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*Published in:*

Physical Review. B, Condensed Matter

*DOI:*

[10.1103/PhysRevB.43.13412](https://doi.org/10.1103/PhysRevB.43.13412)

[Link to publication](#)

*Citation for published version (APA):*

Svistunov, B. V., Hijmans, T. W., Shlyapnikov, G. V., & Walraven, J. T. M. (1991). Resonant-Light Absorption and the Problem of Observing the Kosterlitz-Thouless Transition in Spin-Polarized Atomic-Hydrogen Adsorbed on a Liquid-He Surface. *Physical Review. B, Condensed Matter*, 43, 13412-13416. DOI: 10.1103/PhysRevB.43.13412

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## Resonant-light absorption and the problem of observing the Kosterlitz-Thouless transition in spin-polarized atomic hydrogen adsorbed on a liquid-He surface

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(Received 20 December 1990)

We discuss the conditions for the occurrence of the Kosterlitz-Thouless phase transition in spin-polarized atomic hydrogen, adsorbed on the surface of liquid He, in contact with bulk hydrogen gas. We show that the phase transition will have a marked effect on the density of adsorbed atoms. We present calculations in which we show that determination of this surface density, crucial for detecting the transition, may be done through surface selective absorption of resonant light.

### I. INTRODUCTION

The two-dimensional phase of spin-polarized atomic hydrogen ( $H\downarrow$ ), which is formed on the surface of liquid He due to adsorption of atoms from the bulk, is of special interest as a macroscopic quantum system.<sup>1,2</sup> In accordance with theoretical predictions,<sup>3</sup> this system should remain a rarefield gas at any temperature down to  $T \rightarrow 0$ . Thus we have a near perfect realization of a weakly interacting two-dimensional (2D) Bose gas. At sufficiently low temperatures ( $T \approx 100\text{--}200$  mK) this gas will undergo a Kosterlitz-Thouless (KT) transition<sup>4</sup> at a value of the surface density ( $\hbar=1$ ,  $k_B=1$ )

$$n_{s_c} \approx \frac{2mT}{\pi} \quad (1)$$

substantially lower than the saturation density. Here  $m$  is the mass of the H atom. We have written Eq. (1) in a form tailored to the situation in which  $T$  is kept constant and the surface density  $n_s$  is varied, as this is more convenient from an experimental point of view than the case where the surface density  $n_s$  is constant and  $T$  is variable.

Unfortunately, it is difficult to satisfy the conditions for the occurrence of the KT transition when the adsorbed gas is in equilibrium with a bulk gas at the same temperature. This is related to the fact that at the low temperature at which the transition occurs, three-body dipolar recombination of atoms to molecules on the surface leads to a rapid decay and heating of the system as a whole. This problem can be circumvented by using a small cold surface at temperature  $T$ , in contact with a relatively large buffer volume at a higher temperature  $T_b$ .<sup>5</sup>

The KT transition in a weakly interacting Bose gas is accompanied by the formation of a quasicondensate and should manifest itself as a strong reduction of the three-body dipolar recombination rate constant  $\alpha_s$  of adsorbed atoms.<sup>6</sup> Well inside the KT region,  $\alpha_s$  is reduced by approximately a factor of 6. This effect may be identified experimentally by determining independently  $n_s$  and number of recombination events per unit time

$$\nu_s = \alpha_s n_s^3 A, \quad (2)$$

where  $A$  is the area of the surface. The value of  $\nu_s$  can be easily obtained in the situation when the decay of the H system as a whole is governed by the recombination on the cold surface.

An additional possibility to observe the KT transition through the measurement of the surface density arises from the peculiar behavior of  $n_s$  near the transition point. As we shall show below,  $n_s$  should display an almost jumpwise increase when the KT region is entered through a gradual increase of the density in the buffer volume  $n_b$  at constant  $T$ .

