### Influence on inelastic processes of the phase transition in a weakly collisional twodimensional Bose gas

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It is demonstrated that the phase transition in an interacting two-dimensional Bose gas leads to a strong decrease of the rate of inelastic processes as the temperature decreases, as in the three-dimensional case. It is found that, despite the absence of a true condensate at  $T \neq 0$ , the correlation properties of a condensate with a fluctuating phase that are inherent to the two-dimensional system at temperatures below the phase-transition point  $T_{cS}$  turn out to be sufficient for realization of the effect on the same scale as in the three-dimensional case. In a gas of spin-polarized atomic hydrogen (H1) adsorbed on a surface the probability of three-particle recombination falls as  $T\rightarrow 0$  by a factor of approximately six in comparison with its value at  $T>T_{cS}$ . Both the purely two-dimensional case and the quasi-two-dimensional case with delocalization of the particles in the direction perpendicular to the surface are considered. The phenomenon of the decrease of the rate constant of the reaction at  $T< T_{cS}$  opens up the possibility that the phase transition in the two-dimensional gas H1 can be detected directly from the recombination kinetics.

#### 1. INTRODUCTION

In a previous paper<sup>1</sup> we established that in the phase transition in an interacting Bose gas the rates of inelastic processes are changed substantially. The governing parameter here, in the ordinary three-dimensional case, was the value of the condensate density  $n_0(T)$ . The probabilities of inelastic transitions decrease continuously with increase of  $n_0$ , down to a certain limiting value as  $T \rightarrow 0$ . This effect is easy to understand physically. Considering an individual elementary process in which several identical particles take part, we should perform the corresponding symmetrization of the wavefunction. If the particles are in the condensate, such symmetrization is not necessary and, as a consequence of this, the transition probability changes.

This paper is devoted to an investigation of the analogous phenomenon in a weakly collisional two-dimensional Bose gas. The situation here turns out to be more complicated, since a real condensate exists only at T=0, and at a nonzero temperature the above argument concerning the symmetrization of the wavefunction of the particles is, generally speaking, inadequate.

An important point, however, is that a two-dimensional system at a temperature below the phase-transition point  $T_{cS}$  is characterized by two scales: The scale  $R_c$  of the power-law decay of the density matrix is large in comparison with the ordinary correlation length  $r_c$ . This makes it possible to introduce the concept of a quasicondensate (a condensate with fluctuating phase). In a cell of size L such that  $r_c \ll L \ll R_c$ , we have, as it were, a system with a real condensate. Here, however, the phases of the condensate wavefunctions in cells separated by distances greater than or of the order of  $R_c$  are found to be uncorrelated, and this means that it is not possible to introduce a condensate wavefunction for the entire system as a whole.

As shown in the paper (Sec. 2), the probability of inelastic processes is related to local correlation properties of the system. Because of the presence of a quasicondensate in a two-dimensional Bose gas at  $T < T_{cS}$ , its local correlation

properties are identical to the properties of a system with a real condensate. Thus, the change of the probability of inelastic processes in a two-dimensional Bose gas at  $T < T_{cS}$  will have qualitatively the same character as in the three-dimensional case.

This result has fundamental significance for a system of spin-polarized atomic hydrogen ( $H\downarrow$ ), which remains a gas at temperatures down to T=0. As is well known, in this system a two-dimensional gas phase, formed upon adsorption of hydrogen atoms on the surface of the liquid helium covering the walls of the cell, is always present.

Being metastable, the H1 system decays on account of inelastic processes of depolarization and recombination, which, for  $T < T_{cV}$  ( $T_{cV}$  is the Bose-condensation temperature for the bulk gas), have been considered in detail in Refs. 2 and 3. As has been predicted theoretically<sup>2</sup> and detected experimentally, 4-6 in the H \( \preceq \) gas there exists a nonremovable thresholdless decay channel associated with three-particle recombination via a virtual change of the spin configuration on account of dipole-dipole interaction. In the three-dimensional case, for  $T \rightarrow 0$ , when almost all the particles of the gas are in the condensate, the probability of three-particle recombination falls by a factor of six in comparison with its value at  $T \geqslant T_{cV}$  (Ref. 1). As shown below (Sec. 3), an analogous phenomenon occurs (at a fixed surface density) in the two-dimensional phase as well, despite the absence of a true Bose condensate.

Atomic hydrogen adsorbed on liquid helium actually forms a quasi-two-dimensional gas, in the sense that it is appreciably delocalized in the direction perpendicular to the surface. This implies that the collision of particles acquires a three-dimensional character, but the kinematic properties of the adsorbed gas remain two-dimensional. The latter circumstance means that the temperature dependence of the probability of three-particle recombination remains the same as in the strictly two-dimensional problem. Only the absolute value of the probability changes.

In many cases three-particle dipole recombination in

the adsorbed phase is the leading decay channel for the whole  $H\downarrow$  gas (see Refs. 2, 3, and 5–7). This gives the possibility of detecting the phase transition in the adsorbed gas from the change of the decay time as T decreases, or with increase of the bulk density  $n_{\nu}$  at a fixed T (the surface density  $n_S$  and, consequently,  $T_{cS}$  increase). This prospect seems very alluring, since it is much easier to realize the conditions for this transition than for the phase transition in the bulk (in particular,  $T_{cS}$  is substantially higher than  $T_{cV}$ ). However, it is necessary to note that in the detection of the phase transition in the adsorbed gas one must take account of the fact that the surface density of the atoms increases with decrease of temperature or with increase of  $n_{\nu}$ .

#### 2. THE PROBABILITY OF THREE-PARTICLE **RECOMBINATION AND ITS CONNECTION WITH CORRELATION PROPERTIES OF THE BOSE GAS**

We shall consider a weakly interacting Bose gas of spinpolarized hydrogen atoms in a pure spin state (the spins of the electron and nucleus are parallel). We shall analyze the process of three-particle recombination in such a gas, which leads to the formation of a strongly excited H<sup>\*</sup> molecule and H atom, with kinetic energies that are large in comparison with the characteristic energies of the particles of the gas.

We shall denote by  $\hat{H}$  ' that part of the interaction Hamiltonian which is responsible for the inelastic transition under consideration. Assuming that the rate of the process is small compared with the purely elastic processes, for the number of transitions in unit time we have  $(\hbar = 1)$ 

$$W = 2\pi \sum_{i,f} \rho_i |H'_{if}|^2 \delta(E_f - E_i) = \int_{-\infty}^{\infty} dt \langle \hat{H}'(0) \hat{H}'(t) \rangle,$$

$$\hat{H}'(t) = e^{i\hat{H}_o t} \hat{H}' e^{-i\hat{H}_o t}, \qquad (2.1)$$

where  $\widehat{H}_0$  is the Hamiltonian of the system when inelastic processes are neglected, and  $\rho_i$  is the equilibrium density matrix. In second quantization the Hamiltonian  $\hat{H}'$  has the form

$$\hat{H}' = \int \hat{\varphi}^{+} \left( \frac{\mathbf{r}_{1} + \mathbf{r}_{2}}{2} \right) \hat{\psi}^{+} (\mathbf{r}_{3}) \varphi_{i} (\mathbf{r}_{1} - \mathbf{r}_{2}) V (\mathbf{r}_{1}, \mathbf{r}_{2}, \mathbf{r}_{3})$$

$$\hat{\psi} (\mathbf{r}_{1}) \hat{\psi} (\mathbf{r}_{2}) \hat{\psi} (\mathbf{r}_{3}) d\mathbf{r}_{1} d\mathbf{r}_{2} d\mathbf{r}_{3} + \text{h.c.}$$
(2.2)

The expression (2.2) corresponds to a transition with the formation of an excited molecule in state j and an atom in a definite spin state. Correspondingly, V is the inelastic threeparticle interaction, integrated over the spin variables. In (2.2)  $\hat{\psi}$  and  $\hat{\varphi}$  are the  $\psi$ -operators for the atoms and molecules, defined in the standard manner on the wavefunctions of the free motion, and  $\varphi_i(\mathbf{r}_1 - \mathbf{r}_2)$  is the wavefunction of the molecule in its center-of-mass frame.

Since we assume that the binding energy  $E_i$  of the molecule is large in comparison with the characteristic energies T,  $n_S \tilde{U}$  of the particles of the gas ( $\tilde{U}$  is the effective vertex of the elastic pair interaction of the particles), the motion of the molecule and fast atom that are formed as a result of the reaction can be regarded as free, with energies  $q_1^2/4m - E_i$ and  $q_2^2/2m$ , respectively. Since we consider low temperatures we shall assume that the momenta k of the particles taking part in the reaction certainly satisfy the inequality

$$kR_0 \ll 1$$
 (2.3)

 $(R_0)$  is the effective range of the interaction of the particles).

