APPLICATION OF GAS LASERS TO THE DETERMINATION OF SOME ATOMIC CHARACTERISTICS

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Submitted to JETP editor August 27, 1965; resubmitted March 22, 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) **51**, 121–128 (July, 1966)

An expression is obtained for the dependence of the radiation frequency and power of a gas laser on the resonator tuning and plasma column length under stationary excitation conditions. The expression is deduced by employing an equation for the density matrix in a coordinate system which is not fixed to the moving particles. The result is valid for an arbitrary ratio of the spectral line contour dispersion width to the Doppler width. Degeneracy of the operating levels, relaxation transitions between them, and the self-consistent mechanism of filling the lower operating level from the metastable level are also taken into account. The result is used for estimating some atomic characteristics by comparison with experimental results.

THE comparison of the calculation of the interaction of resonance radiation and a substance with experimental results enables one in a number of cases to calculate certain characteristics of the atomic system that is interacting with the radiation. These characteristics are of interest for the investigation of quantum amplifiers and oscillators and for the study of the interaction of resonance radiation with matter.

Thorough analyses of the peculiarities of generation in gas lasers have been given in a number of papers.[1] However, the use of the results of these papers in a comparison with experiment can be difficult. This is because of certain assumptions made in the calculation to facilitate the investigation of the regime of generation far from threshold and the interaction with several types of oscillations.

The problem can be solved more rigorously by investigating the threshold regime of generation with one type of oscillation. The calculations are also simplified by using an equation for the density matrix in a coordinate system that is not fixed to the moving particles. In addition it is of interest to include in the calculation the fundamental kinetic processes in the plasma of the gas laser which lead to inversion of the populations of the working levels.

1. THE EQUATIONS OF MOTION

The radiation field in the resonator is represented in the form

$$\mathbf{E} = \mathbf{E}^- \cos(\Omega t + \varphi + kz) + \mathbf{E}^+ \cos(\Omega t + \varphi - kz) = {}^1/_2 e^{-i(\Omega t+\varphi)}[\mathbf{E}^+ e^{i\xi} + \mathbf{E}^- e^{-i\xi}] + \text{c.c.} \quad \xi = kz. \tag{1}$$

The axis of the resonator is directed along z. The distribution of the field over the cross-section of the resonator is taken to be nearly homogeneous, so that $E^{-1}dE/dy \ll k$ and $E^{-1}dE/dx \ll k$, where k is the modulus of the wave vector which fits the resonator, and the amplitudes of the field are independent of the coordinates and time.

The Maxwell equation for the field in the medium is represented in the form (c = 1)

$$\left(\Delta - \frac{\partial^2}{\partial t^2}\right)\mathbf{E} = 4\pi\frac{\partial}{\partial t}\left(\sigma\mathbf{E} + \frac{\partial}{\partial t}\mathbf{D}\right), \tag{2}$$

where **D** is the dipole moment induced by the field in a unit volume of the medium, which is under the influence of an external source of excitation. The effective conductivity σ is connected with the Q of the resonator Q_r, its length l_r, the length of the active medium l, and the absorption and transmission coefficients of the mirrors L_1, L_2, and T_1, T_2, respectively, by the relations

$$4\pi\sigma = \Omega(Q_r l / l_r)^{-1} = (T_1 + T_2 + L_1 + L_2)/2l$$

or, more exactly, with diffraction losses taken into account:

$$4\pi\sigma = [{}^1/_2(T_1 + T_2 + L_1 + L_2) + \lambda^2_{mn}\cdot 4.62(l_r\lambda/\pi d^2)^{3/2}]l^{-1}, \tag{3}$$

where d is the diameter of the discharge tube, λ is the wavelength of the radiation, and λ_{mn} is the n-th root of a Bessel function of the m-th order ($I_m(\lambda_{mn}) = 0$). For the fundamental mode, $m = 0$, $n = 1$, $\lambda_{01} = 2.4$.