It seems clear from these arguments that the possibility to determine  $n_s$  independently from  $\nu_s$  is crucial for the experimental observation of the KT transition in adsorbed  $H\downarrow$ . This is however not a trivial matter, particularly as one would like to make the area of the cold surface as small as possible in order to reduce the loss rate of atoms due to surface recombination, and accordingly increase the lifetime of the system.

In this paper we discuss the influence of the KT transition on the function  $n_s(n_b, T)$  and the possibility of detecting the transition through measurement of  $n_s$ . We present calculations of resonant light absorption in adsorbed  $H\downarrow$  and argue that the determination of  $n_s$  can in principle be done by optical means.

### II. INFLUENCE OF THE FORMATION OF THE QUASICONDENSATE ON $n_s(n_b, T)$

Well inside the KT region a quasicondensate is formed which drastically alters the local correlation properties of the system.<sup>6</sup> These correlation properties influence in an essential manner the dependence of  $n_s$  on  $n_b$  and  $T$ . In this section we discuss this dependence in some detail.

Two regimes may be distinguished when considering how the balance between  $n_s$  and  $n_b$  is maintained. At sufficiently low  $T$ , the number of surface recombination events per unit time exceeds the rate of desorption

$[\alpha_s n_s^3 \gg n_s / \tau_d(T)]$ , where  $\tau_d$  is the characteristic desorption time. At higher temperatures the balance is maintained by adsorption and desorption of atoms. As the latter regime seems more favorable from an experimental point of view we shall mainly consider this case. Under these conditions we have

$$\frac{1}{4}\gamma(T_v)\bar{v}n_v(T_v) = \frac{n_s}{\tau_d(T)}. \quad (3)$$

Here  $\gamma$  is the sticking probability which was determined by Berkhout *et al.*<sup>7</sup> and  $\bar{v}$  is the thermal velocity of the atoms in the buffer volume.

In order to derive the expression for  $\tau_d$  we shall first consider the case  $T_v = T$  and equate the chemical potentials  $\mu_v$  and  $\mu_s$  of the particles in the bulk and on the surface. Bearing in mind that  $n_v \lambda^3 \ll 1$  ( $\lambda^2 \equiv 2\pi/mT$ ) in the region of interest, we replace  $\mu_v$  by its classical value  $T \ln(n_v \lambda^3)$ . Above the KT transition ( $n_s < n_{s_c}$ ) we shall use

$$\mu_s = \mu_{id} - \varepsilon_0 + 2T^*, \quad (4)$$

where  $\mu_{id} = T \ln[1 - \exp(-n_s \lambda^2)]$  is the chemical potential of the ideal 2D Bose gas,  $\varepsilon_0$  is the adsorption energy of an H atom,  $T^* = n_s \tilde{U}$ , and  $\tilde{U}$  is the effective elastic interaction between adsorbed H atoms. The adsorption isotherms were obtained in this manner by Goldman and Silvera<sup>8</sup> using a value for  $\tilde{U}$  of  $5 \times 10^{-15}$  K cm<sup>2</sup> taken from Ref. 9. In Fig. 1 we show the isotherm for this value of  $\tilde{U}$  and  $T = 0.1$  K (solid line for  $n_s \lesssim n_{s_c}$ , dotted

