# SCATTERING OF 0.15-1.8-eV ARGON ATOMS FROM A GERMANIUM SURFACE

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The interaction of neutral atoms and molecules with solid surfaces at energies from thermal to a few electron volts has been studied very little, as often noted in the literature [1].

In experiments reported in [2-4], which encompass the energy range indicated, only the angular distributions of the intensity of the scattered atoms were studied; their velocities were not measured. Measured velocities are given in [5] but they have too large a spread to enable any definite conclusions to be drawn. Data on the effect of a layer of adsorbed gas on the angular distributions of intensity and velocity are also lacking.

We report on experimental studies of the scattering of 0.15-1.8-eV argon atoms from a germanium surface for various states of an adsorbed layer. Based on the angular distributions of the intensity and the average velocity of the scattered atoms, estimates were made of the energy accommodation coefficients. Where possible comparisons were made with current simple models of the interaction of atoms with solid surfaces.

The experiments were performed in a vacuum chamber with a gasdynamic molecular beam source at a residual gas pressure of $\sim 5 \cdot 10^{-6}$ mm Hg. The experimental arrangement and the signal storage system are described in [6]. Argon atoms were accelerated in Ar–He and Ar–$H_2$ mixtures to energies of 0.2 to 1.8 eV. The Ar component was separated from the mixture by recording excited $Ar^*$ atoms formed in the bombardment of the molecular beam by a transverse beam of electrons. A secondary-electron multiplier was used to detect the excited atoms. The parameters of the system for exciting and recording $Ar^*$ atoms were chosen so that the main contribution to the recorded signal came from $Ar^*$ atoms [7]. The molecular beam was modulated with a mechanical chopper for time-of-flight measurements of the parameters of the primary and scattered beams. To decrease the error in the determination of the velocity each measurement was performed twice for opposite directions of rotation of the chopper.

The chemically polished (111) surface of a single crystal of Ge was used as a target. The distance from the target to the center of the excitation region, which was 20 mm wide, was 100 mm; the distance from the target to the first VÉU dynode was 200 mm, and the angular resolution of the detector was $\approx 2.5°$.

The results were obtained in the form of angular distributions of intensity (scattering patterns) and the average velocity of the scattered beam. The amplitude of the time-of-flight signal was used as a measure of the intensity, and the average velocity (energy) was determined from the position of the maximum of the signal. All the measurements were performed in the plane passing through the axis of the incident beam and the normal to the target. The angle of incidence $\beta$ and the angular position $\gamma$ of the detector were measured from the normal.

The accelerating gas used in the experiments was $H_2$ or He. Originally, the preference was given to He, since it was assumed that the chemically more active $H_2$ would be better adsorbed on the target and would distort the scattering pattern from the clean crystal. During the experiments, however, it was found that the nature of the scattering for the same Ar energy did not depend on whether an Ar–He or Ar–$H_2$ mixture was used. Therefore, $H_2$ was most frequently used as an accelerating gas since a higher Ar energy was reached with it. In addition, the signal from excited $H_2^*$ was considerably lower than the signal from $He^*$, and at least two orders of magnitude lower than the $Ar^*–H_2^*$ signal. This enabled us to neglect the contribution of the accelerating gas to the total signal from the mixture.

The scattering of Ar from a germanium surface was studied at target temperatures $T_T$ = 20 and 600°C. These correspond to different states of the adsorbed layer: at 20°C the target is covered with an adsorbed lay-

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er, while at 600°C the adsorbed layer exerts a negligible effect on the scattering pattern from a clean target. This conclusion was based on a study of the scattering of thermal beams of Ar and He [6] from germanium for various $T_T$. In particular, it has been established that the shape of the scattering pattern changes from diffuse at 20°C to lobular at 400°C.

A comparison of these data with the results of others (e.g., [8]), in which Auger spectroscopy and low-energy electron-diffraction techniques were used to monitor the state of the surface, shows that diffuse scattering at $T_T$ = 20°C is due to the presence of an adsorbed layer, while lobular scattering at 600°C corresponds to a target without an adsorbed layer. The destruction of the adsorbed layer occurs at lower $T_T$ (300-500°C) also, but the time to reach a stationary scattering regime for $T_T$ < 500°C is rather long (~ 1 h). Therefore, we cleaned the surfaces by heating them to 600°C; at this temperature the time to reach a stationary regime was a few minutes.

The scattering patterns obtained for $T_T$ = 20°C for various beam energies $E_0$ show that as $E_0$ is increased the scattering does not become completely diffuse but shows an appreciable lobular component in a direction between the specular beam and the target surface. As $E_0$ is increased the relative value of the lobular component in the pattern is increased, and its maximum is displaced toward the surface.

