ositron clusters in dense gases

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We demonstrate the possibility of clustering of thermal positrons in dense gases. The region of existence of these self-localized states is adjacent to the "gas-liquid" coexistence curve on the high-temperature side. An expression is obtained for the "critical" temperature of the clustering phenomenon.

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The annihilation of positrons in a medium has been attracting much atten-on.[1] In rarefied gases the annihilation rate λ_e is proportional to the gas den-ty N. In dense gases, however, an increase of N can lead to a jumplike in-'ease of λ_e. In He and Ar, λ_e reached jumpwise values close to those observed a liquid.[2,3] This is connected with the increase of the density of the atoms 'ound the positron—either as a result of the polarization of the medium or as result of the chemical bond. Estimates made in[2,3] do not confirm these ssumptions.

In this paper we demonstrate the existence of positron clusters in He and Ar. hey constitute half-localized states of the fluctuation type,[4] similar to the ubbles" in He[5] and to the electron clusters in Xe.[6] The onset of just clus-rs (and not bubbles as in the case of electron localization in He) is due to the ct that the length L for scattering by atoms of all the inert gases is negative, e., the attraction forces predominate. The region of the existence of clusters limited by its "critical" temperature. Its experimental observation would be undisputed interest.

We assume that localization of positrons e^+ into clusters is possible. Then ıe ratio of the number of free e^+ to the number of localized ones is proportion-l to $\exp(\beta\Delta F)$, where ΔF is the change of the free energy upon localization of ıe positron and $\beta = 1/T$. $\beta\Delta F = -\beta\epsilon + \Delta S$, where ϵ is the e^+ binding energy and S is the change of the entropy of the gas. All the main effects follow from a imple model: 1) the interaction with the atom is described by a Fermi pseudo-otential with scattering length L; 2) a lattice-gas approximation with allowance r the interatomic attraction (the constants b and a) is used for ΔS; 3) the e^+ re localized in a spherical volume of radius R, uniformly filled with atoms of ıaximum possible density equal to $1/b$. For this model we have

$$\beta \Delta F = \pi^2 \lambda_\beta^2 R^{-2} + 4\pi L \lambda_\beta^2 n + \frac{4\pi}{3} R^3 \left[b^{-1} \ln \frac{1}{1-nb} - a n^2 \right]. \qquad (1)$$

'he first term in (1) is the kinetic energy, the second the potential energy, $\beta = \hbar/\sqrt{2mT}$, and $n = b^{-1} - N$ is the excess density of the atoms in the cluster.

The region where the greater part of the positrons is localized is bounded by urve $N_{cl}(T_{cl})$ determined by the system of equations $\partial\Delta F/\partial R = 0$ and $\Delta F = 0$. ỳy solving this equation we obtain $N_{cl}(T_{cl})$ (or $T_{cl}(N_{cl})$) and the dimension R_{cl} f the optimal clusters:

$$\frac{T_{cl}}{T_c} = -\frac{(1-N_{cl}b)^2}{\ln N_{cl}b}\left[\frac{a}{bT_c} + c\frac{|L|^{5/2}\lambda_c^2}{b^{3/2}}\sqrt{1-N_{cl}b}\right],$$ (

$$R_{cl} = \sqrt{\frac{5\pi}{12|L|}\left(\frac{1}{b} - N_{cl}\right)^{-1}},$$

where $c = 16(3/5)^{5/2}\pi^{-3/2}$, $\lambda_c = \hbar/\sqrt{2mT_c}$, and T_c is the critical temperature. Th region of existence of the clusters is bounded, naturally, both on the side of low N and on the side of high ones. It is adjacent to the two-phase region of the gas, having a "critical" temperature $T^* > T_c$. With sufficient accuracy we have

$$\frac{T^*}{T_c} \approx 0.41\left(\frac{a}{bT_c} + 0.85c\,|L|^{5/2}\lambda_c^2 b^{-3/2}\right) \qquad N^* \approx 0.28b^{-1}.$$ (:

The results are valid only for extended clusters, in which the number of atoms is large and the density approaches close-packed, namely when the following conditions are satisfied:

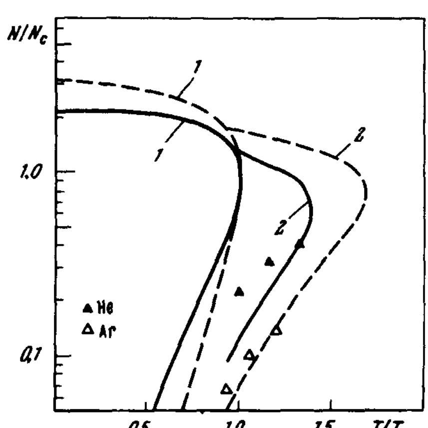
$$|L| << R,\ (4\pi/3)R^3/b >> 1,\quad 1-Nb << Nb\exp[3L\lambda_\beta^2/R^3 - 2a\beta(1-Nb)/b].$$ (4

Let us obtain estimates for Ar and Ne. We express the constant a and b of a lattice gas in terms of N_c and $T_c(b=(2N_c)^{-1},\ a=T_c/N_c)$. We assume $L_{ar} = -3.5a_0$ (according to[7], $-3a \geq L_{Ar} \geq -4a_0$), and $L_{Ne} = -0.6a_0$.[8] For Ar this yields $T^* = 1.6T_c$, i.e., there is a sufficiently wide region of existence of clusters. T^* in Ne practically coincides with T_c, and consequently clusters car exist only in a narrow region. Actually, no anomalies in the rate of annihilation in Ne were observed in[3].

For a quantitative comparison with experiment, calculations were performed on the basis of relations more exact than (1)

$$\Delta F = \frac{1}{2}\int|\nabla\psi|^2 d\mathbf{r} + \int\tilde{V}(\mathbf{r})n(\mathbf{r})d\mathbf{r} + \Delta F_g\{n(\mathbf{r})\}$$ (5

$\psi(\mathbf{r})$ is the wave function of the positron in the cluster, and $\tilde{V}(\mathbf{r}) = \int d\mathbf{r}'V(\mathbf{r}-\mathbf{r}') \times |\psi(\mathbf{r}')|^2$ is the "pseudopotential" of the interaction of e^+ with the atom. In[6],



1—Phase-coexistence curves, 2—clustering curves $N_{cl}(T_{cl})$ solid-He, dashed—Ar. Points—experiment[3].

ıder assumptions that are justified also for the present problem, $\widetilde{V}(\mathbf{r})$ was ›tained with allowance for the short-range and polarization interaction (in (1) was assumed that $\alpha/a_0R|L| \ll 1$, where α is the polarizability of the atom). r) is the local value of n. ΔF_g is the change of the free energy of the medium r fluctuations of $n(r)$. ΔF is first varied with respect to $n(r)$ and then with re- ›ect to $\psi(\mathbf{r})$ on a certain class of trial functions.

The calculation procedure is close to that of[6]. Let us note some differences. F_g was written for the Van der Waals model with experimental values $a = 4$ 10^{-36} erg-cm^3 and $b = 4.5\times10^{-23}$ cm^3 for Ar and with $b = 2.4\times10^{-23}$ cm^3 for He, ith a strongly dependent on the density, $a(N) = 8.4\times10^{-38}\times(1-4.3\times10^{-46}N^2)$:g-cm^3.[9] The polarization of the Ar atoms in the cluster was taken into ac- ›unt by replacing L with $L[1+(8\pi/3)\alpha(n+N)]^{-1}$. The results of the calculation :e compared with experiment in the figure, ($L = -0.44a_0$ for He[10]).

Let us indicate certain characteristics of the position clusters. In He, at $= 7$ °K, their dimension is $25a_0$ and they consist of ~ 300 atoms with a binding ıergy ~ 0.1 eV. The density in the cluster is close to $2.5N_c$, i.e., of the order f the density of a liquid. Allowance for surface effects could change the region ›unded by the $N_{cl}(T_{cl})$ curve, primarily on its lower branch.

We propose that measurement of the annihilation times of slow positrons ver a wider temperature range will reveal the existence of the "critical" tem- erature T^*. For heavy inert gases (Kr, Xe) the values of T^*/T_c should be ırger than for Ar.

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