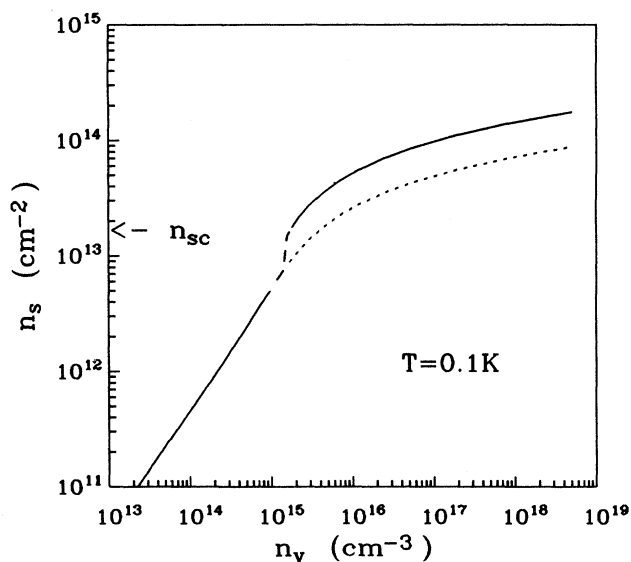


FIG. 1. The adsorption isotherm of H on <sup>4</sup>He for  $T = 0.1$  K and  $\tilde{U} = 5 \times 10^{-15}$  K cm<sup>2</sup>. The physically meaningless continuation of the low-density expression into the KT region, obtained by using Eq. (4) without taking the quasicondensate into account, is denoted by the dotted line. The dashed part of the curve denotes the region where fluctuations may be important. In order to obtain the plot of  $n_s$  vs  $n_v$  for the case  $T = 0.1$  K and  $T_v = 0.3$  K, the values on the  $n_v$  axis should be multiplied by a factor  $\approx 0.2$  (see text).

line for higher  $n_s$ ). Using Eq. (3) with  $T_v = T$  we can convert the adsorption isotherm into an expression for the desorption time  $\tau_d(T)$

$$\frac{1}{\tau_d} = \frac{T}{2\pi} \gamma(T) \frac{1 - \exp(-n_s \lambda^2)}{n_s \lambda^2} \exp\left\{\frac{-\varepsilon_0 + 2T^*}{T}\right\}, \quad n_s < n_{s_c}. \quad (5)$$

Inside the KT region we may use

$$\mu_s = -\varepsilon_0 + T^*. \quad (6)$$

The second term in the rhs of this expression is the chemical potential for the weakly interacting Bose gas at  $T = 0$ . Here we neglect the temperature dependence of  $\mu_s$ . It follows from the theory of the 2D Bose gas<sup>10</sup> that

$$\mu_s(T) - \mu_s(0) = \xi T^* f\left\{\frac{T}{T^*}\right\} \quad (7)$$

where  $\xi = m\tilde{U}/4\pi$  and

$$f(x) = 2x \int_0^\infty dt \frac{1}{\exp\{[t(t+2/x)]^{1/2}\} - 1} \left\{\frac{t}{t+2/x}\right\}^{1/2}. \quad (8)$$

One may easily check that  $\xi \approx 0.1$  for  $\tilde{U} = 5 \times 10^{-15}$  K cm<sup>2</sup>. Furthermore, using Eq. (1) for  $n_s \approx n_{s_c}$  we find  $T/T^* \approx 1$ . Numerical evaluation of Eq. (8) yields  $f(1) = 0.72$ . It is thus clear that the temperature dependent term Eq. (7) is much smaller than  $T^*$  in the entire KT region, justifying its omission in Eq. (6).

The adsorption isotherm in this case assumes the form

$$n_v \lambda^3 = \exp\left\{\frac{-\varepsilon_0 + T^*}{T}\right\}, \quad n_s > n_{s_c}. \quad (9)$$

In Fig. 1 this part of the adsorption isotherm is denoted by a solid line for  $n_s > n_{s_c}$ . Equation (9) has essentially the same form as the adsorption isotherm obtained by Goldman and Silvera,<sup>8</sup> the only difference being the replacement of  $2T^*$  in their expression by  $T^*$ . This change is related to the appearance of the quasicondensate below the KT transition temperature ( $n_s > n_{s_c}$ ). Goldman and Silvera did not take the presence of the quasicondensate into account and used the same expression Eq. (4) for  $\mu_s$  in the entire density range. From Eqs. (9) and (3) with  $T_v = T$  we obtain the expression for  $\tau_d$  for the case  $n_s > n_{s_c}$ :

$$\frac{1}{\tau_d} = \frac{T}{2\pi} \gamma(T) \frac{1}{n_s \lambda^2} \exp\left\{\frac{-\varepsilon_0 + T^*}{T}\right\}, \quad n_s > n_{s_c}. \quad (10)$$

Although the adsorption isotherms are by definition only valid for  $T_v = T$ , the related expressions Eqs. (5) and (10) remain valid for  $T_v \neq T$ , as the desorption rate is clearly independent of  $T_v$ . The plot of  $n_s$  versus  $n_v$  for this case can be directly obtained from the adsorption isotherm by multiplying the scale of  $n_v$  by a factor  $(T/T_v)^{1/2} \gamma(T)/\gamma(T_v)$ , as can be seen from Eq. (3).