In the determination of the interaction vertex (2.2) in the momentum representation the condition (2.3) makes it possible, for small initial particle momenta  $k_i$ , to go over from the true interaction V to an effective interaction  $V_{\chi \mathbf{k}_1 \mathbf{k}_2 \mathbf{k}_3}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ , where the function  $\chi$  takes account of the restructuring of the wavefunction of the initial state in the region of the interaction. For the vertex we then have the following expression:

$$\Gamma(\mathbf{q}_{1}, \mathbf{q}_{2} | \mathbf{k}_{1}, \mathbf{k}_{2}, \mathbf{k}_{3}) = \int d\mathbf{r}_{1} d\mathbf{r}_{2} d\mathbf{r}_{3} \, \varphi_{j}(\mathbf{r}_{1} - \mathbf{r}_{2}) \, V(\mathbf{r}_{1}, \mathbf{r}_{2}, \mathbf{r}_{3})$$

$$\cdot \chi_{\mathbf{k}_{1} \mathbf{k}_{2} \mathbf{k}_{3}}(\mathbf{r}_{1}, \mathbf{r}_{2}, \mathbf{r}_{3}) \exp \left[-i\mathbf{q}_{1} \left(\frac{\mathbf{r}_{1} + \mathbf{r}_{2}}{2}\right)\right]$$

$$-i\mathbf{q}_{2} \mathbf{r}_{3} \exp (i\mathbf{k}_{1} \mathbf{r}_{1} + i\mathbf{k}_{2} \mathbf{r}_{2} + i\mathbf{k}_{2} \mathbf{r}_{3})$$

$$(2.4)$$

(we set the normalization volume equal to unity). Because of the presence of the wavefunction  $\varphi_i(\mathbf{r}_1 - \mathbf{r}_2)$  in the integrand of (2.4), and the scale of the wave vectors in the final state  $(q_1,q_2>1/R_0)$ , the principal contribution to the integral is built up over interparticle distances of the order of  $R_0$ . In the three-dimensional case, in the three-particle system the correlation function  $\gamma$  under the condition (2.3) ceases to depend on the momenta  $k_i$ , and this was used in Ref. 1. In the two-dimensional case, however, the dependence of  $\gamma$  on  $k_i$  turns out to be important. To determine this function we can make use of the results of Appendix II. Taking into account the relation (A20), in this case we have

$$\Gamma(\mathbf{q}_{1},\mathbf{q}_{2}|\mathbf{k}_{1},\mathbf{k}_{2},\mathbf{k}_{3}) = \frac{\tilde{V}(q)}{\ln(2/|\mathbf{k}_{1}-\mathbf{k}_{2}|d)\ln^{2}(6/|\mathbf{k}_{1}+\mathbf{k}_{2}-2\mathbf{k}_{3}|d)}$$
(2.5)

 $(d \sim R_0)$  is the two-dimensional scattering length), where

$$\tilde{V}(q) = \int d\mathbf{R} \, d\mathbf{\rho} \, \varphi_j(\mathbf{R}) \, V(\mathbf{R}, \mathbf{\rho}) \, \Omega(\mathbf{R}, \mathbf{\rho}) \, e^{-i \, \mathbf{q} \, \mathbf{\rho}}. \tag{2.6}$$

Here we have introduced the relative coordinates  $\mathbf{R} = \mathbf{r}_1$  $-\mathbf{r}_2$  and  $\rho = (\mathbf{r}_1 + \mathbf{r}_2)/2 - \mathbf{r}_3$ . In going from (2.4) to (2.5) and (2.6), we have neglected [taking (2.3) and the inequality  $k_i \leqslant q_1, q_2$  into account] the terms  $k_i \cdot r_i$  in the exponent, and, by virtue of the law of conservation of momentum, have set  $\mathbf{q}_1 \cong -\mathbf{q}_2 = \mathbf{q}$ .

At temperatures above the phase-transition point  $T_{cS}$  in the two-dimensional case the effective values of the momenta  $k_i$  appearing in the arguments of the logarithms in (2.5) have values of the order of the thermal momentum  $k_T = (2mT)^{1/2}$ . In an interacting gas of finite density the  $k_i$ remain finite even for  $T\rightarrow 0$ . This is easily understood if we take into account that the particles arrive at the interaction region with an energy equal to the average interaction energy  $n_S \widetilde{U}$  in the gas. Correspondingly, the momentum of the particles will be of the order of  $(2n_s m\widetilde{U})^{1/2}$ . We note that this momentum coincides with the inverse correlation length  $k_c = 1/r_c$  (see the next paragraph), as is natural from a physical point of view (for  $T > n_s \tilde{U}$  the momentum  $k_c$ coincides with  $k_T$ ).

Having made use of these results, we replace the arguments of the logarithms in (2.5) by  $1/k_c d$  and introduce the effective vertex of the inelastic interaction in the form

$$\Gamma_{j} = \Gamma(q_{j}) \approx \widetilde{V}(q_{j}) \ln^{-3}(1/k_{c}d), \qquad (2.7)$$

where  $q_j = (4mE_j/3)^{1/2}$  is a finite momentum determined by the energy-conservation law.

In view of the local character of the interaction, we rewrite the Hamiltonian  $\hat{\mathbf{H}}'$  in the form

$$\hat{H}' \approx \Gamma_i \int \hat{\varphi}^+(\mathbf{r}) \hat{\psi}^+(\mathbf{r}) \hat{\psi}(\mathbf{r}) \hat{\psi}(\mathbf{r}) \hat{\psi}(\mathbf{r}) d\mathbf{r} + \text{h.c.}$$
 (2.8)

Noting that states with the large momenta  $q_1,q_2$  characteristic of the particles formed as a result of the reaction are not populated in the gas, for the correlator  $\langle \hat{H}'(0)\hat{H}'(t)\rangle$  appearing in (2.1) we obtain

$$\langle \hat{H}'(0)\hat{H}'(t)\rangle = |\Gamma_{j}|^{2} \sum_{\mathbf{q}_{1},\mathbf{q}_{2}} \int \langle \hat{\mathbf{\psi}}^{+}(\mathbf{r},0)\hat{\mathbf{\psi}}^{+}(\mathbf{r},0)\hat{\mathbf{\psi}}^{+}(\mathbf{r},0)$$

$$\cdot \hat{\mathbf{\psi}}(\mathbf{r}',t)\hat{\mathbf{\psi}}(\mathbf{r}',t)\hat{\mathbf{\psi}}(\mathbf{r}',t)\rangle$$

$$\cdot \exp\left[i(\mathbf{q}_{1}+\mathbf{q}_{2})(\mathbf{r}-\mathbf{r}')\right] \exp\left[i\left(\frac{q_{1}^{2}}{4m}+\frac{q_{2}^{2}}{2m}-E_{j}\right)t\right]d\mathbf{r}d\mathbf{r}'.$$
(2.9)

The correlator in (2.9) varies over a distance  $|\mathbf{r} - \mathbf{r}'|$  of the order of the correlation length  $r_c$  (see the next section). The integration over the momenta  $\mathbf{q}_1$  and  $\mathbf{q}_2$ , the scale of which is dictated by the large value of  $E_j$ , renders distances  $|\mathbf{r} - \mathbf{r}'|$  that are much shorter than the correlation length effective. Therefore, in the correlator we can set  $\mathbf{r} = \mathbf{r}'$ . Performing the integration over  $\mathbf{r}$  and  $\mathbf{r}'$  in (2.9) in explicit form and returning to the original formula (2.1), for the transition probability we have

$$W_{s} \approx 2\pi |\Gamma_{j}|^{2} \sum_{\mathbf{q}} \int_{-\infty}^{\infty} dt \langle \hat{\psi}^{+}(0,0) \hat{\psi}^{+}(0,0) \hat{\psi}^{+}(0,0) \hat{\psi}(0,t) \\ \hat{\psi}(0,t) \hat{\psi}(0,t) \rangle \exp \left[ i \left( \frac{3q^{2}}{4m} - E_{j} \right) t \right].$$
 (2.10)

The principal contribution to the integral in (2.10) is given by values  $t \sim 1/E_j$ . This time scale is short in comparison with the times  $(1/n_S \tilde{U}, 1/T)_{\min}$  characteristic for time correlations in a weakly interacting Bose gas. This makes it possible to set t=0 in the correlator. As a result, integrating over t and taking (2.7) into account, we find

$$W_s = W_0(q_j)K, \tag{2.11}$$

$$W_{0}(q_{i}) = 12\pi \frac{|\tilde{V}(q_{i})|^{2}}{\ln^{6}(1/k_{c}d)} n_{s}^{3} \sum_{q} \delta\left(\frac{3q^{2}}{4m} - E_{i}\right), \qquad (2.12)$$

$$K = \frac{1}{6n_s^3} \langle \hat{\psi}^+(0,0) \hat{\psi}^+(0,0) \hat{\psi}^+(0,0) \hat{\psi}(0,0) \hat{\psi}(0,0) \hat{\psi}(0,0) \rangle.$$
(2.13)

The correlation function K remains the same for all the reaction channels. The total recombination probability is obtained by summing  $W_S$  (2.11), (2.12) over the final states j of the molecule and the final spin states of the atom.

