DIFFUSION AND TRANSFER OF EXCITATION

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P. L. PAKHOMOV, I. Ya. FUGOL

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IN COLLISIONS OF METASTABLE HELIUM-3 ATOMS

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The study of diffusion and transfer of excitation of metastable atoms in a gas of normal atoms is of great interest in connection with the existence of an unusual long-range repulsion between these atoms. Theoretical calculations ([1–3]) of the interaction potentials V_A and V_S (especially in the region of large internuclear distances $R > 4a_0$, a_0 being the Bohr radius) are not very reliable and do not agree with one another. In the present work an attempt is made to reconstruct the form of the potential-energy curves that best agree with both theoretical and experimental data. The "empirical" potentials constructed in this way are used to calculate the total cross section Q, the diffusion cross section Q_d, and the excitation-transfer cross section Q_{tr}. The calculated values of Q_d are compared with the authors' experimental data for the diffusion coefficient of metastable helium-3 atoms in the temperature range 300–4° K. The calculated cross section Q_{tr} is compared with the experimental data obtained by Coulter et al. ([4]) in the temperature range 500–4° K.

1. It is known that when two atoms with identical nuclei possessing integer spin collide, and two interaction potentials V_S and V_A exist, the amplitude of the scattered wave has the form

$$f_B(\theta) = \frac{1}{2}\left[f_S(\theta) + f_S(\pi-\theta) + f_A(\theta) - f_A(\pi-\theta)\right]. \quad (1)$$

The probability of scattering is determined by the formula

$$|f_B(\theta)|^2 = \frac{1}{4}\left\{|f_S(\theta) + f_A(\theta)|^2 + |f_S(\pi-\theta) - f_S(\pi-\theta)|^2\right\} + R_1. \quad (2)$$

Here the terms $f_S(\theta)$ and $f_A(\theta)$ describe elastic scattering of atoms in the potentials V_S and V_A, while the terms $f_S(\pi-\theta)$ and $f_A(\pi-\theta)$ describe scattering

Fig. 1 and Fig. 2

Figure 1: Fig. 1 and Fig. 2

accompanied by exchange of nuclei, i.e., scattering with transfer of excitation from one nucleus to the other. The term R_1 contains interference terms—products of the type $f_{A,S}(\theta) \cdot f_{A,S}(\pi - \theta)$, which are negligibly small because of the weak overlap of the functions $f(\theta)$ and $f(\pi - \theta)$.

If the colliding atoms have half-integer spin and obey Fermi–Dirac statistics (helium-3), then the scattering probability must be written in the form ([5]):

$$|f(\theta)|^2 = \frac{3}{4}|f_\Phi(\theta)|^2 + \frac{1}{4}|f_B(\theta)|^2, \tag{3}$$

where $f_B(\theta)$ is defined by expression (1) and is symmetric with respect to exchange of nuclei. The function antisymmetric with respect to permutation of the nuclei,

$$f_\Phi(\theta) = \frac{1}{2}\left[f_S(\theta) - f_S(\pi - \theta) + f_A(\theta) + f_A(\pi - \theta)\right], \tag{4}$$

differs from $f_B(\theta)$ in the signs of the terms $f_S(\pi-\theta)$ and $f_A(\pi-\theta)$. Substituting (4) and (1) into (3), we obtain for the scattering probability $|f(\theta)|^2$ an expression analogous to (2), but with another set of interference terms R_2. Po-

since these terms are negligibly small, the expression for the scattering probability of helium-3 atoms has the same form as for helium-4 atoms, which obey Bose-Einstein statistics. Consequently, the basic formulas for the effective scattering cross sections of helium-3 and helium-4 atoms coincide.

