}(t'-t_{01})}\right]\\ &\times\left[\int_{t_{01}}^{t_{01}+\tau}dt''\,e^{-ik_0(t''-t_{01})}e^{i\hat{H}_0^{\text{mol}}(t''-t_{01})}\hat{O}^{\dagger}_{k_0\sigma_1}e^{-i\hat{H}_0^{\text{mol}}(t''-t_{01})}\right]\ ,\end{aligned}$$

where we have defined

$$\hat{O}_{k\sigma}=(2\pi)^{-3/2}(2k)^{-1/2}\boldsymbol{\epsilon}_{k\sigma}\cdot[\hat{\boldsymbol{\mu}}_e,\hat{H}_0^{\text{mol}}]\ .$$

The evaluation of $\langle\chi(t)\,|\,\chi(t)\rangle$ is rather straightforward if the integration over time is performed in a similar way as in Appendix A. Then one gets

$$\text{Tr}[\hat{\rho}(t)\hat{a}^{\dagger}_{k_0\sigma_1}\hat{a}_{k_0\sigma_1}]=\frac{\tau k_0}{2(2\pi)^3}\text{Tr}[\hat{\rho}_0^{\text{mol}}(t_{01})(\Delta\alpha\cos^2\vartheta+\alpha_{\perp})]\ . \tag{B5}$$

Since expression (B5) depends on the time $t_{01}$, when the test pulse appears, it measures the time dependence of the coherence of the molecular state formed at time $t_0$ and described by $\hat{\rho}_0^{\text{mol}}(t_{01})$.

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