It is also obvious that the structure of the results (2.11)-(2.13) will be the same for all inelastic three-particle processes if the kinetic energy of the particles in the final state is large compared with T and  $n_S \tilde{U}$ . The structure of the relations (2.11)-(2.13) is analogous to the results obtained for the three-dimensional case (see Ref. 1). The difference is connected entirely with the form of the matrix element  $\tilde{V}(q_j)$  and with the appearance (specific to the two-dimensional gas) of the factor  $\ln^6(1/k_c d)$ , which depends on  $n_S$  and T, in the denominator of (2.12).

At temperatures above the phase-transition point the calculation of the correlator (2.13) both in the three-dimensional case  $(T>T_{cV})$  and in the two-dimensional case  $(T>T_{cS})$  gives K=1. For  $T< T_{cV}$  in the bulk gas this correlator decreases continuously with decrease of T on account of the increase of the condensate density, and for the ideal gas reaches the value 1/6 at T=0 (Ref. 1). The behavior of the correlator (2.13) in the two-dimensional Bose gas at  $T< T_{cS}$  requires a special analysis, since at  $T\neq 0$  a condensate in the generally accepted sense is absent here.

# 3. CORRELATION PROPERTIES OF THE TWO-DIMENSIONAL BOSE GAS. DETERMINATION OF THE THREE-PARTICLE CORRELATOR $K(n_s, T)$

At temperature T=0 in a two-dimensional interacting Bose gas a Bose condensation of the usual type occurs. The main feature specific to the two-dimensional case is the weak dependence of the effective pair-interaction vertex on the magnitude of the original interaction between the particles:

$$U = \frac{4\pi}{m} \xi, \quad \xi = \ln^{-1} \left( \frac{1}{k_s^2 d^2} \right) \ll 1.$$
 (3.1)

Here  $k_s$  is a quantity close to the inverse correlation length  $k_c = 1/r_c$ , and d is the two-dimensional scattering length, whose value d = 0.9 Å for the H $\downarrow$  gas was found in Ref. 3. The correlation length  $r_c$  can be found from the character of the decay of the one-particle density matrix:

$$\rho(\mathbf{r}) = \langle \hat{\psi}^+(0) \hat{\psi}(\mathbf{r}) \rangle.$$

For this it is sufficient to make use of the well-known technique of u-v transformations of Bogolyubov, susing the effective vertex (3.1). The correlation length at which  $\rho(\mathbf{r})$  falls from the value n at r=0 to the condensate-density value  $n_0$  (and remains constant at larger values of r) is found to be equal to

$$r_c \approx (2mT^*)^{-1/2}, \quad T^* = n_s \tilde{U}.$$
 (3.2)

The assumption of weak interaction in the two-dimensional Bose gas corresponds to the condition  $\xi \leqslant 1$ . This condition essentially replaces the usual criterion for applicability of the gas approximation in the three-dimensional case. Since in this case the inequality  $\xi \ln(1/\xi) \leqslant 1$  holds automatically, for the quantity  $k_S^2$  in (3.1) we can take the value  $4\pi n_S$ . The value of  $\widetilde{U}$  that follows from this was first obtained by Schick<sup>9</sup> for a model of a Bose gas of hard discs.

In the framework of the same technique it is not difficult to find the density of the particles above the condensate (see Ref. 9):

$$n' \approx n_s \xi.$$
 (3.3)

For  $\xi \ll 1$  most of the particles are in the condensate and we need not distinguish  $n_S$  and  $n_0$ .

To describe the properties of the system at T = 0 we can introduce the usual condensate wavefunction in the form

$$\psi_0(\mathbf{r}) = n_0^{1/2} \exp\left[i\Phi(\mathbf{r})\right] \tag{3.4}$$

with a weakly fluctuating phase  $\Phi$ .

At  $T \neq 0$  the picture is radically altered. For the fluctuation of the phase in the two-dimensional case the appearance of an infrared divergence is characteristic, and this leads to destruction of the long-range order. Now, for  $T < T_{cS}$  (and, simultaneously,  $T \leq T^*$ ) the asymptotic be-

havior of the one-particle density matrix is determined by the expression (see Refs. 10-14)

$$\rho(\mathbf{r}) = n_0 \exp\left[-\alpha \ln\left(r/r_{\bullet}\right)\right], \quad r \gg r_{\bullet}, \tag{3.5}$$

where

$$\alpha = \frac{mT}{2\pi\rho_0} = \frac{n_s}{\rho_0} \frac{T}{T} \xi, \tag{3.6}$$

$$r_{\bullet} = (T^{\bullet}/T)r_{c}, \tag{3.7}$$

in which  $\rho_0$  is the density of the superfluid component.

Since  $\alpha \le 1$ , it follows from (3.5) that  $\rho(\mathbf{r})$  decays over a large correlation length

$$R_c = r_* e^{1/\alpha} \tag{3.8}$$

(at T = 0, the correlation length  $R_c \to \infty$ ).

At distances r such that  $r_c \ll r \ll R_c$  the density matrix has a constant value equal to  $n_0$ , as in the usual case when a condensate is present. In this region the phase fluctuations have no effect. Therefore, considering regions of such a size, we can make use of the idea of a quasicondensate, to which a wavefunction of the form (3.4) corresponds. It is not possible to do this in a unified manner over all space, since at distances  $r \gg R_c$  the phase correlation is lost and the average of  $\psi_0$  over the volume is equal to zero.

Thus, in a two-dimensional Bose gas the presence of the two strongly differing scales  $r_c$  and  $R_c \gg r_c$  makes it possible to speak of the presence of a quasicondensate—a condensate with a fluctuating phase. If, conceptually, we divide the space into blocks of size L (cf. Ref. 13), with

$$r_c \ll L \ll R_c, \tag{3.9}$$

then all the correlation properties for which distances much shorter than L and times much shorter than  $\tau_L = L/c$   $(c = (T^*/m)^{1/2})$  is the velocity of sound) are important need be considered only within one block. But within the limits of one block the two-dimensional Bose gas can be regarded as a system possessing a normal condensate with density  $n_0$ . We note that in the volume of an individual block there are a large number of particles and the relative density fluctuations are small.

To determine the densities of the above-condensate (n') and condensate particles in a given block at a nonzero temperature we can make use of the system of temperature Green's functions that is obtained for a weakly interacting Bose gas with a condensate in the three-dimensional case (see, e.g., Ref. 15). The specific nature of the problem under consideration will be manifested only in the change to a two-dimensional phase volume and in the cutoff of the small momenta at a scale 1/L. The explicit expression for n' then has the form

$$n' = -\int_{k>1/L} \frac{d^{2}k}{(2\pi)^{2}} G(\mathbf{k}, \tau = -0)$$

$$= \int_{k>1/L} \frac{d^{2}k}{(2\pi)^{2}} \left\{ \left[ \frac{k^{2}/2m + n_{s}U}{2E(k)} - \frac{1}{2} \right] + \frac{k^{2}/2m}{E(k)} \frac{1}{\exp[E(k)/T] - 1} + \frac{n_{s}U}{E(k)} \frac{1}{\exp[E(k)/T] - 1} \right\},$$
(3.10)

where

$$E(k) = \left[\frac{k^2}{2m} \left(\frac{k^2}{2m} + 2n_s \mathcal{U}\right)\right]^{\prime h}$$

(G is the normal Green's function). For  $T \leqslant T^*$  the principal contribution to the integral (3.10) is given by the first term. This term is built up from momenta  $k \sim k_c \gg 1/L$ , and this makes it possible to set the lower integration limit equal to zero. A direct calculation leads to the relation (3.3).

The results obtained make it possible to determine the value of the correlator  $K(n_S,T)$  (2.13) for  $T \leqslant T^*$ . In fact, as was shown in Sec. 2, in the three-particle correlator in (2.9), which appears in the expression for the recombination probability, distances  $|\mathbf{r}-\mathbf{r}'| \sim R_0$  and times  $t \sim 1/E_j$  turn out to be important. Since these distances and times are small in comparison with L and  $\tau_L$ , respectively, we can use the results pertaining to a single block. Then, in zeroth order in the parameter  $\xi$ , replacing the  $\psi$ -operators in (2.13) by  $\psi_0$  we have

$$K \approx^{1}/_{6} \tag{3.11}$$

(it must be recalled, however, that the very presence of the quasicondensate at  $T \neq 0$  is due to the interaction between the particles, i.e., to the finite value of  $\xi$ ).

