

COHERENCE RELAXATION OF EXCITED ATOMS IN COLLISIONS

M. I. D'YAKONOV and V. I. PEREL'

A. F. Ioffe Physico-technical Institute, Academy of Sciences, U.S.S.R.

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The problem treated is that of the relaxation of the nondiagonal density matrix of excited atoms in a gas owing to collisions with normal atoms. The relaxation term is diagonalized by means of a change to a new representation. The diagonal elements γ_1 and γ_2 have the simple physical meaning of the respective inverse relaxation times of circular and plane polarization. The numerical values of γ_1 and γ_2 are obtained for the most interesting case, that in which the respective total angular momenta of the ground and excited states are zero and unity.

IN a collision of an excited atom with a normal atom of the same element a possible process is resonance transfer of the excitation. For an allowed transition the cross section for such a collision is much larger than the gas-kinetic cross section. Vlasov and Fursov,^[1] studying the effect of this process on the width of the spectral line, showed that the cross section is of the order of $\lambda^2\gamma/\Delta\omega$, where λ is the wavelength of the transition, γ is the natural width of the excited level, and $\Delta\omega$ is the Doppler width of the spectral line. In collisions of this kind there is a redistribution of the populations of the Zeeman sublevels (which can also occur without transfer of the excitation). This manifests itself, for example, in the phenomenon of depolarization of the scattered light owing to collisions. Furthermore, if the excited atom was in a coherent mixture of states (so that the elements of the density matrix that are nondiagonal in the Zeeman sublevels were different from zero), the coherence can be partially destroyed as a result of the collision (relaxation of coherence).

The nondiagonal elements of the density matrix are responsible for a number of resonance effects that appear when light is scattered by a gas in a magnetic field.^[2-5] The line widths of the corresponding resonances are determined at low pressures by the natural lifetime γ^{-1} of the excited state. When the pressure is increased the resonance lines first become narrower owing to capture of radiation,^[6] and then are broadened because of the collisions mentioned above. The collisions must become important when the pressure is so large that the condition $n\lambda^3 \ll 1$ no longer holds, where n is the concentration of normal atoms. Thus there is a need to investigate the relaxation of the nondiagonal density

matrix (the coherence) of an excited atom in collisions with normal atoms.

We have dealt previously^[7] with the problem of the relaxation of coherence owing to the capture of radiation. It was found that the relaxation terms in the equation for the density matrix $f_{mm'}$ are of rather complicated form. It is possible, however, to change to a representation in which there are only two relaxation times γ_1^{-1} and γ_2^{-1} . The first of these is the decay time of circular polarization, and the second is that for plane polarization.

In the present paper it will be shown that there is an analogous situation for the relaxation of coherence owing to collisions. The calculation of the times γ_1^{-1} and γ_2^{-1} is carried to a conclusion for the case in which the total angular momentum of the excited state is $j_1 = 0$ [Eq. (30)]. This case occurs in the scattering of light by vapors of even isotopes of cadmium or mercury.

As is the usual practice in discussing the collisions of heavy particles, we shall treat the translational motion of the atoms classically. Before the collision (at $t = -\infty$) let atom A be excited and have the density matrix $f_{mm'}$, and let atom B be in the normal state with the density matrix $(2j_0 + 1)^{-1} \delta_{\mu\mu'}$.

The indices m and m' number the Zeeman sublevels of the excited state (total angular momentum j_1), and μ and μ' the sublevels of the ground state (total angular momentum j_0). We denote by $f_{mm'}^{(A)}$ the part of the density matrix of atom A that corresponds to its excited state after the collision. We call the corresponding quantity for atom B $f_{mm'}^{(B)}$. It is obvious that the matrices $f_{mm'}^{(A)}$ and $f_{mm'}^{(B)}$ are related linearly to $f_{mm'}$:

$$f_{mm'}^{(A)} = \sum_{m_1 m_1'} A_{mm'}^{m_1 m_1'} f_{m_1 m_1'}, \quad f_{mm'}^{(B)} = \sum_{m_1 m_1'} B_{mm'}^{m_1 m_1'} f_{m_1 m_1'} \quad (1)$$

The coefficients A and B depend on the impact parameter ρ and the relative velocity \mathbf{v} of the atoms (the vectors ρ and \mathbf{v} are of course perpendicular to each other).