Clearly, Eqs. (4) and (5) are valid for  $n_s \ll n_{s_c}$  while Eqs. (6), (9), and (10) apply when  $n_s \gg n_{s_c}$ . The situation near the KT point ( $n_s \sim n_{s_c}$ ) is less obvious in view of the possible importance of fluctuations due to vortices. In relation to this problem we should note that for  $n_s > n_{s_c}$  these fluctuations will be important only in a very narrow region near the KT point because of the low concentration of (bound) vortices. As follows from the considerations of KT,<sup>4</sup> for  $n_s < n_{s_c}$  the number of vortices increases rapidly, implying a destruction of quasicondensate correlation properties. Therefore we are justified in concluding that deviations from Eqs. (4) and (5) will not be very large. Hence, by comparing Eqs. (5) and (10), and using Eq. (3) we see that  $\tau_d$ , and accordingly  $n_s$ , should display an almost jumpwise increase by a factor  $\approx 2$  when entering the KT region. It is worth mentioning that the very occurrence of this change in  $n_s$ , suggests that the region in which fluctuations may be important may be even narrower than in the case of a KT transition at fixed  $n_s$ . In other words, a modest decrease in  $n_v$  may drive the system far from the KT point.

It is interesting to note that at low enough temperature  $T$  when the adsorption rate is balanced by surface recombination rather than by adsorption [ $\frac{1}{4}n_v\gamma(T_v)\bar{v}(T_v) \ll \alpha_s n_s^3$ ], there should be a similar jumpwise increase in  $n_s$  on entering the KT region. In this case this is due to the drop in  $\alpha_s$  by approximately a factor of 6.

### III. THE ABSORPTION PROBABILITY OF RESONANT LIGHT IN ADSORBED H<sub>1</sub>

In order to observe adsorbed H atoms by surface-selective light absorption, one needs a source of frequency  $\omega$  tunable in the vicinity of the resonance frequency  $\omega_0$  of a transition in the free H atom. The detuning  $\Delta\omega = \omega - \omega_0$  is necessary to match the energy shift of the resonance, induced by the interaction with the He substrate. Third-harmonic generation or other four-wave-mixing techniques, offer the possibility of generating powerful and narrow-band light [peak powers of the order of 1 W or more<sup>11</sup> and band widths as low as 150 MHz (Ref. 12)] tunable in the vicinity of the Lyman  $\alpha$  ( $L_\alpha$ ) transition (121.57 nm) in H. With the application of these light sources in mind, we shall analyze the resonance behavior of the adsorbed atoms in the vicinity of  $L_\alpha$ , although the theory is valid for any transition from the atomic ground state. We shall consider values of  $\Delta\omega$  which are substantially larger than the characteristic resonance interaction energy of an excited H atom (H\*) with atoms in the ground state. This interaction  $U_{\text{HH}^*} \sim n_s d^2/l$  where  $d$  is the atomic transition dipole moment and  $l$  ( $\approx 5 \text{ \AA}$ ) is the localization length of an adsorbed H atom in the direction perpendicular to the surface. For  $n_s \sim 10^{13} \text{ cm}^{-2}$  the interaction  $U_{\text{HH}^*}$  is of the order of tens of Kelvins. When  $\Delta\omega$  is substantially larger than this value, the HH\* interaction plays a negligible role and we shall henceforth ignore it.

