Resonantly Enhanced Degenerate Four-Wave Mixing of Millimeter-Wave Radiation in Gas

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Millimeter-wave phase conjugation was realized under resonantly enhanced degenerate four-wave mixing in gaseous carbonyl sulfide. The nonlinear wave interaction was caused by saturation of a transition between rotational levels. For radiation with $\lambda = 1.64$ mm, phase-conjugate reflectivity of about 0.4% has been observed at a gas temperature of 200 K, and under optimal pressure.

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Electromagnetic millimeter-wave radiation is becoming widely used in radar applications, communication, navigation, remote sensing, etc. [1]. However, atmospheric refraction and surface scattering greatly hinder propagation of millimeter waves and result in considerable amplitude and phase fluctuations of signals. To eliminate their negative influence, adaptive [2] and nonlinear [3] phase-conjugating devices can be used, since they perform phase conjugation (PC) of the signal wave on a real time scale. A phase-conjugate signal restores the initial wave front automatically when it propagates backwards through an optically inhomogeneous medium [2–4]. In the dm- and cm-wave regions phase-conjugating antennas are used to correct wave fronts on a real time scale; usually they are made as arrays of nonlinear elements [5]. Use of array antennas for PC in the mm-wave region, however, is prevented because the characteristic scale of wave-front inhomogeneities becomes too small. Therefore, a good prospect is the possibility to perform PC of millimeter radiation by degenerate four-wave mixing (DFWM) in a continuous nonlinear medium. DFWM implies that two quasiplane counterpropagating pump waves (forward pump wave f and backward pump wave b) and a third, signal wave s, of identical frequencies interact within a nonlinear medium and generate the wave c phase conjugate to the signal wave. This method of phase correction is widely used in optics [3,4]. Besides compensation of phase distortions in transparent inhomogeneous media, DFWM can be used in the amplification and generation of microwave radiation, control of spatiotemporal structure of wave fields, interferometry, nonlinear spectroscopy, etc. [3,4].

In spite of the practical importance of various devices operating on the basis of DFWM on the real time scale, four-wave mixing has not yet been experimentally realized in the millimeter-wave region. The most complicated problem was the search for or creation of a suitable nonlinear medium. Artificial Kerr media were proposed as nonlinear media for microwave DFWM [6,7]. Phase conjugation of centimeter waves in such nonlinear media was later demonstrated [8]. Unfortunately, artificial nonlinear media have a slow response time (≥ 1 s), and therefore generally they cannot be used for PC of centimeter or millimeter waves on a real time scale. Of high prospect for PC of the waves in the long millimeter and centimeter wavelength regions on the real time scale is plasma. Theoretical investigations of DFWM in completely [9] and partially [10] ionized plasma showed, however, that an efficient PC of radiation in the short millimeter region ($\lambda < 3$ mm) requires the use of plasma with a high density ($\geq 10^{13}$ cm^{-3}) and a low electron temperature (~ 1 eV). Generation of a completely or partially ionized homogeneous plasma with such parameters in large volume ($\sim 10^3$ cm^3) is a rather complicated problem. These considerations make it necessary to search for other types of nonlinear media for DFWM of millimeter waves.

The present paper gives an experimental demonstration of the possibility of DFWM of millimeter-wave radiation in a gaseous medium, whose mechanism of nonlinearity is caused by power saturation of the rotational transition of dipole molecules. The advantages of such a nonlinear medium are that it is becoming easy to produce, convenient to use, and has a fast response time. These nonlinear media are most efficient in the region of short millimeter and submillimeter waves, where resonance absorption by rotational transitions of molecules is high [11]. Of the gases with strong absorption lines in the millimeter-wave region, one of the most convenient gases for experimental investigation of DFWM is now carbonyl sulfide (OCS). The linear molecule of OCS has relatively low chemical activity and a simple, well-studied rotational spectrum [11]. When the gas pressure is $P > 1$ Torr, rotational lines of OCS are homogeneously broadened. The profile of the spectral line in this case is described by the Lorentz function g, whose width, when the gas is affected by a sufficiently strong and near-resonance electromagnetic field, is determined by both collisions of molecules and power saturation [11,12]:

$$g(\nu - \nu_0, I/I_{\text{sat}}) = (T_2/\pi)[4\pi^2 T_2^2(\nu - \nu_0)^2 + (1 + I/I_{\text{sat}})]^{-1}, \quad (1)$$

where ν is the radiation frequency, ν_0 is the resonance frequency of the rotational transition, I is the intensity of the electromagnetic radiation, and $I_{\rm sat}$ is the saturation intensity of the rotational transition, equal to [12]

$$I_{\rm sat}=\frac{3\epsilon_0 c}{4}\frac{\hbar^2(2J''+1)(2J''+3)}{T_1T_2\mu^2(J''+1)^2}\,. \qquad (2)$$

Here T_2 and T_1 are transverse and longitudinal relaxation times, respectively (T_1 and T_2 are in direct proportion to temperature of the gas T and in inverse proportion to pressure P) [11,13], J'' is the rotational quantum number in the lower state, μ is the dipole momentum of the OCS molecule, and ϵ_0 is the dielectric susceptibility of vacuum. In the range from 150 to 200 GHz the weak-field absorption coefficient at the center of the rotational lines of OCS α_0 at gas temperature $T\approx 200$ K ranges from 0.02 to 0.05 cm^{-1} [11]; this permits one to realize experimentally the situation when the optical thickness of the gas at resonance frequencies of rotational transitions is sufficiently high: $\alpha_0 L\geq 1$.

Resonant DFWM in saturable-absorbing media under the condition of homogeneous line broadening has been theoretically investigated [14–17]. It has been shown that when $\alpha_0 L\sim 1$ the maximal phase-conjugate reflectivity is $R\sim 1\%$, and the maximum reflection is achieved when input intensities of the pump waves I_0 are approximately equal to $I_{\rm sat}$, and when radiation frequency is tuned to the center of the rotational line $\nu-\nu_0<\Delta\nu_L=(2\pi T_2)^{-1}$.

In order to observe and investigate DFWM of millimeter waves in OCS an experimental setup was made, whose scheme is given in Fig. 1. A gyrotron with a pulsed magnetic field [18] produced by a water-cooled solenoid was used as a source of powerful millimeter-wave radiation ($\lambda<2$ mm). The gyrotron pulse duration was 40 μs with a pulse repetition rate of 0.1 Hz. The radiation power at

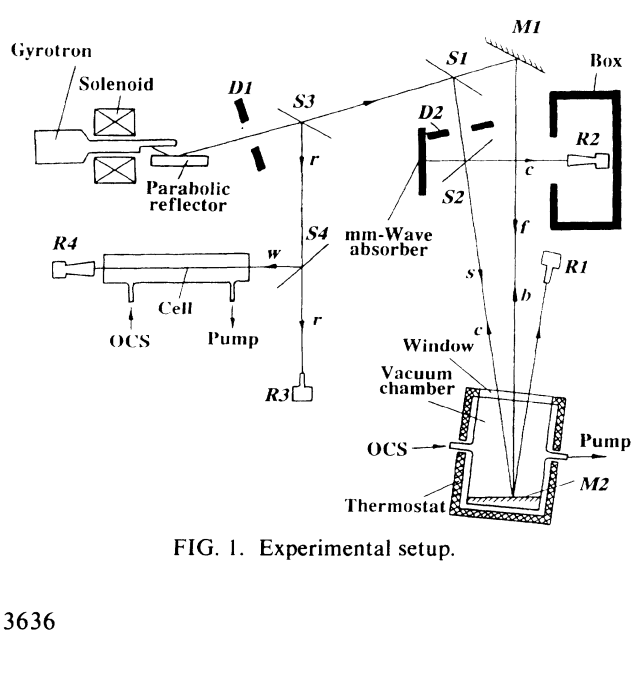

FIG. 1. Experimental setup.

the frequency corresponding to the center of the chosen rotational line was 30 kW. Gyrotron radiation was converted into a quasi Gaussian beam with a parabolic reflector and a diaphragm $D1$ made of a millimeter-wave absorber.

