APPENDIX II

Transfer of Singularity at Arbitrary Saturation Parameters

At $G_{23}^{\alpha}, G_{31}^{\alpha} \gtrsim 1$, the gains in (4.1) are given by the exact expressions $\varkappa_{\alpha} = \varkappa_{\alpha}(I_1, I_2, \Omega_1, \Omega_2)$, which can be obtained, for example, after averaging the expressions obtained in Appendix I over the velocities. The absorption coefficient is $\varkappa = \varkappa(\Delta\omega, I_2)$. Assuming $\varkappa \ll \varkappa_2$, we obtain the rapidly varying increments $\delta I_a(\Delta\omega)$ to the generation intensities, due to the nonlinear absorption:

$$\delta I_1(\Delta\omega) = -\eta_1 \delta I_2(\Delta\omega), \quad \delta I_2(\Delta\omega) = \frac{\varkappa(\Delta\omega, I_2)}{\xi_2(1-\eta_1\eta_2)}, \tag{II.1}$$

where

$$\xi_a = \left(\frac{\partial \varkappa_a}{\partial I_a}\right), \quad \eta_1 = -\left(\frac{\partial I_1}{\partial I_2}\right)_{\varkappa_1}, \quad \eta_2 = -\left(\frac{\partial I_2}{\partial I_1}\right)_{\varkappa_2},$$

I_1 and I_2 are the stationary lasing intensities in the absence of an absorbing cell, with $\varkappa_a$ taken to be functions of only I_a after eliminating the frequencies Ω_a with the aid of the equations for the phases. The contrasts of the singularities

$$h_a = [\delta I_a(\infty) - \delta I_a(0)]/I_a \tag{II.2}$$

have opposite signs in the case of normal competition $\eta_1 > 0$, with

$$h_1 = -\eta_1 \frac{I_2}{I_1}, \quad h_2 = \frac{\varkappa(\infty, I_2) - \varkappa(0, I_2)}{\xi_2(1-\eta_1\eta_2) I_2}. \tag{II.3}$$

From the condition of the stability of the generation when account is taken of the fact that $\xi_1, \xi_2 < 0$ (saturation under the influence of the intrinsic field) it follows that $1 - \eta_1\eta_2 > 0$, so that a peak is formed on the transition $3 \to 2$, and the dip is transferred to the transition $3 \to 1$.

[1)]Neglect of the higher spatial harmonics means smallness of the parameters $(\tilde{\gamma}_{3a}^2/\gamma_{3a}^2) G_{3a} \ll 1$, $a = 1, 2$ (cf. Ref. 12), where the saturation parameters G and $\tilde{\gamma}_{nm}$ are given in (2.8). At $\gamma_{3a}/\gamma_{3a} \ll 1$, which is the case, e.g., for neon,[1,12] neglect of the harmonics is permissible also for a saturation $G \gtrsim 1$ which is not small.

[2)]With the aid of (4.3) it is easy to investigate also the line shape for two-frequency lasing, in analogy with the procedure used by Melekhin[7] for the transition scheme $3 \to 1$, $2 \to 1$.

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Reflection and refraction of a plane wave by the interface of two media, with allowance for positron polarization of the medium

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Integral equations are obtained for the propagation of electromagnetic waves in an atomic system in whose spectrum negative energy states are included as intermediate states (positron polarization of the medium). Additional terms that depend on the coherence properties of the medium are obtained in the Lorentz-Lorenz and Fresnel formulas for a plane wave. It is shown that they are likely to play an important role in systems with small inhomogeneous broadening of the spectral lines (for example, in a rarefied gas).

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1. When optical radiation propagates in a medium, the resultant field acting on an arbitrary j-th dipole is made up of the fields $\vec{H}_j$, $\vec{E}_j$ of the incident wave (which propagates at the speed of light in vacuum), and the fields produced by the surrounding atom at the location of the j-th dipole. Usually in the calculation of the field of the surrounding atoms one confines oneself to the so-called dipole field H_j^e, E_j^e.[1] As shown by us in Ref. 2, the

