Inversion of the population of the rotational levels of molecules by an ultrashort light pulse

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It is shown that a laser pulse of length τ will produce an inversion of the population of rotational levels of molecules when $\omega_r^{-1}\tau \ll 1$, where ω_r is the rotational frequency. When the molecule has an intrinsic dipole moment, it is possible to produce generation at the frequency corresponding to a purely rotational transition.

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Existing optical methods of producing population inversion depend on resonance between the frequency of the pump field and the frequency of the quantum transition in the system to be excited, or on certain definite phase relationships as, for example, in the case of a π-pulse. In this paper, we shall consider the situation in which optical excitation produces a redistribution within the quantum-mechanical system even when neither of these two conditions is satisfied.

We shall be concerned with the excitation of rotational levels of a molecule by an ultrashort pulse of light. We shall consider transitions between the rotational states of linear molecules produced by a laser pulse of length τ much smaller than the reciprocal of the rotational frequency ω_r ($\omega_r \tau \ll 1$, see[1]). For very approximate calculations, let us suppose that $\tau \sim 10^{-12}$ sec. Since the ratio of vibrational and rotational times is large, the quantity $\omega_v \tau$ could be large, where ω_v is the vibrational frequency. Therefore, the vibrations undergo an adiabatic perturbation up to $\tau \sim 10^{-13}$–10^{-14} sec and, therefore, their excitation has a low probability. Since the characteristic wavelength of light is much greater than the linear dimensions of a molecule, the electric field can be regarded as quasistationary, i.e., we can write

$$\mathbf{E}=\mathbf{e}E_0(t)\cos\omega_0 t.$$

where $\mathbf{e}$ is the polarization vector, E_0 is the amplitude, and ω_0 is the wave frequency. In actual fact, ω_0 corresponds to the infrared or red region in the visible part of the spectrum, so that it is appreciably smaller than the electronic frequencies of the molecule. We may therefore use the expression for the energy of interaction between the molecule and the electromagnetic wave averaged over the electron state:

$$V=\tfrac{1}{2}\alpha_{ik}e_i e_k E_0^2\cos^2\omega_0 t. \tag{1}$$

where α_{ik} is the molecular polarizability tensor.

When dispersion is taken into account, α_{ik} must be replaced by

$$\frac{\partial}{\partial\omega_0}[\omega_0\alpha_{ik}(\omega_0)].$$

When the molecule has a dipole moment, the corresponding interaction has the form $\mathbf{d}\cdot\mathbf{e}E_0\cos\omega_0 t$ and its contribution to transitions is negligible because of the rapid oscillations [the characteristic length of $E_0(t)$ is equal to the pulse length τ which, by hypothesis, is much less than ω_r^{-1} but, of course, is much greater than ω_0^{-1}].

In the approximation we are considering, the transition amplitude between states K, M and K', M' is given by[1]

$$a_{K,M;K',M'}=\int_0^{\pi}\int_0^{2\pi}\Psi_{K,M}(\theta,\varphi)e^{-ip\cos\theta}\Psi_{K',M'}(\theta,\varphi)\sin\theta\,d\theta\,d\varphi,$$

$$p=\int_{-\infty}^{+\infty}V\,dt. \tag{2}$$

where θ and φ are angles defining the rotation of the molecule, K and M are rotational quantum numbers, and $\Psi_{K,M}$ are the rotational wave functions.

For a linear molecule, the tensor α_{ik} referred to the principal axes has only two independent components, namely, $\alpha_{11}=\alpha_{22}=\alpha_\perp$, $\alpha_{33}=\alpha_\parallel$. Transforming to the laboratory frame, and assuming that light is linearly polarized, we obtain

$$p=\frac{\alpha_{ik}}{4}e_i e_k\int_{-\infty}^{+\infty}E_0^2\,dt=\gamma\cos^2\theta,$$

$$\gamma=\frac{1}{4}(\alpha_\parallel-\alpha_\perp)\int_{-\infty}^{+\infty}E_0^2\,dt. \tag{3}$$

The term independent of θ is omitted from (3).

Only those transitions are possible for which the selection rules $\Delta M=0$, $\Delta K=\pm 2, \pm 4, \ldots$ are satisfied.

Henceforth, we shall be interested in the population of the rotational levels of the molecule, $f(K, M)$, after the application of the pulse. Equation (2) shows that the relation between the density matrix $\hat{\rho}_+$ after the pulse is related to the initial density matrix $\hat{\rho}_-$ by the formula

$$\hat{\rho}_+=e^{i\hat{p}^*}\hat{\rho}_-e^{-i\hat{p}}, \tag{4}$$

where $\hat{p}^*$ is the Hermitian conjugate operator. We shall suppose that the initial matrix is diagonal and

corresponds to the equilibrium Boltzmann distribution, i.e.,

$$f_-(K, M)=Z^{-1}\exp\{-\varepsilon K(K+1)\}. \quad (5)$$

where $\varepsilon = B/kT$, B is the rotational constant, T is the temperature, and Z is the rotational partition function. The occupation numbers after the application of the pulse are then given by