The change of the density matrix of the excited atoms in one collision is obviously

$$\Delta f_{mm'} = f_{mm'} - f_{mm'}^{(A)} - f_{mm'}^{(B)}. \quad (2)$$

The increment of the density matrix per unit time, referred to one excited atom, is

$$\frac{\partial f_{mm'}}{\partial t} = -n \int \Delta f_{mm'} \rho d\rho v^3 dv \Phi(v) d\Omega, \quad (3)$$

$$d\Omega = \sin \theta d\theta d\varphi d\psi,$$

where n is the concentration of normal atoms, θ and φ are the polar angles of the vector \mathbf{v} , ψ is the azimuthal angle of the vector ρ in a plane perpendicular to \mathbf{v} , and $\Phi(\mathbf{v})$ is the distribution of relative velocities, normalized to unity (a Maxwell distribution with the mass $M/2$, where M is the mass of an atom). The integration over angles in (3) can be performed in the following way.

In the equations (1) we go over to a system of coordinates connected rigidly to ρ and \mathbf{v} . Then the equations (1) take the form

$$\tilde{f}_{mm'}^{(A)} = \sum_{m_1 m_1'} \tilde{A}_{mm'}^{m_1 m_1'} \tilde{f}_{m_1 m_1'}, \quad \tilde{f}_{mm'}^{(B)} = \sum_{m_1 m_1'} \tilde{B}_{mm'}^{m_1 m_1'} \tilde{f}_{m_1 m_1'}. \quad (4)$$

The quantities A and B are connected with the quantities \tilde{A} and \tilde{B} by the relations

$$A_{mm'}^{m_1 m_1'} = \sum_{r r' s s'} D_{m r}^{* j_1}(\Omega) D_{m' r'}^{j_1}(\Omega) D_{m_1 s}^{j_1}(\Omega) D_{m_1' s'}^{* j_1}(\Omega) \tilde{A}_{r r'}^{s s'},$$

$$B_{mm'}^{m_1 m_1'} = \sum_{r r' s s'} D_{m r}^{* j_1}(\Omega) D_{m' r'}^{j_1}(\Omega) D_{m_1 s}^{j_1}(\Omega) D_{m_1' s'}^{* j_1}(\Omega) \tilde{B}_{r r'}^{s s'}. \quad (5)$$

The matrices \tilde{A} and \tilde{B} are of course independent of the angles Ω . Here $D_{mm'}^{j_1}(\Omega)$ is the matrix for a finite rotation.^[8]

The integral of the four matrices D can be calculated by expanding the product of two D matrices in a Clebsch-Gordan series and using the orthogonality property. After the integration over the angles we get finally

$$\frac{\partial f_{mm'}}{\partial t} = - \sum_{m_1 m_1'} \Gamma_{mm'}^{m_1 m_1'} f_{m_1 m_1'}, \quad (6)$$

$$\Gamma_{mm'}^{m_1 m_1'} = \sum_{r r' s s'} (-1)^{m-m_1+s-r} \sum_{j p p'} (2j+1) \begin{pmatrix} j_1 & j_1 & j \\ m-m' & p & p \end{pmatrix} \begin{pmatrix} j_1 & j_1 & j \\ m_1 & m_1' & p \end{pmatrix} \times \begin{pmatrix} j_1 & j_1 & j \\ r-r' & p' & p \end{pmatrix} \begin{pmatrix} j_1 & j_1 & j \\ s-s' & p' & p \end{pmatrix} L_{r r'}^{s s'}, \quad (7)$$

$$L_{r r'}^{s s'} = 8\pi^2 n \int \Phi(v) v^3 dv \rho d\rho [\delta_{s r} \delta_{s' r'} - \tilde{A}_{r r'}^{s s'} - \tilde{B}_{r r'}^{s s'}]. \quad (8)$$

We expand the density matrix $f_{mm'}$ in terms of irreducible tensor operators^[8,7]:

$$\hat{f} = \sum_{\kappa=0}^{2j_1} \sum_{q=-\kappa}^{\kappa} \hat{T}_{-q}^{\kappa} f_q^{\kappa} (-1)^q. \quad (9)$$