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dipole field can be calculated if the interactions of the atoms in the external radiation field are regarded as effects of third order of perturbation theory. The physical meaning of this description is illustrated by Fig. 1, where m and n are the initial states of the l-th and j-th atoms. As a result of the interactions of the atoms via the field of the virtual photons, the j-th atom goes over into a certain final state p, while the l-th atom goes into an intermediate state l_+ with positive energy. Under the influence of the external radiation field, the l-th atom then goes from the intermediate state l_+ to the initial state m, as a result of which an energy quantum $\hbar\omega$ is emitted (absorbed). Simultaneously with these processes, we took into account in Ref. 2, the interactions between the atoms in the radiation field with allowance for the intermediate positronic states l_-. This led to the appearance of additional internal field acting on the fixed j-th atom (positronic polarization of the medium). It then becomes possible to describe processes in which intermediate states l_- participate within the framework of first rather than third-order perturbation theory, if one uses the Hamiltonian of the interaction of a system of N atoms with the radiation field in the following form ($r_{lj} \gg \lambda$):

$$\mathscr{H}=\mathscr{H}_1+\mathscr{H}_2, \quad \mathscr{H}_1=-\frac{e}{mc}\sum_{j=1}^{N}(\mathbf{p}_j\mathbf{A}_j),$$
$$\mathscr{H}_2=\frac{e^2}{m^2c^3}\sum_{l\neq j}^{N}\frac{b_{lj}}{r_{lj}}[(\tilde{\mathbf{A}}_l\mathbf{p}_j)-(\tilde{\mathbf{A}}_l\mathbf{n}_{lj})(\mathbf{n}_{lj}\mathbf{p}_j)], \tag{1.1}$$

where r_{lj} is the distance between the atoms, $\mathbf{p}_{l(j)}$ are the operators of the momenta of the $l(j)$-th atom, $\tilde{\mathbf{A}}_{(j)}$ is the vector potential of the radiation field $\mathbf{n}_{lj}=\mathbf{r}_{lj}/r_{lj}$, λ is the wavelength of the external radiation field, $b_{lj}=\exp(ic^{-1}\omega_{pn}r_{jl})$, ω_{pn} is the natural frequency in the spectrum of the atom and is equal to $|\omega_p-\omega_n|$, and ω_p, ω_n are the frequencies of the states p and n of the atom.

A similar interaction Hamiltonian was obtained also by Drake[3] for the case of one helium-like atom, in which account was taken of the interaction between the electrons in the external-radiation field within the framework of third-order perturbation theory, with satisfaction of the obvious condition $r_{lj} \ll \lambda$, where r_{lj} is the distance between the electrons.[3] Thus, allowance for the negative-energy intermediate states of the atoms with interactions between a system of atoms and an external radiation field leads to the need for considering the resultant field acting on an arbitrary j-th dipole of the system, in the form of three components: the external radiation field, the internal field of the dipoles, and the internal field due to allowance for the negative-energy intermediate states in the atoms.

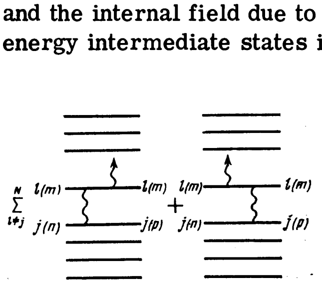

FIG. 1.

In the present paper we derive integral equations for the propagation of electromagnetic waves, with allowance for the positronic polarization of the medium. We obtain the additional terms in the Lorentz-Lorenz formula and in the Fresnel formulas for the reflectivity (transmittivity) of the interface between two media. These terms are due to positronic polarization of the medium and depend on the coherence properties of the medium.

2. The resultant field acting on the j-th dipole in an optical medium is given by

$$\mathbf{E}_j=\tilde{\mathbf{E}}_j+\sum_l{}'\mathbf{E}_{jl}^{e}+\sum_l{}'\mathbf{E}_{jl}^{p}, \tag{2.1}$$

where the superscript $e(p)$ denotes the electronic (positronic) component of the electric-field intensity. To calculate E_{jl}^e we start out with retarded potentials. Recognizing that the displacements ξ_l of the electrons from the equilibrium positions of the l-th atoms are small compared with the distance between the nuclei of the j-th and l-th atoms, and performing a gauge transformation of the potentials, we obtain the scalar (φ_{jl}^e) and the vector (A_{jl}^e) potentials of the field of the l-th dipole at the location of the j-th dipole.