The angular distributions of the average velocity $v(\gamma)$ depend critically on $E_0$. Figure 1 shows curves for various values of $E_0$: 4) 0.18 eV; 2) 1.4 eV for $T_T$ = 20°C and $\beta = 45°$. For small $E_0$ corresponding to a diffuse scattering pattern the value of the average velocity is practically the same for all angles. A pronounced difference from a uniform distribution begins to appear for $E_0 > 0.3$ eV.

We estimate the loss of energy of Ar atoms scattered in various directions by using the differential accommodation coefficient $\alpha(\gamma)$,

$$\alpha(\gamma) = [v_0^2 - v^2(\gamma)]/[v_0^2 - v_p^2],$$

where $v_0$ is the average velocity of the incident atoms, $v(\gamma)$ is the average velocity of the scattered atoms and depends on direction, $v_p = \sqrt{2kT_T/m}$ is the most probable velocity corresponding to the target temperature $T_T$, and m is the mass of an Ar atom. Two values of $\alpha$ were determined: $\alpha_m$ for the direction corresponding to the maximum of the scattering pattern and $\alpha_n$ corresponding to the normal to the target. Curves 1 and 3 of Fig. 2 show the dependence of $\alpha_n$ and $\alpha_m$ on $E_0$ for $\beta = 45°$ and $T_T$ = 20°C. Near the normal to the target $\alpha(\gamma) \sim 1$ for all $E_0$, and the scattering patterns for $\gamma < 20°$ are closely described by cos $\gamma$. It follows from this that close to the normal direction the Ar atoms are scattered diffusely. Consequently, at least for a qualitative explanation of scattering phenomena, we are justified in dividing the scattering into diffuse and lobular components with different accommodation coefficients $\alpha_m$ and $\alpha_n$.

For $T_T$ = 600°C and $\beta = 45°$ lobular scattering patterns with a negligible diffuse component are found for all $E_0$. Curves 3 and 1 of Fig. 1 show v as a function of $\gamma$ for $E_0$ = 0.40 and 1.7 eV, respectively. It is clear that for small $E_0$ there is a maximum of the velocity in the direction of the maximum of the scattering pattern. For $E_0 > 0.5$ eV there is no such maximum and the velocity constantly increases as the detector is displaced toward the target surface.

Curves 2 and 4 of Fig. 2 show the dependence of $\alpha_n$ and $\alpha_m$ on $E_0$ for $T_T$ = 600°C and $\beta = 45°$. The relatively large spread of points for $\alpha_m(E_0)$ is due to the small difference in times of flight of the direct and scattered beams. The value of $\alpha_n$ is close to unity as it is for $T_T$ = 20°C. This means that even in this case there is a diffuse component in the scattering pattern.

The difference in the values of $\alpha_m$ for $T_T$ = 600 and 20°C can be explained by the following model of the interaction of Ar atoms with a target covered by an adsorbed layer. We assume that an incident atom first collides with one adsorbed particle and loses part of its energy and then is scattered from the surface of the lattice, losing the same fraction of its energy as in scattering from a clean surface, i.e., at $T_T$ = 600°C. We consider a collision of an incident and an adsorbed particle under the following assumptions: 1) both particles are structureless; 2) the adsorbed particle is free; 3) the velocity of the incident particle before collision is much larger than the velocity of thermal motion of the adsorbed particle. After collision with an adsorbed particle an atom of Ar has an energy [9] $E_1 = (1-\xi)E_0$, where $\xi = 2\ mm_1/(m+m_1)^2$, m is the mass of the incident atom, $m_1$ is the mass of the adsorbed particle, and $E_0$ is the energy of the incident atom. After scattering from the crystal target with an energy accommodation coefficient $\alpha_m$ the energy of the scattered atom is

$$E = E_0(1-\xi)(1-\alpha_m) + \alpha_m E_s.$$

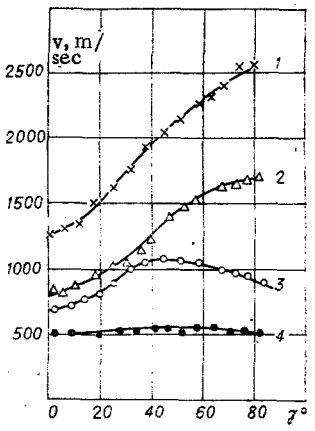


Fig. 1

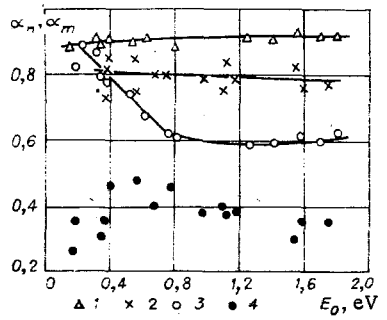


Fig. 2

where $E_s$ is the energy of atoms scattered in equilibrium with the target temperature $T_T$. The effective accommodation coefficient $\alpha'$ for successive interactions of an Ar atom with an adsorbed molecule and with the target has the form

$$\alpha' = (E_0 - E)/(E_0 - E_s) = [1 - (1 - \xi)(1 - \alpha_m) - \alpha_m E_s E_0]/(1 - E_s E_0). \quad (1)$$