Here the operators \hat{T}_q^{κ} are normalized so that

$$(T_q^{\kappa})_{mm'} = \frac{2\kappa+1}{(2j_1+1)^{1/2}} (-1)^{j_1-m'} \begin{pmatrix} j_1 & \kappa & j_1 \\ -m & q & m' \end{pmatrix}. \quad (10)$$

Furthermore, owing to the orthogonality property of the 3j symbols

$$f_q^{\kappa} = (-1)^q \frac{2j_1+1}{2\kappa+1} \sum_{mm'} (T_{-q}^{\kappa})_{mm'} f_{mm'}, \quad f_0^0 = \text{Sp } \hat{f}. \quad (11)$$

Using Eqs. (6), (7), (11), we can get for the quantities f_q^{κ} the equations

$$\partial f_q^{\kappa} / \partial t = -\gamma_{\kappa} f_q^{\kappa}, \quad (12)$$

$$\gamma_{\kappa} = \sum_p \sum_{r r' s s'} (-1)^{s-r} \begin{pmatrix} j_1 & j_1 & \kappa \\ r & -r' & p \end{pmatrix} \begin{pmatrix} j_1 & j_1 & \kappa \\ s & -s' & p \end{pmatrix} L_{r r'}^{s s'}. \quad (13)$$

It can be seen that as in the case of relaxation owing to the diffusion of radiation^[7] the equations for different f_q^{κ} are separated, and the relaxation times depend only on κ , not on q .

As will be shown below, $\gamma_0 = 0$; this expresses the conservation of the number of excited atoms. The physical meaning of the quantities γ_1 and γ_2 has been made clear in earlier papers,^[7,9] where it was shown that these quantities are the respective inverse relaxation times for circular and plane polarization.

We now turn to the calculation of the quantities $L_{r r'}^{s s'}$ [Eq. (8)]. For this we must prescribe a concrete form of the interaction which leads to the transfer of excitation and to the redistribution over the Zeeman sublevels. If there is an allowed dipole transition between the ground and excited states, the main interaction between the normal and excited atoms is the dipole-dipole interaction

$$\hat{V}(t) = r^{-5} [3(\hat{\mathbf{d}}^A \mathbf{r})(\hat{\mathbf{d}}^B \mathbf{r}) - (\hat{\mathbf{d}}^A \hat{\mathbf{d}}^B) r^2]. \quad (14)$$

Here $\hat{\mathbf{d}}^A$ and $\hat{\mathbf{d}}^B$ are the operators of the dipole moments of atoms A and B, $\mathbf{r} = \rho + \mathbf{v}t$ is the radius vector drawn from one atom to the other, ρ is the impact parameter, and \mathbf{v} is the relative velocity.

Let $\varphi_{m\mu}^A$ be the eigenfunction of the unperturbed Hamiltonian of the two atoms that corresponds to the state in which atom A is in sublevel m of the excited state and atom B in sublevel μ of the ground state. $\varphi_{m\mu}^B$ describes the state in which atom B is excited. The same value ϵ_0 of the total energy of the system corresponds to the two functions $\varphi_{m\mu}^A$ and $\varphi_{m\mu}^B$. We shall regard these functions as chosen in a coordinate system

rigidly connected with the vectors ρ and \mathbf{v} . For definiteness we direct the x axis along \mathbf{v} and the y axis along ρ .

The wave function of the system can be written in the form

$$\Psi = \sum_{m\mu} (a_{m\mu}\varphi_{m\mu}^A + b_{m\mu}\varphi_{m\mu}^B) e^{-ie_0t}$$

We can derive from the Schrödinger equation equations for the coefficients $a_{m\mu}$ and $b_{m\mu}$ ($\hbar = 1$)

$$\begin{aligned} i\dot{a}_{m\mu} &= \sum_{m_1\mu_1} V_{mm_1}^{\mu\mu_1} b_{m_1\mu_1}, & i\dot{b}_{m\mu} &= \sum_{m_1\mu_1} V_{mm_1}^{\mu\mu_1} a_{m_1\mu_1}; \\ V_{mm_1}^{\mu\mu_1} &= \int (\varphi_{m\mu}^A)^* \hat{V} \varphi_{m_1\mu_1}^B d\tau = \int (\varphi_{m\mu}^B)^* \hat{V} \varphi_{m_1\mu_1}^A d\tau. \end{aligned} \quad (15)$$