We change now from potentials to the corresponding electric field intensities:

$$\mathbf{E}_{jl}^{e}=-\frac{e}{c^2}\frac{[\ddot{\xi}_l]-\mathbf{n}_{jl}(n_{jl}[\ddot{\xi}_l])}{r_{jl}}-\frac{e}{c}\frac{[\dot{\xi}_l]-3\mathbf{n}_{jl}(\mathbf{n}_{jl}[\dot{\xi}_l])}{r_{jl}^2}+\frac{3en_{jl}([\xi_l]\mathbf{n}_{jl})}{r_{jl}^3}-\frac{e[\xi_l]}{r_{jl}^3}=\operatorname{rot}\operatorname{rot}\frac{\mathbf{d}_l(t-r_{jl}/c)}{r_{jl}}. \tag{2.2}$$

Here $\mathbf{d}_l=e\xi_l$ is the dipole moment of the l-th atom, and the symbol $[\ldots]$ denotes that the corresponding quantity depends on the time $t-r_{jl}/c$. The expression for the field intensities E_{jl}^p is obtained from (1.1):

$$\mathbf{E}_{jl}^{p}=-\frac{e^2}{mc^2}b_{jl}[\tilde{\mathbf{E}}_l-\mathbf{n}_{jl}(\mathbf{n}_{jl}\tilde{\mathbf{E}}_l)]/r_{jl}.$$

Since the external radiation field $\mathbf{E}_j$ is replaced in the medium by the field $\mathbf{E}$ we obtain, taking (2.2) into account, the following integral equation

$$\mathbf{E}(r,t)=\tilde{\mathbf{E}}(r,t)+\int\operatorname{rot}\operatorname{rot}\frac{\mathbf{D}(r',t-R/c)}{R}dV'$$
$$-a\int N(\mathbf{r}')\frac{b(R)}{R}\left[\mathbf{E}\left(\mathbf{r}',t-\frac{R}{c}\right)-\mathbf{n}_0\left(\mathbf{n}_0\mathbf{E}\left(\mathbf{r}',t-\frac{R}{c}\right)\right)\right]dV', \tag{2.3}$$

where $\mathbf{D}$ is the electric dipole moment per unit volume of the medium, $N(\mathbf{r}')$ is the concentration of the atoms as a function of the coordinates,

$$R=|\mathbf{r}-\mathbf{r}'|, \quad \mathbf{n}_0=\mathbf{R}/R, \quad a=e^2/mc^2.$$

The obtained integral equation of propagation of the electromagnetic waves in the medium differs from the known equations[1] in that it contains terms due to the positronic polarization of the medium. We examine their influence on the dielectric constant of the medium and on the reflection (refraction) of a plane wave by the interface between two media.

3. We consider the case $\mathbf{D}=N\alpha\mathbf{E}$, where α is the polarizability of the atom. Let Σ be the boundary of the considered media. Then, for the observation points located inside the medium, the integration in (2.3) is carried out over the entire volume bounded by $\Sigma+\sigma$, where σ is a sphere of small radius δ enclosing the atom at the point of observation with a radius vector $\mathbf{r}$. The external radiation field is assumed to be a plane monochromatic wave

$$\tilde{\mathbf{E}}=\tilde{\mathbf{E}}_0(\mathbf{r})e^{-i\omega t},$$

propagating with velocity c, and the solution of (2.3) is sought in the form of a plane monochromatic wave

$$\mathbf{E}(\mathbf{r},t)=\mathbf{E}_0(\mathbf{r})e^{-i\omega t}, \tag{3.1}$$

propagating with velocity c/n, where n is the refractive index of the medium, with

$$\nabla^2\mathbf{E}_0+n^2k^2\mathbf{E}_0=0,\quad k=\omega/c. \tag{3.2}$$

The electric-polarization wave propagates in the medium with velocity c/n and is given by

$$\mathbf{D}(r,t)=\mathbf{Q}(r)e^{-i\omega t},\quad \nabla^2\mathbf{Q}+n^2k^2\mathbf{Q}=0. \tag{3.3}$$

