# Mixing of the frequencies 88.37 THz ($\lambda = 3.39$ $\mu$), 125.13 THz ($\lambda = 2.39$ $\mu$), and 260.1 THz ($\lambda = 1.15$ $\mu$) in a gas, and production of continuous coherent emission with combined frequency 473.6 THz ($\lambda = 0.63$ $\mu$)

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We report, for the first time, the mixing of three infrared frequencies corresponding to wavelengths 3.39, 2.39, and 1.15 $\mu$, and production of coherent continuous emission at the combined frequency of wavelength 0.63 $\mu$ in a gas. Effective continuous frequency conversion was realized via nonlinear interaction of three fields that are resonant with adjacent transitions in neon. Owing to the resonance conditions, the addition of the frequencies in the gas turned out to be more effective by 8–10 orders of magnitude than in nonlinear optical crystals.

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1. Until recently, continuous coherent conversion (addition and substraction) of infrared and optical frequencies was effected with the aid of nonlinear optical crystals and diodes based on metal-metal point contacts. The production of harmonics and addition of frequencies in metal vapors under pulsed conditions was reported in[1] and[2] respectively. We have carried out the first successful experiments on the addition of the frequencies 88.37 THz ($\lambda = 3.39$ $\mu$), 125.13 THz ($\lambda = 2.39$ $\mu$), and 260.1 THz ($\lambda = 1.15$ $\mu$) and obtained continuous coherent emission at the summary frequency 473.6 THz ($\lambda = 0.63$ $\mu$) in a gas under

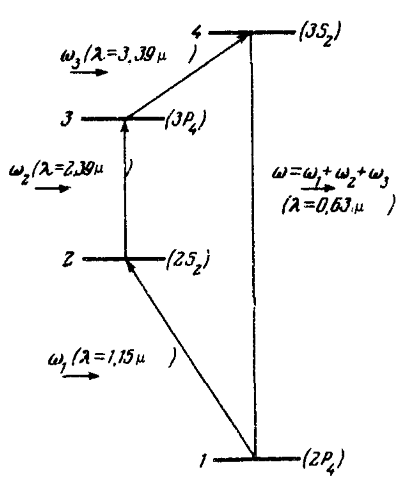


FIG. 1. Working-level scheme in neon.

resonance conditions. The wavelengths 3.39 μ, 2.39 μ, and 1.15 μ correspond to the coupled transitions $3s_2$—$3p_4$, $3p_4$—$2s_2$, and $2s_2$—$2p_4$ in neon (see[3]).

2. Effective continuous frequency conversion in a gas is realized by nonlinear interaction of three fields with amplitudes $E_1$, $E_2$, and $E_3$ at frequencies $\omega_1$, $\omega_2$, and $\omega_3$ resonant to the transitions $1\to 2$, $2\to 3$, and $3\to 4$, respectively (see Fig. 1). The appearance of radiation at the summary frequency $\omega_4=\omega_1+\omega_2+\omega_3$ is usually connected with the onset of polarization at this frequency.[4] It is more convenient to attribute this polarization under resonance condition in a gas to microscopic characteristics of the medium. We write down only those equations for the density matrix, which enable us to obtain directly the polarization $P_{41}$ of interest to us

$$\left(\frac{d}{dt}+i\omega_{21}+\Gamma_{21}\right)\rho_{21}=V_{21}n_1e^{-i\omega_1t+ik_1z}, \tag{1}$$

$$\left(\frac{d}{dt}+i\omega_{31}+\Gamma_{31}\right)\rho_{31}=V_{32}\rho_{21}e^{-i\omega_2t+ik_2z}, \tag{2}$$

$$\left(\frac{d}{dt}+i\omega_{41}+\Gamma_{41}\right)\rho_{41}=V_{43}\rho_{31}e^{-i\omega_3t+ik_3z}. \tag{3}$$

Here $V_{ik}=id_{ik}E_k/2\hbar$, $d_{ik}$ are the matrix elements of the $i\to k$ transitions, $\omega_{ik}$ are the frequencies of the $i\to k$ transitions, and $\Gamma_{ik}$ are the damping constants of the off-diagonal elements. We have assumed for simplicity that in the absence of fields the populations of all the levels except the first are equal to zero. The polarization per unit volume is equal to $P_{41}=d_{14}\rho_{41}$. From (1) we determine $\rho_{21}$. Substituting the obtained $\rho_{21}$ in the second equation, we obtain $\rho_{31}$. We then use (3) to determine $\rho_{41}$:

$$\rho_{41}=\tilde{\rho}_{41}e^{-i(\omega_1+\omega_2+\omega_3)t+i(k_1+k_2+k_3)z}$$

$$\tilde{\rho}_{41}=n_1\frac{V_{21}}{[-i(\omega_1-\omega_{21})+\Gamma_{21}]}\,\frac{V_{32}}{[-i(\omega_1+\omega_2-\omega_{31})+\Gamma_{31}]}$$

$$\times\frac{V_{43}}{[-i(\omega_1+\omega_2+\omega_3-\omega_{41})+\Gamma_{41}]}. \tag{4}$$

Polarization at the frequency $\omega$ is thus the result of the following processes: the field of transition $1\to 2$ and the population $n_1$ of level 1 produce polarization at the frequency $\omega_1$, the field at the frequency $\omega_2$ and the polarization at the frequency $\omega_1$ produce polarization at a frequency $\omega_1+\omega_2$, and finally, the polarization at the frequency $\omega_1+\omega_2$ and the field at the frequency $\omega_3$ produce the polarization at the frequency $\omega_4=\omega_1+\omega_2+\omega_3$. We can obtain analogously the polarization $P_{41}$ following initial excitation of other levels. The emission power at the frequency $\omega$ is proportional to the product $(V_{21}V_{32}V_{43})^2$ and depends on the deviations of the frequencies from resonance and on the statisfaction of the synchronism conditions. We note that allowance for the motion of the atoms changes the quantitative estimate of the process when $d_{ik}E_k/2\hbar\sim\Gamma_{ik}$.

