Resonance charge exchange in high density gases

R. S. Vitlina and A. M. Dykhne
Institute of Semiconductor Physics, Siberian Division, USSR Academy of Sciences
(Submitted June 21, 1972)
Zh. Eksp. Teor. Fiz. **64**, 510-514 (February 1973)

It is demonstrated that even at sufficiently low densities, when the formally calculated gas parameter nf^3 is small, the medium may affect pair resonance collisions appreciably. The effect of the medium is considered in detail for the case of resonance charge exchange.

1. INTRODUCTION

As is well known, a gas can be regarded as rarefied with respect to some process if $\beta = nf^3 \ll 1$, where f is the amplitude of this process. In connection with various applications, researchers recently have become interested in relatively dense gases, where β is not too small or is of the order of unity. An important role is then assumed by collective effects in the gas, particularly ternary collisions.

We wish to call attention to the fact that multiparticle effects can appear in a gas at much lower densities ($\beta \ll 1$), if the processes under consideration have a resonant character. We illustrate such an effect in this paper, using the influence of the medium on resonant charge exchange in a gas as an example.

2. QUALITATIVE CONSIDERATION

Our remark reduces qualitatively to the following. In order for the charge-exchange process to be resonant, it is necessary that the electronic levels of both atomic particles participating in the collision process coincide. A third particle, even if its distance $n^{-1/3}$ from the two colliding particles greatly exceeds the charge-exchange amplitude, can upset the resonance condition and by the same token alter significantly the charge-exchange cross section in pair collision. The cross section is effectively dependent on the density of the scatterers, and although the gas is rarefied ($\beta \ll 1$), the inverse proportionality of the mean free path to the gas density gives way to another more complicated dependence.

We calculate the charge-exchange cross section in two stages. We first calculate the charge-exchange probability in the presence of a given deviation from resonance. The cross section obtained in this manner is then averaged, for which purpose it is necessary to calculate the distribution function of the deviations from resonance.

3. CHARGE-EXCHANGE PROBABILITY IN THE PRESENCE OF WEAK DEVIATION FROM RESONANCE

In the case of resonance charge exchange, as shown by Firsov[1], there exists a characteristic impact distance $\rho_0 \sim a\ln(e^2/\hbar v)$ (a is the atomic dimension and v is the relative collision velocity), such that the charge-exchange probability is

$$W = {}^1/_2 \text{ if } \rho \leqslant \rho_0, \quad W \ll 1 \text{ if } \rho > \rho_0.$$

The influence of the deviation from resonance on the probability of the resonance charge exchange was investigated by Demkov[2]. It was shown that the transition occurs effectively in relatively small vicinities of two critical points, one of which occurs as the colliding particles come closer together, and the other as they move apart. The deviation (according to Demkov) leads to the appearance of an additional factor (smaller than unity) in the expression for the charge-exchange probability, $\cosh^{-2}(\pi\Delta/2\gamma v_{cr})$, where $\gamma^2/2 \equiv E$ is the energy of the electronic level, v_{cr} is the radial component of the relative velocity at the critical point, and Δ is the detuning.

In the case of interest to us, however, the situation is somewhat different from that considered by Demkov[2]. Since the deviation from resonance is now produced by factors that are external with respect to the colliding particles, it can be different at the two critical points r_{cr1} and r_{cr2}. A simple generalization of Demkov's calculations to this case leads to the result

$$d\sigma = 2\pi\rho\, d\rho W = \pi\rho\, d\rho \left(1 - \operatorname{th}\frac{\pi|\Delta_1|}{2\gamma v_{\text{кр}1}} \operatorname{th}\frac{\pi|\Delta_2|}{2\gamma v_{\text{кр}2}}\right) \qquad (1)$$

at $\rho \leq 0$.

4. ESTIMATE OF THE EFFECT

The concrete mechanism responsible for the deviation from resonance can vary from system to system. We estimate the magnitude of the effects using as an example the charge exchange of an ion A^+ in a neutral gas A. In this case, the polarization mechanism of deviation from resonance is effective. The charge-exchange system (ion + the atom with which the charge exchange takes place) polarize the medium, and the polarized medium produces an electric field in the region where the charge-charge-exchange system is located. The magnitude of this field depends on the charge-exchanging atomic particle in which the electron is located.

