Dynamic-interaction effects in the decay of fast molecules in thin films

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(Submitted 20 July 1977)
Zh. Eksp. Teor. Fiz. **74**, 288–299 (January 1978)

The problem of the ions that fly apart in a "Coulomb explosion" of a fast diatomic molecule passing through a solid film is analyzed. The relative roles of the stopping forces and of the "wake" forces due to the polarization of the medium are assessed. It is shown that in the case of a film of small thickness all the singularities of the particle distribution in angle and in energy are determined by the specific angular momentum acquired under the influence of these forces by the particles that move apart, as well as by the non-inertial character of the motion of the mass center of the cluster in the medium. The result is a strong anisotropy of the angular distribution, with peaks of varying intensities for particles emitted parallel and anti-parallel to the primary beam in the c.m.s. Analytic expressions in closed form are obtained, which demonstrate in explicit form the dependence on the masses and the charges of the decay particles, and explain the pattern of the phenomena observed in experiments by Gemmell *et al.* [Phys. Rev. Lett. **34**, 1420 (1975); Nucl. Instrum. Methods. **132**, 61 (1976); Phys. Rev. A14, 638 (1976); Phys. Rev. Lett. **37**, 1352 (1976); Informal Workshop on Wake Phenomena, Summary Report by G. Lapitcki and S. Stern, New York University, 1977, p. 3].

PACS numbers: 61.80.Lj, 68.90.+g

1. INTRODUCTION

Many interesting experiments have been recently performed[1-4] on the correlated motion of the decay products of fast molecular ions passing through thin films. The least trivial of the observed effects turned out to be the singularities of the angular distribution, with a clearly pronounced asymmetry for particles emitted parallel and antiparallel to the motion of the primary beam in the c.m.s. The physics of phenomena of this type can be understood in principle within the framework of the picture of the dynamic interaction of diverging particles via the so-called "wake" potential produced as a result of polarization of the solid-state electron plasma.[1-4]

Interest in this group of problems has greatly increased recently, particularly in connection with problems of the energy loss of proton clusters[5,6] and bound states of electrons in the wake potential.[7-11] However, no direct analysis has been made so far of the correlated dispersal of the ions that fly apart upon decay of a fast diatomic molecule in a thin film. This is the subject of the present paper.

When a molecule of energy $E \gtrsim 100$ keV/nucleon enters a solid film, the bound electrons are stripped at atomic thicknesses[1,2] and the so-called "Coulomb explosion" of the molecule sets in, i.e., the ions move apart under the influence of the repulsion forces that act between them. The main physical feature of the problem, which determines to a substantial degree all the angular dependences, is that the pair of diverging particles acquires an angular momentum in the film.

Assume that at the instant that an ion pair starts to fly apart it has no angular momentum and the axis of the cluster makes an angle φ_0 with the direction of the motion (the z axis). The angular distribution behind the film will then be determined by the dependence of the final l.s. angle ψ of the particle emission on φ_0, and by the fact that the distribution of the molecules over the initial angle φ_0 is equally probable. After a time t of travel through the film, the angle φ of the cluster orientation in the c.m.s. changes:

$$\varphi(t)=\varphi_0+\Delta\varphi_{in}(\varphi_0, t)$$

and the pair of particles acquires simultaneously, a certain angular momentum $M=M(\varphi_0, t)$, as the result of which the angle φ along the path from the film to the detector again increases by an amount $\Delta\varphi_{out}(\varphi_0, t)$. For bare particles with atomic numbers Z_1 and Z_2 this amount is determined by the known formula from the theory of the motion of a particle in a Coulomb field (see, e.g., [12]):

$$\Delta\varphi_{out}(\varphi_0, t)=\int_{r_1}^{\infty}\frac{M\,dr}{r^2}\left[2\mu\left(E_1-\frac{Z_1Z_2e^2}{r}\right)-\frac{M^2}{r^2}\right]^{-1/2}, \tag{1.1}$$

where μ is the reduced mass and E_1 and r_1 are respectively the energy of the relative motion and the distance between the particles at the exit from the film.

Another important circumstance is that on changing to the l.s. it is necessary to take into account, besides the trivial change in the angle scale, also the non-inertial character of the motion of the mass center of the cluster in the medium under the influence of the unequal forces that act on the different particles of the pair.

2. EQUATIONS OF MOTION

If the particles that fly apart are bare nuclei with masses m_1 and m_2, then the equations of the relative motion ($r=r_2-r_1$) and of the motion of the mass center $\mathbf{R}$ in the film are

$$\mu\ddot{\mathbf{r}}=\frac{Z_1Z_2e^2}{r^2}\frac{\mathbf{r}}{r}+\mu\left(\frac{Z_2^2}{m_2}-\frac{Z_1^2}{m_1}\right)\mathbf{F}^s+\mu Z_1Z_2\left[\frac{\mathbf{F}_{12}^w(\mathbf{r})}{m_2}-\frac{\mathbf{F}_{21}^w(\mathbf{r})}{m_1}\right] \tag{2.1}$$

$$(m_1+m_2)\ddot{\mathbf{R}}=(Z_1^2+Z_2^2)\mathbf{F}^s+Z_1Z_2[\mathbf{F}_{12}^w(\mathbf{r})+\mathbf{F}_{21}^w(\mathbf{r})]. \tag{2.2}$$

 0038-5646/78/4701-0148$02.40

Here $\mathbf{F}^s$ is the stopping force that acts on the particle with a unity atomic number in the direction of the negative z axis, $Z_1Z_2\mathbf{F}^w_{ik}(\mathbf{r})$ is the wake force produced by the ith particle and acting on the kth particle ($F^w_{12} \neq -F^w_{21}$).