The metallic mirror $M1$ was used to direct the pump wave into a vacuum chamber (with an internal diameter of 18 cm and length $L_{\rm ch}=30$ cm) through a plane-parallel quartz window. The polished surface of the chamber back wall served as a plane mirror $M2$ ($R=100\%$) set perpendicularly to the incident pump beam and was used to form the backward pump beam. The chamber was filled with gaseous OCS, whose pressure within the chamber was measured with mechanical and oil pressure gauges. The chamber was placed in a thermostat and cooled with dry ice to a temperature of ≈ 200 K to increase resonance absorption. Signal beam s was directed into the chamber at a 6° angle to the pump beam f. The signal beam was formed by means of beam splitter $S1$ (a plane-parallel quartz plate with reflection coefficient 50%), and diaphragm $D2$, 60 mm in diameter. Within the chamber, at the boundary of the gaseous medium, the ratio of maximum intensities of the pump and signal beams was approximately 1.5, and their widths (full width at half maximum) were 7.5 and 6.5 cm, respectively. The optical axes of the signal and pump beams were crossed at the surface of mirror $M2$; hence the length of the wave interaction volume was approximately equal to the doubled length of the vacuum chamber: $L\approx 2L_{\rm ch}=60$ cm. After double pass through the chamber, the signal wave reached receiving system $R1$. The reflected wave resulting from DFWM in the gas was directed to receiving system $R2$ by the plane-parallel quartz plate $S2$ with a reflection coefficient of 15%; system $R2$ was placed in a box made of a millimeter-wave absorber. Beam r was branched from the main beam with Mylar-film beam splitter $S3$ (reflection coefficient 7%). It was directed to receiving system $R3$ and used to control the regime of gyrotron operation and radiation power in each pulse. Beam r was split with Mylar-film beam splitter $S4$ (reflection coefficient 7%) and the obtained beam w passed through a cylindrical quartz cell filled with OCS at a temperature of 300 K, and then reached receiving system $R4$. The internal diameter of this cell was 8 cm and the length $L_c=40$ cm. Since power saturation of the rotational transition in OCS does not occur when the weak beam w propagates through the gas in the cell, this beam was used to measure the radiation frequency detuning from the center of the rotational line. As receiving systems $R1$, $R2$, $R3$, $R4$, we used mm-wave packaged diodes at whose input there was set either a section of a rectangular waveguide for the two-millimeter region ($R1$,$R3$) or a horn rectangular antenna ($R2$,$R4$), depending on the intensity of the received wave.

The wavelength of the gyrotron radiation measured with the Michelson interferometer proved to be 1.65 ± 0.04 mm. The rotational transition $J'=15 \leftarrow J''=14$, whose resonance frequency is $\nu_0=182.43$ GHz [11], is the closest according to its frequency. For this rotational transition a line-center saturation intensity is easy to evaluate using Eq. (2); it is $I_{sat} \approx 2(PT_0/P_0T)^2$, where I_{sat} is measured in W/cm^2, P in Torr, and T in K, with $P_0=1$ Torr and $T_0=300$ K. Under the experimental conditions, the pump wave intensity in the center of the beam did not exceed 300 W/cm^2, and, therefore, noticeable saturation ($I/I_{sat} \geq 1$) of the rotational transition at $T=200$ K is possible only at $P \leq 10$ Torr. Under this condition, the collision half-width of the rotational line $\Delta\nu_L$ [$\Delta\nu_L(\mathrm{MHz}) \approx 6PT_0/P_0T$) [11]] does not exceed 100 MHz; hence for the condition $\nu - \nu_0 \ll \Delta\nu_L$ to be valid gyrotron radiation frequency tuning to the center of the spectral line with precision ≤ 30 MHz is necessary. Such tuning was performed by selecting an appropriate gyrotron cavity from a set of cavities with slightly differing diameters, and by smooth frequency tuning by changing the value of magnetic field produced by the solenoid. The detuning $\delta\nu = \nu - \nu_0$ was determined by the dependence of the gas transmissivity in the cell $T_w = I_w/I_{w0}$ for a weak beam w on the gas pressure. Its intensity at the cell input was $I_{w0} \approx 1$ W/cm^2; therefore, a line broadening caused by power saturation of the rotational transition by this beam at $P>1$ Torr can be neglected. In this case, according to Eq. (1) and the Bouguer-Lambert-Beer law, the dependence T_w on the gas pressure has the following form:

$$T_w = \frac{I_w(P)}{I_{w0}} = \exp\left\{ -\alpha_0 L_c \left[1 + \left(\frac{\delta\nu}{\Delta\nu_1 P/P_0} \right)^2 \right]^{-1} \right\}, \tag{3}$$

where $\Delta\nu_1(\mathrm{MHz}) \approx 6T/T_0$. From Eq. (3) it is easy to find that the function $Y = \{-[1+(\alpha_0 L_c/\ln T_w)]\}^{-1/2}$ depends linearly on the gas pressure: $Y(P) = \Delta\nu_1 P/\delta\nu P_0$. Therefore, using the dependence $T_w(P)$ given in Fig. 2, one can determine the tangent of the angle between the straight line $Y(P)$ and the horizontal axis P, and, hence, find that detuning $\delta\nu$ in our experiments is about 15 MHz.

To verify that saturation influences propagation of intensive radiation with the frequency near to the OCS rotational transition frequency, we measured the pressure dependence of the signal beam transmissivity, $T_s = I_s(P)/I_{s0}$, through gas filling the vacuum chamber. The intensity of the signal wave at the entrance to the gas medium, I_{s0}, was 2 orders of magnitude larger than that of the beam w. The pump beam in this series of experiments was blocked by an absorbing screen behind mirror $M1$. The experimental results for two values of the gas temperature (200 and 300 K) are given in Fig. 2. It is difficult to determine precisely the value of I_{sat} by the experimental dependences, because of the low accuracy of

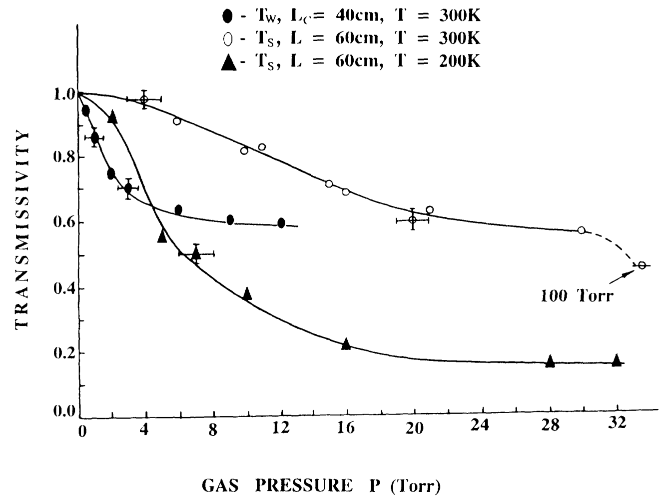

FIG. 2. Dependence of transmissivity of radiation, with the frequency close to the resonance frequency of the rotational transition $15 \leftarrow 14$, on the gas pressure. T_w, transmissivity of the weak beam w through gas in the quartz cell; T_s, transmissivity of the signal beam s through gas in the vacuum chamber; $I_{w0}/I_{s0} \approx 10^{-2}$.

measurements of absolute values of I_{s0}, as well as the complex structure of the field within the chamber, caused by interference of two counterpropagating and partially overlapped beams. However, these results give us the opportunity to evaluate the gas pressure P_{sat}, under which the saturation parameter $I_{s0}/I_{sat}(P)$ equals 1. At 200 K, P_{sat} is about 10 Torr.

