"Jarring" of a quantum system and the corresponding stimulated transitions

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A systematic theory of sudden perturbations is derived for quantum systems whose states are described both by wave functions (a pure ensemble) and by a quantum density operator (a mixed ensemble). A perturbation series is written in powers of the parameter $\omega\tau$, which is small when the perturbation is "sudden"; $\hbar\omega$ is the typical eigenvalue of the unperturbed system; and τ is the characteristic collision time. When the perturbation $\hat{V}(t)$, taken at different times, commutes with itself, the theory yields a compact analytic expression for the probabilities for stimulated transitions for any value of $V\tau/\hbar$. The results of many cross-section calculations for atomic collision processes are discussed from a common standpoint: the processes are treated as "jarring" processes which stimulate transitions in the quantum system. If a momentum δ_p is rapidly transferred to the system in a collision, regardless of the physical nature of the "jarring," the probabilities for the stimulated transitions are governed by the parameter $N \sim \delta_p \cdot \delta R/\hbar$ where δR is a measure of the uncertainty in the coordinates which is due to the relatively slow motions in the unperturbed system.

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1. INTRODUCTION

The probability for any transition in a quantum-mechanical system which is subjected to some external agent is governed primarily by the duration of the interaction. In general, the interaction can vary with the time in an arbitrary manner. If an atom, for example, is excited by a passing heavy charged particle, the rate of change of the interaction between the particles depends on the velocity of the incident particle. In nonresonant γ scattering the interaction time is governed by the frequency of the γ ray, while in resonant scattering it is governed by the lifetime of the excited state, and so forth.

This characteristic time τ, which we call the "collision time," is always important not in itself but in comparison with the characteristic period $2\pi/\omega$ of the quantum system in whose transitions we are interested. Adiabatic interaction ($\omega\tau \gg 1$) lead to small probabilities for the quantum transitions in states with a discrete spectrum (the probabilities decrease in an exponential or power-law manner). The "most suitable" type of interaction for excitation is the opposite case of a sudden perturbation ($\omega\tau \ll 1$), for which the probability for a transition to any other state can approach unity.

For convenience, we will single out the two extreme cases of the various possible "jarring" processes. In the first case, the Hamiltonian of the quantum system alters rapidly in a time τ short in comparison with $1/\omega$ (a jarring of the "turn-on" type; Fig. 1). In the other case, a perturbation $V(t)$ acts for a short time τ, and in the limits $t \to \pm\infty$ the total Hamiltonian of the system is the unperturbed Hamiltonian $\hat{\mathcal{H}}_0$ (a jarring of the "scattering" type; Fig. 2). In general, of course, there can also be a jarring of the most complicated type, involving both a sudden jump of the Hamiltonian and a perturbation $\hat{V}(t)$ (Fig. 3).

The simplest example of "turn-on" jarring is the en-

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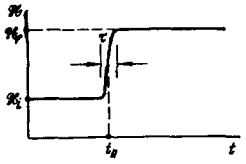

FIG. 1. "Jarring" of the "turn-on" type.

trance of an electron into a strong external field.[1] If the probabilities for quantum transitions are to be significant in the case of "turn-on" jarring, the change in the Hamiltonian must be comparable to the Hamiltonian itself, while in a jarring of the scattering type the necessary condition is that the perturbation $\hat{V}(t)$ be large enough to satisfy the condition $\hat{V}\tau/\hbar \gtrsim 1$.

The major thrust in the classical papers on jarring processes was to determine the effects of atomic structure in fast nuclear reactions.[2,3] Other interesting processes are the fast processes at the atomic and molecular level, where the collisions come in a wider variety than in nuclear reactions, since the atomic and molecular processes are more susceptible to all types of external fields and may be strongly affected by them.

Despite the differences in the physical nature of the various processes which are interpreted as jarring processes, there are many common features. The most important feature is that most of these processes can be assigned a "jarring parameter" N, whose meaning will be discussed in detail below.

The general behavior and characteristic features of the real jarring effects (the list is endless, of course) can be studied in detail in the example of the interaction of a weakly bound electron with hard electromagnetic radiation. Of primary importance here are the Compton effect at an atomic electron and the absorption, emission, and scattering of light in which molecules are involved. The interaction with radiation takes up a large part of the present paper, but we will also describe the most general features of the jarring parameter in collisions of molecules with fast charged particles. We will make no effort to study any of these effects in detail, since a detailed study would not be necessary for our purposes; we simply wish to point out the jarring interpretation of a broad range of problems, and we will approach each separate problem with this goal in mind.