From the form of Eqs. (2.1) and (2.2) we can draw the following conclusions:

(a) The action of the wake potential on the relative motion is not symmetrical to the interchange $m_1 \rightleftarrows m_2$. An analysis of the angular dependences of the wake forces (see Sec. 4 for details) shows that the corresponding torque is due primarily to the force $\mathbf{F}^w$ that acts in the wake of the front particle. Combining these facts, we see that the torque of the wake forces is stronger the smaller the mass of the rear particle.

(b) The action of the stopping forces as a function of the ratio of the parameters Z_1^2/m_1 and Z_2^2/m_2 can both help and hinder the rotation due to the weight potential. Since $\mathbf{F}^s$ has the same origin and is of the same order as $\mathbf{F}^w$, a suitable choice of the parameters can enable the resultant torque, in principle, to counteract the wake forces and increase the angle between $\mathbf{r}$ and the z axis. In particular cases, as for the $^4HeH^+$ ions, the term with the stopping force drops out completely from (2.1) and the cluster rotates only under the influence of the wake potential.

(c) Since the wake forces $\mathbf{F}^w_{ik}$ have components perpendicular to the z axis, the solution of Eq. (2.2) leads to the appearance of a transverse velocity component of the cluster mass center. Allowance for this component on changing to the l.s. corresponds, in the case of the real particle, to additional rotation towards the z axis, and in the case of the front particle, on the contrary, to repulsion away from this axis, (we consider films that are thin enough so that the time of flight t is too short for the cluster axis to turn parallel to the z axis).

3. APPROXIMATION LINEAR IN t. THE OLD SYSTEM OF COORDINATES

We trace the action of the effects listed above, using as an example motion in a homogeneous and isotropic electron plasma. In this case the problem, in view of the axial symmetry, is greatly simplified and reduces to the problem of particle motion in a plane passing through the symmetry axis z. Assume that at the instant of Coulomb explosion we have $r=r_0$ and $\dot{r}=0$. For films of sufficiently small thickness, corresponding to a time of flight

$$t \ll t^* = 2r_0/v^*, \tag{3.1}$$

where

$$v^* = (2Z_1Z_2e^2/\mu r_0)^{1/2}, \tag{3.2}$$

all the effects can be considered in an approximation linear in t.

The equation for the angular momentum $M=\mu r^2\dot{\varphi}$ of the pair particles moving in a film takes the form

$$dM/dt = -\partial U(r,\varphi)/\partial\varphi, \tag{3.3}$$

where

$$U(r,\varphi)=\mu\left(\frac{Z_2^2}{m_2}-\frac{Z_1^2}{m_1}\right)F^s r\cos\varphi+\mu Z_1Z_2\left[\frac{U_{21}^w(r,\varphi)}{m_1}+\frac{U_{12}^w(r,\varphi)}{m_2}\right] \tag{3.4}$$

is the effective potential of the dynamic polarization interaction between the particles that move apart, and depends on the polar coordinates r and φ in the plane of particle motion (a direct Coulomb interaction obviously does not lead to a change in the angular momentum). Here $F^s=|\mathbf{F}^s|=\text{const}$, and $U^w_{ik}(r,\varphi)$ are the potentials corresponding to the wake forces $\mathbf{F}^w_{ik}$; the angle φ is reckoned from the direction of the z axis.

Eq. (3.3) shows that at small t we have $\Delta\varphi_{in}(\varphi_0,t)\sim t^2$ and should be omitted. For the angular momentum $M(\varphi_0,t)$ acquired by the particle pair on moving through the film we have, on the other hand,

$$M(\varphi_0,t)=-t\frac{\partial}{\partial\varphi_0}U(r_0,\varphi_0).$$

From (1.1) it follows then directly that in the approximation linear in t we have

$$\Delta\varphi_{out}(\varphi_0,t)=-\frac{2t}{\mu r_0 v^*}\frac{\partial}{\partial\varphi_0}U(r_0,\varphi_0) \tag{3.5}$$

and consequently the connection between the final (φ) and initial (φ_0) orientation angles of the pair of particles in the c.m.s. will be determined by the relation

$$\varphi(\varphi_0,t)=\varphi_0+\Delta\varphi_{out}(\varphi_0,t)=\varphi_0-\frac{2t}{\mu r_0 v^*}\frac{\partial}{\partial\varphi_0}U(r_0,\varphi_0). \tag{3.6}$$

Determination of $\varphi_0(\varphi)$ from (3.6) and its substitution in the formula $df=d\Omega_{\varphi_0}/4\pi$ would make it possible to calculate the distribution of the particles with respect to the angle φ. However, the distribution obtained in this manner cannot be interpreted as the angular distribution of the particles in the c.m.s., because the noninertial character of the mass-center motion in the medium causes the velocity of the c.m.s. to be dependent on 0 after the passage through the film, so that c.m.s. that correspond to different initial values of φ_0 are not identical.