In order to find the temperature dependence of the correlator K at low T and to determine at the same time the correction to (3.11) on account of the interaction between the particles, we write the operator  $\hat{\psi}$  appearing in (2.13), as usual, in the form

$$\hat{\psi} = \psi_0 + \hat{\psi}'$$

where the operator  $\hat{\psi}'$  pertains to particles above the condensate. Then, confining ourselves to terms bilinear in  $\hat{\psi}'$ , we find

$$K(T) = \frac{1}{6} + \frac{1}{n_s} (n' + n_a), \tag{3.12}$$

where  $n_a = \langle \hat{\psi}' \hat{\psi}' \rangle$  is the anomalous average. Using the well-known expression for the anomalous Green's function of a Bose gas (see, e.g., Ref. 15) and determining the value of  $n_a$  in terms of it, in the two-dimensional case, with allowance for (3.10), we have

$$\frac{n'+n_a}{n_s} = \frac{1}{2n_s} \int \frac{d^2k}{(2\pi)^2} \left(\frac{k^2/2m}{E(k)} - 1\right) + \frac{1}{n_s} \int \frac{d^2k}{(2\pi)^2} \frac{k^2/2m}{E(k) \left\{ \exp[E(k)/T] - 1 \right\}}.$$
(3.13)

The temperature dependence of the correlator K is determined by the second term in (3.13), which does not have a singularity at small k. For  $T \leqslant T^*$ 

$$K(T) - K(0) \approx 2.4 \xi (T/T^*)^3$$
. (3.14)

The correction to K(0) (3.11) is determined by the first term in (3.13), which also is nonsingular at small k. However, this expression formally diverges at large k. Here we encounter the same problem as in the three-dimensional case, in which, when the contribution of the anomalous averages is taken into account, it becomes necessary, when going over from the bare vertex in the interaction Hamiltonian to the scattering amplitude a, to take into account terms not only of first order in a but also of the next order (see Ref. 8).

The bare three-particle-recombination amplitude  $\Gamma_j^{(0)}$  is given by the expression (2.4) with  $\chi_{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)=1$ . This amplitude is practically independent of the momenta  $k_i$  of the colliding particles. The vertex renormalization that occurs on account of elastic rescattering of the three parti-

cles by each other is determined by the function  $\chi$ , which, in perturbation theory in the effective pair interaction  $\widetilde{U}$  (3.1), can be written in the form

$$\chi_{\mathbf{k}_{1}\mathbf{k}_{2}\mathbf{k}_{3}}(\mathbf{r}_{1}, \mathbf{r}_{2}, \mathbf{r}_{3}) = 1 + \mathcal{U} \sum_{\mathbf{k}} \left[ \frac{e^{i(\mathbf{k}-\mathbf{k}_{1})(\mathbf{r}_{1}-\mathbf{r}_{2})}}{\varepsilon_{\mathbf{k}_{1}} + \varepsilon_{\mathbf{k}_{2}} - \varepsilon_{\mathbf{k}} - \varepsilon_{-\mathbf{k}+\mathbf{k}_{1}+\mathbf{k}_{2}}} \right.$$

$$+ \frac{e^{i(\mathbf{k}-\mathbf{k}_{1})(\mathbf{r}_{1}-\mathbf{r}_{3})}}{\varepsilon_{\mathbf{k}_{1}} + \varepsilon_{\mathbf{k}_{3}} - \varepsilon_{\mathbf{k}} - \varepsilon_{-\mathbf{k}+\mathbf{k}_{1}+\mathbf{k}_{3}}}$$

$$+ \frac{e^{i(\mathbf{k}-\mathbf{k}_{2})(\mathbf{r}_{2}-\mathbf{r}_{3})}}{\varepsilon_{\mathbf{k}_{2}} + \varepsilon_{\mathbf{k}_{3}} - \varepsilon_{\mathbf{k}} - \varepsilon_{-\mathbf{k}+\mathbf{k}_{2}+\mathbf{k}_{3}}} \right].$$
 (3.15)

In obtaining this expression we have used for the elastic pair interaction not the true potential but, in fact, the pseudopotential, the Fourier component of which at small momenta coincides with  $\widetilde{U}$ . We shall retain this value for larger momenta as well, keeping in mind that the final results will still be determined by the small momentum values, as has already been assumed implicitly in the derivation of the expressions (3.10) and (3.13).

Confining ourselves to the linear approximation in the parameter  $\xi$  and, correspondingly,  $\widetilde{U}$ , we should renormalize the vertices only for the dominant first term in the correlator (3.12), corresponding to the interaction of three quasicondensate particles. These particles have energy  $\varepsilon_{\mathbf{k}_i} = n_S \widetilde{U} = k_c^2/2m$ . The expression for the function  $\chi$  in this case acquires the form

$$\chi_0(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) = 1 + \frac{\tilde{U}}{2} \sum_{\mathbf{k}} \left[ \frac{e^{i\mathbf{k}(\mathbf{r}_1 - \mathbf{r}_2)} + e^{i\mathbf{k}(\mathbf{r}_1 - \mathbf{r}_3)} + e^{i\mathbf{k}(\mathbf{r}_2 - \mathbf{r}_3)}}{k_c^2 / 2m - k^2 / 2m} \right].$$
(3.16)

We now substitute this expression into (2.4). Then, assuming that the sum over k in (3.16) is bounded from above by values  $kd \le 1$  (and thereby separating out from the integral the bare vertex  $\Gamma_j^{(0)}$ ), and taking into account that the integral over k in (3.16) is taken in the principal-value sense, we have

$$\Gamma_{j} = \Gamma_{j}^{(0)} \left[ 1 - \frac{3\tilde{U}}{2} \sum_{k>k} \frac{1}{k_{c}^{2}/2m} \right].$$
 (3.17)

In fact, in going over to the formula (2.7) we used the replacement  $\Gamma_j^{(0)} \to \Gamma_j$ . In fact, to go over from the bare vertex to the true vertex it would be necessary to make use of the inverse series for  $\Gamma_j^{(0)}$  in terms of  $\Gamma_j$  (and  $\widetilde{U}$ ), the first two terms of which can be determined from the expression (3.17). Accordingly,

$$\Gamma_i^{(0)} \rightarrow \Gamma_i \left( 1 + \frac{3U}{2} \sum_{k > k_c} \frac{1}{k_c^2 / 2m} \right).$$
 (3.18)

Replacing  $\Gamma_j$  in (2.7) and, correspondingly, in (2.12) by the expression in the right-hand side in (3.18), with allowance for the first term in the correlator K (3.12), we obtain a correction linear in  $\xi$ ; this correction is equivalent to the appearance in (3.12) of a term equal to

$$\frac{\mathcal{U}}{2n_s} \int_{k>k_c} \frac{d^2k}{(2\pi)^2} \frac{1}{k^2/2m}.$$
 (3.19)

The sum of (3.13) and (3.19) no longer diverges at large momenta, and the expression for K(0) turns out to be finite. Direct calculation gives

$$K(0) = {}^{1}/_{6} + b\xi, \tag{3.20}$$

where  $b = (1 - \ln 2)$ .

The relations (3.14), (3.20) permit us to draw the important conclusion that in the two-dimensional case, at a low but nonzero temperature, when the true condensate disappears, the presence of the quasicondensate ensures that the correlator (2.13) falls to a value close to (3.11). In other words, the general result predicted earlier for the three-dimensional case remains valid.

The phase transition, or Kosterlitz-Thouless transition, in a two-dimensional Bose gas occurs at the temperature (see Ref. 16)

$$T_{cs} \approx \pi \rho_0 / 2m$$
, (3.21)

where  $\rho_0$  is the density of the superfluid component at  $T = T_{cS} - 0$ . For the real scale of the surface density the parameter  $\xi$  has magnitude  $\sim 10^{-1}$ . Comparing the expressions (3.21) and (3.1), (3.2), it is not difficult to convince oneself that  $T_{cS}$  and  $T^*$  are quantities of the same order and are even comparatively close to each other (the density  $\rho_0$ , defined in the same way as in the case of the three-dimensional Bose liquid (see, e.g., Ref. 8), turns out to be comparable with the total density). Therefore, as the temperature is lowered the correlator K(T) will experience a discontinuity upon passage through the point  $T_{cS}$  and will then go over comparatively rapidly to the value determined by the expression (3.14). To make a reliable determination of the magnitude of the discontinuity, and of the behavior of K(T) in the crossover region, presents substantial difficulties.

We note that because the quantities  $T_{cS}$  and  $T^*$  are comparable the inverse correlation length  $k_c$  appearing in the argument of the logarithm in (2.7) and (2.12) essentially retains its low-temperature value  $(2mT^*)^{1/2}$  near the phase transition as well.