The adsorbed layer can come from the Ar gas under study, the He or $H_2$ used as an accelerating gas, or the residual gas in the working chamber. According to [10], He is hardly adsorbed on germanium at $T_T > 25°C$, and $H_2$ is very weakly adsorbed. The mass spectrum of the residual gas has the following components in order of decreasing peaks: $H_2O$, CO, $N_2$, $CO_2$, etc. It follows from [9] that CO and $N_2$ are hardly adsorbed on germanium at room temperature, and $CO_2$ is adsorbed very weakly. $O_2$ is adsorbed well on germanium but there is very little of it in the residual gas. Molecules of $H_2O$ are also well adsorbed; the energy of adsorption of $H_2O$ in the formation of a monolayer is 0.74 eV and the desorption temperature is about 350°C. Consequently, the adsorbed layer is most probably composed of $H_2O$ molecules from the residual gas. This is confirmed by the value of the temperature $T_T \sim 300$-500°C at which the adsorbed layer is destroyed.

Assuming that the adsorbed layer is formed of $H_2O$ molecules ($m_1 = 18$ amu), and using $\alpha_m = 0.4$ for a clean target (curve 4 of Fig. 3) we find from Eq. (1) $\alpha' = 0.65$ for an incident Ar atom (m = 40 amu). The calculated value of $\alpha'$ is close to the value $\alpha_m = 0.60$ measured for $T_T = 20°C$ for $E_0 > 0.8$ eV. It would probably be more correct to take $\alpha_m < 0.4$ for the calculation, since the energy accommodation coefficient on a clean target decreases as $T_T$ is decreased. This leads to a certain decrease in the calculated value of $\alpha'$.

If we assume that $O_2$ is adsorbed on the target, $\alpha' = 0.7$, and for $H_2$, $\alpha' = 0.45$.

For $E_0 < 0.8$ eV an Ar atom may make more than one collision with adsorbed $H_2O$ molecules, resulting in an increase in the relative loss of energy of Ar atoms. This can be accounted for by the increase in $\alpha_m$ for a decrease in $E_0$ from 0.8 to 0.2 eV for $T_T = 20°C$.

We note that the nature of the curve for $\alpha_m(E_0)$ shown in Fig. 2 for $T_T = 20°C$ agrees qualitatively with the relation calculated in [11] for the energy accommodation coefficient as a function of $E_0$ for particles interacting with a solid surface covered with an adsorbed layer.

The basic parameters of the scattering pattern from a clean target as a function of $E_0$ were investigated also. These can be used to determine the interaction regimes. In [2-4] the correlation parameters for determining the beginning of the transition from thermal to structural scattering were taken as the position of the maximum of the $\Delta\gamma(E_0)$ curve or the position of the minimum of the $\Delta\varphi(E_0)$ curve, where $\Delta\gamma = \gamma_m - \beta$ is the angle by which the maximum of the pattern is displaced from the specular beam, and $\Delta\varphi$ is the width of the pattern at half-maximum, henceforth called the half-width. Figure 4 shows $\Delta\gamma$ and $\Delta\varphi$ as functions of $E_0$ for $\beta = 45°$ and $T_T = 600°C$. The graph of $\Delta\gamma$ as a function of $E_0$ shows a transition from supraspecular (between the specular beam and the normal to the target) to subspecular scattering. The maximum value of $\Delta\gamma$ is $\approx 19°$, and in the energy range investigated shows no tendency to decrease with increasing $E_0$ as in [2-4] in the study of the scattering of Ar from the (111) surface of Ag. In addition, our value of $\Delta\gamma$ is somewhat larger than the maximum values $\Delta\gamma = 5$-6° obtained in [2-4]. This seems to be related to the larger transfer of normal momentum in the

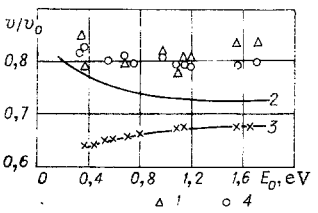


Fig. 3

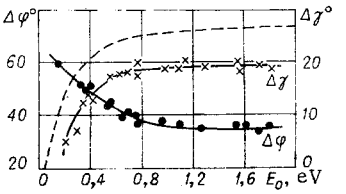


Fig. 4

Ar–Ge system than in the Ar–Ag system, since the atomic weight of Ge (72.6 amu) is less than that of Ag (108 amu).