Measurements of transmissivity of the signal beam through the vacuum chamber at $P \gg P_{sat}$ permit us to determine the double-path optical thickness of the gas at the center of the unsaturated rotational transition $\alpha_0 L$, which was equal to 1.9 ± 0.1 at $T=200$ K and to 0.85 ± 0.05 at $T=300$ K.

If the signal and pump beams were directed into the vacuum chamber simultaneously, then at pressures lower than 30 Torr ($T=200$ K) the movable receiving system $R2$ detected a reflected wave formed by DFWM in the gas. The optical path from the chamber window to receiving system $R2$ was ≈ 1.5 m. With the aid of this moving receiver we found that the reflected beam propagating towards the signal beam had a horizontal width of less than 5 cm, and their maximums coincided with an experimental accuracy of ≈ 2 cm. The fact that the reflected wave was produced by DFWM in gas was confirmed, besides by the opposite direction of propagation, by the disappearance of this wave when either of the two beam incident at the gas chamber was blocked by either the signal beam or the pump beam. To measure the absolute value of the phase-conjugate reflectivity, the receiving system $R2$ was calibrated with the aid of a Mylar-film mirror with a thickness of 10 μs. During calibration this mirror was set in place of the chamber per-

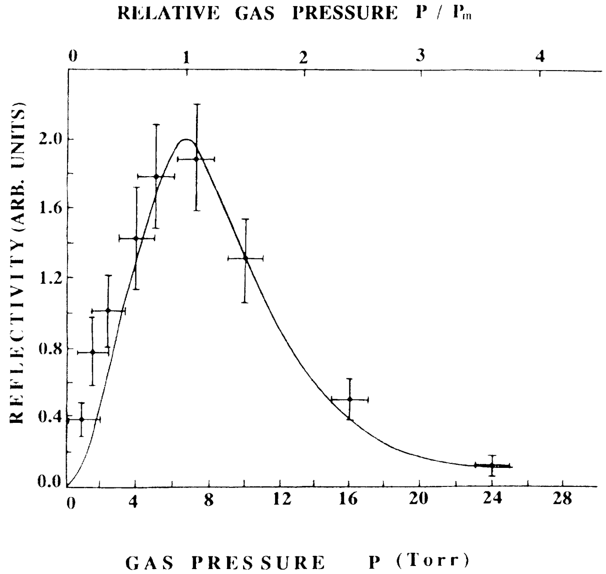

FIG. 3. Dependence of phase-conjugate reflectivity on the gas pressure. The solid line shows the results of calculations of reflectivity $0.2R_c$ vs relative pressure P/P_m.

pendicularly to the axis of the signal beam, and its reflection coefficient was 0.2%.

The dependence of the phase-conjugate reflectivity R_c on gas pressure at $T=200$ K ($\alpha_0 L=1.9$) was experimentally investigated. The results of these measurements are presented in Fig. 3. The reflection was maximal, with the pressure close to P_{sat}, under which the saturation parameter is optimal, and approximately equals 1. Figure 3 also shows the theoretical dependence of $0.2R_c$ on the relative pressure P/P_m; this dependence was derived from calculations given in [15] of the reflectivity via DFWM in a two-level saturable absorber on the saturation parameter $I_0/I_{\text{sat}}=(P_m/P)^2$, for the case of $\alpha_0 L=2$. The coefficient $P_m \approx 7$ Torr was chosen such that the maximum R positions at theoretical and experimental graphs would coincide. Maximum R_c by DFWM in gas was about 0.4%. In accord with calculations in [15], when $\alpha_0 L=2$, maximal reflectivity $\approx 2\%$. The difference between experimental and theoretical values of the maximal reflectivity is caused, apparently, by the fact that real pump waves had limited widths, and were not plane waves as was assumed in [14–16].