Taking (3.3) and the fact that div $\mathbf{Q}=0$ into account, we obtain[1]

$$\int_\sigma^\Sigma \operatorname{rot}\operatorname{rot}\mathbf{Q}(r')G(R)\,dV'=\frac{4\pi}{3}\left(\frac{n^2+2}{n^2-1}\right)\mathbf{Q}$$

$$+\frac{1}{(n^2-1)k^2}\operatorname{rot}\operatorname{rot}\int_\Sigma\left\{\mathbf{Q}\frac{\partial G}{\partial\nu'}-G\frac{\partial\mathbf{Q}}{\partial\nu'}\right\}dS', \tag{3.4}$$

$$G=e^{ikR}/R,$$

where $\partial/\partial\nu'$ denotes differentiation along the outward normal to the boundary Σ. Taking (3.2) into account, we obtain the value of the following integral in (2.3):

$$\int_\sigma^\Sigma\frac{b(R)}{R}\mathbf{E}\left(r',t-\frac{R}{c}\right)dV'=\int_\sigma^\Sigma G_0(R)\mathbf{E}_0(r')\,dV'$$

$$=\frac{1}{n^2k^2-(k+k_0)^2}\int_\sigma^\Sigma(\mathbf{E}_0\nabla^2G_0-G_0\nabla^2\mathbf{E}_0)\,dV',$$

where ω_0 is the natural frequency of the atom,

$$G_0=\exp[i(k+k_0)R]/R,\quad k_0=\omega_0/c.$$

Integration with the aid of Green's theorem yields

$$\int_\sigma^\Sigma\frac{b(R)}{R}\mathbf{E}\left(\mathbf{r}',t-\frac{R}{c}\right)dV'=\frac{1}{n^2k^2-(k+k_0)^2}\Big[4\pi\mathbf{E}_0(\mathbf{r})$$

$$+\int_\Sigma\left\{\mathbf{E}_0\frac{\partial G_0}{\partial\nu'}-G_0\frac{\partial\mathbf{E}_0}{\partial\nu'}\right\}dS'\Big]. \tag{3.5}$$

Since the obvious approximate equality

$$\int_\sigma^\Sigma\frac{b(R)}{R}\mathbf{n}_0\left(\mathbf{n}_0\mathbf{E}\left(\mathbf{r}',t-\frac{R}{c}\right)\right)dV'\approx\frac{1}{3}\int_\sigma^\Sigma\mathbf{E}_0G_0(R)\,dV' \tag{3.6}$$

is satisfied, the last integral of (2.3) can also be calculated. In real systems the natural frequencies ω_0 of individual atoms differ from the mean-statistical value $\omega_0^{(0)}$ because of the inhomogeneous broadening of the spectral lines, due to the scatter of the local fields, to the motion of the atoms, etc.

If the condition $|\omega^0-\omega|\gg\Delta\omega_0$ is satisfied, where $\Delta\omega_0$ is a certain shift of the natural frequency of the atom from the mean-statistical value, then the influence of the inhomogeneous broadening of the spectral lines on the dipole field (3.4) is negligible and we shall disregard it. The positronic polarization, as seen from (3.5) and (3.6), depends more strongly on the inhomogeneous broadening of the spectral lines and will be taken into account by us with the aid of a Gaussian distribution of the frequencies $g(\Delta\omega)$. We then obtain in place of (3.5)

$$\left\langle\int_\sigma^\Sigma\frac{b(R)}{R}\mathbf{E}\left(\mathbf{r}',t-\frac{R}{c}\right)dV'\right\rangle\approx\frac{1}{n^2k^2-(k+k_0^{(0)})^2}\Big[\frac{4\pi\gamma_0}{\overline{\Delta\omega_0}}\mathbf{E}_0(\mathbf{r})$$

$$+\int_\Sigma\beta_0(R)\left\{\mathbf{E}_0\frac{\partial G_0^{(0)}(R)}{\partial\nu'}-G_0^{(0)}(R)\frac{\partial\mathbf{E}_0}{\partial\nu'}\right\}dS'\Big], \tag{3.7}$$

where the angle brackets $\langle\ldots\rangle$ denote averaging over $\Delta\omega_0$, γ_0 is the constant of the radiative damping of the atom, $\overline{\Delta\omega_0}$ is the inhomogeneous line width at half-height, $k_0^{(0)}=\omega_0^{(0)}/c$, and $G_0^{(0)}$ is obtained from G_0 by replacing k_0 by $k_0^{(0)}$

$$\beta_0(R)=\exp(-R^2\overline{\Delta\omega_0}^2/2c^2).$$

We now separate in the right-hand side of Eq. (2.3), in which we substitute the obtained values of the integrals (3.4) and (3.5), the terms corresponding to waves propagating with velocity c/n. We then obtain a formula that connects the refractive index of the medium with the polarizability of the atoms and with their concentration when a monochromatic plane wave propagates in the medium:

$$\frac{n^2-1}{n^2+2}=\frac{4\pi}{3}N\alpha-\frac{8\pi}{3}\frac{aN\gamma_0/\overline{\Delta\omega_0}}{n^2k^2-(k+k_0^{(0)})^2}\frac{n^2-1}{n^2+2}. \tag{3.8}$$