These equations must be solved with the initial conditions

$$\begin{aligned} b_{m\mu}(-\infty) &= 0, & a_{m\mu}(-\infty) &= a_{m\mu}^0, \\ \langle a_{m\mu}^0 (a_{m'\mu'}^0)^* \rangle &= \tilde{f}_{mm'} (2j_0 + 1)^{-1} \delta_{\mu\mu'}. \end{aligned} \quad (16)$$

It is obvious that

$$\begin{aligned} \tilde{f}_{mm'}^{(A)} &= \sum_{\mu} \langle a_{m\mu}(\infty) a_{m'\mu'}^*(\infty) \rangle, \\ \tilde{f}_{mm'}^{(B)} &= \sum_{\mu} \langle b_{m\mu}(\infty) b_{m'\mu'}^*(\infty) \rangle. \end{aligned} \quad (17)$$

Angle brackets denote averaging over the initial state.

The result of the solution of the equations (15) is to express the coefficients $a_{m\mu}(t)$ and $b_{m\mu}(t)$ linearly in terms of the $a_{m\mu}^0$:

$$a_{m\mu}(t) = \sum_{m_1\mu_1} P_{mm_1}^{\mu\mu_1}(t) a_{m_1\mu_1}^0, \quad b_{m\mu}(t) = \sum_{m_1\mu_1} Q_{mm_1}^{\mu\mu_1}(t) a_{m_1\mu_1}^0. \quad (18)$$

The matrices P and Q satisfy the system of equations

$$i\dot{P}_{mm'}^{\mu\mu'} = \sum_{m_1\mu_1} V_{mm_1}^{\mu\mu_1} Q_{m_1m'}^{\mu_1\mu'}, \quad i\dot{Q}_{mm'}^{\mu\mu'} = \sum_{m_1\mu_1} V_{mm_1}^{\mu\mu_1} P_{m_1m'}^{\mu_1\mu'} \quad (19)$$

with the initial conditions

$$P_{mm'}^{\mu\mu'}(-\infty) = \delta_{mm'} \delta_{\mu\mu'}, \quad Q_{mm'}^{\mu\mu'}(-\infty) = 0.$$

The matrices \tilde{A} and \tilde{B} that appear in the expression (8) can be expressed in terms of these quantities. On the basis of Eqs. (4), (16)–(18) we get

$$\begin{aligned} \tilde{A}_{mm'}^{m_1m_1'} &= (2j_0 + 1)^{-1} \sum_{\mu\mu_1} P_{mm_1}^{\mu\mu_1}(\infty) [P_{m'm_1'}^{\mu\mu_1}(\infty)]^*, \\ \tilde{B}_{mm'}^{m_1m_1'} &= (2j_0 + 1)^{-1} \sum_{\mu\mu_1} Q_{mm_1}^{\mu\mu_1}(\infty) [Q_{m'm_1'}^{\mu\mu_1}(\infty)]^*. \end{aligned} \quad (20)$$

We introduce instead of P and Q more convenient quantities R and S :

$$R_{mm'}^{\mu\mu'} = P_{mm'}^{\mu\mu'} + Q_{mm'}^{\mu\mu'}, \quad S_{mm'}^{\mu\mu'} = P_{mm'}^{\mu\mu'} - Q_{mm'}^{\mu\mu'}. \quad (21)$$

From (19) we get for these coefficients the equations

$$i\dot{R}_{mm'}^{\mu\mu'} = \sum_{m_1\mu_1} V_{mm_1}^{\mu\mu_1} R_{m_1m'}^{\mu_1\mu'}, \quad i\dot{S}_{mm'}^{\mu\mu'} = - \sum_{m_1\mu_1} V_{mm_1}^{\mu\mu_1} S_{m_1m'}^{\mu_1\mu'} \quad (22)$$

with the initial conditions

$$R_{mm'}^{\mu\mu'}(-\infty) = S_{mm'}^{\mu\mu'}(-\infty) = \delta_{mm'} \delta_{\mu\mu'}.$$