Consider for the sake of argument particles of mass m_2 which have already passed through the film, and let E_{min} and E_{max} be respectively the minimal and maximal energy of the particles of this type in the l.s. We separate from them a group of particles that have in the l.s. an energy $\bar{E}=\frac{1}{2}(E_{min}+E_{max})$, and introduce a reference frame in which the particles of the separated group travel perpendicular to the direction of incidence of the primary beam of molecules. We shall call this reference frame the old-system (o.s.). The annular distributions of the decay particles as functions of the energy E and of the emission angle ψ in the l.s., which were experimentally investigated in [2-4], are precisely the ones that reveal the properties of the angular distributions of the particles in the o-systems (see Sec. 5 below).

It can be shown (and this will be seen from the results of Sec. 5) that within the framework of the approximation linear in t the o-system coincides with the c.m.s. of a cluster oriented from the very outset along the incidence direction (along the z axis). Operating in this system, we designate the emission angle of the particles with mass m_2 relative to z by χ. Let furthermore $p_{\parallel c}(\varphi_0, t)$ and $p_{\perp c}(\varphi_0; t)$ be respectively, in the o-system, the longitudinal and transverse (relative to the same axis) components of the momentum of the mass center of the cluster whose initial orientation is given by the angle φ_0. Then the connection between the angles χ and φ_0 will obviously be

$$\tan\chi=\frac{m_1 v_r(\varphi_0, t)\sin\varphi(\varphi_0, t)+p_{\perp c}(\varphi_0, t)}{m_1 v_r(\varphi_0, t)\cos\varphi(\varphi_0, t)+p_{\parallel c}(\varphi_0, t)}. \tag{3.7}$$

Here $v_r(\varphi_0, t)$ is the relative velocity of the diverging particles at infinity, and $\varphi(\varphi_0, t)$ is the angle between the vector $\mathbf{v}_r(\varphi_0, t)$ and the z axis.

In the approximation linear in t, the velocity $v_r(\varphi_0, t)$ turns out to be independent of φ_0 or of t, and is simply given by

$$v_r(\varphi_0, t)=v^* \tag{3.8}$$

[see (3.2)], while the angle $\varphi(\varphi_0, t)$ is defined by (3.6). For the quantity $\Delta\varphi_{\text{out}}(\varphi_0, t)$ (3.5) we then get the inequality

$$|\Delta\varphi_{out}(\varphi_0, t)|\ll\varphi_0. \tag{3.9}$$

The validity of this relation in the region $\varphi_0 \gtrsim 1$ is ensured by the fact that $\Delta\varphi_{\text{out}} \sim t/t^*$. On the other hand, in the region of small angles φ_0 the properties of the wake potentials (see Sec. 4 below) lead to $\Delta\varphi_{\text{out}}(\varphi_0, t) \sim \varphi_0 t/t^*$ and, consequently, the inequality (3.9) is satisfied in that region, too.

Substituting (3.6) and (3.8) in (3.7) and taking (3.9) into account, we have

$$\tan\chi=\frac{m_1 v^*[\sin\varphi_0+\Delta\varphi_{out}(\varphi_0, t)\cos\varphi_0]+p_{\perp c}(\varphi_0, t)}{m_1 v^*[\cos\varphi_0-\Delta\varphi_{out}(\varphi_0, t)\sin\varphi_0]+p_{\parallel c}(\varphi_0, t)}. \tag{3.10}$$

It follows from (3.10) that in the considered approximation we have

$$\varphi_0(\chi)=\chi+\delta(\chi, t), \tag{3.11}$$

where

$$\delta(\chi, t)=-\Delta\varphi_{out}(\chi, t)-\frac{1}{m_1 v^*}[p_{\perp c}(\chi, t)\cos\chi-p_{\parallel c}(\chi, t)\sin\chi]. \tag{3.12}$$

The quantity $p_{\perp c}(\chi, t)\cos\chi - p_{\parallel c}(\chi, t)\sin\chi$, in the last expression can be expressed directly in terms of the derivatives of the wake potentials, we use an equation analogous to (2.2) for the motion of the mass center of the cluster in the o.s. Simple calculations yield

$$p_{\perp c}(\chi, t)=-t\left[\sin\chi\frac{\partial V(r_0,\chi)}{\partial r_0}+\frac{\cos\chi}{r_0}\frac{\partial V(r_0,\chi)}{\partial\chi}\right], \tag{3.13}$$

$$p_{\parallel c}(\chi, t)=-t\left[\cos\chi\frac{\partial V(r_0,\chi)}{\partial r_0}-\frac{\sin\chi}{r_0}\frac{\partial V(r_0,\chi)}{\partial\chi}\right] \tag{3.14}$$

whence

$$p_{\perp c}(\chi, t)\cos\chi-p_{\parallel c}(\chi, t)\sin\chi=-\frac{t}{r_0}\frac{\partial}{\partial\chi}V(r_0,\chi). \tag{3.15}$$