2. A quantum-mechanical treatment leads to the following expressions for the total elastic scattering cross section Q, the diffusion cross section Q_d, and the cross section for excitation transfer from a metastable atom to a normal one Q_{tr}:

Fig. 1. Energy of the long-range repulsion $V_A(R)$ and $V_S(R)$ of metastable and normal helium atoms (empirical potentials)

Fig. 2. Calculated scattering cross sections of metastable and normal helium-3 atoms.
1—total cross section Q; 2—diffusion cross section Q_d; 3—excitation-transfer cross section Q_{tr}

$$Q = 4\pi k^{-2} \sum_{l=0}^{\infty} (2l+1)\sin^2\delta_l, \tag{5}$$

$$Q_d = 4\pi k^{-2} \sum_{l=0}^{\infty} (l+1) \sin^2(\delta_l - \delta_{l+1}), \tag{6}$$

$$Q_{tr} = \pi k^{-2} \sum_{l=0}^{\infty} (2l+1) \sin^2(\beta_l - \gamma_l). \tag{7}$$

Here $k = \mu v/\hbar$ is the wave vector, μ is the reduced mass, and v is the relative velocity of the atoms; the even phases $\delta_{2r} = \beta_{2r}$ are determined by the symmetric interaction potential V_S, while the odd phases $\delta_{2r+1} = \gamma_{2r+1}$ are determined by the antisymmetric potential V_A. The scattering phases were calculated in the Jeffreys quasiclassical approximation with the Langer correction, by a numerical method; its details are presented in Refs. ([6,7]). To compute the phases it is necessary to know the interaction potentials V_S and V_A.

Figure 1 shows the potentials $V_A(R)$ and $V_S(R)$ used by us at distances $R > 4a_0$. These potentials were constructed so as to ensure the best agreement of the calculation results with the experimental data. The asymptotic form of the curves $V_A(R)$ and $V_S(R)$ for $R > 7a_0$ was chosen as

$$V_{A,S}(R) = AR^2 \exp(-BR), \tag{8}$$

where $A = 21.115$ a.u., $B = 2.04$ a.u.*

* The asymptotic curve for $R > 7a_0$ is also well described by the power function $75R^{-12}$ (10^5 atomic units).

The curve $V_A(R)$ in the interval $5a_0 < R < 6a_0$ is "joined" to the potential of Madsen et al. ([3]). The curve $V_s(R)$ in the interval $6a_0 < R < 7a_0$ is "joined" to the potential $V_s(R)$ calculated by Beckingham and Dalgarno ([1]). The calculated values Q, Q_d, and Q_{tr} are given in Fig. 2. They differ appreciably from those calculated by Beckingham and Dalgarno ([6]). At high energies of the colliding atoms, the cross sections satisfy the limiting relations $Q \simeq 2Q_d$, and $Q_{tr} \simeq \frac{1}{2}Q_d$.

3. To compare with experiment, we determine the diffusion coefficient D of metastable helium-3 atoms.

$$D = \frac{3\pi}{32} \left(\frac{8\chi T}{\pi\mu}\right)^{1/2} \frac{1}{n\overline{Q}_d}, \tag{9}$$

where

$$\overline{Q}_d = \left(\frac{\mu}{2\chi T}\right)^3 \int_0^\infty v^5 Q_d(v) \exp\left(-\frac{\mu v^2}{2\chi T}\right) dv, \tag{10}$$

n is the density of normal atoms. The quantity $\overline{Q}_d$ can be calculated using the interpolation formula for the diffusion cross section $Q_d(v)$ of the form ([7])

$$Q_d(v) = \alpha + \beta v^{-1}. \tag{11}$$

The coefficients α and β in formula (11) for helium-3 are found to be $\alpha = 2.6 \cdot 10^{-15}$ cm^2, $\beta = 2.3 \cdot 10^{-10}$ cm$^3 \cdot$ s^{-1}. As a result we obtain the relation

$$Dn = \frac{3\pi}{32} \frac{(8\chi T/\pi\mu)^{1/2}}{\alpha + 0.75\,\beta(8\chi T/\pi\mu)^{-1/2}}. \tag{12}$$

If the reduced pressure $P = n\chi \cdot 300$ (mm Hg) is introduced, expression (12) can be written in the form

$$DP = 0.31 \cdot 10^{-16} Dn \quad (\text{cm}^2 \cdot \text{s}^{-1} \cdot \text{mm Hg}).$$

The calculated dependence of DP on T is shown in Fig. 3.