Without the second term in the right-hand side, Eq. (3.8) is the Lorentz–Lorenz formula. For gases, Eq. (3.8) is somewhat simplified, since $n^2+2\approx3$, and takes the form

$$n^2=\varepsilon\approx1+4\pi N\left[\alpha-\frac{2}{3}\frac{a(n^2-1)\gamma_0/\overline{\Delta\omega_0}}{n^2k^2-(k+k_0^{(0)})^2}\right]. \tag{3.8a}$$

4. The processes of reflection and refraction of a plane wave by an interface between two media depend on the properties of the surface and are described by the surface integrals that enter in (3.4) and (3.5). Allowance for the positronic polarization of the medium changes the field that participates in the extinction of the external wave on the surface of the optical medium, and the extinction theorem takes the form

$$\tilde{\mathbf{E}}+N\alpha\frac{1}{k^2(n^2-1)}\operatorname{rot}\operatorname{rot}\mathbf{J}-\frac{2aN\mathbf{J}_0}{3[n^2k^2-(k+k_0)^2]}=0, \tag{4.1}$$

where $\mathbf{J}$ is the surface integral that enters in (3.4) and is cited in Ref. 1,

$$\mathbf{J}_0=i\mathbf{E}_{0t}\int_\Sigma\left\{(k+k_0)\frac{\partial R}{\partial\nu'}-nk\left(\frac{\partial\mathbf{r}'}{\partial\nu'}\mathbf{s}_t\right)\right\}\frac{\exp\{i[(k+k_0)R+kn(\mathbf{r}'\mathbf{s}_t)]\}}{R}dS', \tag{4.2}$$

and $\mathbf{s}_t$ is a unit vector along the direction of propagation of the refractive wave with amplitude $\mathbf{E}_{0t}$. We calculate (4.2) by using the stationary-phase method for the case of normal incidence of the plane wave on the interface. At a sufficiently large distance in the optical medium from the interface, such that $k_0r\gg1$, and at a sufficiently large distance above the surface, the forms of the transmittivity and reflectivity of the interface of the two media, as well as the laws of reflection and refraction, do not differ from those given in Ref. 1. The influence of the electronic polarization of the medium on the refraction and reflection enters via the value of the refractive index n in accordance with (3.8).

5. DISCUSSION OF RESULTS AND CONCLUSIONS

A consistent analysis of the processes of interaction with the radiation field of a system of interacting atoms, including the intermediate states of the atoms with negative energy, leads to the need for taking into account, besides the field of the dipoles (the electronic polarization of the medium) also the positronic polarization of the medium. This means that in a medium each atom "feels" a resultant field consisting of three components. In the present paper we consider the effects of coherent optical radiation incident in the form of a plane monochromatic wave on the interface between two media.

We compare the quantities $\mathbf{E}^{e}_{jl}$ and $\mathbf{E}^{p}_{jl}$ at an arbitrary j-th atom, assuming

$$\mathbf{d}_l=\alpha\tilde{\mathbf{E}}_l=\frac{e^2}{m[(\omega^2-\omega_0^2)-i\omega\gamma_0]}\tilde{\mathbf{E}}_{0l}(\mathbf{r}_l)e^{-i\omega t}. \tag{5.1}$$

At small interatomic distances $r_{jl}\ll\lambda$ there is no retardation in the interaction between the atoms, and

$$|\mathbf{E}_{jl}^{e}|\approx|\mathbf{d}_l|/r_{jl}^3. \tag{5.2}$$

Then the ratio of (5.2) and $|\mathbf{E}^{p}_{jl}|\approx a|\mathbf{E}_l|/r_{jl}$ takes the form

$$\frac{|\mathbf{E}_{jl}^{e}|}{|\mathbf{E}_{jl}^{p}|}=\frac{c^2}{[(\omega^2-\omega_0^2)-i\omega\gamma_0]r_{jl}^2}. \tag{5.3}$$