Here

$$V(r, \varphi)=Z_1Z_2[U_{12}^w(r, \varphi)-U_{21}^w(r, \varphi)+F^w r\cos\varphi], \tag{3.16}$$

$$F^w=-\frac{\partial}{\partial r_0}[U_{12}^w(r_0, 0)-U_{21}^w(r_0, 0)]=\text{const}. \tag{3.17}$$

The use of relations (3.5), (3.15), (3.4), and (3.16) makes it possible to express $\delta(\chi, t)$ [Eq. (3.12)] in the following final form:

$$\delta(\chi, t)=\frac{t}{\mu r_0 v^*}\left[2\frac{\partial U(r_0,\chi)}{\partial\chi}+\frac{\mu}{m_1}\frac{\partial V(r_0,\chi)}{\partial\chi}\right]$$
$$=\frac{t}{r_0 v^*}\left\{Z_1Z_2\left[\frac{1}{m_1}\frac{\partial U_{21}^w(r_0,\chi)}{\partial\chi}+\left(\frac{2}{m_2}+\frac{1}{m_1}\right)\frac{\partial U_{12}^w(r_0,\chi)}{\partial\chi}\right]\right.$$
$$\left.-r_0\sin\chi\left[2\left(\frac{Z_2^2}{m_2}-\frac{Z_1^2}{m_1}\right)F^s+\frac{Z_1Z_2}{m_1}F^w\right]\right\}. \tag{3.18}$$

Knowledge of the function $\varphi_0(\chi)$ (3.11) enables us to obtain directly the angular distribution $df/d\Omega_\chi$ of the decay particles with mass m_2 in the o.s. In the general case

$$\frac{df}{d\Omega_\chi}=\frac{1}{4\pi}\frac{\sin\varphi_0(\chi)}{\sin\chi}\left|\frac{d\varphi_0(\chi)}{d\chi}\right|.$$

In the approximation linear in t, this formula becomes

$$\frac{df}{d\Omega_\chi}=\frac{1}{4\pi}\left[1+\delta(\chi, t)\operatorname{ctg}\chi+\frac{\partial}{\partial\chi}\delta(\chi, t)\right]. \tag{3.19}$$

Substitution of (3.18) and (3.19) yields in fact the sought particle angular distribution.

4. ANGULAR DISTRIBUTIONS OF THE DECAY PARTICLE FOR CASES OF FORWARD AND BACKWARD EMISSION IN THE o.s.

To reveal the singularities and the differences of the angular distributions of the decay particles emitted in the o.s. parallel and antiparallel to the direction of motion of the primary molecule beam, we consider the distribution of $df/d\Omega_\chi$ in two regions, $\chi \ll 1$ and $\pi-\chi \ll 1$. From the symmetry of the problem it follows that the wake potential $U_{21}^w(r_0, \chi)$ can be represented in these regions by a series of the form

$$U_{21}^w(r_0, \chi)=U_{21}^w(r_0, 0)+r_0F^s\chi^2(\alpha_1+\beta_1\chi^2)+\ldots \quad \text{if} \quad \chi\ll 1, \tag{4.1}$$

$$U_{21}^w(r_0, \chi)=U_{21}^w(r_0, \pi)+r_0F^s(\pi-\chi)^2[\alpha_2+\beta_2(\pi-\chi)^2]+\ldots \text{if} \quad \pi-\chi\ll 1. \tag{4.2}$$

The factor r_0F^s is introduced in (4.1) and (4.2) from symmetry considerations.

The expansions for the potential $U_{12}^w(r_0, \chi)$ in the considered regions of χ follow from (4.1) and (4.2) if we use the relation

$$U_{12}^w(r, \chi)=U_{21}^w(r, \pi-\chi).$$

Using these expansions in (3.18) and (3.19), we easily obtain for the angular distribution of the particles of mass m_2 in the region $\chi \ll 1$

$$\frac{df}{d\Omega_\chi}=\frac{1}{4\pi}(1+\lambda_1+\nu_1\chi^2),\quad \chi\ll 1, \tag{4.3}$$

where

$$\lambda_1=4\gamma\frac{t}{t^*}\mu\left[\frac{\alpha_1}{m_1}+\alpha_2\left(\frac{1}{m_1}+\frac{2}{m_2}\right)-\frac{1}{Z_1Z_2}\left(\frac{Z_2^2}{m_2}-\frac{Z_1^2}{m_1}\right)-\frac{F^w}{2F^s m_1}\right], \tag{4.4}$$

$$\nu_1=2\gamma\frac{t}{t^*}\mu\left[\frac{1}{m_1}\left(8\beta_1-\frac{\alpha_1}{3}\right)+\left(\frac{1}{m_1}+\frac{2}{m_2}\right)\left(8\beta_2-\frac{\alpha_2}{3}\right)+\frac{1}{Z_1Z_2}\left(\frac{Z_2^2}{m_2}-\frac{Z_1^2}{m_1}\right)+\frac{F^w}{2F^s m_1}\right]. \tag{4.5}$$

