

Spectral shape of a signal in light-induced diffusive pulling (pushing) of particles into a light beam

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Abstract. We study theoretically how the dependences of transport collision frequencies ν_i , collision broadening γ and collision shift Δ of the levels on the velocity v of resonant particles influence light-induced diffusive pulling (pushing) (LDP) effects in the framework of a generalised model of strong collisions in the case of velocity-dependent collision rates (so-called kangaroo model). It is found that allowance for the dependences $\nu_i(v)$, $\gamma(v)$ and $\Delta(v)$ does not change the spectral shape of an LDP signal. In particular, in the case of low-intensity radiation, the spectral dependence of the LDP signal coincides with the absorption line shape. It is shown that the magnitude of the LDP effect is proportional to the difference between the diffusion coefficients of particles in the excited and ground states. It is found that the spectral anomalies previously predicted in the LDP effect [Gel'mukhanov F.Kh. *JETP Lett.*, 55, 214 (1992)] for an idealised model of the Lorentz gas (the limiting case of heavy buffer particles), which arise due to the dependences $\nu_i(v)$, $\gamma(v)$ and $\Delta(v)$, are typical only for this gas. At a realistic ratio of the masses of absorbing and buffer particles, spectral anomalies do not occur in the LDP effect.

Keywords: kinetic equations, collisions, transport rate, impact width, collision drift.

1. Introduction

The effect of light-induced diffusive pulling (pushing) (LDP) [1, 2] refers to the strongest gas-kinetic effects in a laser field and to date is well studied both theoretically and experimentally [2–10]. The essence of the effect is that particles, absorbing resonantly radiation and being in a mixture with a buffer gas, can be pulled into or pushed out of a light beam. The LDP effect is caused by the spatial inhomogeneity of concentrations (due to the spatial inhomogeneity of the radiation intensity in the direction transverse to the direction of the light beam) and the difference in the diffusion coefficients of excited and unexcited particles of an absorbing gas. With this effect, the diffusion coefficients of Li, Na, K and Rb atoms (in the ground and excited states) were measured in various buffer gases [5, 6, 8–10].

In the case of the LDP effect, the spatial distribution of a light-induced nonequilibrium increment ΔN to the concentration of absorbing particles in the light beam ‘keeps track of’ the local radiation intensity. In LDP theory, it is usually assumed that the collision broadening $\gamma(v)$ and collision shift $\Delta(v)$ of the levels do not depend on the velocity v of the resonant particles:

$$\gamma(v) = \gamma_0 = \text{const}, \quad \Delta(v) = \Delta_0 = \text{const}. \quad (1)$$

In this case, the spectral dependence of the LDP signal (ΔN) at a low radiation intensity coincides with the absorption line shape [2–5, 7].

Theoretical studies [11, 12] for the Lorentz gas (the limiting case of heavy buffer particles: $M \ll M_b$, where M and M_b are the masses of resonant and buffer particles) have shown that taking into account the dependences $\gamma(v)$ and $\Delta(v)$ can significantly alter the spectral profile of the LDP signal right up to the appearance of a deep dip near the centre of the absorption line or even a sign-alternating dependence on the frequency detuning Ω of radiation. The strong influence of dependences $\gamma(v)$ and $\Delta(v)$ on the shape of the LDP signal may occur when the difference between the transport collision frequencies of the resonant particles in the ground and excited states with buffer particles, $\Delta\nu(v)$, changes its sign as a function of v [11, 12].

The sign-alternating dependence $\Delta\nu(v)$ is not at all uncommon and can be observed for a variety of atoms and molecules. It is well known that this dependence results in the appearance of so-called anomalous light-induced drift (LID), experimentally measured for atoms [13] and molecules [14–21]. Therefore, the spectral anomalies in the LDP effect predicted in papers [11, 12] can actually arise under the same conditions in which anomalous LID is observed.

Apart from papers [11, 12], no other studies of the influence of dependences $\gamma(v)$, $\Delta(v)$ and $\Delta\nu(v)$ on the spectral shape of the LDP signal have been carried out yet. A natural question arises about whether spectral anomalies in the LDP effect are possible for the general case of an arbitrary ratio of masses of buffer and absorbing particles or they are specific to the Lorentz gas. In this regard, it is interesting to consider the LDP effect in the framework of a universal collisional model that would describe the possible impact of the velocity dependence of the collision rates on the LDP effect for any interaction potential of the colliding particles and any ratio of their masses. We believe that as such, use can be made of the so-called kangaroo model [22–24], which is a generalisation of the model of strong collisions to the case of velocity-dependent collision rates.