The amplitudes of the field oscillations at frequencies Ω, $-\Omega$ are obtained from Eq. (2):

$$(\Omega^2 - k^2 + i4\pi\sigma\Omega)\mathbf{E}_{-\Omega}{}^{\pm}e^{-i\varphi} = 8\pi\Omega^2\mathbf{D}_{-\Omega}{}^{\pm},$$
$$(\Omega^2 - k^2 - i4\pi\sigma\Omega)\mathbf{E}_{\Omega}{}^{\pm}e^{i\varphi} = 8\pi\Omega^2\mathbf{D}_{\Omega}{}^{\pm}, \tag{4}$$

where $D_{\pm\Omega}{}^{\pm}$ are the coefficients of the corresponding exponentials in the formula for the dipole moment per unit volume induced by the field.

The terms of interest to us are calculated with the help of the density matrix ρ, the equation for which in the interaction representation has the form ($\hbar = 1$)

$$\left(\frac{\partial}{\partial t} + \mathrm{v}\nabla\right)R_{mn} = i\sum_{s,\,\Omega=\pm\Omega}\{R_{ms}(U_\Omega)_{sn}\exp[i(\omega_{sn}+\Omega)t]$$
$$- R_{sn}(U_\Omega)_{ms}\exp[i(\omega_{ms}+\Omega)t]\} + [\Gamma(R)]_{mn}, \tag{5}$$

where $R = e^{iH_0t}\rho e^{-iH_0t}$, H_0 is the unperturbed Hamiltonian, $\omega_{sn} = \mathscr{E}_s - \mathscr{E}_n$, $\mathscr{E}_{s,n}$ are the eigenvalues of the Hamiltonian H_0. The operator $\Gamma(R)$ describes the processes of excitation of the radiating particles by the external source and their relaxation:

$$[\Gamma(R)]_{mn} = \begin{cases} \sum_k (R_{kk}\gamma_{km} - R_{mm}\gamma_{mk}, & m = n \\ -\gamma^{mn}R_{mn}, \quad \gamma^{mn} = \gamma^{nm}, & m \neq n \end{cases}, \tag{6}$$

where γ_{ik} are the probabilities of relaxation and excitation transitions per unit time and γ^{mn} is the decrement of the decay of an off-diagonal element of R. It follows from Eq. (5) that if the frequency Ω is close to the frequency of only one transition $m \leftrightarrow n$, then for an off-diagonal element of the density matrix $R_{mn}(\omega_{mn} > 0)$ only the perturbation Hamiltonian is "resonant":

$$(W_{-\Omega})_{mn} = (W_\Omega)_{nm}{}^* = (U_{-\Omega})_{mn}e^{-i\Omega t}$$
$$= e^{-i\Omega t}(U_{mn}{}^+e^{i\xi} + U_{mn}{}^-e^{-i\xi}),$$
$$\text{where } U_{mn}{}^{\pm} = -{}^1\!/\!_2\mathbf{E}^{\pm}\mathbf{d}_{mn}e^{-i\varphi}. \tag{7}$$

For gas lasers the energy level scheme shown in Fig. 1 is typical. Generation takes place in the transition $3 \leftrightarrow 2$. Level 3 is populated from the ground level 0 and by collisions of the second kind with impurity particles. Level 2 can be populated by relaxation transitions from level 3 and on account of exciting transitions from the metastable level 1, the population of which is in turn determined mainly by relaxation transitions from level 2 and the relaxation of metastables on the walls of the discharge tube. The lower levels may also be de-excited by collisions of the second kind. For real rates of excitation of level 3, the population of the ground level 0 may be assumed to be independent of this characteristic.