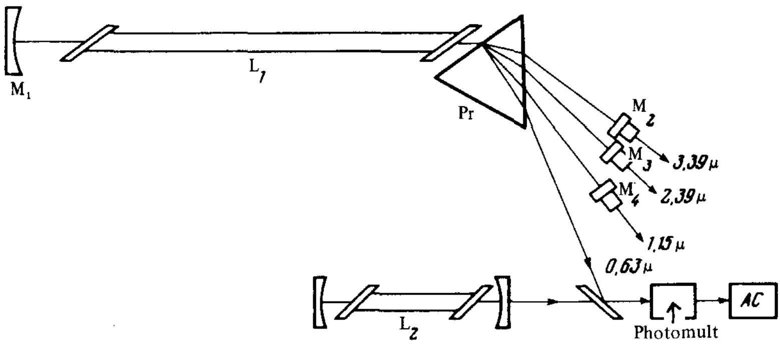


FIG. 2. Diagram of experimental setup.

3. To observe the effect of frequency mixing in a gas, it was simplest to use a cascade He–Ne laser at the three wavelengths 3.39, 2.29, and 1.15 $\mu$ [see (3)] corresponding to the cascade transition $3s_2 \rightarrow 3p_4 \rightarrow 2s_2 \rightarrow 2p_4$ in neon. The three fields produced as a result of the lasing interact with the excited neon atoms and induce polarization at the combined frequency, as a result of which coherent emission is produced at this frequency ($\lambda = 0.63$ $\mu$).

The cascade laser $L_1$ (Fig. 2) had a compound resonator consisting of one common aluminum-coated spherical mirror ($R = 20$ m), a prism ($P_r$), and three flat mirrors $M_2$, $M_3$, and $M_4$ forming three independent resonators at the wavelengths 3.39, 2.39, and 1.15 $\mu$, respectively. The transmissions of the interference mirrors $M_2$ for $\lambda = 3.39$ $\mu$, $M_3$ for $\lambda = 2.39$ $\mu$, and $M_4$ for $\lambda = 1.15$ $\mu$ were respectively 30%, 20%, and 1.5%. Each of the resonators could be tuned independently in frequency with the aid of the piezoelectric elements to which the mirrors were secured. We take special note of the absence of a resonator for $\lambda = 0.63$ $\mu$, so that ordinary lasing was completely excluded.

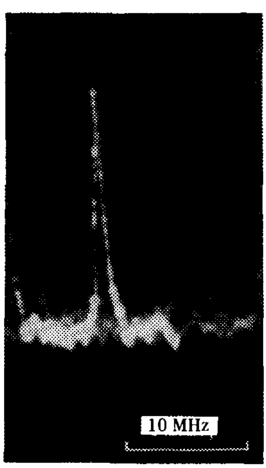


FIG. 3. Spectrum of beat signal produced when the emission at the summary frequency was mixed with emission of an He—Ne laser at $\lambda = 0.63$ $\mu$.

All three fields overlapped in the region of the discharge tube, and thus interacted with the same active medium. The discharge tube was 5 m long and had an inside diameter 15 mm.

Lasing at 3.39 μ and 1.15 μ took place in a wide range of pressures of the He—Ne mixture (0.1—3 Torr) and discharge currents (40—100 mA). Lasing at 2.39 μ was produced only in the presence of lasing at 3.39 and 1.15 μ, and was present in a rather narrow pressure interval (0.2—0.5 Torr at a mixture ratio $P_{Ne} : P_{He} = 1 : 10$). The input lasing power at the wavelengths 3.39, 2.39, and 1.15 μ was respectively 20, 12, and 7 mW. Coherent emission at $\lambda = 0.63$ μ without a resonator was observed only when lasing took place at the three wavelengths simultaneously. It was sufficient to interrupt the lasing at any of the wavelengths to cause the emission at $\lambda = 0.63$ μ to vanish. The power at 0.63 μ was of the order of $10^{-6}$ W. It started smoothly upon appearance of the simultaneous lasing at the three infrared wavelengths. At a certain distance from the prism (2 m) it was possible to observe on a shaded screen the structure of the spot produced by the radiation. The spot was sharply outlined and had an approximate diameter 5 mm. The structure of the spot changed with the adjustment of the resonator mirrors.

The coherent properties of the emission were investigated by mixing it with emission from an additional He—Ne laser $L_2$ at $\lambda = 0.63$ μ (Fig. 3), and by observing the signal at the intermode beat frequency. The cascade laser operated in the multimode regime and the emission at the summary frequency had therefore a complicated frequency structure. It was easy to see on the spectrum-analyzer screen the spectrum of the beats between the individual components of the structure. When the lasing frequency at one of the infrared wavelengths was varied, the emission at $\lambda = 0.63$ μ changed accordingly.

The observed phenomenon can be regarded as resonant multiphoton undisplaced scattering. The difference between the frequencies of the incident and scattered radiation provides unique opportunities for investigating resonant coherent scattering. The new method of frequency mixing in a gas is important for direct measurements of optical-band frequencies and for the development of a unified time and length standard. In the described cascade system of frequency mixing, two transitions are already used to develop lasers of highest frequency stability, at $\lambda = 3.39$ μ and $\lambda = 0.63$ μ.

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