The time t in expressions (4.4) and (4.5) is the time of flight of the cluster through the film, while t^* is given by formula (3.1) and the dimensionless constant γ is defined by the relation

$$\gamma=(r_0^2/e^2)F^s. \tag{4.6}$$

Similarly, in the region $\pi-\chi\ll 1$ we have

$$\frac{df}{d\Omega_\chi}=\frac{1}{4\pi}[1+\lambda_2+\nu_2(\pi-\chi)^2],\quad \pi-\chi\ll 1, \tag{4.7}$$

$$\lambda_2=4\gamma\frac{t}{t^*}\mu\left[\alpha_1\left(\frac{1}{m_1}+\frac{2}{m_2}\right)+\frac{\alpha_2}{m_2}+\frac{1}{Z_1Z_2}\left(\frac{Z_2^2}{m_2}-\frac{Z_1^2}{m_1}\right)+\frac{F^w}{2F^s m_1}\right], \tag{4.8}$$

$$\nu_2=2\gamma\frac{t}{t^*}\mu\left[\left(\frac{1}{m_1}+\frac{2}{m_2}\right)\left(8\beta_1-\frac{\alpha_1}{3}\right)+\frac{1}{m_1}\left(8\beta_2-\frac{\alpha_2}{3}\right)-\frac{1}{Z_1Z_2}\left(\frac{Z_2^2}{m_2}-\frac{Z_1^2}{m_1}\right)-\frac{F^w}{2F^s m_1}\right]. \tag{4.9}$$

The corresponding formulas for the particles of mass m_1 follows from (4.3)–(4.9) with the substitutions $m_1 \rightleftarrows m_2$, $Z_1 \rightleftarrows Z_2$.

The obtained expressions, which describe the angular distributions of the particles in the considered regions of χ, demonstrate explicitly both the dependence on the masses and charges of the diverging particles, and the differences between particles emitted forward ($\chi \ll 1$) and backward ($\pi-\chi\ll 1$) in the o.s. (we emphasize that the constants $\alpha_i, \beta_i, F^s, F^w$, and γ do not depend on the masses or charges of the particles). Of course, the values of the constants λ_i and ν_i in (4.3) and (4.7) depend substantially on the values of the constants α_i and β_i in (4.1) and (4.2). An exact calculation of the latter is a rather complicated task and is outside the scope of the present paper. However, from the general properties of wake potentials of fast charged particles in solids, which are clearly manifest even in the simplest cases, such, for example, as in [3], we can deduce that

$$|\alpha_2|\ll|\alpha_1|,\quad |\beta_2|\ll|\beta_1|,$$

wherein α_1 is positive and can reach several units in magnitude (for the typical values $r_0 \approx 1$ Å of interest to us), while β_1 is apparently negative.

We now analyze the angular distribution of the ions that result from the decay of fast diatomic molecules in a thin film. We consider first the lighter component, i.e., we assume that $m_2 < m_1$, and let for the sake of argument $Z_2^2/m_2 \geq Z_1^2/m_1$. Comparing the expressions for λ_2 (4.8) and λ_1 (4.4), we see that because of the rotation of the cluster under the influence of the wake forces (the first two terms in the square brackets of the two expressions) the particles are "raked together" in the region of small angles and in the region of angles χ close to π, and this effect is much more strongly pronounced for the rear peak ($\pi-\chi\ll 1$) then for the front one ($\chi \ll 1$). We are concerned primarily with the peaks, inasmuch as by virtue of $\beta_1 < 0$ the coefficients $\nu_1 < 0$ and $\nu_2 < 0$, and the particle distribution has maxima at $\chi = 0$ and $\chi=\pi$. The intensities of the two maxima exceed the intensity corresponding to a uniform angular distribution of the particles in the c.m.s. in the case of the Coulomb explosion of a molecule in vacuum.

The physical reason for the difference between the intensities of the rear and front peaks is that the gradient of the wake potential produced by the front particle at the location of the rear particle is much larger than the corresponding gradient produced by the rear particle at the location of the front particle. Therefore the force that presses the rear particle towards the flight axis z is therefore much larger than for the front particle. Since Newton's third law does in fact not apply here, the "pure" rotation of the cluster takes place in a certain coordinate frame that has a velocity component perpendicular to the initial direction of motion, thereby enhancing the raking effect for the rear particle and, conversely, weakening this effect for the front particle. We note that the stopping of the particles (the terms proportional to $Z_2^2/m_2 - Z_1^2/m_1$ in (4.4) and (4.8)) only increases the difference between the intensities of the front and rear peaks in this case. A certain decrease in the anisotropy of the angular distribution is due to those terms in λ_1 and λ_2 which are proportional to

$$F^w/2F^s m_1<0,$$

and which appear as a result of the non-inertial character of the longitudinal motion of the mass center of the cluster in the film, and have a tendency to enhance the raking of the front particles and weaken the raking of the rear particles. These terms can become significant precisely in the opposite case, when the heavier component of the decay is considered (m_1 designates in this case the lighter particles). If it is recognized that in this case the difference between the raking effects can substantially be decreased for the rear and front particles, due to the rotation of the cluster, then we can arrive at the conclusion that the difference between the intensities of the front and rear peaks for the heavy particles may turn out to be small and the sign of the anisotropy may even be reversed. We note that this is precisely the pattern observed in the first experiments[2-4] on the correlated motion of the decay products of the $^4HeH^+$ ions ($Z_2^2/m_2 = Z_1^2/m_1$) in thin films.