### 4. QUASI-TWO-DIMENSIONAL GAS OF SPIN-POLARIZED ATOMIC HYDROGEN ON A LIQUID HELIUM SURFACE

We now consider the surface phase of atomic hydrogen adsorbed on liquid helium, making use of the results of the preceding sections. As has been elucidated previously, for all accessible values of the surface density of atoms this phase remains a gas at all temperatures (see Ref. 3). However, it is not purely two-dimensional, in the sense that, by virtue of the low energy  $\varepsilon_0$  of adsorption of a hydrogen atom, its characteristic localization length  $l=(2m\varepsilon_0)^{-1/2}$  in the direction perpendicular to the surface is appreciably greater than the effective range of the interaction of the atoms with each other. Because of this, the atomic-collision process has, to an important extent, a three-dimensional character, and this gives rise to a change of the effective interaction vertex in comparison with the purely two-dimensional case.

As we shall see below,  $T_{cS} \ll \varepsilon_0$  holds practically always, and in this case the density of the H $\downarrow$  gas adsorbed on the surface is small in comparison with the density for maximum occupation. In these conditions the inequality  $kl \ll 1$  is valid for the characteristic value of the momenta of the particles along the surface. By considering, in this situation, the collision of two adsorbed particles with allowance for their strong delocalization along the normal to the surface, we can solve the corresponding Schrödinger equation systematically and find the effective vertex of the pair interaction. The

solution of this problem, obtained in Appendix I, leads, for particles with relative two-dimensional momentum k, to an effective vertex in the form (see (A11))

$$\bar{U} = \frac{4\pi}{m} \frac{a}{e} \left[ 1 + \frac{2a}{l} \ln \frac{2}{kl} \right]^{-1}, \tag{4.1}$$

where a = 0.72 Å is the three-dimensional scattering length for the collision of hydrogen atoms in the triplet state.

In the present case of a quasi-two-dimensional gas of finite density at  $T < T_{cS}$  the effective momentum k of the colliding particles, as in the purely two-dimensional case, is close to  $k_c = 1/r_c$  (3.2). In the quasi-two-dimensional case the small parameter replacing  $\xi$  in (3.1) is the quantity  $\left[l/a + 2\ln(2/k_c l)\right]^{-1}$ . If the logarithmic term is smaller than l/a, as is the case at the usual experimental densities, the appropriate small parameter is the ratio a/l. The smallest value  $a/l \approx 0.08$  is achieved on a <sup>3</sup>He film ( $\varepsilon_0 \approx 0.35$  K; see Refs. 17 and 18). On a <sup>4</sup>He film this parameter is greater:  $a/l \approx 0.14$  ( $\varepsilon_0 \approx 1$  K; see Refs. 17 and 18).

The delocalization of the adsorbed particles changes the effective vertex  $\Gamma_i$  (2.7) of the inelastic three-particle interaction in two aspects. First, for  $l \gg R_0$  the wavefunction  $\chi^{(3)}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$ , given in (A24) (the  $\mathbf{r}_i$  here are three-dimension vectors) of the three colliding particles, which appears in the definition of  $\Gamma_i$ , will be the same, over short distances in the center-of-mass frame, as in the three-dimensional case. On the other hand, the rescattering that arises for  $kl \le 1$ , which is specific for the two-dimensional motion and determines the decrease of the wavefunction in the region of direct interaction of the three particles, is now characterized by a factor  $A_*$  (A25), in contrast to the factor A (A19) corresponding to the purely two-dimensional case. In addition, a further integration should be performed along the center-of-mass coordinate in the direction perpendicular to the surface. As a result, for the vertex of the three-particle interaction in the quasi-two-dimensional case we find

 $\Gamma(\mathbf{q}_1,\mathbf{q}_2|\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)$ 

$$= \left(1 + \frac{2a}{l} \ln \frac{2}{kl}\right)^{-1} \left(1 + \frac{2a}{l} \ln \frac{4}{kl}\right)^{-2} \frac{2}{3^{1/2}} \Gamma_j^{(3)}, \qquad (4.2)$$

where  $\Gamma_j^{(3)}$  is the three-dimensional vertex of the inelastic three-particle interaction,  $\mathbf{k}=(\mathbf{k}_1-\mathbf{k}_2)/2$ , and  $\tilde{\mathbf{k}}=(\mathbf{k}_1+\mathbf{k}_2-2\mathbf{k}_3)/3$ . Using the same arguments as in the purely two-dimensional case, we can approximate the relative momenta in the arguments of the logarithms by  $k_c$ . The transition from the Hamiltonian  $\hat{H}'$  (2.8) to the transition probability in the quasi-two-dimensional case is carried out in the same way as in Sec. 2. As a result, for the transition probability we have

$$W_s = \frac{4}{3l^2} \frac{\alpha_v n_s^3}{[1 + (2a/l)\ln(2/k_c l)]^6} K,$$
 (4.3)

where  $\alpha_V$  is the rate constant of three-particle recombination in the bulk at a temperature above the Bose-condensation point  $T_{cV}$  for the bulk gas.

The three-particle correlator K in (4.3) is defined on purely two-dimensional  $\psi$ -operators and in this sense does not differ from the correlator that was analyzed in the preceding section. Therefore, its entire temperature dependence is preserved, as, in particular, is the main result—the decrease of K below  $T_{cS}$  to a value close to 1/6. As regards the low-temperature behavior of K, it is determined as before by

the formulas (3.14), (3.20), provided that we make the replacement

$$\xi \to [l/a + 2 \ln(2/k_c l)]^{-1}$$
.

We note that the above properties of the correlator K follow in a natural manner from the condition  $r_c=1/k_c\gg l$ , which essentially reflects the two-dimensional character of the correlation properties of the quasi-two-dimensional gas under consideration. Correspondingly, the purely two-dimensional asymptotic behavior of the density matrix (3.5) and the idea of the quasicondensate are preserved. Naturally, the phase-transition temperature  $T_{cS}$  will be determined as before by the relation (3.21).

For  $T > T_{cS}$  the correlator K = 1, and formula (4.3) refines the result obtained in Ref. 3, in which the logarithmic term in the denominator was absent.

In the real geometry of the experiment it is easy to realize conditions in which the governing factor for the decay of the entire  $H\downarrow$  system (the bulk gas plus the gas on the surface) is three-particle dipole recombination on the surface. Here, from the magnitude of the inverse decay time

$$\frac{1}{r} \approx \frac{W_{s}S}{(n_{v}V + n_{s}S)} \tag{4.4}$$

( $n_V$  is the bulk density, V is the volume of the system, and S is the area of the surface) we can determine the value of  $W_S$  and, at the same time, the state of the adsorbed phase. However, the actual density of the adsorbed  $H\downarrow$  phase increases with decrease of the temperature, and this, naturally, leads to an increase of  $W_S$ . Therefore, to detect the phase transition in the adsorbed  $H\downarrow$  gas it is necessary not only to measure the lifetime of the system but also to determine independently the surface density of hydrogen atoms. In this case the detection of the phase transition should rest on the temperature dependence of the rate constant  $\alpha_S = W_S/n_S^3$  of the recombination process, which depends very weakly on  $n_S$ .

The phase-transition temperature  $T_{cS}$  for the adsorbed  $H\downarrow$  gas, in accordance with (3.21), is proportional to the density  $\rho_0$  of the superfluid component of this gas, which, in turn, depends on  $n_S$  and T. In conditions of equilibrium between the bulk and the surface, the value of  $n_S$  is dictated by the value of the temperature and of the bulk density  $n_V$ , and, thus,  $T_{cS}$  becomes, in fact, a function of  $n_V$ . The chemical potentials of the bulk and adsorbed phases in equilibrium are connected by the relation

$$\mu_{s} = \mu_{v} + \epsilon_{0}. \tag{4.5}$$

It is not difficult to convince oneself that at  $T = T_{cS} + 0$  we have

$$\mu_s \approx 2n_s(T_{cs})\tilde{U}. \tag{4.6}$$

The average distance between the particles in the adsorbed  $H\downarrow$  gas is much smaller than in the bulk gas. Therefore,  $T_{cS}$  substantially exceeds the Bose-condensation temperature  $T_{cV}$  for the gas in the bulk, and in the calculation of  $\mu_V(T_{cS})$  this gas can be regarded as a Boltzmann gas. Substituting (4.6) now into (4.5) with  $\tilde{U}$  (4.1) and taking (3.21) into account, we obtain for the phase transition temperature the equation

$$T_{cs} = \varepsilon_0 \left\{ \ln \left[ \frac{1}{n_{v}} \left( \frac{mT_{cs}}{2\pi} \right)^{\gamma_0} \right] + \gamma \right\}^{-1}, \tag{4.7}$$

where

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$$\gamma = 16 \frac{a}{l} \left( \frac{n_s}{\rho_0} \right) \left( 1 + \frac{2a}{l} \ln \frac{2}{k_c l} \right)^{-1}$$
.