In the optical frequency band, even far from resonance, this quantity is much larger than unity, i.e., in this case it is meaningless to speak of positronic polarization of the medium. The situation is entirely different at $r_{jl}\gg\lambda$ if the retardation in the interatomic interaction is taken into account. In this case there remain in $\mathbf{E}^{e}_{jl}$ only terms inversely proportional to the first power of r_{jl}. Then

$$|\mathbf{E}_{jl}^{e}|/|\mathbf{E}_{jl}^{p}| =\omega^2/[(\omega^2-\omega_0^2)-i\omega\gamma_0]. \tag{5.4}$$

As seen from (5.4), in the case of resonance ($\omega=\omega_0$) this quantity is much larger than unity, a fact noted by us back in Ref. 2. Far from resonance, the ratio (5.4) is of the order of unity. Thus, in coherent optics, where the main contribution to the polarization of the medium is made by atoms located in the wave zone, the influence of the positronic polarization of the medium may turn out to be of the same order as the electronic polarization.

We analyze now Eq. (3.8), which is a generalization of the Lorentz–Lorenz formula for a gas medium, in which positronic polarization is also taken into account. The frequency dependence of the dielectric constant of such a medium is shown in Fig. 2, where the solid curve corresponds to the known dispersion curve for a gas.[1] The dashed curve corresponds to allowance for the positronic polarization of the medium. The change of the dispersion curve is more substantial the larger $\gamma_0/\overline{\Delta\omega_0}$. In rarefied gases it is apparently possible to have a situation wherein $\gamma_0/\overline{\Delta\omega_0}\sim 1$ and the change of the dielectric constant can be determined experimentally

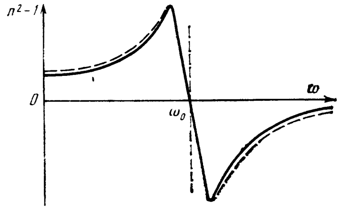

FIG. 2.

from the reflection spectra. In fact, in the case of normal incidence, the reflectivity $\mathcal{R}$ does not depend on the distance r and takes the usual form:

$$\mathcal{R}=\frac{(n-1)^2+\varkappa^2}{(n+1)^2+\varkappa^2}, \tag{5.5}$$

where n and $\varkappa$ are respectively the refractive index and the absorption coefficient of the medium, with account taken of the positronic polarization of the medium in accordance with (3.8). Far from resonance we have $n\gg\varkappa$, and we obtain the dependence of the width of the spectral line with center at ω_0 on the concentration of the atoms and on the reflectivity of the interface (between a vacuum and a rarefied gas) on which a plane monochromatic wave of frequency ω is incident:

$$\overline{\Delta\omega_0}=\frac{8\pi N}{3}\frac{e^2\gamma_0}{m[(\omega+\omega_0)^2-\omega^2]} \times\left\{1+\frac{2\pi Ne^2}{m(\omega_0^2-\omega^2)}\left[1-\left(1+\frac{(1-R)^2}{4R}\right)^{1/2}\right]\right\}^{-1}. \tag{5.6}$$

We estimate now the value of (5.6) at the numerical values of the physical quantities given in Ref. 4, where an experimental investigation was made of the broadening of a number of spectral lines of krypton at various concentrations of the atoms in the region $(0.025-20)\times10^{17}$ atoms/cm^3. Let $\omega_0=2.483\times10^{15}$ rad/sec, $\omega=16.35\times10^{14}$ rad/sec, $N=20\times10^{17}$ atoms/cm^3, and $\mathcal{R}=0.00002\%$. Then $\overline{\Delta\omega_0}/\gamma_0\sim1$. Allowance for the positronic polarization of the medium changes the concentration dependence of the refractive index and the coefficient $\mathcal{R}$ of reflection from the interface. This circumstance can apparently lead to a change in the linear dependence of the width of the spectral line on the concentration, as was the case in the experiments of Ref. 4. For a detailed comparison of the proposed theoretical interpretation of the anomalous broadening of the lines at small concentrations[4] we need data on the dependence of the reflection coefficients $\mathcal{R}$ on the concentration of the atoms. These can be obtained by investigating the reflection of laser radiation from the interface between a vacuum and a rarefied gas.

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