The deviation from resonance produced in this manner is equal to

$$\frac{\alpha e^2}{4} \sum_i \left(\frac{1}{R_{1i}^4} - \frac{1}{R_{2i}^4}\right),$$

where α is the polarizability of the atom, R_{1i} and R_{2i} are the distances from the i-th particle of the medium to the first and second-charge-exchange particles, respectively. This deviation is of the order of $\overline{\Delta} \sim \alpha e^2\rho_0/R^5$, where ρ_0 is the resonant-scattering amplitude and $R \sim n^{-1/3}$ is the average distance between the particles of the medium.

As seen from the results given in Sec. 3, the charge-exchange cross section decreases exponentially when the deviation from resonance exceeds values of the order of $\Delta_0 = \gamma v$. Let $\overline{\Delta} \gg \Delta_0$ (for example, $\epsilon = \overline{\Delta}/\Delta_0 \sim 10$); then the charge-exchange cross section is as a rule exponentially small (of the order of the non-resonant value). It is of the order of the resonant value in those infrequent cases when the surrounding medium leads accidentally to a small deviation between the level energies of the colliding particles.

An estimate for the effective charge-exchange cross section can be obtained by multiplying the resonance charge-exchange cross section $\pi\rho_0^2/2$ by the probability of the collision occurring in the case of small deviations ($\Delta \lesssim \Delta_0$). The latter probability is of the order of $\Delta_0/\overline{\Delta}$. We thus obtain for the charge-exchange cross section the formula

$$\sigma = A\rho_0\gamma v/\alpha e^2 n^{5/3}, \tag{2}$$

where A is a number on the order of unity.

5. RESULTS

To obtain quantitative data on the charge-exchange cross section it is necessary to calculate the distribution function of the detuning. Indeed, as seen from (1), it is necessary to know the joint distribution of the detunings at two critical points:

$$f(\Delta_1, \Delta_2) = \Big\langle \delta\Big(\Delta_1 - \sum_i [U(\mathbf{R}_i + \mathbf{r}_{\text{кр}1}) - U(\mathbf{R}_i)]\Big) \times \delta\Big(\Delta_2 - \sum_i [U(\mathbf{R}_i + \mathbf{r}_{\text{кр}2}) - U(\mathbf{R}_i)]\Big)\Big\rangle. \tag{3}$$

U is the interaction potential of the i-th particle of the medium with the charge-exchange system. One of the charge-exchange particles is at the origin. This mean value is calculated in standard fashion (see, for example, [3]). A detailed calculation of the distribution function is given in the Appendix.

With logarithmic accuracy, the charge-exchange cross section is equal to

$$\sigma = 8\pi \int_0^\infty d\Delta_1 \int_0^\infty d\Delta_2 \int_0^{\rho_0} f(\Delta_1, \Delta_2, \rho) W(\Delta_1, \Delta_2, \rho)\rho\, d\rho. \tag{4}$$

The asymptotic form of the cross section at $\epsilon = \overline{\Delta}/\Delta_0 \ll 1$ is given by

$$\sigma = \frac{\pi\rho_0^2}{2}\left[1 - C\left(\frac{\alpha e^2\rho_0}{\gamma v}\right)^{3/4} n\right]. \tag{5}$$

The second term is a small density-dependent correction to the cross section.

A large change in the cross section takes place at $\epsilon = \overline{\Delta}/\Delta_0 \gg 1$. In this case we have

$$\sigma = 0.03\rho_0\gamma v/\alpha e^2 n^{5/3}. \tag{6}$$

As seen from (6), the influence of the medium becomes stronger if the atoms of the medium have high polarizability. Thus, a noticeable effect is obtained for Cs ($\alpha = 360$) at a relative colliding-particle velocity $v \sim 10^{-4}e^2/\hbar$ and $n \sim 10^{18}$ cm^{-3}.