As estimates show,  $\rho_0$  at the point  $T_{cS}$  is comparable with  $n_S$ , and the parameter  $\gamma \sim 1$ .

From (4.7) it follows that the transition temperature depends only logarithmically on  $n_{\nu}$ . For a limited bulk density ( $n_{\nu} \leq 10^{17} \, \mathrm{cm}^{-3}$ ) the logarithmic term in the denominator of (4.7) is large and  $T_{cS} \ll \varepsilon_0$ . In these conditions, at  $T \sim T_{cS}(n_{\nu})$  the surface density, as follows from (4.5) and (4.6), changes substantially over a temperature interval

$$\delta T \sim T_{cs}/\ln\left[\frac{1}{n_{rr}}\left(\frac{mT_{cs}}{2\pi}\right)^{\eta_{r}}\right]$$
.

Because of this, as the temperature is lowered through the point  $T_{cS}(n_V)$  there is a rapid increase of the transition temperature  $T_{cS}$  (3.21). As a consequence, the correlator K very rapidly reaches a value close to the limiting value (3.20) (here  $n_S$  still remains substantially smaller than the limiting-occupation density  $n_S^0 \cong 10^{14} \text{ cm}^{-2}$ ). Thus, the decrease of the rate constant  $\alpha_S$  at the phase transition is found to be effectively very sharp. In fact, if we take into account that the momentum  $k_c$  appearing in the argument of the logarithm in (4.3) is  $\sim n_S^{1/2}$  (see (3.2)), then for the rate constant we have the following expression:

$$\alpha_s(T, n_v) = \alpha_s(T_{cs}(n_v)) \left\{ \frac{1}{6} + \frac{a}{l} \left[ b + \ln \frac{n_s(T, n_v)}{n_s(T_{cs}(n_v))} \right] \right\}.$$

$$(4.8)$$

In another way of reaching the point  $T_{cS}$ , when the system is compressed at T= const, the surface density  $n_S$  changes substantially only when the bulk density changes by an amount of the order of  $n_V$  itself. In this case, as  $n_V$  increases from the critical value corresponding to the phasetransition point, the rate constant  $\alpha_S(T,n_V)$  goes over smoothly to its limiting value (4.8).

#### **5. CONCLUDING REMARKS**

Although the conditions for achieving a phase transition in an adsorbed  $H\downarrow$  gas are considerably easier to realize than in the bulk, it is evident, nevertheless, that appreciable experimental difficulties remain. In particular, these are connected with the necessity of having limited heating of the gas and, correspondingly a limited Kapitsa discontinuity  $\Delta T$  on the gas-liquid-helium boundary. Since  $\Delta T$  and  $1/\tau$  (4.4) are proportional to  $n_S^3(n_V V \gg n_S S)$ , and  $T_{cS} \sim n_S$  (see (3.21)), when surface recombination plays the dominant role, it may turn out to be optimal to search for the phase transition at low values of  $n_S$ . Then, however, the phase-transition temperature may be shifted into the millikelvin region. In this case the lifetime of the system, with the gas now concentrated practically entirely on the surface, is of the order of 10-100 sec  $(n_S S \gg n_V V)$  and  $1/\tau \sim n_S^2$ .

For high surface densities  $n_s \sim 10^{13}$  cm<sup>-2</sup>, corresponding to a temperature  $T_{cs} \sim 50$  mK, the equilibrium bulk density  $n_V$  in the case of a <sup>3</sup>He surface is  $\sim 10^{14}-10^{17}$  cm<sup>-3</sup>, and it becomes necessary to impose conditions on the relative magnitudes of the mean free path of a particle in the bulk and the linear dimensions of the system. Essentially, this is the condition that the excited molecules formed in recombina-

tion, and, under certain conditions, hot atoms as well, succeed in "diving" into the helium before they transfer energy to the atoms of the gas. With allowance for the restrictions that arise on the linear dimensions of the system, the lifetimes turn out to be of the order of seconds. These time scales for closed systems make it reasonable to use a pulse technique. The conditions are simplified appreciably in the case of so-called open systems (see Ref. 7), when the recombination energy is drawn off through a magnetic wall.

It should be noted that one further restriction on the attainment of the phase transition arises because of the need to have a sufficiently long lifetime of an adsorbed atom on the surface—this time should exceed the characteristic correlation time of the superfluid motion of the two-dimensional H $\downarrow$  gas (see Ref. 21). As estimates show, for  $T < T_{cS} \le 50$  mK this condition is certainly fulfilled both on a <sup>4</sup>He surface and on a <sup>3</sup>He surface.

We note in conclusion that the use of the more accurate relation (4.3) for the rate constant of three-particle dipole recombination on the surface changes the quantitative estimate of this quantity from that given in Ref. 3 for  $T > T_{cS}$ (K = 1). Taking into account that the momentum  $k_c$  should be replaced by the thermal momentum  $k_T = (2mT)^{1/2}$ , for adsorption on a <sup>4</sup>He surface ( $l \approx 5$  Å) we have  $\alpha_S$  $\approx 0.5 \cdot 10^{-24} \text{ cm}^4/\text{sec}$  [in (4.3),  $W_S = \alpha_S n_S^3$  is the number of recombination events per unit area in unit time]. This number corresponds to  $T \approx 0.3$  K, but the dependence  $\alpha_s(T)$  is very weak. The value found is five times smaller than that which is obtained without the logarithmic term in the denominator. The experimentally obtained values of  $\alpha_s$ lie in the range  $(0.5-1)\cdot 10^{-24}$  cm<sup>4</sup>/sec (Refs.6, 22-24). This good agreement between theory and experiment should not be overvalued, since, strictly speaking, for the case of <sup>4</sup>He the parameter  $R_0/l$  is not sufficiently small, although the ratio a/l amounts to 0.14.

#### **APPENDIX I**

### Effective vertex of the elastic interaction of a pair of particles in the quasi-two-dimensional case

We shall consider the idealized quasi-two-dimensional case, when the localization length  $l=(2m\varepsilon_0)^{-1/2}$  of an adsorbed atom in the direction perpendicular to the surface is much greater than the effective range  $R_0$  of the interaction of the particles. Actually,  $R_0$  is close in magnitude to the effective range of the interaction of an adsorbed atom with particles of the surface. Therefore, at distances  $z \gg R_0$  from the surface, at which the wavefunction of the adsorbed atom is mainly localized, the latter is determined by the expression

$$\psi_0(z) \approx (2/l)^{1/2} \exp(-z/l)$$
. (A1)

The Schrödinger equation for a pair of adsorbed atoms has the form (in the plane of the surface we have selected the center-of-mass system)

$$\left[ -\frac{1}{m} \Delta_{\rho} - \frac{1}{2m} \frac{d^{2}}{dz_{1}^{2}} - \frac{1}{2m} \frac{d^{2}}{dz_{2}^{2}} + U_{0}(z_{1}) + U_{0}(z_{2}) + U(r) \right] \psi$$

$$= (-2\varepsilon_{0} + k^{2}/m) \psi. \tag{A2}$$

Here  $\rho$  is the projection of the vector  $\mathbf{r}$  of the interatomic spacing on the plane of the surface,  $U_0(z)$  is the potential of the interaction of the adsorbed atom with the surface, U(r) is the potential of the interaction of the adsorbed atoms with

each other, and k is the wave vector of the relative motion of the atoms in the plane of the surface. For  $\rho \to \infty$  we have

$$\psi = \frac{2}{l} \exp \left[ -\frac{(z_1 + z_2)}{l} \right] e^{ik\rho}.$$

We shall be interested in momenta  $k \le 1/R_0$ . Then for  $kl \le 1$  ( $\varepsilon_0 \ge k^2/2m$ ), at interatomic distances  $\rho \le 1/k$  the wavefunction  $\psi$  is determined by Eq. (A2) with k=0, and the dependence of  $\psi$  on k can be factored out. If  $\rho \le l$  also, we can regard the relative motion of the atoms as three-dimensional and represent  $\psi$  in the form of a product:

$$\psi = B(k) \eta(\mathbf{r}) (2/l) \exp[-(z_1 + z_2)/l], \tag{A3}$$

where  $\eta(\mathbf{r})$  is the wavefunction of the three-dimensional relative motion for  $k \to 0$ , determined by the equation

$$-\frac{1}{m} \left[ \frac{d^2 \eta}{dr^2} + \frac{2}{r} \frac{d\eta}{dr} \right] + U(r) \eta = 0$$

 $(\eta(\mathbf{r}) \to 1$  as  $r \to \infty$ ), and the function  $(2/l)\exp[-(z_1+z_2)/l]$  describes the motion of the center of mass of the pair along the z axis.