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The aim of this work is to study, within the framework of the kangaroo model, the influence of the velocity dependence of the collision rate on the LDP effect.

2. General relations

Consider the interaction of a travelling monochromatic electromagnetic wave

$$\mathcal{E} = [E \exp(i\mathbf{k}\mathbf{r} - i\omega t) + \text{c. c.}] / 2$$

with two-level absorbing particles in a mixture with buffer particles. We neglect collisions between absorbing particles, assuming the concentration N_b of the buffer gas to be much higher than that of the absorbing gas, N . Interaction of particles with radiation in stationary and spatially inhomogeneous conditions is described by the equations for the density matrix [7, 25]:

$$\begin{aligned} (\mathbf{v}\nabla + \Gamma_m)\rho_m(\mathbf{v}) &= S_m(\mathbf{v}) + NP(\mathbf{v}), \\ \mathbf{v}\nabla\rho_n(\mathbf{v}) &= S_n(\mathbf{v}) + \Gamma_m\rho_m(\mathbf{v}) - NP(\mathbf{v}), \\ \{\mathbf{v}\nabla + \Gamma(v) - i[\Omega(v) - \mathbf{k}\mathbf{v}]\}\rho_{mn}(\mathbf{v}) &= iG[\rho_n(\mathbf{v}) - \rho_m(\mathbf{v})], \end{aligned} \quad (2)$$

where

$$\begin{aligned} NP(\mathbf{v}) &= -2\text{Re}[iG^*\rho_{mn}(\mathbf{v})]; \quad G = \frac{Ed_{mm}}{2\hbar}; \\ \Gamma(v) &= \frac{\Gamma_m}{2} + \gamma(v); \quad \Omega(v) = \omega - \omega_{mn} - \Delta(v); \end{aligned} \quad (3)$$

$\rho_n(\mathbf{v})$ and $\rho_m(\mathbf{v})$ are the particle velocity distributions in the ground (n) and excited (m) states; $N = N_m + N_n$ is the concentration of absorbing particles ($N_i = \int \rho_i(\mathbf{v}) d\mathbf{v}$, $i = m, n$); $S_m(\mathbf{v})$ and $S_n(\mathbf{v})$ are the collision integrals; d_{mm} is the matrix element of the dipole moment of the transition $m-n$; ω and \mathbf{k} are the frequency and the wave vector of radiation; ω_{mn} is the frequency of the transition $m-n$; Γ_m is the rate of spontaneous relaxation of the excited state m ; and $P(\mathbf{v})$ is the probability of radiation absorption by a particle with a specified velocity \mathbf{v} per unit time.

We take into account the fact that in the LDP experiments [5, 6, 8–10], the characteristic size of the spatial inhomogeneity (light beam radius a) is significantly greater than the mean free path of particles, l :

$$a \gg l. \quad (4)$$

When condition (4) is met, in the last equation in (2) one can neglect the term with the spatial derivative. To this end, for the absorption probability $P(\mathbf{v})$ (3) we find from the last equation in (2)

$$\begin{aligned} NP(\mathbf{v}) &= 2|G|^2 Y(\mathbf{v})[\rho_n(\mathbf{v}) - \rho_m(\mathbf{v})], \\ Y(\mathbf{v}) &= \frac{\Gamma(v)}{\Gamma^2(v) + [\Omega(v) - \mathbf{k}\mathbf{v}]^2}. \end{aligned} \quad (5)$$

Note also that in the LDP experiments, the homogeneous half-width of the absorption line $\Gamma(v)$ is large compared with the Doppler broadening kv_T , which allows us to make use of the homogeneous broadening approximation:

$$\Gamma(v) \gg kv_T, \quad (6)$$

where $v_T = (2k_B T/M)^{1/2}$ is the most probable velocity of absorbing particles; T is the temperature; and k_B is the Boltzmann constant.