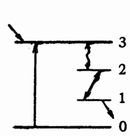

FIG. 1

Let q_3, q_2, and q_1 be the probabilities of excitation per unit time of the levels 3, 2, and 1, respectively, due to collisions of the second kind and from the ground state. In the case of de-excitation of some of the levels, q can be negative. Keeping all of this in mind, we represent the equations for the interesting components of the density matrix in steady state in the form

$$(-iw\partial/\partial\xi - 2\zeta_3)n_3 = -(VR^* - \text{c.c.}) - iq_3/ku,$$
$$(-iw\partial/\partial\xi - 2\zeta_2)n_2 = (VR^* - \text{c.c.}) - iq_2/ku - iy_{32}n_3 - iy_{12}n_1, \tag{8}$$

$$(-iw\partial/\partial\xi - 2\zeta_1)n_1 = -iq_1/ku - iy_{21}n_2;$$
$$(-iw\partial/\partial\xi - \zeta)R = \Delta nV, \tag{9}$$

where the following dimensionless quantities have been introduced: $w = v/u$, where u is the width of the Maxwellian distribution of the particles over velocity, $\zeta = x + iy$, where $x = (\Omega - \omega_{32})/ku$, $y = \gamma^{32}/ku$, $\zeta_i = iy_i/2$, $y_{ik} = \gamma_{ik}/ku$, $n_i = R_{ii}$, $\Delta n = n_3 - n_2$, $R_{32} = R_{23}^* = R\exp[-i(\Omega - \omega_{32})t]$,

$$V = (U_{-\Omega})_{32}/ku = V_+e^{i\xi} + V_-e^{-i\xi}, \tag{10}$$
$$V_{\pm} = -\mathbf{d}_{32}\mathbf{E}^{\pm}e^{-i\varphi}/2ku. \tag{11}$$

We shall solve Eqs. (8) and (9) by perturbation theory.

2. COHERENT MOTIONS IN THE MEDIUM

In zeroth approximation Eqs. (8) give the unsaturated difference of the populations of levels 3 and 2, i.e., the difference in the populations created by the excitation source at the initiation of generation:

$$\Delta n^0 = (1 - \alpha\gamma_{32})\frac{q_3}{\gamma_3} - \left(\alpha q_2 + \alpha\frac{\gamma_{12}}{\gamma_1}q_1\right) \tag{12}$$

$$\alpha = \gamma_1(\gamma_1\gamma_2 - \gamma_{21}\gamma_{12})^{-1}. \tag{13}$$

The second approximation brings in the action of the field, which decreases the population difference of the working levels created by the excitation source:

$$\Delta n^{(2)} = \Delta n^0[1 - \eta_1|V_+|^2 - \eta_2|V_-|^2$$
$$- {}^1\!/\!_2(\eta_3e^{i2\xi}V_+V_-{}^* + \text{c.c.}], \tag{14}$$

where

$$\eta_1 = 2\varkappa \operatorname{Im} \mathscr{L}_+, \quad \eta_2 = 2\varkappa \operatorname{Im} \mathscr{L}_-, \tag{15}$$

$$\eta_3 = (\mathscr{L}_-^* - \mathscr{L}_+)[\mathscr{L}_3 + \mathscr{L}_2 + i(y_{32}/2)\mathscr{L}_3\mathscr{L}_2];$$
$$\varkappa = y_3^{-1} + y_2^{-1}[(\gamma_3\gamma_2 - \gamma_{32}\gamma_2)/(\gamma_3\gamma_2 - \gamma_{12}\gamma_{21})]$$
$$= y_3^{-1} + \tilde{y}_2^{-1},$$
$$\mathscr{L}_\pm = (\pm w - \zeta)^{-1}, \quad \mathscr{L}_i = (w - \zeta_i)^{-1}. \tag{16}$$

In the third approximation the formula for the off-diagonal element R_{32} of the density matrix has the form

$$R_{32} = r_+ e^{i\xi} + r_- e^{-i\xi} + \tilde{r}_+ e^{i3\xi} + \tilde{r}_- e^{-i3\xi}. \tag{17}$$

