neton,[6] it follows that $\mu_{\rm eff}$ for N_2 is ≈ 1.5 nuclear magnetons. Moreover, since the nuclear magnetic moment of the N_2 molecule is ≈ 0.4 nuclear magnetons, we can regard it as highly likely that the effect observed in nitrogen is due to the rotational as well as the nuclear magnetic moments. The more accurate measurements which are proposed should give us quantitative data on the rotational magnetic moment of N_2. It is also proposed to carry out measurements on other gases.

The authors are grateful to I. K. Kikoin, Yu. M. Kagan, A. A. Sazykin, and L. A. Maksimov for their valuable discussions and advice, to V. Kh. Volkov for his interest and help in this work, and to V. I. Nikolaev for his constant help in the making of the instruments and carrying out of the measurements. The authors are also grateful to the late L. D. Puzikov for his valuable discussions and advice.

---

[1] The layout was similar to the oxygen gas analyzer with a constant magnetic field, described in [8].

---

[1] H. Senftleben and J. Pietzner, Ann. Physik **16**, 907 (1933); **27**, 108, 117 (1936); **30**, 541 (1937).

[2] H. Senftleben and H. Gladisch, Ann. Physik **30**, 713 (1947); **33**, 471 (1938).

[3] E. Rieger, Ann. Physik **31**, 453 (1938).

[4] C. J. Gorter, Naturwissenschaften **26**, 140 (1938).

[5] F. Zernike and C. van Lier, Physica **6**, 961 (1939).

[6] Yu. M. Kagan and L. A. Maksimov, JETP **41**, 842 (1961), Soviet Phys. JETP **14**, 604 (1962).

[7] Beenakker, Scoles, Knaap, and Jonkman, Phys. Letters **2**, 5 (1962).

[8] L. L. Gorelik, ZhTF **33**, No. 12, (1963), Soviet Phys. Tech. Phys. **8**, No. 12 (1964).

Translated by A. Tybulewicz
58

EXCITATION AND DISSOCIATION OF MOLECULES IN AN INTENSE LIGHT FIELD

G. A. ASKAR'YAN

P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor November 16, 1963

J. Exptl. Theoret. Phys. (U.S.S.R.) **46**, 403-405 (January, 1964)

THE purpose of the present note is to indicate the possibility of and conditions for the efficient direct dissociation of molecules in a very intense light field and to estimate the magnitude of the effect by means of simple quasiclassical calculations, since the classical energy transfer (cf., for example,[1]) determines the excitation probability of a quantum oscillator. This direct dissociation process is related to the resonance excitation of the vibrational oscillations of atoms in a molecule or to the tunnel breakup of a molecule on the appearance of pulsating interaction forces between atoms polarized by the wave field (this mechanism applies in the case of homopolar molecules) or on the direct action of the field on atoms considered as ionic fractions of molecules (in the case of heteropolar molecules or molecular ions).

First, we shall consider homopolar molecules. The short-range interaction force between atoms, polarized by a light-wave field $E = E_0 \sin \omega t$ and located in a molecule at distances r comparable with atomic dimensions a, is equal to

$$f(t) \approx q_{\rm eff}^2(t)\, r^{-2} \approx \alpha^2 E_0^2 a^{-4} \sin^2 \omega t \approx \tfrac{1}{2} a^2 E_0^2 (1 - \cos 2\omega t),$$

where α is the polarizability of the atoms; $\alpha \approx q_{\rm eff} a/E \approx e^2/m_e\omega_{0e}^2 \approx a^3$ for frequencies ω which are small compared with the electronic frequencies ω_{0e}. The appearance of this pulsating force may excite the vibrational oscillations of the molecule. The buildup of these oscillations is strongest when the pulsation frequency of the force is $\Omega_f = 2\omega \rightarrow \Omega_a$, where Ω_a is the natural vibrational frequency of the atoms in the molecule (Ω_a lies in the infrared region). In this case, the amplitude of the oscillations rises linearly with time:

$$x_0(t) = f_0 t/2M_a\Omega_a = (a^2E_0^2/4M_a\Omega_a)\, t.$$

When the oscillation amplitude is close to critical ($x_c \gtrless a$), the molecule will break up. For this, we require a time $\tau \approx M_a\Omega_a/aE_0^2$. For example, when the atomic mass is $M_a \approx 10^{-24}$ g, we find that $\tau \approx 10^{-10}$ sec for the vibrational frequency $\Omega_a < 10^{15}$ sec^{-1} and $E_0 \approx 10^7$ V/cm, i.e., the breakup of the molecule will occur very quickly when the frequencies become equal at resonance: $\Omega_f \rightarrow \Omega_a$.

