PROPAGATION OF BIHARMONIC LASER RADIATION RESONANT TO SPIN FREQUENCIES THROUGH ATMOSPHERIC AND INERT GASES. PART 2

A.V. En'shin

Tomsk State University
Received March 5, 1994

*The extent to which the nitrogen and argon participate in the process of the resonance biharmonic scattering is investigated. The results are compared to those for the case of oxygen and air. It is shown that nitrogen is most active in the process under study. The mechanism of creating polaritons in the atmosphere is considered. It is shown to involve the cascade of three–photon processes of the second order, the development of which is conditioned by breaking down of the inversion symmetry of the medium. Such a sharp change of the macroscopic characteristics of the medium is interpreted as a result of appearance of spin waves in the medium. This macroscopic quantum effect manifests itself when the nonequilibrium population of spin states of particles is generated in the medium. This conclusion is supported by the measurements in air under pressures from*760 *down to* 10^{-4} mm Hg.

This paper continues discussion of experimental results on atmospheric scattering of laser biharmonics resonant to spin frequencies.[1] The data obtained with nitrogen and argon are presented. Since nitrogen molecule has nuclear spin $I = 2$ and rotational Raman spectrum whereas argon is inert, the comparison of the data presented in this paper with those of Ref. 1 enables us to estimate the cross section of the scattering process under study and its physical grounds.

EXPERIMENT

Experiments have been carried out using special test bench described in detail in Ref. 2. Working chamber was filled with air, nitrogen, or argon. Signals from Stokes spectral region[2] separated from the basic wavelength of incident radiation by about 500 Ð were monitored. Spectral width of the recorded interval was 10 Ð. The detailed description of the experimental procedure is presented in Refs.1 and 2.

RESULTS

1. Typically signals obtained from the reference and test channels of the setup, when the chamber was filled with air, were 460 and 500 mV, respectively, gas pressure being 760 mm Hg at a temperature of 300 K. Then the chamber was pumped out and filled with nitrogen with the purity not worse than 99%. After the chamber was filled with nitrogen up to 760 mm Hg the swing of the M–1400 microammeter pointer of two divisions to the right from zero was observed. This indicated that the input signal from the test channel increased by 22 mV.

2. In experiments with argon the initial values of signals from reference and test channels of the setup measured with the chamber filled with air were 320 and 640 mV, respectively. After the chamber was filled with argon with the purity not less than 99.9% the swing of microammeter pointer by two divisions to the left from zero was observed. Hence, the input signal from the test channel decreased by 30 mV.

3. The previous experiments with oxygen[1] demonstrated relative decrease of the input signal by 15 mV. When the air pressure was decreased from 760 down to 10^{-4} mm Hg the signal was found to be constant.

DISCUSSION

1. The results presented show that nitrogen is the most active gas in the scattering process studied. Oxygen is more active than argon (the signal drop of 15 and 30 mV, respectively, was observed) but its relative contribution to the signal is lower than in the case of nitrogen that demonstrated increase of the input signal by 22 mV. In order to understand this result, consider molecular structure of nitrogen and possible mechanism of interaction between gas molecules and biharmonic resonant to spin frequencies. Since the spectrum observed[1] exhibits clearly pronounced polariton character,[3] let us consider polariton formation in the medium.

It is known that in conventional active Raman scattering (ARS) the signal is generated via intermediate states. However, in the media without center of symmetry ARS by polaritons is also possible.[3] Natural frequencies of those composite quasiparticles depend on the magnitude and direction of wave vector.[3] The intensity of ARS signal from polaritons depends not only on a biharmonic frequency difference $\omega_1 - \omega_2$ but also on the wave vector difference $\mathbf{K}_1 - \mathbf{K}_2$. Hence, in the media without a center of symmetry both direct four–photon processes caused by cubic nonlinear susceptibility $\chi^{(3)}$ and cascade three–photon processes involving polariton states and related to quadratic nonlinearity $\chi^{(2)}$ contribute to ARS signal. Three–photon processes involved in ARS by polaritons are to be considered as two successive three–photon processes schematically shown in Fig. 1*a*: 1) excitation of polariton at a frequency $\omega_p = \omega_1 - \omega_2$ in the field of two laser beams with frequencies ω_1 and ω_2 and 2) scattering of probing wave with the frequency ω_1 by the polaritons thus excited.

0235-6880/95/05 363-03 $02.00

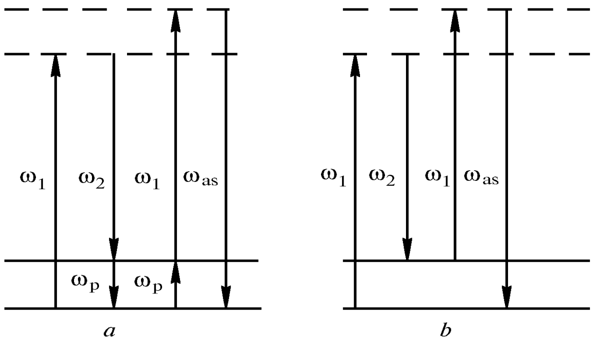

FIG. 1. Diagram illustrating cascade three–photon (a) and direct four–photon (b) processes involved in ARS by polaritons.