However, to solve Eqs. (4) it is necessary to determine only the amplitudes r_+ and r_-. The formula for r_+ has the form

$$r_+ = \Delta n^0 V_+ \mathscr{L}_+ \{1 - \eta_1 |V_+|^2 - (\eta_2 + {}^1/_2\eta_3)|V_-|^2\}. \tag{18}$$

The formula for r_- is obtained from the solution (18) by replacing the subscripts + by − and vice versa. The last term in Eq. (18), proportional to η_3, describes the contribution to the traveling wave of polarization that arises as a result of the interaction in the medium of the second spatial harmonic of the population difference with the harmonic of the field traveling in the opposite direction. However, in most cases this term can be neglected, as will be shown below, so that it is of higher order with respect to Γ/ku compared with the other terms. Thus the discriminating factor is the smallness of Γ/ku—the ratio of the dispersion width to the Gaussian width of the spectral lines of the transition.

Assuming that a Maxwellian velocity distribution of the radiating particles is realized in the discharge, we average (18) over a distribution with half-width u. We obtain

$$\langle r_+\rangle_v = \Delta n^0 V_+ \{\mathscr{H} - 2\varkappa\mathscr{H}_1 |V_+|^2 - 2\varkappa\mathscr{H}_2 |V_-|^2 + \mathscr{H}_3 |V_-|^2\}, \tag{19}$$

where

$$\mathscr{H}_1 = \langle \mathscr{L}_+ \operatorname{Im} \mathscr{L}_+\rangle_v, \quad \mathscr{H}_2 = \langle \mathscr{L}_+ \operatorname{Im} \mathscr{L}_-\rangle_v,$$
$$\mathscr{H}_3 = {}^1/_2\langle \mathscr{L}_+(\mathscr{L}_+ - \mathscr{L}_-^*)[\mathscr{L}_3 + \mathscr{L}_2 + {}^1/_2 i y_{32}\mathscr{L}_3\mathscr{L}_2]\rangle_v. \tag{20}$$

As is shown in the Appendix (Eqs. (A6)–(A8)), all the integrals $\mathscr{H}_1$, $\mathscr{H}_2$, $\mathscr{H}_3$ can be expressed in terms of a single tabulated[2, 3] function $\mathscr{H}(\zeta)$, used in the theory of spectral line shapes and plasma theory:

$$\mathscr{H}(\zeta) = \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} \frac{e^{-w^2}}{\pm w - \zeta} dw = 2i \int_0^\infty dw \exp(-w^2 + 2iw\zeta). \tag{21}$$

As already mentioned, $\mathscr{H}_3$ gives the weight with which the interference of the harmonic of the field with the second harmonic of the population difference enters into the traveling wave of polarization.

With the aid of Eq. (19) it is possible to calculate $D_{-\Omega}^+$, the amplitude of the traveling wave of polarization arising as a result of the interaction of a coherent field with a medium that is under the influence of a source of excitation. If the energy levels of the medium are degenerate, the result must be summed over all states of the levels:

$$\mathbf{D}_{-\Omega}^+ = \sum_{a,\,2m,\,3m} \mathbf{d}_{2m,\,3m}^a (\langle r_+\rangle_v)_{3m,\,2m}^a. \tag{22}$$

In the phenomenological calculation, the summation over a in the relaxation times of the particle interaction reduces to a multiplication by the N-density of the working substance. For simplicity we shall assume that in the discharge plasma the relaxation and excitation of all states of a given level by the source are the same, which is apparently actually the case. Then from Eqs. (19) and (22) we get

$$D_{-\Omega}^+ = e^{-i\varphi}E^+ \frac{\Delta N^0 S}{2ku}\Big\{G_1\mathscr{H} - \frac{GS}{(2ku)^2}[2K(\mathscr{H}_1 E^{+^2} + \mathscr{H}_2 E^{-^2}) - \mathscr{H}_3 E^{-^2}]\Big\}, \tag{23}$$

where

$$\Delta N^0 = N[1 - (\Gamma_{32}/\Gamma_2)\beta](Q_3/g_3\Gamma_3) - \beta[(Q_2/g_2\Gamma_2) + (\Gamma_{12}/\Gamma_2)(Q_1/g_1\Gamma_1)], \tag{24}$$