For sufficiently large detuning ( $\Delta\omega \sim 1000 \text{ K}$ ), radiative transitions occur at short distances from the surface

( $z < 4 \text{ \AA}$ ). In this case the excited H atom interacts effectively with a single He atom, and we may consider that a HeH\* quasimolecule is formed in the photon absorption process. As we consider sufficiently high-frequency detuning

$$|\Delta\omega| \gg T, \varepsilon_c, \quad (11)$$

where  $\varepsilon_c$  is the characteristic potential energy of the He atoms, we may neglect the energy of the initial state in the energy conservation law of the absorption process. Furthermore, the relative motion of the nuclei in the HeH\* quasimolecule is quasiclassical. Hence,<sup>13</sup> although the motion of the particle in the initial state is clearly quantum mechanical, the radiative transitions will occur in a narrow range of internuclear distances around  $r_{v*}$  given by

$$\omega_0 + \Delta\omega = U_v(r_{v*}). \quad (12)$$

Here  $U_v(r)$  is the potential curve of HeH\* in the electronic state  $v$ . Then we may introduce the transition probability at a fixed distance  $\mathbf{r}$

$$W(\mathbf{r}) = \frac{4\pi^2\omega_0}{c} \sum_v |\mathbf{d}_v(\mathbf{r}) \cdot \mathbf{e}_{p\beta}|^2 \delta(\omega_0 + \Delta\omega - U_v(r)), \quad (13)$$

and write the following expression for the total probability of photon absorption:

$$W = \int W(\mathbf{r}) G(\mathbf{R}, \mathbf{r}) d\mathbf{R} d\mathbf{r}. \quad (14)$$

Here  $\mathbf{d}_v(\mathbf{r})$  is the electronic dipole moment for the transition of the HeH quasimolecule from the ground state to state  $v$ ,  $\mathbf{e}_{p\beta}$  is the polarization vector of a photon with wave vector  $\mathbf{p}$  and polarization  $\beta$ , and  $c$  is the speed of light. The correlator  $G(\mathbf{R}, \mathbf{r})$  describes the motion of the He and the H atom in the initial state,  $\mathbf{R}$  is the center of mass coordinate of the quasimolecule. The normalization surface area has been set equal to unity.

We use the following approximate representation for the correlator  $G$ :

$$G(\mathbf{R}, \mathbf{r}) = n_{\text{He}} n_s f(r) g_1(z - r\chi) g_2(z), \quad (15)$$

where  $n_{\text{He}}$  is the He bulk density,  $z$  is the component of  $\mathbf{R}$  perpendicular to the surface,  $\chi = \cos(\theta)$ ,  $\theta$  is the angle between  $\mathbf{r}$  and the  $z$  direction, and  $f(r) = \varphi^2(r)/r^2$  is a pair correlation function for the ground state of the HeH quasimolecule. This function may be found from Schrödinger's equation:

$$-\frac{1}{2m_*} \varphi''(r) + U_{\text{HeH}} \varphi(r) = 0$$

with the boundary conditions  $\varphi(0) = 0$ ,  $\varphi'(0) = 0$ , and  $\varphi'(\infty) = 1$ . Here  $m_* = m_{\text{He}} m / (m_{\text{He}} + m)$  and  $U_{\text{HeH}}$  is the ground-state potential energy of the quasimolecule. The functions  $g_1(z)$  and  $g_2(z)$  describe the density profile in the  $z$  direction of He and H atoms, respectively. Substituting Eqs. (13) and (15) in Eq. (14) and integrating over  $\mathbf{r}$  we obtain

$$W = \frac{8\pi^3 d^2 \omega_0 n_{\text{He}} n_s}{c} \sum_{\nu} \frac{r_{\nu*}^2 f(r_{\nu*}) |\alpha_{\nu}(r_{\nu*})|^2 \Lambda(r_{\nu*})}{|U'_{\nu}(r_{\nu*})|}. \quad (16)$$