4. A study of the decay kinetics of metastable 2^3S atoms of helium-3 in the afterglow makes it possible to determine the diffusion coefficient of 2^3S atoms of helium-3 ([8]). At gas densities not exceeding $n \simeq 6{\cdot}10^{16}$ cm^{-3}, the change in the volume-averaged concentration of metastable atoms $M(t)$ follows an exponential dependence. The exponent contains

$$M(t) = M_0 \exp(-Dt/\lambda^2) \tag{13}$$

the diffusion collision frequency D/λ^2, where λ is the diffusion length of the vessel, and M_0 is the volume-averaged concentration at the initial instant of the afterglow. The concentration of metastable atoms was determined from the magnitude of the resonant absorption of the 3888.6 Å line from an external source, which was a discharge lamp filled with helium-3. The experimental conditions are described in detail in the authors' work ([7]), where experimental data are given for the diffusion of metastable helium-4 atoms.

Measurements of the diffusion coefficient in helium-3 were carried out over a wide temperature interval $T = 300, 77, 20$, and 4°K. The experimental values of DP are presented in Fig. 3. The diffusion coefficients of metastable helium-3 atoms exceed the diffusion coefficients of helium-4 by $(\mu_4/\mu_3)^{1/2} = 1.16$ times. The measured values of DP, as is seen from Fig. 3, agree well with the values of DP calculated with the aid of the empirical potential.

5. Measurement of the effective cross section for excitation transfer from a metastable helium-3 atom to a normal one was carried out in the work of Kulgav and coauthors ([4]). The experiments were performed in the temperature interval from 4 to

500°K. The resonance width of the sublevel of metastable atoms in the state with $m_j = 1/2$ was measured. The lifetime of an atom on this sublevel is determined entirely by the probability of transfer of excitation to a normal helium-3 atom. The experimentally measured quantity is

$$\Delta\nu \equiv \frac{1}{\pi\tau} = \frac{1}{\pi} nvQ_{tr}. \tag{14}$$

In Fig. 4 the dashed line shows the experimentally obtained temperature dependence of the averaged cross section $\overline{Q}_{tr}(T) = \overline{vQ_{tr}}/\overline{v}$ (the vertical lines indicate the experimental error). The measured quantity $\overline{Q}_{tr}(T)$ is expressed in terms of the calculated cross section $Q_{tr}(v)$ by the formula

$$\overline{Q}_{tr}(T) = \frac{\mu^2}{2\chi^2 T^2} \int_0^\infty v^3 Q_{tr}(v) \exp\left(-\frac{\mu v^2}{2\chi T}\right) dv. \tag{15}$$

Using the calculated values of $Q_{tr}(v)$, we determined the dependence $\overline{Q}_{tr}(T)$. The integral was evaluated graphically for $T = 100, 200, 300, 400$, and 500°K. The results of these calculations are shown in Fig. 4 by the solid line. It is seen that the theoretical values lie within the limits of the experimental error.

Thus, the experimental data on diffusion and excitation transfer of metastable helium-3 atoms agree well with the results of the calculation. This agreement confirms the validity of the empirical interaction potentials $V_A(R)$ and $V_S(R)$ at large distances.

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Physical-Technical Institute
of Low Temperatures
Academy of Sciences of the Ukrainian SSR

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Fig. 3. Temperature dependence of the diffusion coefficient DP of metastable helium-3 atoms. The curve is the calculated dependence; the points are the experimental results.

Fig. 4. Temperature dependence of the mean excitation-transfer cross section $\overline{Q}_{tr}$. 1 —experimental results ([4]); 2 —calculated dependence.

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Note: Figure translations are in progress. See original paper for figures.