If the stopping forces of the cluster particles are greatly different, i.e., if the parameters Z_i^2/m_i differ greatly, these forces can in principle alter qualitatively the pattern of the angular distribution of the decay particles. Thus, if the difference $Z_2^2/m_2 - Z_1^2/m_1$ becomes large enough, then when light particles emitted forward are examined, the intensity may reveal a dip rather than a peak, whereas the rear peak of the distribution will remain strongly pronounced as before. If we follow the behavior of the heavy component under the same conditions (stronger stopping of the light particles), then an analogous "unraking" effect can appear already for the particles emitted backward, while the angular distribution retains a maximum for the particles moving forward. We note that the described pattern can be observed most effectively in the case of a large difference between the masses of the ions in the decaying molecule, if the heavy particle is only partially ionized as it moves in the medium.

Attention must be called to the fact that simultaneous measurement of the intensities of the light and heavy components emitted forward and backward in the o-system in the decay of fast diatomic molecules in a thin film (at $t \ll t^*$ (3.1), when expressions (4.3)–(4.9) are valid) would make it possible to determine experimentally the value of the constants $\alpha_1, \alpha_2, \beta_1, \beta_2$, and F^w which characterize the wake potentials of the fast charge particles in such a film.

To get an idea of the order of magnitude of the described effect, we consider the decay of the molecular ion $^4HeH^+$ in a carbon film. Using the value $r_0 = 0.8$ Å,[1-3] we easily obtain the corresponding value $t^* = 1.7 \cdot 10^{-15}$ sec. Assuming by way of an estimate

$$\alpha_1=2, \quad \alpha_2=0, \quad F^w=-2F',$$

we get for the angular distribution of the decay protons from (4.4) and (4.8)

$$\lambda_1=2.4\gamma t/t^*, \; \lambda_2=13.6\gamma t/t^*. \tag{4.10}$$

if we approximate the quantity γ (4.6) by the formula

$$\gamma=\left(r_0\frac{\omega_p}{v_0}\right)^2 \ln\frac{2mv_0^2}{\hbar\omega_p}, \tag{4.11}$$

where m is the electron mass, ω_p is the plasma frequency of the electron subsystem of the film ($\hbar\omega_p = 25$ eV for carbon), and v_0 is the velocity of the primary molecule beam, then for molecules of energy $E = 2$ MeV/nucleon incident on a carbon film of thickness 100 Å ($\gamma \approx 0.13, t \approx 0.3t^* \approx 0.52 \cdot 10^{-15}$ sec) relation (4.10) yields

$$\lambda_1\approx0.094, \quad \lambda_2\approx0.53. \tag{4.12}$$

Similarly, for quantities λ_1 and λ_2 that enter in the angular distribution of the helium nuclei we obtain the expressions

$$\lambda_1=9.6\gamma t/t^*, \quad \lambda_2=6.4\gamma t/t^*, \tag{4.13}$$

which yield, at the same molecule energies and film thicknesses as above,

$$\lambda_1\approx0.37, \quad \lambda_2\approx0.25. \tag{4.14}$$

The foregoing relations show clearly the great difference between the intensities of the front and rear peaks for light particles and the small difference, furthermore of opposite sign, in the case of the heavy ones.

Thus, even under the conditions when the approximation linear in t is applicable, the dynamic-interaction effects in the angular distributions of the decay particles can be appreciable. At a fixed film thickness and at a constant incident direction we have

$$\lambda_i, \nu_i \sim E^{-1/2} \ln E$$

and consequently the peaks in the angular distributions of the particles should increase quite rapidly with decreasing energy E.

To conclude this section, we note that a direct quantitative comparison of our results with the experimental results of [1-4] is difficult, because the experimental parameters corresponded to rather large values of γ and to flight times $t \gtrsim 0.5t^*$ that made it necessary to go outside the framework of the approximation linear in t.

5. DISTRIBUTION OF DECAY PARTICLES IN THE LAB SYSTEM AS A FUNCTION OF THE ENERGY AND OF THE EMISSION ANGLE

A study of the fast-molecule decay-product distribution as a function of the energy and of the emission angle in the l.s. is of great interest primarily because this distribution contains direct information on the angular distribution of the particles in the o.s.

Let ϵ be the l.s. energy of the particle with mass m_2 reckoned from the value $\bar{E} = \frac{1}{2}(E_{min} + E_{max})$ (see Sec. 3). If V_0 is the velocity of the o.s. relative to the l.s. and $v(\chi)$ is the o.s. velocity of the particles of the considered type and depends on the emission angle χ, then we have for the distributions of these particles in the energy ϵ and in the emission angle ψ in the l.s.

$$\frac{d^2f}{d\Omega_\psi d\varepsilon}=\frac{df}{d\Omega_\chi}\frac{V_0}{v(\chi)}\frac{\delta(\varepsilon-m_2V_0v(\chi)\cos\chi)}{|\psi'(\chi)|}. \tag{5.1}$$