Here  $U'_{\nu} = dU_{\nu}/dr$  and

$$\Lambda(r_{\nu*}) = \int \frac{d\Omega}{2\pi} |\mathbf{e}_{\nu}(\mathbf{r}_{\nu*}) \cdot \mathbf{e}_{p\beta}|^2 Q(r_{\nu*}\chi), \quad (17)$$

$$Q(h) = \int_{-\infty}^{\infty} g_1(z-h) g_2(z) dz. \quad (18)$$

We used the representation  $\mathbf{d}_{\nu}(\mathbf{r}) = d\mathbf{e}_{\nu}(\mathbf{r})\alpha_{\nu}(\mathbf{r})$  where  $\mathbf{e}_{\nu}(\mathbf{r})$  is the unit polarization vector of the  $\text{HeH}^*$  molecule. As we have  $\varepsilon \gg |\Delta\omega|$ , where  $\varepsilon$  is the separation between the  $L_{\alpha}$  level and any other excited states of the H atom,  $a_{\nu}$  is essentially the coefficient determining the admixture of the  $2P$  state of H in the LCAO expression for the electronic wave function of the  $\text{HeH}^*$  molecule.

In the above treatment we implicitly replaced the energy of the  $\text{HeH}^*$  molecule by the potential  $U_{\nu}(r)$  [see Eq. (13)]. This is clearly allowed in the case of positive detuning, when only unbound states of  $\text{HeH}^*$  are formed and Eq. (12) may be fulfilled for any  $\Delta\omega > 0$ . For  $\Delta\omega < 0$  the states of  $\text{HeH}^*$  are bound which suggests that absorption of light will occur only at certain discrete values of  $\Delta\omega$ . However, the bound states are closely spaced and consequently they will be mixed by interaction of the  $\text{HeH}^*$  molecule with other He atoms and smeared out into a continuum. Hence the theory is approximately applicable to the case  $\Delta\omega < 0$ .

#### IV. FEASIBILITY OF OBSERVING THE KT TRANSITION

In this section we shall apply the results obtained above for an experimentally reasonable range of the parameters and discuss the possibility to determine  $n_s$  through the absorption of resonant light. For  $T=0.1$  K ( $n_s \sim 10^{13}$  cm $^{-2}$ ) and  $T_v \sim 0.3$  K, using<sup>7</sup>  $\gamma(T)/T \approx 0.3$  K $^{-1}$ , we can obtain from Eqs. (3), (5), and (10) that both the classical ( $n_s < n_{s_c}$ ) and the quantum ( $n_s > n_{s_c}$ ) regimes are accessible while maintaining  $n_v \sim 10^{14} - 10^{15}$  cm $^{-3}$ . It may be easily verified in this case, with  $\alpha_s \sim 10^{-24}$  cm $^4$ /s (see Ref. 2) that  $\tau_d^{-1} \gg \alpha_s n_s^2$  and hence Eq. (3) is applicable. In order to perform meaningful measurements, the lifetime  $\tau$  of the  $\text{H}_1$  system has to be at least several seconds. Under the conditions discussed, and using reasonable values for  $V$  ( $\sim 1$  cm $^3$ ) and  $A$  ( $\sim 1-3$  mm $^2$ ) one has  $\tau \approx n_v V / \alpha_s n_s^3 A \sim 10-100$  s.

In order to obtain numerical values for the absorption probability  $W$ , we need the explicit form of the potential curves  $U_{\nu}(r)$ . These curves taken from Theodorakopoulos *et al.*<sup>14</sup> are shown in Fig. 2. At sufficiently high positive frequency detuning, only transitions to the  $C^2\Sigma^+$  state occur, while in the case of negative  $\Delta\omega$  we have transitions to the  $A^2\Sigma^+$  and the  $B^2\Pi$  states. As can be seen from the potential curves of Fig. 2, for the same value of  $|\Delta\omega|$ , condition Eq. (12) is satisfied at larger values of  $r_*$  for positive detuning than in the case of negative detuning. One can easily check that the former situation leads to higher values of  $W$ . We shall therefore confine ourselves here to this case and consequently omit the sum-