When condition (6) is met, the velocity distribution of the absorbing particle in each state differs little from the Maxwellian distribution. This allows one to obtain from kinetic equations (2) closed equations of hydrodynamics. In the case of elastic collisions, for diagonal collision integrals equation (2) contains the relations ($i = m, n$) [7, 26]:

$$\begin{aligned} \int S_i(\mathbf{v}) d\mathbf{v} &= 0, \\ \int \mathbf{v} S_i(\mathbf{v}) d\mathbf{v} &= -v_i^{\text{tr}} \mathbf{j}_i, \quad \mathbf{j}_i = \int \mathbf{v} \rho_i(\mathbf{v}) d\mathbf{v}. \end{aligned} \quad (7)$$

Here, \mathbf{j}_i is the particle flux density in the state i ; v_i^{tr} is the mean transport collision rate of absorbing particles with buffer ones. Collision rates v_i^{tr} can be considered independent of the coordinates, since the concentration of the absorbing gas, N , is assumed to be significantly lower than the concentration of the buffer gas, N_b .

In the general case, each of the flows \mathbf{j}_i has two components: a transverse component with respect to the wave vector \mathbf{k} associated with the concentration gradient, and a longitudinal component arising due to the velocity-selective excitation. The presence of the longitudinal components of the flow leads to the LDP effect [7, 26]. Not to introduce new notations, by flows \mathbf{j}_i are meant the transverse components and we consider the problem in the plane orthogonal to the vector \mathbf{k} .

Integrating the first equation in (2) in velocity, with allowance for (7) we obtain

$$\Gamma_m N_m + \text{div} \mathbf{j}_m = NP, \quad (8)$$

where $P = \int P(\mathbf{v}) d\mathbf{v}$ is the velocity integrated probability of radiation absorption, which determines the absorption line contour. Multiplying equation (2) by \mathbf{v} and integrating in velocity, we obtain the equations for the flows:

$$\begin{aligned} (v_m^{\text{tr}} + \Gamma_m) \mathbf{j}_m + \frac{v_T^2}{2} \nabla N_m &= 0, \\ v_n^{\text{tr}} \mathbf{j}_n + \frac{v_T^2}{2} \nabla N_n &= \Gamma_m \mathbf{j}_m. \end{aligned} \quad (9)$$

When conditions

$$v_m^{\text{tr}}, v_n^{\text{tr}} \gg \Gamma_m \quad (10)$$

are satisfied, we find from (9) the relation between a partial flow and a gradient of the corresponding population, similar to the ordinary relationship in the theory of diffusion:

$$\mathbf{j}_i = -\frac{v_T^2}{2v_i^{\text{tr}}} \nabla N_i = -D_i \nabla N_i, \quad (11)$$

where $D_i = v_T^2/(2v_i^{\text{tr}})$ is the diffusion coefficient of the absorbing particles in the state i . Under steady-state conditions, the total diffusion flux of particles across the light beam is equal to zero:

$$\mathbf{j}_m + \mathbf{j}_n = 0. \quad (12)$$

After substituting (11) into (12), we obtain the equation that establishes a relationship between the concentration of particles in the ground and excited states:

$$\frac{\nabla N_m}{v_m^{\text{tr}}} + \frac{\nabla N_n}{v_n^{\text{tr}}} = 0. \quad (13)$$

This relation is of differential nature where spatial inhomogeneity manifests itself.

A necessary additional relation between the concentrations N_m , N_n and the integral absorption probability P is found using equation (8). Let the condition

$$\Gamma_m \gg D_m/a^2 \quad (14)$$

be met. Inequality (14) means that the distance, over which an atom diffuses into the excited state $1/\Gamma_m$ during the lifetime, is significantly less than the light beam radius a . In condition (14) the second term of equation (8) can be neglected; as a result, taking into account the normalisation condition ($N = N_m + N_n$) we have from (8)

$$N_m = \frac{NP}{\Gamma_m}; \quad N_n = N - N_m = N \left(1 - \frac{P}{\Gamma_m}\right). \quad (15)$$

After substituting (15) into (13), we obtain a differential equation for the concentration of absorbing particles, N :

$$\frac{\nabla N}{N} \left(1 + \frac{v_n^{\text{tr}} - v_m^{\text{tr}}}{v_m^{\text{tr}}} \frac{P}{\Gamma_m}\right) = \frac{v_m^{\text{tr}} - v_n^{\text{tr}}}{v_m^{\text{tr}}} \frac{\nabla P}{\Gamma_m}. \quad (16)$$