We now find the factor B(k). For this we multiply Eq. (A2) by  $\psi_0(z_1)\psi_0(z_2)$  and integrate over  $z_1$  and  $z_2$ . From the resulting equation we subtract the equation

$$\left[ -\frac{1}{2m} \frac{d^2}{dz_1^2} - \frac{1}{2m} \frac{d^2}{dz_2^2} + U_0(z_1) + U_0(z_2) \right] \psi_0(z_1) \psi_0(z_2)$$

$$= -2\varepsilon_0 \psi_0(z_1) \psi_0(z_2),$$

multiplied by  $\psi$  and also integrated over  $z_1$  and  $z_2$ . As a result, at distances  $\rho \gg R_0$  for which in (A2) the potential U(r) can be neglected, we obtain

$$(\Delta_{\mathbf{\rho}} + k^2) \varphi(\mathbf{\rho}) = 0, \tag{A4}$$

where

$$\varphi(\boldsymbol{\rho}) = \int_{1}^{\infty} dz_{1} \int_{1}^{\infty} dz_{2} \psi_{0}(z_{1}) \psi_{0}(z_{2}) \psi(z_{1}, z_{2}, \boldsymbol{\rho}). \tag{A5}$$

The two-dimensional free-motion equation (A4) is, naturally, also valid at distances  $\rho \gg l$ , at which the relative motion of the particles is almost purely two-dimensional and  $\psi = \psi_0(z_1)\psi_0(z_2)\varphi(\rho)$ . The solution of this equation that at infinity becomes the plane wave  $e^{i\mathbf{k}\cdot\mathbf{p}}$ , at distances  $\rho \ll 1/k$  at which we can confine ourselves to the S-wave has the form

$$\varphi(\rho) = \left[ 1 - \frac{\ln(2e^{-c}/k\rho)}{\ln(1/kd^*)} \right], \quad R_0 \ll \rho \ll \frac{1}{k}$$
(A6)

(C = 0.577 is the Euler constant). The quantity  $d^*$  is determined by the form of the wavefunction at distances  $r \sim R_0$ .

Thus, the formula (A6) should be obtained from (A5) with  $\psi(z_1,z_2,\rho)$  (A3) with  $R_0 \ll \rho \ll l$ , 1/k. The three-dimensional wavefunction  $\eta(\mathbf{r})$  of the relative motion for  $\rho \gg R_0$  can be written in the form

$$\eta(\mathbf{r}) = 1 - a/r,\tag{A7}$$

where a is the three-dimensional scattering length. Substituting (A7) into (A5), changing to the integration variables  $z_1 - z_2$  and  $(z_1 + z_2)/2$ , and performing the integration between the limits  $-(z_1 + z_2) \le z_1 - z_2 \le z_1 + z_2$ ,  $z_1 + z_2 \ge 0$ , we find

$$\varphi(\rho) = B(k) \left( 1 - \frac{2a}{l} \ln \frac{l}{\rho} + \frac{2a}{l} C \right). \tag{A8}$$

Equating now in (A8) and (A6) the terms that depend lo-

garithmically on  $\rho$ , and also equating the terms that do not depend on  $\rho$ , we obtain B(k) and  $D^*$ :

$$B(k) = [1 + (2a/l)\ln(2/kl)]^{-1}, \tag{A9}$$

$$d^* = (l/2) \exp(-l/2a).$$
 (A10)

The expression for the effective vertex of the elastic interaction of two particles in the quasi-two-dimensional case under consideration has the form (we set the normalization volume equal to unity)

$$U = \int \psi(z_1, z_2, \rho) U(r) \psi_0(z_1) \psi_0(z_2) dz_1 dz_2 d\rho.$$

Substituting into this the true wavefunction  $\psi$  (A3) with B(k) (A9) and integrating, taking the relation

$$\int \eta(r) U(r) d\mathbf{r} = 4\pi a/m$$

into account, we find

$$U = \frac{4\pi}{m} \frac{a}{l} \left( 1 + \frac{2a}{l} \ln \frac{2}{kl} \right)^{-1}.$$
 (A11)

The formula (A11) without the logarithmic term in the denominator was obtained earlier in Ref. 3. By making use of the parameter d \* (A10), we can represent  $\tilde{U}$  in the form

$$U = \frac{4\pi}{m \ln(1/kd^*)}.$$

This result differs from the formula (3.1) for  $\widetilde{U}$  in the purely two-dimensional case (see Refs. 3 and 9) only in the replacement of the two-dimensional scattering length d by the parameter  $d^*$ .

We note that for  $kl \gg 1$  we ought to regard the collisions of the particles as purely three-dimensional. In this case, in (A3) the coefficient B=1, and the averaging of the interaction energy of a pair of particles over their motion along the z axis gives the result (A11), but without the logarithmic term in the denominator.

#### **APPENDIX II**

## Determination of the three-particle correlation function $\chi(R,\rho)$

We first consider the purely two-dimensional case. The Hamiltonian of three hydrogen atoms in the center-of-mass frame has the form

$$\hat{H} = -\frac{1}{2m} \left( 2\Delta_{\mathbf{R}} + \frac{3}{2} \Delta_{\mathbf{\rho}} \right) + U(R) + U\left( \left| \mathbf{\rho} - \frac{\mathbf{R}}{2} \right| \right) + U\left( \left| \mathbf{\rho} \pm \frac{\mathbf{R}}{2} \right| \right), \tag{A12}$$

where  $\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3$  are the coordinates of the atoms,  $\mathbf{R} = \mathbf{r}_1 - \mathbf{r}_2$ ,  $\rho = (\mathbf{r}_1 + \mathbf{r}_2)/2 - \mathbf{r}_3$ , and U(r) is the potential of the pair elastic interaction of the particles.

The initial state corresponds to three slow atoms, and at infinity the wavefunction is

$$\psi = e^{i\mathbf{k}\mathbf{R}}e^{i\mathbf{k}\rho}, \quad R \to \infty, \ \rho \to \infty.$$

Here  $\mathbf{k} = (\mathbf{k}_1 - \mathbf{k}_2)/2$  is the momentum of the relative motion of particles 1 and 2, and  $\mathbf{k} = (\mathbf{k}_1 + \mathbf{k}_2 - 2\mathbf{k}_3)/3$  is the momentum of the motion of particle 3 relative to the quasimolecule 1–2. We shall be interested in particle monenta k,  $\tilde{k} \leqslant 1/R_0$ . We represent the wavefunction  $\psi$ , with allowance for rescattering of the particles, in the form  $\psi = \chi(\mathbf{R}, \rho)$ 

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 $\times \exp(i\mathbf{k}\cdot\mathbf{R}+i\tilde{\mathbf{k}}\cdot\boldsymbol{\rho})$ . Then at interatomic distances  $R,\rho \leqslant 1/k,1/\tilde{k}$ , the three-particle correlation function  $\chi$  will be a solution of the Schrödinger equation  $\hat{H}\chi=0$  and can be written in the form

$$\chi(\mathbf{R}, \, \mathbf{\rho}) = A(k, \, \tilde{k}) \, \Omega(\mathbf{R}, \, \mathbf{\rho}), \tag{A13}$$

where the function  $\Omega(\mathbf{R}, \rho)$  does not depend on the momenta of the particles.

In the Hamiltonian  $\hat{H}$  (A12) we go over from the variables  $\mathbf{R}$ ,  $\boldsymbol{\rho}$  to the variables  $R, \varphi, r_1 = |\boldsymbol{\rho} + \mathbf{R}/2|$ ,  $r_2 = |\boldsymbol{\rho} - \mathbf{R}/2|$  ( $\varphi$  is the angle of the vector  $\mathbf{R}$  relative to the stationary axis). We obtain  $\hat{H} = \hat{H}_0 + \hat{H}_1$ , where

$$\hat{H}_{0} = -\frac{1}{m} \left[ \frac{d^{2}}{dR^{2}} + \frac{1}{R} \frac{d}{dR} + \frac{1}{R^{2}} \frac{d^{2}}{d\phi^{2}} + \frac{d^{2}}{dr_{1}^{2}} + \frac{1}{r_{1}} \frac{d}{dr_{1}} \right]$$

$$+ \frac{d^{2}}{dr_{2}^{2}} + \frac{1}{r_{2}} \frac{d}{dr_{2}} + U(R) + U(r_{1}) + U(r_{2}), \quad (A14)$$

$$\hat{H}_{1} = -\frac{1}{m} \left[ \frac{r_{1}^{2} + r_{2}^{2} - R^{2}}{r_{1}r_{2}} \frac{d^{2}}{dr_{1}dr_{2}} + \frac{r_{1}^{2} - r_{2}^{2} + R^{2}}{r_{1}R} \frac{d^{2}}{dr_{1}dR} + \frac{r_{2}^{2} - r_{1}^{2} + R^{2}}{r_{2}R} \frac{d^{2}}{dr_{2}dR} + \frac{1}{r_{1}R^{2}} \left[ 4r_{1}^{2}R^{2} - (r_{1}^{2} + R^{2} - r_{2}^{2})^{2} \right]^{\eta_{1}} \frac{d^{2}}{dr_{1}d\phi} - \frac{1}{r_{2}R^{2}} \left[ 4r_{2}^{2}R^{2} - (r_{2}^{2} + R^{2} - r_{1}^{2})^{2} \right]^{\eta_{1}} \frac{d^{2}}{dr_{2}d\phi}. \quad (A15)$$

To find  $A(k, \tilde{k})$  in  $\chi$  (A13) it is sufficient to consider the region of interparticle distances in which at least two of the three distances R,  $r_1$ ,  $r_2$  are large in comparison with  $R_0$ . In this case, as will be established below, in the determination of  $\chi$  the Hamiltonian  $\hat{H}_1$  can be neglected.