Both these processes are caused by quadratic nonlinearity of the medium $\chi^{(2)}$ and the highest efficiency of ARS is reached under conditions of synchronism[3]:

$$\mathbf{K}_p - (\mathbf{K}_1 - \mathbf{K}_2) \equiv D\,\mathbf{K}_p = 0\ ;$$
$$\mathbf{K}_p - (\mathbf{K}_{as} - \mathbf{K}_1) \equiv D\,\mathbf{K} = 0\ , \qquad (1)$$

where $\mathbf{K}_1$, $\mathbf{K}_2$, $\mathbf{K}_p$, $\mathbf{K}_{as}$ are corresponding wave vectors.

Direct four–photon process occurs when the fields of three lasers mix: $\omega_{as} = 2\omega_1 - \omega_2$ (see Fig. 1*b*). This process is most efficient under the following condition:

$$\mathbf{K}_{as} - (2\,\mathbf{K}_1 - \mathbf{K}_2) \equiv \Delta\,\mathbf{K}_{as} = 0. \qquad (2)$$

Since the degree of nonlinearity decreases with increasing order of process,[4] the third–order processes are much weaker than the allowed second–order processes. Indeed, we have:

$$\chi^{(3)} / \chi^{(2)} \sim 1/E_{at}\ , \qquad (3)$$

where E_{at} is intratomic field whose typical strength for gases is $\sim 3 \cdot 10^8$ V/cm.

Thus, the second–order nonlinear processes in the atmosphere discovered in our experiments are characterized by magnitude of susceptibility $\chi^{(2)}$ being 10^8 times higher than for the early known third–order processes. It is evident (see Ref. 4) that second–order processes in the media with center of symmetry (gases) are forbidden by symmetry rule. When constant electric or magnetic field is applied to a medium the inversion symmetry is broken. Since electric and magnetic fields used in our experiments being about 100 V/cm were very low as compared to the intratomic field of gas molecules, their direct influence could not change the symmetry of the medium. The break of inversion symmetry observed in the experiments can be explained in the following way. Biharmonics with frequency difference of 500 MHz can resonantly influence elementary excitations of the medium caused by the motion of nuclear and electron magnetic moments in nitrogen and oxygen molecules. That results, as was noted in Ref. 1, in nonequilibrium population of spin states of nitrogen molecules or nuclear and electron polarization with respect to spin direction.

In the space exposed to biharmonic radiation the motion of all nuclear (electron) spins of molecules proves to be correlated. As shown in Ref. 6, the macroscopic quantum effects related to spin polarizations of gas particles, such as spin waves, are observed. These effects lead to dramatic changes in macroscopic properties of the system. Spin waves appear as a result of collective interaction between the particles causing the occurrence of a peculiar self–consistent field that influences each gas particle.

The existence of collective spin waves in a polarized gas mean that additional Bose excitation branch related to spin is present in the system. It is essential that such a field is formed on a path much shorter than the free path.

Following the theory presented in Ref. 5, in this case there exists the direct exchange mechanism of inelastic scattering of particles by thermal fluctuations of transverse magnetization. It results in spin flip of the particle scattered and emission or absorption of a collective spin wave referred to as magnon. The cross section of such a process is on the same order as that of conventional gas kinetic elastic scattering of particles by each other but under certain conditions it can be significantly higher. The latter fact is in a good agreement with our results.

Thus, the thermal molecular motion is superimposed by regular flow caused with a momentum exchange between photons and particles of the medium. The momentum transfer is usually characterized by internal friction coefficient η. According to the elementary kinetic theory η is expressed as

$$\eta = \frac{1}{3}\,\bar{u}\,\rho\,\bar{\lambda}, \qquad (4)$$

where $\bar{u}$ is the mean velocity of thermal motion, ρ is the density of the particles, $\bar{\lambda}$ is the mean free path.

Equation (4) includes product $\rho\,\bar{l}$ which is independent of gas pressure. Hence, signal values recorded in this case may not depend on gas pressure too, because they are proportional to the coefficient η. The experimental data obtained with air within the range of pressure between 760 and 10^{-4} mm Hg are also in agreement with this conclusion of the kinetic theory.

CONCLUSION

From analysis performed in this paper one can conclude that results of the experiments agree with the idea on Raman scattering of light by magnon polaritons formed due to mixing of nuclear spin oscillations with the polarization oscillations of electron shell influenced by resonant biharmonic.

The scattering process discovered is governed by the second–order susceptibility and is caused by collective interaction between particles, namely, spin waves of quantum origin. In our opinion, very high cross section of the process and its low threshold make it promising for remote sensing of the atmosphere using ARS techniques.

Certainly, only further experimental and theoretical study will enable the description of polariton formation dynamics and specify conditions for break of inversion symmetry of the medium as well as determine all macroscopic quantum effects accompanying this process.

REFERENCES

1. A.V. En'shin, Atmos. Oceanic Opt. **8**, No. 4, 532–539 (1985).
2. A.V. En'shin, Atmos. Oceanic Opt. **7**, No. 10, 767–769 (1994).

3. Yu.I. Polivanov and A.T. Sukhodol'skii, Pis'ma Zh. Eksp. Teor. Fiz. **25**, No. 5, 240–244 (1977).
4. I.R.Shen, *Principles of Nonlinear Optics* [Russian translation] (Nauka, Moscow, 1989), 560 pp.
5. E.P. Bashkin, Pis'ma Zh. Eksp. Teor. Fiz. **49**, No. 6, 320–322 (1989).
6. E.P. Bashkin, Usp. Fiz. Nauk **148**, No. 3, 433–471 (1986).