Equation (16) has the solution:

$$\frac{N - N_0}{N_0} = \frac{v_m^{\text{tr}} - v_n^{\text{tr}}}{v_m^{\text{tr}}} \frac{P}{\Gamma_m} \left(1 + \frac{v_n^{\text{tr}} - v_m^{\text{tr}}}{v_m^{\text{tr}}} \frac{P}{\Gamma_m}\right)^{-1}, \quad (17)$$

where N_0 is the concentration of absorbing particles outside the light beam. The concentration is spatially inhomogeneous due to the spatial inhomogeneity of absorption probability of radiation (radiation intensity) in the transverse direction of the light beam. Due to condition (14) the concentration of absorbing particles is determined by the local value of the radiation intensity at a given point of the light beam.

As can be seen from formula (17), if $v_m^{\text{tr}} \neq v_n^{\text{tr}}$ and $P \neq 0$, then the change in the concentration $\Delta N = N - N_0$ is nonzero. When $v_m^{\text{tr}} > v_n^{\text{tr}}$, absorbing particles are pulled into the light beam ($N > N_0$), but when $v_m^{\text{tr}} < v_n^{\text{tr}}$, they are pushed out of it ($N < N_0$). With increasing radiation intensity (with increasing P) the concentration gradient increases. At a high intensity the absorption probability P tends to $\Gamma_m/2$ (see, e.g., [7]), and in this case, the LDP effect is maximal: $(N - N_0)/N_0 = (v_m^{\text{tr}} - v_n^{\text{tr}})/(v_m^{\text{tr}} + v_n^{\text{tr}})$. Here, if $v_m^{\text{tr}} \gg v_n^{\text{tr}}$, the concentration in the beam is doubled. In the opposite limiting case ($v_m^{\text{tr}} \ll v_n^{\text{tr}}$), absorbing particles are almost completely pushed out of the beam: $N/N_0 = 2v_m^{\text{tr}}/v_n^{\text{tr}}$.

In the case of low intensity, such that

$$P/\Gamma_m \ll 1, \quad (18)$$

formula (17) takes the simple form:

$$\frac{N - N_0}{N_0} = \frac{v_m^{\text{tr}} - v_n^{\text{tr}}}{v_m^{\text{tr}}} \frac{P}{\Gamma_m}. \quad (19)$$

If the mean transport collision rates v_i^{tr} of absorbing particles with buffer particles do not depend on the frequency detuning Ω of radiation, the spectral dependence of the LDP signal $\Delta N \equiv N - N_0$ coincides with the shape of the absorption line, determined by the radiation absorption probability P . If the rates v_i^{tr} depend on the detuning Ω , then spectral anomalies are possible in the LDP effect.

3. Collision kangaroo model

Let us find the mean transport collision rate v_i^{tr} of absorbing particles with buffer ones, defined by (7). For the collision integrals $S_i(\mathbf{v})$ we will use the collision kangaroo model [22–24]:

$$S_i(\mathbf{v}) = -v_i(\mathbf{v})\rho_i(\mathbf{v}) + \frac{\langle v_i(\mathbf{v})\rho_i(\mathbf{v}) \rangle}{\langle v_i(\mathbf{v})W(\mathbf{v}) \rangle} v_i(\mathbf{v})W(\mathbf{v}), \quad (20)$$

where $v_i(\mathbf{v})$ is the transport collision rate; $W(\mathbf{v})$ is the Maxwell distribution; and the angle brackets denote velocity integration. The kangaroo model (20) is a generalisation of the model of strong collisions to the case of velocity-dependent collision rates.