In the equation  $\widehat{H}_0 \chi = 0$  the variables can be separated. In the variables  $r_1$  and  $r_2$  we have equations describing s-scattering of two particles. By virtue of the condition  $kR_0 \leqslant 1$ , for the motion in the R direction we should also confine ourselves to the s-wave. As a result, we have

$$\chi = \psi(R)\tilde{\psi}(r_1)\tilde{\psi}(r_2), \tag{A16}$$

where all three functions correspond to s-scattering. From the solution of the pair problem in the two-dimensional case it is known that for  $R_0 \ll R \ll 1/k$  the wavefunction corresponding to momentum k is (see Appendix I)

$$\psi(R) = \left(1 - \frac{\ln(2e^{-c}/kR)}{\ln(1/kd)}\right) = \frac{\ln(gR/d)}{\ln(1/kd)},$$
 (A17)

where g = 0.89 and d is the two-dimensional scattering length, determined by the shape of the potential U(r). Analogously,

$$\tilde{\psi}(r_i) = \frac{\ln(gr_i/d)}{\ln(2/\tilde{k}d)}, \quad R_0 \ll r_i \ll 1/\tilde{k}$$
(A18)

(each of the functions  $\tilde{\psi}(r_i)$  corresponds to momentum  $\tilde{k}/2$ ).

Comparing the function (A16) obtained on the basis of (A17), (A18) with the expression (A13), for the coefficient  $A(k, \tilde{k})$  we find

$$A(k,\tilde{k}) = \frac{1}{\ln(1/kd)\ln^2(2/\tilde{k}d)}.$$
 (A19)

We now consider the Hamiltonian  $\hat{H}_1$ . As regards the terms containing derivatives with respect to  $\varphi$ , they can give a contribution only when p-scattering is taken into account, and are small in proportion to the parameter  $kR_0 \leqslant 1$ . The three other terms in the expression for  $\hat{H}_1 \chi$  at distances  $R \sim r_i \sim r \gg R_0 \sim d$  are of the order of  $\sim Ar^{-2} \ln(r/d)$ . The

ratio of these terms to the terms appearing in  $\widehat{H}_0 \chi$  is of the order of  $\sim \ln^{-1}(r/d) \ll 1$ . The ratio of any of the terms in  $\widehat{H}_1 \chi$  to the largest term in  $\widehat{H}_0 \chi$  can only decrease when the distances R,  $r_1$ , and  $r_2$  are different in scale (including the case when one of them becomes  $\sim R_0$ ). The above estimates and arguments justify the neglect of the Hamiltonian  $\widehat{H}_1$  in the determination of the factor  $A(k, \tilde{k})$ .

Thus, the three-particle correlation function  $\chi(\mathbf{R}, \rho)$  appearing in the definition of the vertex (2.4) of the inelastic transition, in the region of interparticle distances  $R, \rho \leqslant 1/k$ ,  $1/\tilde{k}$ , has the form

$$\chi(\mathbf{R}, \mathbf{\rho}) = \frac{\Omega(\mathbf{R}, \mathbf{\rho})}{\ln(1/kd)\ln^2(2/\tilde{k}d)}, \tag{A20}$$

where the function  $\Omega(R,\rho)$  for  $R,\rho \geqslant R_0$  has the asymptotic form

$$\Omega(\mathbf{R}, \mathbf{\rho}) \rightarrow \ln\left(\frac{gR}{d}\right) \ln\left(\frac{gr_1}{d}\right) \ln\left(\frac{gr_2}{d}\right).$$

We consider now the quasi-two-dimensional case for  $l \gg R_0$ . We first carry out the same procedure as in the derivation of formula (A4), but in a system of three particles. As a result we find

$$(2\Delta_{\mathbf{R}} + \frac{3}{2}\Delta_{\rho} + 2k^{2} + \frac{3}{2}\tilde{k}^{2})\tilde{\varphi}(\mathbf{R}, \, \rho) = 0, \quad R, \, \rho \gg R_{0},$$

$$\tilde{\varphi}(\mathbf{R}, \, \rho) = \int \tilde{\gamma}(\mathbf{R}, \, \rho, \, z_{1}, \, z_{2}, \, z_{3}) \, \psi_{0}(z_{1}) \, \psi_{0}(z_{2}) \, \psi_{0}(z_{3}) \, dz_{1} \, dz_{2} \, dz_{3},$$

$$(A21)$$

where  $\widetilde{\chi}$  is the quasi-two-dimensional three-particle correlation function. For  $R_0 \ll R \ll 1/k$ ,  $1/\tilde{k}$ , in correspondence with the results obtained above we have

$$\tilde{\varphi}(\mathbf{R}, \mathbf{\rho}) = \frac{\ln(gR/d_0)\ln(gr_1/d_0)\ln(gr_2/d_0)}{\ln(1/kd_0)\ln^2(2/\tilde{k}d_0)}.$$
 (A22)

In the region of interparticle distances R,  $\rho \leqslant 1/k$ ,  $1/\tilde{k}$ , l, the relative motion of the particles is purely three-dimensional and the quasi-two-dimensional function  $\tilde{\chi}$  for k,  $\tilde{k} \leqslant 1/l$  can be represented in the form

$$\tilde{\chi}(\mathbf{R}, \, \boldsymbol{\rho}, \, z_{\scriptscriptstyle 1}, \, z_{\scriptscriptstyle 2}, \, z_{\scriptscriptstyle 3})$$

$$=A_{\star}(k, \tilde{k})\chi^{(3)}(\mathbf{R}, \boldsymbol{\rho}, z_1, z_2, z_3)\psi_0(z_1)\psi_0(z_2)\psi_0(z_3), \quad (\mathbf{A23})$$

where  $\chi^{(3)}$  is the three-dimensional correlation function. If in this case we have  $R, \rho \gg R_0$ , the function  $\chi^{(3)}$  can be written in the form

$$\chi^{(3)} = \{1 - a[R^2 + (z_1 - z_2)^2]^{-1/2}\} \{1 - a[r_1^2 + (z_1 - z_3)^2]^{-1/2}\}$$

$$\times \{1 - a[r_2^2 + (z_2 - z_3)^2]^{-1/2}\} \cdot$$
(A24)

Substituting (A23) with  $\chi^{(3)}$  (A24) into (A20), confining ourselves to terms linear in the scattering length a, and performing the integration over  $z_1$ ,  $z_2$ , and  $z_3$ , we obtain an expression for  $\tilde{\varphi}(\mathbf{R}, \boldsymbol{\rho})$  that should coincide with (A22). Comparing the two expressions, for  $d_0$  we find a value coinciding with d \* (A10), while for the coefficient  $A_*(k, \tilde{k})$  we obtain

$$A_{\bullet}(k,\tilde{k}) = \left\{ \left( 1 + \frac{2a}{l} \ln \frac{2}{kl} \right) \left( 1 + \frac{2a}{l} \ln \frac{4}{\tilde{k}l} \right)^{2} \right\}^{-1}. \quad (A25)$$

In the calculation of the vertex (2.4) of the inelastic three-particle transition in the quasi-two-dimensional case it is necessary to note that the final wavefunction of the system of three particles is now represented in the form of a product of the three-dimensional wavefunctions of the atom and molecule in the center-of-mass frame and the wavefunction

(normalized to unity) of the motion of the center of mass of the three particles along the z axis:

$$\hat{\mathbf{\psi}}_{j} = \varphi_{j}(\mathbf{R}) \exp(i\mathbf{q}_{j}\mathbf{\rho}) (6/l)^{1/2} \exp\left[-(z_{1}+z_{2}+z_{3})/l\right] \quad (\mathbf{A26})$$

(**R** and  $\rho$  are three-dimensional vectors).

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