In writing down this formula we used the fact that in the case of fast molecules the formula is valid with high accuracy in the case of fast molecules.

$$\sin\psi(\chi)=\frac{v(\chi)}{V_0}\sin\chi=\psi(\chi).$$

We introduce on the $\epsilon\psi$ plane polar coordinates with χ as the azimuthal angle. The polar radius ρ is defined by the relations

$$\psi=\rho\sin\chi, \quad \varepsilon/m_2V_0^2=\rho\cos\chi. \tag{5.2}$$

It is then easy to show that

$$\frac{1}{|\psi'(\chi)|}\delta(\varepsilon-m_2V_0v(\chi)\cos\chi)=\frac{1}{m_2V_0^2\rho}\delta\left(\rho-\frac{v(\chi)}{V_0}\right). \tag{5.3}$$

Therefore relation (5.1) can be written in the following final form:

$$\frac{d^2f}{d\Omega_\psi d\varepsilon}=\frac{df}{d\Omega_\chi}\frac{\delta(\rho-v(\chi)/V_0)}{m_2V_0^2\rho^2}. \tag{5.4}$$

It is clear from this expression that in the case of

Coulomb explosion of a molecule in vacuum, when $df/d\Omega_\chi = \text{const}$ and the velocity $v(\chi)$ is also independent of the emission angle χ, a uniformly populated ring of radius $\rho = v/V_0$ is produced on the $\psi\epsilon$ plane if the units of ψ and ϵ are suitably chosen (see formulas (5.2)). When the molecules in the film decay, the pattern of the $\psi\epsilon$ distribution undergoes changes of two types. On the one hand, it follows from (5.3) that the curve on which $d^2f/d\Omega_\psi d\epsilon$ differs from zero is no longer a circle but a more complicated curve defined by the equation $\rho(\chi) = v(\chi)/V_0$. On the other hand, the population of this curve turns out to be directly proportional to $df/d\Omega_\chi$. Thus, measurement of the population-curve radius ρ corresponding to the angle χ on the $\psi\epsilon$ plane, and measurement of the corresponding population intensity, make it possible to determine directly the particle angular distribution $df/d\Omega_\chi$ in the o.s. Of course, in a real experimental situation, when the curve populated by the particles has on the $\psi\epsilon$ plane a finite width, to obtain information on $df/d\Omega_\chi$ it is necessary to integrate a distribution of the type (5.4), multiplied by ρ^2, with respect to the radius ρ.

Turning to the velocity $v(\chi)$ of the particles of mass m_2 in the o.s., we have (cf. 3.7)

$$v(\chi)=\frac{1}{m_1+m_2}\{[m_1v_r(\varphi_0,t)]^2 +2m_1v_r(\varphi_0,t)[p_{\perp c}(\varphi_0,t)\sin\varphi(\varphi_0,t)+p_{\parallel c}(\varphi_0,t)\cos\varphi(\varphi_0,t)]+p_c^2(\varphi_0,t)\}^{1/2},$$

where the angle φ_0 is assumed to be a known function of the angle χ. It is easily seen that in the approximation linear in t we have

$$v(\chi)=\frac{1}{m_1+m_2}[m_1v^*+p_{\perp c}(\chi,t)\sin\chi+p_{\parallel c}(\chi,t)\cos\chi].$$

Using relations (3.13) and (3.14) for the transverse and longitudinal components $p_{\perp c}$ and $p_{\parallel c}$ of the cluster mass-center momentum in the o.s., we obtain ultimately in the approximation linear in t

$$v(\chi)=\frac{m_1}{m_1+m_2}\left[v^*-\frac{t}{m_1}\frac{\partial V(r_0,\chi)}{\partial r_0}\right] \tag{5.5}$$

The velocity v^* and the potential $V(r,\chi)$ in this equation are defined respectively by (3.2) and (3.16).

Let us analyze the obtained expression (5.5). From the definitions (3.16) and (3.17) and from the symmetry conditions of the problem it follows that at fixed r the potential $V(r,\chi)$ is antisymmetrical with respect to the value $\chi=\pi/2$, i.e.,

$$V(r,\pi/2+\chi)=-V(r,\pi/2-\chi).$$

It is clear therefore that the same property is possessed also by the derivative $\partial V(r,\chi)/\partial r$

$$\frac{\partial V(r_0,\pi/2+\chi)}{\partial r_0}=-\frac{\partial V(r_0,\pi/2-\chi)}{\partial r_0}. \tag{5.6}$$

On the other hand, it follows also from (3.16) and (3.17) that

$$\frac{\partial V(r_0,0)}{\partial r_0}=0 \tag{5.7}$$

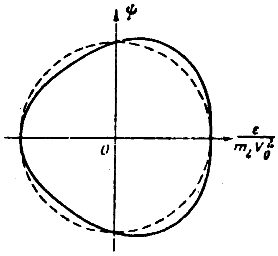

FIG. 1. View, on the $\psi\epsilon$ plane, of the pear-shaped curve along which the distribution of the decay particles differs from zero under conditions when the approximation linear in t is applicable. The dashed circle corresponds to Coulomb explosion of the molecule in vacuum.