$$f_+(K, M)=\sum_{K', M'} |a_{K, M; K', M'}|^2 f_-(K', M'). \quad (6)$$

Let us consider in greater detail the population of the two lowest-lying rotational levels. They correspond to $K=0$, $M=0$ and $K=1$, $M=\pm 1, 0$. We shall suppose that the electronic ground state of the molecule corresponds to the term $^1\Sigma$. The rotational wave functions can then be expressed in terms of the associated Legendre polynomials. To determine the occupation numbers for the above states, we must have the amplitudes $a_{0,0;K,0}$; $a_{1,0;K,0}$; $a_{1,\pm 1;K,\pm 1}$. Calculations based on (2) yield

$$a_{0,0;K,0}=(2i\gamma)^{K/2}\sqrt{2K+1}\,\frac{(K-1)!!}{(2K+1)!!}F\left(\frac{K+1}{2}, K+\frac{3}{2}; i\gamma\right),$$

$$a_{1,0;K,0}=(2i\gamma)^{(K-1)/2}\sqrt{3(2K+1)}\,\frac{K!!}{(2K+1)!!}F\left(\frac{K}{2}+1, K+\frac{3}{2}; i\gamma\right),$$

$$a_{1,\pm 1;K,\pm 1}=(2i\gamma)^{(K-1)/2}\sqrt{\frac{3}{2}\frac{K(K+1)}{(2K+1)}}\,\frac{(K-2)!!}{(2K-1)!!}F\left(\frac{K}{2}, K-\frac{3}{2}; i\gamma\right). \quad (7)$$

where F is the confluent hypergeometric function.

For realistic values of the field E_0 in the pulse, the parameter γ may turn out to be substantially greater than unity. For example, for the HCN molecule, we have $\alpha_\parallel - \alpha_\perp = 16$ a.u., and for $\tau = 10^{-12}$ sec we find that γ becomes greater than unity when $E_0 \gtrsim 10^7$ V/cm. The case $\gamma \gg 1$ is, of course, of particular interest. The Boltzmann distribution is then substantially distorted, and it can be shown[1] that the most populated states are those with $K^2 \sim \gamma$. Therefore, the population looked upon as a function of K has a maximum and, consequently, the lower levels are overpopulated.

When $\varepsilon\gamma \gg 1$, we can use the asymptotic behavior of the hypergeometric function, and obtain

$$f_+(1, \pm 1)-f_+(0, 0) > \frac{\sqrt{\pi}}{8\gamma^{3/4}}\sqrt{\varepsilon}. \quad (8)$$

The populations of sublevels with different M and given K exhibit a peculiar modulation in this limit: $f_+(K, M)$ is proportional to γ^{-1} or γ^{-2}, depending on whether $K+M$ is even or odd.

To determine the populations in the region $\gamma\varepsilon \lesssim 1$, we have carried out a numerical calculation for the $HC^{12}N^{14}$ molecule. The results for $\varepsilon = 0.02$ were as follows:

γ:	1	5	10	15	20
$f_+(1,\pm1)-f_+(0,0)$:	$-7.4\cdot10^{-4}$	10^{-4}	$1.1\cdot10^{-3}$	$1.3\cdot10^{-3}$	$1.1\cdot10^{-3}$

When the molecule has an intrinsic dipole moment, these states are connected by a dipole transition. We therefore have the possibility of generation at frequency corresponding to a purely rotational transition of the molecule.

Let us estimate the gain β for the HCN molecule (dipole moment $d = 2.9$ Debye units, $B = 44\,316$ MHz):

$$\beta = \frac{\lambda^2 W}{4\Gamma} n[f_-(1, \pm 1)-f_-(0, 0)].$$

where λ, W, and Γ are, respectively, the wavelength, probability, and linewidth of the given transition, and n is the gas density. The probability W of a purely rotational transition is given by the well-known formula[2]

$$W = \frac{4}{3\lambda^3} d^2 \frac{K}{2K-1} = \frac{4d^2}{9\lambda^3}. \quad (10)$$

We shall now estimate the gain for $n \sim 10^{18}$ cm^{-3}, $T \sim 10^2$ °K, $\tau = 10^{-12}$ sec, $\gamma = 10$ (which corresponds to $E_0 = 4\times10^7$ V/cm, i.e., the current density is $Q = 4\times10^{12}$ W/cm^2).

For these values of the parameters, the quantity Γ must be interpreted as the collisional width, since it is the largest width. In fact, according to[3], the collisional width for HCN is 2.5×10^9 sec^{-1} for $n = 10^{18}$ cm^{-3}, whereas the Doppler width is $kv \sim 6\times10^5$ sec^{-1}. The value obtained for the gain is $\beta \approx 0.3$ cm^{-1}. We note that this estimate is not very dependent on the gas density and on the dipole moment of the molecule since, so far, the predominant broadening mechanism is provided by collisions between molecules, and the quantity Γ is proportional to nd^2 which cancels out with the corresponding factors in the overpopulation and the spontaneous transition probability.