By means of (8), (20) and (21) we can transform the expression (13) for γ_K into the form

$$\gamma_K = 8\pi^2 n \int \Phi(v) v^3 dv \rho d\rho \tau_K(\rho, v), \quad (23)$$

$$\begin{aligned} \tau_K(\rho, v) &= 1 - \frac{1}{2(2j_0 + 1)} \sum_{rr's's'} (-1)^{s-r} \sum_p \begin{pmatrix} j_1 & j_1 & \kappa \\ r & -r' & p \end{pmatrix} \\ &\times \begin{pmatrix} j_1 & j_1 & \kappa \\ s & -s' & p \end{pmatrix} \sum_{\mu\mu_1} (R_{rs}^{\mu\mu_1} R_{r's'}^{\mu_1\mu} + S_{rs}^{\mu\mu_1} S_{r's'}^{\mu_1\mu}). \end{aligned} \quad (24)$$

Here and hereafter it is understood that the matrices R and S are taken at $t = \infty$.

Using the properties of 3j symbols, we write τ_K in a form more convenient for practical calculations:

$$\begin{aligned} \tau_K &= \frac{1}{2} \sum_j \left[1 - (-1)^{j+\kappa+2j_1} (2j_1 + 1) \begin{Bmatrix} j_1 & j_1 & \kappa \\ j_1 & j_1 & j \end{Bmatrix} \right] \\ &\times \sum_{\mu\mu_1 m} [|M_{jm}^{\mu\mu_1}|^2 + |N_{jm}^{\mu\mu_1}|^2], \end{aligned} \quad (25)$$

$$\begin{aligned} M_{jm}^{\mu\mu_1} &= \left[\frac{2j+1}{(2j_1+1)(2j_0+1)} \right]^{1/2} \\ &\times \sum_{rs} (-1)^{j_1-r} \begin{pmatrix} j_1 & j_1 & j \\ r & -s & m \end{pmatrix} R_{rs}^{\mu\mu_1}. \end{aligned} \quad (26)$$

The matrix N is expressed in an analogous way in terms of the matrix S . The quantity in curly brackets in Eq. (25) is a Wigner 6j symbol. In writing Eq. (25) we have used the fact that

$$\sum_{\substack{jm \\ \mu\mu_1}} |M_{jm}^{\mu\mu_1}|^2 = (2j_0 + 1)^{-1} (2j_1 + 1)^{-1} \sum_{\substack{mm' \\ \mu\mu_1}} |R_{mm'}^{\mu\mu_1}|^2 = 1; \quad (27)$$

there is a precisely similar relation for the matrices N and S . Equation (27) follows from (22) when we use the Hermitian character of the matrix V and the initial conditions for the matrix R . It follows from (25) and the properties of 6j symbols that $\tau_0 = 0$.

We now note that for the dipole-dipole interaction (14) the quantities R and S that satisfy the equations (22), and consequently also the τ_K , depend on the impact parameter ρ , the relative velocity v , the wavelength λ of the transition, and

the lifetime γ of the excited level only through the single parameter α :

$$\alpha = (\lambda/2\pi)^3 \gamma / \rho^2 v.$$

This can easily be seen if we use the fact that

$$\gamma = \frac{4}{3} \left(\frac{2\pi}{\lambda} \right)^3 \sum_{\mu} |d_{m\mu}|^2,$$

and go over in (22) to the dimensionless variable $x = vt/\rho$.

Introducing in (23) the new variable α instead of ρ , and carrying out the integration over the velocity, we get the final expression for γ_K :

$$\gamma_K = \pi n \left(\frac{\lambda}{2\pi} \right)^3 \gamma \int_0^{\infty} \frac{d\alpha}{\alpha^2} \tau_K(\alpha). \quad (28)$$

The form of the function $\tau_K(\alpha)$, and consequently also the numerical value of the integral in (28), depends only on j_1 and j_0 . We have carried out the numerical integration of the system (22) for the case of greatest practical importance, $j_1 = 1$, $j_0 = 0$ (the resonance lines of mercury, cadmium, and so on). For this case the equations (22) can be put in the form

$$\begin{aligned} i \frac{dC_{1m}}{d\theta} &= C_{1m} + \frac{9}{8} \alpha \sin \theta (C_{1m} + C_{-1m}); \\ i \frac{dC_{0m}}{d\theta} &= 0, \quad i \frac{dC_{-1m}}{d\theta} = -C_{-1m} + \frac{9}{8} \alpha \sin \theta (C_{1m} + C_{-1m}), \\ C_{m'm}(\alpha, 0) &= \delta_{m'm}. \end{aligned} \quad (29)$$