The drift away from resonance may be quite important unless special measures are taken: the resonance frequencies are selected to cover several levels, the frequency is varied with time, the buildup time is short, etc.

If the conditions are not exactly those of resonance, beats are observed with the frequency $\Delta\Omega \sim \Omega - \Omega_a$ and the probability of excitation increases strongly at the instant of maximum buildup. Close to resonance, the amplitude

$$x_0(t) \sim \frac{f_0}{M_a(\Omega_f^2 - \Omega_a^2)} \sin\ (\Omega_f - \Omega_a)\, t$$

does not exceed the value

$$x_{0max} = a^2 E_0^2 / M_a (\Omega_f^2 - \Omega_a^2)$$

Far from resonance, the forced vibration amplitude is given by the same expression.

In strong fields or in the case of strong excitation, the interaction forces between polarized atoms may cause sub-barrier breakup of the molecule by the tunnel effect, with an exponential dependence of the breakup probability on the polarization force.

It is interesting to note the difference between the nonresonance and resonance interactions as a function of the mutual positions of the interacting atoms and the direction of the electric vector of the light wave. If the interacting atoms lie in a plane perpendicular to the direction of the field, the polarized atoms always repel one another, which favors the breakup of the molecule. However, if the atoms lie along the direction of the field, then, in the nonresonance case, the polarized atoms attract one another while close to resonance the buildup of oscillations again occurs and there is a tendency for the molecule to break up.

Because of the quadratic dependence of the force on the field intensity, the average force is not equal to zero. This allows us to use light fields of high frequencies $\omega \gg \Omega_a$, modulating their amplitude or rotating the electric field vector at frequencies which build up the resonance oscillations (the modulation frequency of the field amplitude $\Omega_{mod} \sim \Omega_a/2$). Such modulated or rotating light fields can be obtained by superimposing waves with close but different frequencies or polarizations, or with a phase shift which ensures rotation. For the oscillations with $\omega \gg \Omega_{mod}$, $\dot{\Omega}_{mod}/\Omega = \dot{\omega}/\Omega \gg \dot{\omega}/\omega$; therefore, rapid variation of the frequency Ω_{mod} with time can be obtained, thus ensuring synchronization of the nonlinear oscillations and the force.

We shall consider now the case of heteropolar molecules or molecular ions. Here, the force acting on the atoms, $f(t) \approx eE_0 \sin \omega t$, depends linearly on the field intensity, and therefore the resonance condition is $\omega_a \rightarrow \Omega_a$ 1) and the resonant rise of the amplitude is $x_0(t) = (eE_0/2M_a\Omega_a)t$, which gives the resonance buildup time

$$\tau \sim aM_a\Omega_a / eE_0 \approx 3 \cdot 10^{-13} \text{ sec},$$

i.e., at the same values of the various quantities involved, the dissociation of heteropolar and ionic systems should be more intense. The strongest dissociation effect appears when the atoms lie along the direction of the electric field. For the dissociation of homopolar molecular ions we can use light fields of higher frequencies, modulated or rotating at a frequency close to the vibration resonance.

It follows from the above treatment that it is possible to use an intense light field with resonance carrier or modulation frequencies for effective excitation and dissociation.

The effects considered may be used, for example, to intensify the effect of light on molecular media or on micro-objects, to dissociate beams of fast atoms or ions entering a trap,[2] to obtain an atomic beam from a molecular beam, to heat directly the molecules of a medium, etc.

---

1) G. S. Voronov drew the author's attention to the fact that exchange electron transitions in a homopolar molecular ion may alter the resonance conditions.

[1] V. I. Kogan and V. M. Galitskiĭ, Sbornik zadach po kvantovoĭ mekhanike (Collection of Problems on Quantum Mechanics), GITTL, 1956. Ch. V and VI.

[2] Askar'yan, Delone, and Rabinovich, JETP 46, 814 (1964).

Translated by A. Tybulewicz
59