$\beta = \Gamma_1\Gamma_2(\Gamma_1\Gamma_2 - \Gamma_{21}\Gamma_{12})^{-1}$, g is the statistical weight of the levels, Q is the probability of excitation of the levels per unit time by the source of excitation, Γ are the relaxation characteristics of the levels,

$$K = ku[\Gamma_3^{-1} + \Gamma_2^{-1}(\Gamma_2\Gamma_3 - \Gamma_{32}\Gamma_2)(\Gamma_2\Gamma_3 - \Gamma_{21}\Gamma_{12})^{-1}]$$
$$= ku(\Gamma_3^{-1} + \tilde{\Gamma}_2^{-1}), \tag{25}$$

$\mathscr{H}$, $\mathscr{H}_1$, $\mathscr{H}_2$, $\mathscr{H}_3$ are the corresponding integrals with γ replaced by Γ, S is the strength of the transition:

$$S = \sum_{3m,2m} |d_{3m,2m}|^2, \tag{26}$$

$$G_1 = \sum_m \begin{pmatrix} j & 1 & j' \\ -m & 0 & m \end{pmatrix}^2 = \frac{1}{3}, \quad G = \sum_m \begin{pmatrix} j & 1 & j' \\ -m & 0 & m \end{pmatrix}^4, \tag{27}$$

(⋯) are Wigner 3j-symbols. Here we have made use of the rule for summing matrix elements of tensor operators.[3]

In a previous paper[4] an attempt was made to take into account the degeneracy of the levels for particles in which the dipole moments can be considered as firmly coupled to the orientation of the particles (molecules). However, the limits of applicability of the results obtained are not clear, since the picture of the phenomenon in molecules is considerably more complex.

3. RADIATION POWER

The power loss in the resonator is determined by its Q:

$$P_{\text{loss}} = \Omega Q_r^{-1} W_{\text{stor}}, \quad (28)$$

where W_{stor} is the energy stored in the resonator:

$$W_{\text{stor}} = \int_0 \frac{\overline{E}^2}{4\pi} dv = s \int_0^{l_r} \frac{\overline{E}^2}{4\pi} dz = \frac{sl_r}{16\pi} E_0^2, \quad (29)$$

E_0 is the amplitude of the electromagnetic wave averaged over the cross section of the beam, s is the beam cross section. The averaging is carried out over a period of the oscillation, the integration over the resonator volume. In the case of a nearly homogeneous distribution of the field over the cross section of the beam, the integration over volume reduces to an integration over the length of the resonator. As follows from (3) and (28), the power of the radiation from one end of the laser with transmission coefficient T is

$$P = (T/2)s(E_0^2/16\pi). \quad (30)$$

Equations (4), (23), and (30) permit the calculation of the frequency and power of generation as a function of the characteristic frequency of the resonator and the rate of excitation of the working levels, with allowance for the relaxation processes and filling of the metastable level 1.

It is probably simpler from the experimental point of view to study the dependence of the power on the discharge length for constant excitation conditions rather than on the rates of excitation of the levels. The dependence of the power on the discharge length l is connected with the dependence of the equations on σ.

For a traveling-wave laser ($E^- = 0$, $E^+ = E$), we obtain from Eqs. (4), (23), and (30), by expressing the transition strength through the probability of spontaneous emission in the transition $3 \rightarrow 2$:

$$P_t = \frac{scT}{32\pi}\left[\frac{9}{2}\frac{g_3 G A_{32}}{\hbar k^3 (2ku)^2} K \frac{\ln \mathcal{H}_1}{\mathcal{H}(iy)} \frac{l}{l_0}\right]^{-1} \times \left(\frac{l}{l_0}\frac{\operatorname{Im}\mathcal{H}}{\mathcal{H}(iy)} - 1\right),$$

$$A_{32} = 4k^3 S_{32}/3\hbar g_3, \quad (31)$$

where l_0 is the threshold length of discharge for generation in the line center for a given set of excitation conditions:

$$l_0 = \sigma l k^3 u [c g_3 \Delta N^0 A_{32} \mathcal{H}(iy)]^{-1}, \quad (32)$$