6. DISCUSSION

We can indicate a number of physical causes for which the influence of the medium on resonance charge exchange may be stronger than in the simplest case considered above.

A. The magnitude of the described effect can depend strongly on whether the atoms of the medium are in the ground state or in an excited one. Thus, for hydrogen-like states the parameter $\epsilon \sim \alpha e^2\rho_0 n^{5/3}/\gamma v$, which characterizes the influence of the medium, is proportional to the eighth power of the principal quantum number n (the polarizability is proportional to n^6, $\gamma \propto 1/n$, and $\rho_0 \propto 1/\gamma \propto n$); for example, for n = 2 the influence sets in at densities one-tenth as large as for n = 1.

B. If charged particles are present in the system, then the Coulomb deviation from resonance can greatly exceed the polarization deviation, even if the degree of ionization is small. In complete analogy with the procedure in Sec. 2, we find that in such systems $\epsilon = \overline{\Delta}/\Delta_0 \sim e^2\rho_0 n^{2/3}/\gamma v$. At the same value $v \sim 10^{-4}e^2/\hbar$, a noticeable effect sets in at lower densities, $n \sim 10^{16}$ cm^{-3}.

We note that the presence of charged particles can affect the resonant transfer of excitation between S-states. In this case the characteristic defect is of the order of $\alpha e^2\rho_0 n^{5/3}$, where α is the larger of the polarizabilities of the two states.

C. Let us consider the charge exchange of ions A^+ in a mixture of gases A and B, with $n_A \ll n_B$. The ions exchange charge resonantly with the atoms A ($n_A\rho_A^3 \ll 1$) and nonresonantly with atoms B (with cross section $\sigma \sim a^2 \ll \rho_A^2$). In this case the influence of the medium comes into play in purest form. In fact, the mean free path with respect to charge exchange depends on the partial pressure of the gas B (in accordance with formula (2)), in spite of the fact that the direct charge exchange with the atoms B can be neglected (if $n_A\rho_A^2 \gg n_B a^2$).

APPENDIX

Calculation of the probability distribution of the deviations from resonance

The defect produced by the particles of the medium in the resonant system $A + A^+$, at $n\rho_0^3 \ll 1$ and $R \sim n^{-1/3}$, is equal to

$$\Delta = 2\alpha e^2\Big[\rho\sum_i \frac{\sin\theta_i \sin\varphi_i}{R_i^5} - x\sum_i \frac{\sin\theta_i\cos\varphi_i}{R_i^5}\Big],$$

where r is the distance between the charge-exchange particles, ρ is the impact parameter, $x = (r^2 - \rho^2)^{1/2}$ when the particles approach each other and $x = -(r^2 - \rho^2)^{1/2}$ when they move apart; R_i, θ_i, and φ_i are the coordinates of the i-th particle of the medium, and α is the polarizability of the atoms of the medium.

The critical points of charge exchange with deviation from resonance as is well known, are the solutions of the equation $H_{12}(r) = |\Delta(r)|$, where H_{12} is the overlap integral of the wave functions ($H_{12} \sim E_0 e^{-\gamma r}$). Let Δ_1 and Δ_2 be the deviations at the critical points. We can assume with logarithmic accuracy that $r_{1cr} = r_{2cr} = \rho_0$, and that the critical points lie on both sides of the closest-approach point ($r = \rho$). Then

$$\Delta_1(r_{1cr}) = 2\alpha e^2\sum_i \frac{\sin\theta_i}{R_i^5}(\rho\sin\varphi_i - x_0\cos\varphi_i),$$

$$\Delta_2(r_{2cr}) = 2\alpha e^2\sum_i \frac{\sin\theta_i}{R_i^5}(\rho\sin\varphi_i + x_0\cos\varphi_i);$$

$$f(\Delta_1, \Delta_2) = \Big\langle \delta\Big(\Delta_1 - 2\alpha e^2\sum_i \frac{\sin\theta_i}{R_i^5}(\rho\sin\varphi_i - x_0\cos\varphi_i)\Big) \times \delta\Big(\Delta_2 - 2\alpha e^2\sum_i \frac{\sin\theta_i}{R_i^5}(\rho\sin\varphi_i + x_0\cos\varphi_i)\Big)\Big\rangle$$