As will be seen below, the parameter N is very sensitive to the uncertainty in the coordinates of the unperturbed system, δR, which is due to the relatively slow internal motions. We emphasize that the reason for this uncertainty with respect to the coordinates is physically unimportant; the reason could be the classical motion of an electron in a laser wave; quantized motion in the field of an atom, a molecule, or a crystal lattice; etc. The only differences in the different situations are in the equations for the transition probabilities; the way in

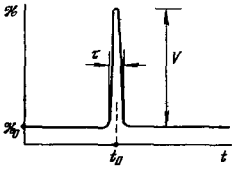

FIG. 2. "Jarring" of the scattering type.

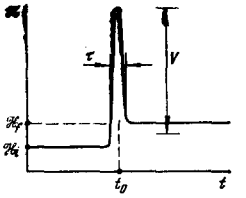

FIG. 3. "Jarring" in the general case.

which these probabilities vary with the parameter N does not change.

Before we can demonstrate all these arguments, however, we need a systematic theory for sudden perturbations (based on, for example, a power series in $\omega\tau$) so that we can clearly specify the range of applicability of the jarring approach and, in particular, so that we can see in which collisions we should expect such a characteristic as this parameter N to arise. We will first review the previous work on this subject.

The theory of sudden perturbations has been derived for only a few model problems (see, for example, Refs. 2 and 4), and the analysis has usually been restricted to the zeroth approximation for the transition amplitude in the parameter $\omega\tau$. In the scattering type of jarring we are most interested in arbitrary values of $V\tau/\hbar$. In the existing theory,[5-8] two distinct commutation relations are adopted to find the transition amplitudes for this type of jarring:

$$[\hat{V}, \hat{\mathcal{H}}_0] \equiv 0, \quad [\hat{V}(t), \hat{V}(t')] \equiv 0$$

(Vitlina and Chaplik[9] derive an equation for the amplitude in zeroth order in $\omega\tau$ by a method based on only the second of these commutation relations). Despite these stringent requirements (the condition $[\hat{V}, \hat{\mathcal{H}}_0] \equiv 0$ never holds), the relative simplicity of the resulting equations for the scattering amplitudes has led to the widespread use of this approximation to study the vibrational and rotational excitation of molecules in collisions with electrons and heavy particles. In the absence of a full-fledged theory of sudden perturbations, the only way to evaluate the results has been to compare them numerically with other calculations, carried out on a firmer basis.

In Section 2 below we set forth a theory of sudden perturbations which does not suffer from these shortcomings. We will show that the condition $[\hat{V}, \hat{\mathcal{H}}_0] \equiv 0$, which is not satisfied, is completely unnecessary at small values of $\omega\tau$. Whether the equation for the transition amplitude is simple or complicated for an arbitrary value of $V\tau/\hbar$ depends strongly on whether the commutator

$$\hat{\xi}(t, t') \equiv [\hat{V}(t), \hat{V}(t')] \tag{1}$$

is small or large. The equation for the amplitude is simplest in the limit $\xi \to 0$, which corresponds to the collision models most commonly used. If, on the other hand, the commutator in (1) is not small, the theory becomes far more complicated, and in general the theory cannot be formulated in finite form for arbitrary values of $V\tau/\hbar$ (see the Note Added in Proof at the end of this paper).

From nuclear physics we have a good example of a situation in which the interaction potential is large and the

commutator in (1) is by no means small. We have in mind the tensor part of the two-nuclear interaction,[10,11] which is proportional to the operator

$$\frac{3}{r^2}(\hat{\sigma}_1\mathbf{r})(\hat{\sigma}_2\mathbf{r})-\hat{\sigma}_1\hat{\sigma}_2,$$

where $\hat{\sigma}_{1,2}$ are spinors, and $\mathbf{r}$ is the radius vector between the two nucleons. Choosing any arbitrary classical trajectory for one of the nucleons as it is scattered by the other, we easily see that the operators $(\hat{\sigma}_1\mathbf{n})(\hat{\sigma}_2\mathbf{n})$ corresponding to different times do not commute (the direction of the unit vector $\mathbf{n}=\mathbf{r}/r$ varies with the time).