For mean transport collision rates v_i^{tr} , from (7) with allowance for (20) we obtain the expression:

$$v_i^{\text{tr}} = \frac{\langle (\mathbf{v}\mathbf{j}_i)v_i(\mathbf{v})\rho_i(\mathbf{v}) \rangle}{\langle (\mathbf{v}\mathbf{j}_i)\rho_i(\mathbf{v}) \rangle}. \quad (21)$$

Here, $\rho_i(\mathbf{v})$ is velocity distribution of particles in the state i in terms of spatial inhomogeneity that is characteristic of the LDP effect. It is quite simple to find an expression for $\rho_i(\mathbf{v})$. Let $\rho_i^0(\mathbf{v})$ be the solution to the kinetic equation (2) under spatially homogeneous conditions. Because of the presence of the diffusion flux of particles \mathbf{j}_i across the light beam, the distribution $\rho_i(\mathbf{v})$ will almost not differ from the distribution $\rho_i^0(\mathbf{v})$. This difference is most easily taken into account by the approximate Grad method, widely used for the solution of the kinetic equations [27, 28]. According to this method, the dependence $\rho_i(\mathbf{v})$ is given by the sum of the distribution $\rho_i^0(\mathbf{v})$ and antisymmetric increment:

$$\rho_i(\mathbf{v}) = \rho_i^0(\mathbf{v}) \left(1 + \frac{2}{v_T^2} \frac{\mathbf{v}\mathbf{j}_i}{N_i}\right). \quad (22)$$

Next, to find the velocity distribution of populations, $\rho_i^0(\mathbf{v})$, we limit our consideration to the condition of the low radiation intensity (18), assuming that the fraction of particles in the excited state m is negligible ($N_m \ll N$), and the velocity distribution of populations in the ground state n is close to Maxwellian. Expression (5) for the absorption probability $P(\mathbf{v})$ takes the simplest form:

$$P(\mathbf{v}) = 2|G|^2 Y(\mathbf{v})W(\mathbf{v}). \quad (23)$$

Under spatially homogeneous conditions, from kinetic equations (2) with allowance for expressions (20) and (23) we find the velocity distribution of populations, which can be conveniently represented as

$$\frac{\rho_m^0(\mathbf{v})}{N} = \tau_{1m}(\mathbf{v})P(\mathbf{v}) + \langle \tau_{2m}(\mathbf{v})P(\mathbf{v}) \rangle \frac{\tau_{2m}(\mathbf{v})W(\mathbf{v})}{\langle \tau_{2m}(\mathbf{v})W(\mathbf{v}) \rangle},$$

$$\begin{aligned} \frac{\rho_n^0(\mathbf{v})}{N} = & W(\mathbf{v}) - \tau_{1n}(\mathbf{v})P(\mathbf{v}) \\ & - \left\{ \frac{P}{\Gamma_m} - \langle \tau_{1n}(\mathbf{v})P(\mathbf{v}) \rangle + \frac{\langle \tau_{2m}(\mathbf{v})P(\mathbf{v}) \rangle}{\langle \tau_{2m}(\mathbf{v})W(\mathbf{v}) \rangle} \right. \\ & \left. \times [\langle \tau_{1n}(\mathbf{v})W(\mathbf{v}) \rangle - \tau_{1n}(\mathbf{v})] \right\} W(\mathbf{v}), \end{aligned} \quad (24)$$

where

$$\begin{aligned} \tau_{1m}(\mathbf{v}) = & \frac{1}{\Gamma_m + v_m(\mathbf{v})}; \quad \tau_{2m}(\mathbf{v}) = \frac{v_m(\mathbf{v})}{\Gamma_m[\Gamma_m + v_m(\mathbf{v})]}, \\ \tau_{1n}(\mathbf{v}) = & \frac{v_m(\mathbf{v})}{v_n(\mathbf{v})}\tau_{1m}(\mathbf{v}). \end{aligned} \quad (25)$$

From (24) it follows that the velocity distributions of populations, $\rho_m^0(\mathbf{v})$ and $\rho_n^0(\mathbf{v})$, represent the sum of the anisotropic parts directly induced by radiation [function $P(\mathbf{v})$] and isotropic parts generated by the arrival term in the collision integral (20).