The properties (5.6) and (5.7) allow us to conclude that in the approximation linear in t the populated curve on the plane (ϵ is in units of $m_2V_0^2$) will change with increasing t from a circle into a certain pear-shaped curve (see the figure), and furthermore in such a way that those points on the curve which correspond to $\chi=0$, $\chi=\pi/2$, and $\chi=\pi$ remain immobile. The last circumstance follows from the condition

$$v(0)=v(\pi/2)=v(\pi), \tag{5.8}$$

which follows directly from (5.5)–(5.7). The relative deformation of the populated curve

$$\eta(\chi)=-\frac{v(0)-v(\chi)}{v(0)}=-\frac{t}{m_1v^*}\frac{\partial V(r_0,\chi)}{\partial r_0}$$

turns out to be inversely proportional to the mass m_1 of the first particle at the given value of χ.

It can be shown that the maximum value of the deformation of the curve can be estimated at

$$|\eta|_{max}=\xi\frac{tZ_1Z_2F^*}{m_1v^*}=\xi\gamma\frac{\mu}{m_1}\frac{t}{t^*}$$

where the constant ξ is of the order of unity and γ is defined in accordance with (4.11). For the α particles produced as a result of $^4HeH^+$ ions with energy 2 MeV/nucleon in a carbon film 100 Å thick we have

$$|\eta|_{max}\approx 3.1\cdot 10^{-2}.$$

The corresponding value of $|\eta|_{max}$ for protons is smaller by a factor of four.

Thus, the influence of the dynamic interaction of the dispersing particles on the shape of the populated curve in the $\psi\epsilon$ plane is much weaker in the approximation linear in t than the corresponding effects in the intensity of the population of this curve (cf. (4.10), (4.12)–(4.14)).

We note in conclusion that relation (5.8) allows us to verify directly that the particles emitted at an angle $\chi=\pi/2$ in the reference frame considered by us, which coincides with the c.m.s. of the cluster, carry in the l.s. an energy $E=\frac{1}{2}(E_{min}+E_{max})$. It is this which proves that the reference frame chosen by us is indeed the o.s.

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Translated by J. G. Adashko

External photoeffect in the diffraction of x rays in a crystal with a perturbed layer

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(Submitted 20 July 1977)
Zh. Eksp. Teor. Fiz. 74, 300–313 (January 1978)

A general theory is constructed of the emission of secondary rays in Bragg diffraction of x rays in an ideal crystal and in a crystal with a perturbed surface layer. In the case of photoemission, the angular dependence of the photoelectron emission is extremely sensitive to weak displacements of the atoms in the surface layer. The question of the possibility of extracting information on the structure of the perturbed layer is analyzed in detail. It is shown that in a number of situations the aggregate of the data provided by the photoemission curve and the reflection curve permits a complete reconstruction of the structure of the perturbations in the surface layer, including both the averaged and random displacements of the atoms from their positions in an ideal crystal.

PACS numbers: 61.10.Dp, 68.20.+t, 79.60.−i

1. INTRODUCTION

The diffraction scattering of x rays in thick crystals of high degree of perfection, in the so-called dynamic regime, is characterized by the formation of a single wave field by coherent superposition of the incident and diffracted waves. The structure of this field, i.e., the distribution of its nodes and antinodes, depends strongly on the angle of incidence of the x rays on the crystal. This leads in turn to a strong angular dependence of the cross sections of the inelastic processes such as the photoeffect, fluorescence, Compton scattering, and others, which decrease strongly when the field nodes are at the crystal atoms, and conversely increase when the crystal lattice sites correspond to antinodes of the combined electric field. A reflection of this circumstance is the sharp decrease of the x-ray absorption coefficient in diffraction in the Laue geometry, observed by Borrmann in 1941—the so-called anomalous passage effect (see, e.g.,[1]).

The changes of the intensities of the inelastic processes can be investigated also directly, by studying the angular dependence of the emission of secondary rays. The first to choose this procedure, namely measurement of the angular dependence of the yield of the n–γ-reaction, was Knowles,[2] in an investigation of the anomalous passage of thermal neutrons in perfect calcite crystals. In the Sixties, starting with the pioneering work of Batterman,[3] extensive investigations have been made of secondary processes (fluorescence,[3-5] thermal diffuse scattering[6,7] Compton scattering[8]) that accompany the diffraction of x rays. In all cases, strongly pronounced anomalies were observed in the angular dependence of the secondary emission near the Bragg angle. However, in view of the large depths L of emergence of the secondary radiations investigated in,[3-7] which greatly exceed the extinction length L_{ex} of the x rays, the observed anomalies took mainly the form of dips on the yield curves, and only a small asymmetry of the curve reflected the structure of the wave field.

The situation changes radically if one registers the emission from the crystal of photoelectrons for which L is always much less than L_{ex}. Such investigations were first initiated in the early Seventies at the initiative of O. N. Efimov at the Leningrad University. It was shown even in the first papers[8,9] that the photoelectron yield curve reflects primarily the structure of the wave field in the crystal, while the extinction of the x rays has little effect. This circumstance was the basis of a new method of investigating structural imperfections produced in a crystal by various types of external action.[10-12]

Later on Golovchenko, Batterman, and Brown[13] found a method of decreasing the parameter L also in