The graph of the half-width of the scattering pattern $\Delta\varphi$ as a function of $E_0$ approaches a straight line asymptotically, in contrast with the results in [2-4] where after reaching a minimum the half-width again increases. This may be a consequence of the fact that structure scattering is not achieved for the Ar–Ge system in the range of $E_0$ values investigated.

A number of experiments on clean targets were performed for angles of incidence $\beta = 30$ and 60°. The nature of the $\Delta\varphi(E_0)$ and $\Delta\gamma(E_0)$ relations remains the same as for $\beta = 45°$. As $\beta$ is increased at constant $E_0$ the values of $\Delta\varphi$ and $\Delta\gamma$ decrease. The experiments showed that as the angle of incidence is increased $\alpha_n$ remains constant, while $\alpha_m$ decreases for all $E_0$. This agrees qualitatively with calculations given in [10].

Our experimental results were compared with those given by the hard-cube model [12] and the hard-sphere model [13]. We chose the following parameters for the comparison: the half-width of the scattering pattern $\Delta\varphi(E_0)$, the angle of deviation of the maximum from the specular direction $\Delta\gamma(E_0)$, and the value of the velocity in the direction of the maximum of the scattering pattern $v_m$. Comparison showed that neither of these models gives a complete explanation of all the results obtained, although the hard-cube model describes some of the results qualitatively correctly.

The open curve of Fig. 4 shows $\Delta\gamma(E_0)$ for $\beta = 45°$ calculated with the hard-cube model. The calculated curve has nearly the same shape as the corresponding experimental curve, although there is an appreciable quantitative difference between them.

Figure 3 shows the dependence of the velocity ratio $v_m/v_0$ on $E_0$; points marked 1 are the experimental results; curve 2 was calculated with the hard-cube model; curve 3 was calculated with the hard-sphere model; $v_0$ is the velocity of the incident beam. For all values of $E_0$ the measured values $v_m/v_0$ are close to those calculated with the hard-cube model. Better agreement between the measured and calculated velocities can be obtained by taking the position of the maximum of the scattering pattern from experiment and using the fact that in the hard-cube model the tangential component of momentum is conserved in reflection. In this case $v_m/v_0 = \sin\beta/\sin\gamma_m$, where $\gamma_m$ is the angular position of the maximum of the pattern. The values of $v_m/v_0$ shown by points marked 4 in Fig. 3 were calculated with the above formula for $\beta = 45°$ and are in satisfactory agreement with experiment. Similar agreement is found for $\beta = 30$ and 60°.

We note that the half-width of the scattering pattern for both models is appreciably different from the experimental value. Neither model gives the evolution of the angular distribution of the average velocity with increasing energy of the incident atoms.

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# ESTIMATE OF THE AMPLITUDES OF THE FIELDS CREATED BY AN UNSTEADY GAMMA SOURCE

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It is known [1-4] that an unsteady gamma source gives rise to an electromagnetic field in the surrounding space. Most of the studies of the characteristics of such fields have been performed in the approximation which is linear in the field [1-3]. An exception is [4] in which the slowing down of Compton electrons by the electric field is taken into account. It follows from [1, 2] that the characteristic scale of the fields created close to the source is of the order of $3 \cdot 10^4$ V/m.* Although this value is appreciably lower than the value of breakdown fields in air, electric discharges are observed [5] in the vicinity of a gamma source, indicating the presence of substantially larger fields. One effect not taken into account in the latter approximation which could lead to an increase in the field is the increase in electron termperature due to the electric field [6]. On the one hand, this decreases the electron mobility and consequently also the conductivity of the system. On the other hand, it is known that the electron attachment coefficient $\gamma$ for electronegative molecules strongly affects the characteristics of electric fields and depends on the electron energy. Therefore, the electron balance equation must take account of the dependence of $\gamma$ on the electric field through the electron energy, and this leads to a further change in conductivity. We take account of these effects on the shaping of electric fields in air in the vicinity of the source. It is assumed that electron lifetimes are determined solely by their attachment to molecules. This is a good approximation for air pressures near normal [1-3].

Let us consider the dependence of the electron energy and mobility on the intensity of the electric field. It is shown in [4] that if the electron thermalization time $\tau = 1/\nu\delta$, where $\nu$ is the frequency of collisions of electrons with gas molecules and $\delta$ is the average relative loss of energy of an electron in a collision, is very much shorter than the characteristic times determining the shaping of electric fields, the processes are quasistatic. In this case one can assume that the electron energy $\varepsilon$ at a given instant is determined by the electric field E at that same instant. The relation between these quantities for $\delta = \text{const}$ and $\nu = \nu_0\sqrt{\varepsilon/\varepsilon_0}$ is derived in [6] and has the form

*A similar value is obtained also from the results of [1] for proper values of the physical constants.

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