In the millimeter-wave region the majority of applications of PC devices require $R_c \sim 100\%$. For short millimeter region such a growth in R_c in saturable-absorbing gases may be provided by an increase in $\alpha_0 L$ [3]. It is possible to significantly increase both L and α_0, first realizing the DFWM in oversized waveguides [3], and second using gases whose linear molecules have a large dipole momentum (BrCN, ClCN, etc.) [11]. However, even at $R_c \lesssim 1\%$, realized in our experiments, the resonantly enhanced DFWM seems to be an effective tool for nonlinear molecular rotational spectroscopy as well as for diagnostic use for gases and flames. Note that the response time of saturable-absorbing gaseous media is determined by the relaxation time of the population difference of rotational levels T_1 of OCS molecules. In our case this time is less than 10^{-8} s. As a result, such nonlinear media can be used as PC mirrors in resonators of powerful short-pulse millimeter-wave generators, e.g., for free electron lasers.

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[1] *Infrared and Millimeter Waves*, edited by K. J. Button and J. C. Wiltse (Academic, New York, 1981), Vol. 4; *ibid.*, edited by K. J. Button (Academic, New York, 1983), Vol. 9.
[2] D. L. Fried, J. Opt. Soc. Am. **67**, 422 (1991).
[3] *Optical Phase Conjugation*, edited by R. A. Fischer (Academic, New York, 1983).
[4] B. Ya. Zel'dovich, N. F. Pilipetsky, and V. V. Shkunov, *Principles of Phase Conjugation* (Springer, Heidelberg, 1985).
[5] Special Issue on Active and Adaptive Antennas. IEEE Trans. Antennas Propag. **12**, 140 (1964).
[6] B. Bobbs *et al.*, Appl. Phys. Lett. **52**, 4 (1988).
[7] D. K. Mansfield and J. F. Federici, Int. J. Infrared Millimeter Waves **9**, 419 (1988).
[8] R. Shin *et al.*, Phys. Rev. Lett. **65**, 579 (1990).
[9] I. Nebenzahl, A. Ron, and N. Rostocker, Phys. Rev. Lett. **60**, 1030 (1988); J. F. Federici and D. K. Mansfield, J. Opt. Soc. Am. B **2**, 1588 (1986); E. A. Williams, D. M. Lininger, and M. V. Goldman, Phys. Fluids B **1**, 1561 (1989).
[10] N. A. Bogatov, M. S. Gitlin, and S. V. Golubev, in *Proceedings of the Twentieth International Conference on Phenomena in Ionized Gases, Il Giorgio, Italy, 1991*, edited by V. Palleschi and M. Vaselli (Istituto di Fisica Atomica e Molecule, Pisa, 1991), p. 1174.
[11] W. Gordy and R. L. Cook, *Microwave Molecular Spectra* (Wiley, New York, 1984).
[12] R. H. Pantell and H. E. Puthoff, *Fundamentals of Quantum Electronics* (Wiley, New York, 1969).
[13] H. Mäder, Z. Naturforsch. **34a**, 1170 (1979).
[14] W. P. Brown, J. Opt. Soc. Am. **73**, 629 (1983).
[15] M. T. Gruneisen, A. L. Gaeta, and R. W. Boyd, J. Opt. Soc. Am. B **2**, 1117 (1985).
[16] A. A. Betin *et al.*, Kvantovaya Electron. (Moscow) **13**, 1975 (1986) [Sov. J. Quantum Electron. **16**, 1304 (1986)].
[17] R. L. Abrams and R. C. Lind, Opt. Lett. **2**, 94 (1978).
[18] V. A. Flyagin, A. G. Luchinin, and G. S. Nusinovich, Int. J. Infrared Millimeter Waves **4**, 629 (1983).