(σl) is independent of l. The frequency of generation is determined by the real part of the saturated susceptibility of the medium:

$$\frac{\Omega^2 - (kc)^2}{\Omega^2} = 4\pi \frac{\Delta N^0 S_{32}}{3\hbar k u}\left\{\operatorname{Re}\mathcal{H} - \frac{GS}{(2\hbar k u)^2} 6K \operatorname{Re}\mathcal{H}_1 E^2\right\}. \quad (33)$$

Similarly, we obtain for a standing-wave laser ($E^+ = E^- = E$):

$$P_{\text{st}} = \frac{csT}{8\pi}\left[\frac{9}{4}\frac{g_3 G A_{32}}{\hbar k^3 (2ku)^2} \frac{2K \operatorname{Im}(\mathcal{H}_1 + \mathcal{H}_2) - \operatorname{Im}\mathcal{H}_3}{\mathcal{H}(iy)} \frac{l}{l_0}\right]^{-1} \times \left(\frac{l}{l_0}\frac{\operatorname{Im}\mathcal{H}}{\mathcal{H}(iy)} - 1\right), \quad (34)$$

l_0 is given by Eq. (32), and the frequency of generation by the following formula:

$$\frac{\Omega^2 - (kc)^2}{\Omega^2} = 4\pi \frac{\Delta N^0 S_{32}}{3\hbar k u} \times \left\{\operatorname{Re}\mathcal{H} - \frac{3GS}{(2\hbar k u)^2}[2K \operatorname{Re}(\mathcal{H}_1 + \mathcal{H}_2) - \operatorname{Re}\mathcal{H}_3] E^2\right\}. \quad (35)$$

As follows from the Appendix, Re $(\mathcal{H}_1, \mathcal{H}_2, \mathcal{H}_3)$ together with Re$\mathcal{H}$ vanish when $x = 0$. This means that tuning the resonator to the center of the line of an atomic transition corresponds to generation at the proper frequency of the transition at any radiation intensity. Otherwise the frequency of generation varies with the level of excitation in accordance with Eqs. (33) and (35). However, these assertions are true only for homogeneous broadening in the absence of saturation of the line shape. In the case of inhomogeneous broadening (e.g., when isotopes are present in the discharge), it is necessary to average over the distribution of the proper frequencies of the atomic transitions. In averaging over the velocities of the particles, no kind of limitation was imposed on the magnitude of the ratio Γ/ku; hence the results may turn out to be useful for longwave transitions of lasers using heavy inert gases ($\Gamma/ku \sim 1$) and for the case of solids ($\Gamma/ku \gg 1$).

If the results of a detailed calculation of the interaction of quantum systems with radiation are compared with corresponding experiments, it is possible in a number of cases to derive unknown atomic characteristics from known ones. Such an experiment, for example, could be frequency measurements relating to Eqs. (31)–(35). In addition, as follows from Eqs. (31) and (34), the slope of the curve of generated power in the line center versus the length of the discharge column for constant excitation conditions is determined only by the relaxation characteristics of the system and the known parameters of the resonator. Further, by measuring the threshold lengths of the plasma column for different excitation levels, it is possible to determine from Eq. (32) the unsaturated population difference of the operating levels in a given discharge tube for corresponding excitation conditions. In this connection stabilization of the frequency of generation (against thermal and mechanical fluctuations of the proper frequency of the resonator) can be accomplished by a number of known methods, as was done, for example in [5].