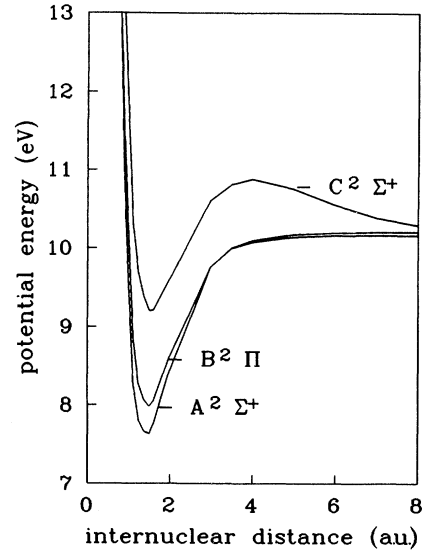


FIG. 2. The potential curves of the excited  $\text{HeH}$  quasi-molecule (from Theodorakopoulos *et al.*, Ref. 14).

mation over the index  $\nu$  in Eq. (16).

For  $\Delta\omega \approx 1000$  K the distance  $r_* \approx 8a_0$  where  $a_0$  is the Bohr radius. Then using the ground-state potential curve from,<sup>15</sup> we obtain  $f(r_*) \approx 0.5$ . As  $\Delta\omega$  is much larger than the fine structure splittings within the  $L_{\alpha}$  manifold we have  $|a(r_*)|^2 \approx 1/2$ . For  $g_1$  and  $g_2$  we shall use the form given by Mantz and Edwards.<sup>16</sup> Then with  $\mathbf{e}(r_*) = \mathbf{r}_*/r_*$  we find for the case when  $\mathbf{p}$  is perpendicular to the surface:  $\Lambda \approx 6 \times 10^{-2}$ . Finally using  $n_{\text{He}} = 2 \times 10^{22}$  cm $^{-3}$  and the  $L_{\alpha}$  transition dipole moment  $d = 0.745ea_0$ , we obtain

$$W \approx 10^{-17} n_s, \quad (19)$$

where  $n_s$  is given in cm $^{-2}$ .

The value of  $W$  may be increased by decreasing  $\Delta\omega$ . Although the quasimolecular approximation for  $\text{HeH}^*$  is no longer strictly valid in this case,  $W$  will remain proportional to  $n_s$  as long as  $\Delta\omega$  is much larger than  $U_{\text{HH}^*}$ . Because the absorption of light leads to the formation of unbound states of  $\text{HeH}^*$  for  $\Delta\omega > 0$ , the  $\text{H}^*$  atom is repelled from the surface and will subsequently radiate luminescence  $L_{\alpha}$  photons. If we assume an incident flux of  $10^9$  photons/10 ns pulse (see Ref. 12) and take  $n_s \approx 10^{13}$  cm $^{-2}$ , corresponding to a KT transition at 0.1 K, it follows from Eq. (19) that the number of luminescence photons will be  $\sim 10^5$ /pulse. It should be possible to detect this luminescence, particularly because the incident light is not scattered by atoms in the bulk due to the large detuning. The luminescent light has a frequency much closer to  $\omega_0$  as the repelled atoms acquire a kinetic energy of the order of  $\Delta\omega$  corresponding to a Doppler shift from  $\omega_0$  of only  $\approx 1$  K. To observe the luminescence, one requires the density in the buffer volume  $n_v$  to be  $\lesssim 10^{15}$  cm $^{-3}$  in order to avoid scattering of these photons from atoms in the bulk. As was shown above this requirement is easily satisfied and hence the surface density can be directly measured through detection of luminescence.

## ACKNOWLEDGMENTS

We wish to acknowledge Professor Yu. Kagan, Professor T. Mizusaki, Dr. H.G.C. Werij, and I. D. Setija for

fruitful discussions. This work was financially supported in part by the 'Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO) and the 'Stichting voor Fundamenteel Onderzoek der Materie (FOM), which is financially supported by NWO.

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