The resulting emission is associated with the rotational transition $1, \pm 1 \to 0, 0$ ($\lambda \approx 0.3$ cm), i.e., two circularly polarized waves are generated. The quantization axis for the angular momentum is the direction of the wave vector of the generated wave, whereas, in population calculations, we assumed that the quantization axis was the direction of the electric field in the exciting pulse. It follows that the generated radiation originating from the rotational transition is propagated at right-angles to the direction of propagation of the exciting pulse.

It is clear from the foregoing estimates that the necessary field E_0 is very high, and the problem therefore arises as to whether the ionization of the molecule is possible under the action of the laser pulse. It is clear that, if a substantial fraction of the molecules is ionized during the operation of the pulse, this will lead to strong excitation of rotational and vibrational degrees of freedom which, in turn, will highly distort the distribution over the rotational sublevels, which we have established.

The molecule can be ionized in two ways: either by a cascade breakdown due to the acceleration of electrons in the field of the laser wave, or directly by field ionization through tunnel or multiphoton effects. To

estimate the efficiency of the cascade mechanism, we must compare the energy $\mathscr{E}$ given to the electron during the operation of the pulse with the ionization potential I_0. Assuming that the electron velocity is of the order of 10^8 cm/sec, and that the cross section for a collision between the electron and the molecule is of the order of 10^{-16} cm^2, we find that $\nu \sim 10^{10}$ sec^{-1} when $n \sim 10^{18}$ cm^{-3}. Suppose that the frequency ω_0 of the laser pulse corresponds to a 1.2-eV photon. In this situation, we have $\omega_0 \gg 1/\tau \gg \nu$, and elementary calculations show that $\mathscr{E} = e^2 E_0 v \tau / 4m\omega_0^2 \sim 3 \times 10^{-3}$ eV, where e, m are, respectively, the charge and mass of the electron. The ionization potential I_0 of the HCN molecule is 13.9 eV, i.e., $\mathscr{E} \ll I_0$, so that the cascade does not succeed in developing during the operation of the pulse.

The field ionization probability can be estimated from the results reported by Keldysh.[4] Estimates show that the largest contribution is provided by multiphoton ionization through an intermediate level.

The probability of ionization during the pulse is of the order of[4]

$$10^{12}\tau\,[\text{sec}] \cdot (6 \cdot 10^{-15} Q\,[\text{W/cm}^2])^{n_s - 1},$$

where n_s is the number of photons in the energy interval between the ground and intermediate levels. Hence, it is clear that, for our values of τ and Q, the fraction of ionized molecules is negligible.

We note in conclusion that, since the above mechanism of producing overpopulation is not selective, it should, at least in principle, enable us to achieve generation simultaneously on a number of rotational transitions in a given molecule, or simultaneously on different molecules, when a gas mixture is employed.

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De-excitation of metastable nuclei during negative-muon capture

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A negative muon captured by a metastable nucleus can accelerate the de-excitation of the latter by several orders of magnitude. For a certain relationship between the nuclear and mesonic level spacing, this de-excitation is accompanied by the ejection of a meson, which can then participate in the de-excitation of other nuclei. When a specimen with sufficiently high density of metastable nuclei and a meson beam of sufficiently high intensity are used, this process leads to a rapid increase in the γ activity of the specimen, and can be used as a basis for a powerful source of monochromatic γ rays. The single-particle model is used in this paper to calculate the decay probability for different channels of metastable nuclear states in the presence of a meson. The conditions under which an experimental realization of the de-excitation acceleration effect may be possible are discussed.

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I. INTRODUCTION. FORMULATION OF THE PROBLEM

The possibility in principle of accumulating considerable quantities of metastable nuclei during the processes involved in nuclear technology, followed by concentration through chemical[1,2] and laser[3] methods, leads naturally to the question as to whether the rate of decay of such nuclei could be controlled. The possibility of controlling the rate of radiative decay of metastable nuclei through stimulated γ emission in a laser-type device (the current state of the γ laser problem is reviewed in[7]) was discussed in[4–6]. The possibility of influencing nuclear decay involving the participation of atomic-shell electrons (K capture and internal conversion) through ionization was considered in[8]. The possible acceleration of the de-excitation of a metastable nucleus through the transfer of some of its angular momentum to the atomic-shell electrons was recently considered in[9]. The influence of the electron shell turned out to be negligible because the ratio r_n/r_a of the nuclear to atomic radius was very small. In this respect, the mesonic atom is more attractive than the ordinary atom because the ratio r_n/r_a can then vary in a broad range of values, depending on the nuclear charge.

In this paper, we consider the possibility of de-excitation of metastable nuclei by negative-muon bombardment. We shall not consider muon capture as such, since it has been discussed in detail in the fundamental papers.[10–12] The γ-ray spectrum emitted during cap-