In the limit of large transport collision rates (these are typical conditions in LDP experiments),

$$v_m(\mathbf{v}) \gg \Gamma_m, \quad (26)$$

and at low intensity (18), the velocity distributions of populations (24) contain only the isotropic parts and coincide with the Maxwell distribution:

$$\rho_m^0(\mathbf{v}) = \frac{P}{\Gamma_m}NW(\mathbf{v}), \quad \rho_n^0(\mathbf{v}) = NW(\mathbf{v}). \quad (27)$$

As a result, for mean transport collision rates, from relation (21) with formulas (22) and (27) taken into account we find the expression:

$$v_i^{\text{tr}} = \frac{8}{3\sqrt{\pi}} \frac{1}{v_i^{\text{tr}}} \int_0^\infty v^4 \exp\left(-\frac{v^2}{v_i^{\text{tr}}}\right) v_i(v) dv. \quad (28)$$

As can be seen, mean transport collision rates v_i^{tr} of absorbing particles with buffer particles, appearing in LDP theory, do not depend on the frequency detuning Ω of radiation. This means, according to (19) that the spectral dependence of the LDP signal ΔN at a low radiation intensity always coincides with the shape of the absorption line, determined by the radiation absorption probability P .

Thus, the spectral anomalies in the LDP effect predicted by theoretical studies [11, 12] for an idealised model of the Lorentz gas (the limiting case of $M \ll M_b$) are typical only for this gas. At a realistic ratio of the masses of absorbing and buffer particles the spectral anomalies do not arise in the LDP effect.

The reason behind the appearance of spectral anomalies in the LDP effect when it is considered in the Lorentz gas model is as follows. In the case of heavy buffer particles, the condition $M \ll M_b$ allows one to distinguish between two scales of the collision relaxation rate: the direction $[v_i(v)]$ and the magnitude $[v_i(v)M/M_b]$ of the velocity v of resonant particles. Given that

$$\Gamma_m M_b / M \gg v_m(\mathbf{v}) \gg \Gamma_m, \quad (29)$$

which can only be fulfilled in the Lorentz gas (this condition was used in calculations of Refs [11, 12]), the atom during its lifetime in the excited state $1/\Gamma_m$ does not change the modulus

of its velocity $v = |v|$. Therefore, the non-equilibrium of the velocity distribution of particles, directly induced by radiation [function $P(\mathbf{v})$], is effectively transferred to the orthogonal direction of the wave vector \mathbf{k} . These non-equilibrium structures in the distributions of populations in velocity modulus v at the levels m and n determine the dependence of the mean of transport collision rates v_i^{tr} on the frequency detuning Ω of the radiation, which leads to the appearance of the spectral anomalies in LDP effect for the Lorentz gas.

For the non-Lorentz gas, the effect of collisional transfer of non-equilibrium in the velocity distribution of resonant particles in a laser field is weak and can be neglected [24, 29, 30]. In view of this, the mean transport collision rates v_i^{tr} do not depend on the frequency detuning Ω of radiation, and therefore the spectral anomalies in the LDP effect do not arise.

4. Conclusions

The analysis conducted in the present study has shown that the mean transport collision rates v_i^{tr} of absorbing particles with buffer particles appearing in the LDP theory do not depend on the frequency detuning Ω of radiation. This means in particular that the spectral dependence of the LDP signal $\Delta N \equiv N - N_0$ at a low radiation intensity always coincides with the absorption line shape. Spectral anomalies in the LDP effect predicted in [11, 12] are only possible in an idealised model of the Lorentz gas (the limiting case of heavy buffer particles: $M \ll M_b$).

We note the following fact. The magnitudes of the LDP effect and of the so-called normal LDP effect are proportional to the factor $\Delta v/v \equiv (v_m^{\text{tr}} - v_n^{\text{tr}})/v_n^{\text{tr}}$, which is equal to a relative change in the mean transport collision rates of absorbing particles in the excited and ground states with buffer particles. A comparison of the theory with the experimental results on the LID or LDP allows one to define the parameter $\Delta v/v$. This fact forms the basis of effective, independent methods for measuring the diffusion coefficient of the particles in short-lived excited states (with the help of LDP [5, 6, 8–10] and LID effects [7, 31–33]). For a number of atoms and molecules the so-called anomalous LID was experimentally observed [13–21]. In the case of the anomalous LID, the magnitude of the effect is not proportional to the factor $\Delta v/v$, and thus, it cannot be used to measure the diffusion coefficients of the particles in the short-lived excited states. We have shown that the magnitude of the LDP effect, as opposed to that of the LID, is always proportional to the factor $\Delta v/v$. Therefore, the measurement of the diffusion coefficients of excited particles by using the LDP effect as an independent method is a good addition to the method based on the LID effect.

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