Here a new variable and new unknown functions have been introduced according to the formulas

$$\begin{aligned} \cos \theta &= -vt / (\rho^2 + v^2 t^2)^{1/2}, \\ S_{m'm}^{00}(-\alpha, \theta) &= R_{m'm}^{00}(\alpha, \theta) \end{aligned}$$

$$= C_{m'm}(\alpha, \theta) \exp \left\{ im'\theta + \frac{3}{4} i\alpha(1 - \cos \theta) \right\}.$$

The integrals that appear in (28) can be expressed in terms of the coefficients $C_{m'm}(\alpha, \pi)$ in the following way:

$$\begin{aligned} \int_0^{\infty} \frac{d\alpha}{\alpha^2} \tau_1(\alpha) &= \frac{1}{12} \int_{-\infty}^{\infty} \frac{d\alpha}{\alpha^2} \{ 9 - |C_{11} + C_{-1,-1} - 1|^2 \\ &\quad - |C_{11} - C_{-1,-1}|^2 \}, \\ \int_0^{\infty} \frac{d\alpha}{\alpha^2} \tau_2(\alpha) &= \frac{1}{20} \int_{-\infty}^{\infty} \frac{d\alpha}{\alpha^2} \{ 9 - |C_{11} + C_{-1,-1} - 1|^2 \\ &\quad + |C_{11} - C_{-1,-1}|^2 \}, \end{aligned}$$

Here all of the $C_{m'm}$ are taken at the value $\theta = \pi$. Numerical integration of (29) and numerical

calculation of the integrals gives for γ_1 and γ_2 the values

$$\gamma_1 = 0.035 n \lambda^3 \gamma, \quad \gamma_2 = 0.028 n \lambda^3 \gamma. \quad (30)$$

We recall that these values are for the case $j_1 = 1$, $j_0 = 0$.

In resonance phenomena occurring in the scattering of plane-polarized or unpolarized light (parametric resonance, and so on), at large pressures the width of the resonance curve will be determined by the quantity γ_2 . When there is capture of radiation the total inverse relaxation time, which determines the width of the resonance line, is composed of the quantities given by (30) and of the natural line width, diminished on account of diffusion of radiation.^[7] When there is complete imprisonment (for $j_1 = 1$, $j_0 = 0$)

$$\gamma_1 = \gamma^{1/2} + 0.035 n \lambda^3, \quad \gamma_2 = \gamma^{3/10} + 0.028 n \lambda^3.$$

Let us point out the limits of applicability of the treatment given here. In order to regard the interaction as binary collisions, it is necessary that the cross section be smaller than the square of the mean distance between atoms; that is, $n\lambda^3(\gamma/\Delta\omega)^{3/2} \ll 1$ ($\Delta\omega$ is the Doppler width of the line). The treatment has actually been carried out for zero magnetic field. It remains valid, however, in fields in which the Zeeman splitting is small in comparison with the inverse of the effective time of flight, i.e., the quantity $\Delta\omega(\Delta\omega/\gamma)^{1/2}$. Clearly this condition is satisfied under the usual experimental conditions, in which the Zeeman splitting is smaller than the Doppler width of the line.

The collision broadening of double-resonance lines has been calculated recently by Byron and Foley.^[10] Their calculations contain errors, however. They regarded the density matrix as diagonal. This would be justified if the relaxation of coherence occurred more rapidly than the equalization of the populations in the Zeeman sublevels. As can be seen from our treatment, these two processes go with the same speed. Besides this, Eq. (9) of the paper of Byron and Foley is erroneous, since in it the chronological product of noncommuting operators is dealt with as if they commuted. Despite these errors, their result is only 15 percent larger than the exact value of γ_2 [Eq. (30)]. For the most interesting case, that of mercury, there are no reliable experimental data. The recently measured collision broadening of the double resonance lines^[11] of cadmium is about 2.5 times the value of γ_2 given by Eq. (30). Byron and Foley have pointed out that for cadmium and zinc, for which the intercombination transitions

are rather strongly forbidden, interaction through intermediate states may be of importance. Their calculation of the corresponding cross section is also affected, however, by the same mistakes as mentioned above.

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