$$= \frac{1}{4\pi^2}\iint_{-\infty} \exp\{i(\Delta_1 t_1 + \Delta_2 t_2)\}\, dt_1\, dt_2 \Big\langle \exp\Big\{-2i\alpha e^2 t_1\sum_i \frac{\sin\theta_i}{R_i^5} \times (\rho\sin\varphi_i - x_0\cos\varphi_i) - 2i\alpha e^2 t_2\sum_i \frac{\sin\theta_i}{R_i^5}(\rho\sin\varphi_i + x_0\cos\varphi_i)\Big\}\Big\rangle;$$

Here $x_0 = (\rho_0^2 - \rho^2)^{1/2}$.

The expression for the mean value can be rewritten as follows:

$$\left\langle \exp\left\{-2i\alpha e^2 t_1 \sum_i \frac{\sin\theta_i}{R_i^5}(\rho\sin\varphi_i - x_0\cos\varphi_i)\right\}\right.$$
$$\left.\times\exp\left\{-2i\alpha e^2 t_2 \sum_i \frac{\sin\theta_i}{R_i^5}(\rho\sin\varphi_i + x_0\cos\varphi_i)\right\}\right\rangle$$
$$=\left\langle \exp\left\{-2i\alpha e^2 t_1 \frac{\sin\theta}{R^5}(\rho\sin\varphi - x_0\cos\varphi)\right\}\right.$$
$$\left.\times\exp\left\{-2i\alpha e^2 t_2 \frac{\sin\theta}{R^5}(\rho\sin\varphi + x_0\cos\varphi)\right\}\right\rangle^N$$
$$=\exp\left\{-n\iiint\left[1-\exp\left\{-2i\alpha e^2 t_1(\rho\sin\varphi - x_0\cos\varphi)\frac{\sin\theta}{R^5}\right.\right.\right.$$
$$\left.\left.\left.-2i\alpha e^2 t_2\frac{\sin\theta}{R^5}(\rho\sin\varphi + x_0\cos\varphi)\right\}\right]dV\right\}.$$

The calculation of the integral in the exponential leads to the result

$$J = A(\alpha e^2)^{3/5}[\rho^2(t_1+t_2)^2 + x_0^2(t_1-t_2)^2]^{3/10},$$
$$A = \frac{4\pi}{5}2^{3/5}\int_0^\infty \frac{dz}{z^{8/5}}\left(1-\frac{\sin z}{z}\right) \approx 7{,}91.$$

Thus,

$$f(\Delta_1,\Delta_2) = \frac{1}{4\pi^2}\iint_{-\infty}^{+\infty}\exp\{i(\Delta_1 t_1 + \Delta_2 t_2)\}$$
$$\times\exp\{-nA(\alpha e^2)^{3/5}[\rho^2(t_1+t_2)^2 + x_0^2(t_1-t_2)^2]^{3/10}\}\,dt_1\,dt_2.$$

We present some asymptotic values of $f(\Delta_1, \Delta_2)$:

1) in the case $\Delta_1 \sim \Delta_2 \ll \bar{\Delta}$

$$f(\Delta_1,\Delta_2) \sim \frac{1}{\bar{\Delta}^2}\left(1 + C\frac{\Delta_1\Delta_2}{\bar{\Delta}^2}\right), \quad C \sim 1;$$

2) in the case $\Delta_1 \sim \Delta_2 \gg \Delta$

$$f(\Delta_1,\Delta_2) \sim \frac{\Delta^{8/5}}{\Delta_1^{13/10}\Delta_2^{13/10}}.$$

[1] O. B. Firsov, Zh. Eksp. Teor. Fiz. **21**, 1001 (1951).
[2] Yu. N. Demkov, Zh. Eksp. Teor. Fiz. **45**, 195 (1963) [Sov. Phys.-JETP **18**, 138 (1964)].
[3] S. Chandrasekhar, Revs. Mod. Phys. **15**, 1 (1943).

Translated by J. G. Adashko
55