Unfortunately, we do not have available any results of a detailed experiment on the threshold regime of emission. However, for the sake of illustration, we shall attempt to make some estimates by means of the formulas obtained, with no pretensions as to how accurate they might be. When $x = 0$, $y \ll 1$, we find from Eqs. (32), (34), and (A11)

$$l_0 = \sigma l k^3 u (c\sqrt{\pi} g_3 \Delta N^0 A_{32})^{-1}, \tag{36}$$

$$P_{\mathrm{st}} = \frac{csT}{8\pi}\left[\frac{9}{16}\frac{g_3 G A_{3^n}}{\hbar k^3 \Gamma}(\Gamma_3^{-1} + \tilde{\Gamma}_2^{-1})\frac{l}{l_0}\right]^{-1}\left(\frac{l}{l_0} - 1\right), \tag{37}$$

where $\tilde{\Gamma}_2$ is the effective total probability of escape of a particle from the lower level; obviously, the Γ's can take into account certain collective processes such as the elastic collisions considered by Weisskopf.

For estimates pertaining to the transition $3s_2-2p_4$ in Ne we make use of the results of [6], in which was studied the dependence of the power of a Ne-He laser on discharge length and the dependence of threshold plasma lengths on current. However, in this paper no information was given about the cross section of the generated beam of radiation, and the number of measurements in the threshold region was small. Assuming the cross section of the beam to be equal to the area of the first Fresnel zone, we obtain the following starting data for our calculation: $s \approx 5 \times 10^{-2}$ cm², $T = 1.5 \times 10^{-2}$, $L = 2.5 \times 10^{-2}$, $[dP/d(l/l_0)]_{l=l_0} \approx 3.5$ mW. Further, assuming $\Gamma_3 \approx \Gamma_2 \approx 8 \times 10^7$ sec⁻¹, $A_{32} \approx 8 \times 10^6$ sec⁻¹, [7] $ku = 6 \times 10^9$ sec⁻¹, $j_3 = 1$, $j_2 = 2$, we obtain by means of Eq. (36) and the results of [6], the dependence of the population difference of levels $3s_2$ and $2p_4$ on current shown in Fig. 2, and for Γ we obtain the value $\approx 8 \times 10^8$ sec⁻¹.

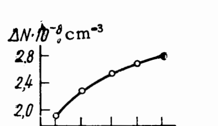

FIG. 2

In conclusion, the author thanks V. A. Ignatchenko for his interest and helpful discussions.

APPENDIX

We expand the integrands of the functions $\mathscr{H}_1$, $\mathscr{H}_2$, and $\mathscr{H}_3$ in simple fractions. We obtain

$$\mathscr{H}_1 = \langle \mathscr{L}_+ \operatorname{Im} \mathscr{L}_+ \rangle = (2i)^{-1}[\langle \mathscr{L}_+^2 \rangle - (2iy)^{-1}(\langle \mathscr{L}_+ \rangle - \langle \mathscr{L}_+^* \rangle)], \tag{A.1}$$

$$\mathscr{H}_2 = \langle \mathscr{L}_+ \operatorname{Im} \mathscr{L}_- \rangle = (2i)^{-1}[(2x)^{-1}(\langle \mathscr{L}_+ \rangle + \langle \mathscr{L}_-^* \rangle) - (2\zeta)^{-1}(\langle \mathscr{L}_+ \rangle + \langle \mathscr{L}_- \rangle)], \tag{A.2}$$

$$\begin{aligned}\mathscr{H}_3 = &-{}^1/_2[\Delta_3^2 + \Delta_2^2 + (iy_{32}/2)(\Delta_3\Delta_2^2 + \Delta_2\Delta_3^2)]\langle \mathscr{L} \rangle_+ \\ &+ (4x)^{-1}(\Delta_2 + \Delta_3)\langle \mathscr{L}_+ \rangle - (4x)^{-1}(\Delta_2^* + \Delta_3^*)\langle \mathscr{L}_-^* \rangle \\ &+ (iy_{32}/8x) \times \Delta_3\Delta_2\langle \mathscr{L}_+ \rangle - (iy_{32}/8x)\Delta_3^*\Delta_2^*\langle \mathscr{L}_-^* \rangle \\ &+ {}^1/_2[\Delta_3 + \Delta_2 + (iy_{32}/2)\Delta_3\Delta_2] \\ &\times \langle \mathscr{L}_+^2 \rangle + {}^1/_2(\Delta_3^2 - |\Delta_3|^2)[1 - (iy_{32}/4)\Delta_{23}]\langle \mathscr{L}_3 \rangle \\ &+ {}^1/_2(\Delta_2^2 - |\Delta_2|^2)[1 + (iy_{32}/4)\Delta_{23}]\langle \mathscr{L}^2 \rangle.\end{aligned} \tag{A.3}$$

The angle brackets denote an average over velocities, and

$$\Delta_i = (\zeta - \zeta_i)^{-1}, \quad \Delta_{23} = (\zeta_2 - \zeta_3)^{-1} = i(y_2 - y_3). \tag{A.4}$$

From Eq. (21) it follows that $\langle \mathscr{L}_+ \rangle = \langle \mathscr{L}_- \rangle = \mathscr{H}(\zeta)$. From this same equation it is easily established that

$$\langle \mathscr{L}_+^2 \rangle = \frac{d}{d\zeta}\mathscr{H}(\zeta) = -2[1 + \zeta\mathscr{H}(\zeta)]. \tag{A.5}$$

Thus, we obtain

$$\mathscr{H}_1 = i(1 + \zeta\mathscr{H}(\zeta) + (2y)^{-1}\operatorname{Im}\mathscr{H}(\zeta)], \tag{A.6}$$

$$\mathscr{H}_2 = (2i)^{-1}[x^{-1}\operatorname{Re}\mathscr{H}(\zeta) - \zeta^{-1}\mathscr{H}(\zeta)], \tag{A.7}$$

$$\begin{aligned}\mathscr{H}_3 = &-(\Delta_3 + \Delta_2 + iy_{32}\Delta_3\Delta_2/2) - \{(\Delta_3 + \Delta_2 \\ &+ iy_{32}\Delta_3\Delta_2/2]\zeta + {}^1/_2[\Delta_3^2 + \Delta_2^2 + iy_{32}(\Delta_3 \\ &+ \Delta_2)\Delta_3\Delta_2/2]\}\mathscr{H}(\zeta) - (i2x)^{-1}\operatorname{Im}[(\Delta_3 + \Delta_2 \\ &+ iy_{32}\Delta_3\Delta_2/2)\mathscr{H}(\zeta)] + {}^1/_2\Delta_3^2 - |\Delta_3|^2) \\ &\times [1 - iy_{32}\Delta_{23}/2]\mathscr{H}(\zeta_3) + {}^1/_2(\Delta_2^2 - |\Delta_2|^2) \\ &\times [1 + iy_{32}\Delta_{23}/2]\mathscr{H}(\zeta_2).\end{aligned} \tag{A.8}$$

Thus, all the integrals of interest to us are expressed in terms of one, the expansion for which in powers of y under the condition $y \ll 1$ has the form

$$\mathscr{H}(x + iy) = [-2F(x) + 2\sqrt{\pi}xye^{-x^2} + \ldots] + i\{\sqrt{\pi}e^{-x^2} - 2y[1 - 2xF(x)] + \ldots\}, \tag{A.9}$$

where

$$F(x) = e^{-x^2}\int_0^x dt e^{t^2}. \tag{A.10}$$

When $x = 0$, $y \ll 1$, we obtain, up to terms in y^{-1}

$$\mathcal{H}_1 = \mathcal{H}_2 = i(\sqrt{\pi}/2)y^{-1}, \tag{A.11}$$

$\mathcal{H}_3$ contains only higher powers of y.

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Translated by L. M. Matarrese
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