An analogous example from atomic physics is the interaction of a magnetic moment $\hat{\mu}$ with a strong magnetic field. If the field is variable, and the vector $\mathbf{H}(t)$ changes direction in a time τ, then the potential $\hat{V}=\hat{\mu}\mathbf{H}(t)$ does not commute with itself at different times. In this case the exact equation for the transition amplitude has nothing in common with the simplest jarring result, which is valid only in the limit $\xi\to 0$.

2. THEORY OF SUDDEN PERTURBATIONS

a. "Jarring" of the scattering type with $\hat{\xi}\equiv 0$

We assume that the total Hamiltonian of the quantum-mechanical system $\hat{\mathscr{H}}$, is broken up into the sum of a time-independent part $\hat{\mathscr{H}}_0$ (the unperturbed Hamiltonian) and a part which is due to the interaction with the external field, $\hat{V}(t)$:

$$\hat{\mathscr{H}}=\hat{\mathscr{H}}_0+\hat{V}(t).$$

We assume that $\hat{V}(t)$ vanishes outside a time interval τ near the instant t_0 and that the operators $\hat{V}(t)$ taken at different times commute.

Below we will distinguish among the state vectors of the quantum system which are written in the interaction picture, $|\Psi\rangle$ and those which are written in the Schrödinger picture $|\Phi\rangle$; and the corresponding unitary evolution operators $\hat{S}(t,t')$ and $\hat{U}(t,t)$, which relate the corresponding vectors at different times:

$$|\Psi(t)\rangle=\hat{S}(t,t')\,|\Psi(t')\rangle, \tag{2}$$
$$|\Phi(t)\rangle=\hat{U}(t,t')\,|\Phi(t')\rangle. \tag{3}$$

In the general case, we find the most informative possible description of the quantum-mechanical system when we replace the state vectors by the quantum density operator $\hat{\rho}$ (this operator is the "statistical operator," and the set of elements $\langle s'|\hat{\rho}|s\rangle$ forms the density matrix). For our purposes it will be convenient to use the density operator written in the interaction picture,

$$\hat{\rho}=\sum_s |\Psi_s\rangle w_s\langle\Psi_s|, \tag{4}$$

(the weight factors w_s determine the probabilities for finding the system in the different quantum states $|\Psi_s\rangle$). The time evolution of the operator in (4) is evidently described by

$$\hat{\rho}(t)=\hat{S}(t,t')\,\hat{\rho}(t')\,\hat{S}^{-1}(t,t'). \tag{5}$$

The time-evolution operator $\hat{S}(t,t')$ satisfies the differential equation

$$i\hbar\frac{\partial\hat{S}(t,t')}{\partial t}=\hat{W}(t)\,\hat{S}(t,t') \tag{6}$$

with the boundary condition $\hat{S}(t,t)=\hat{I}$, or, equivalently, the integral equation

$$\hat{S}(t,t')=\hat{I}-\frac{i}{\hbar}\int_{t'}^{t}dt\,\hat{W}(t)\,\hat{S}(t,t'); \tag{7}$$

here $\hat{W}(t)$ is the operator $\hat{V}(t)$ written in the interaction picture,

$$\hat{W}(t)=\exp\left(\frac{i}{\hbar}\hat{\mathscr{H}}_0t\right)\hat{V}(t)\exp\left(-\frac{i}{\hbar}\hat{\mathscr{H}}_0t\right). \tag{8}$$

In terms of the state vectors describing the pure ensemble, the scattering problem can be formulated as follows. We denote by $|i\rangle$ and $|f\rangle$ the initial and final states of the system, which are eigenfunctions of the Hamiltonian $\hat{\mathscr{H}}_0$:

$$\hat{\mathscr{H}}_0|i\rangle=E_i|i\rangle,\quad \hat{\mathscr{H}}_0|f\rangle=E_f|f\rangle. \tag{9}$$

The probability for a transition from state $|i\rangle$ to the state $|f\rangle$ as the result of the perturbation $\hat{V}(t)$ is governed by the square of the modulus of the matrix element

$$\mathfrak{M}_{fi}=\langle f|\hat{S}(+\infty,-\infty)|i\rangle. \tag{10}$$

For the case of a mixed ensemble the scattering problem cannot be formulated as a quantum-transition problem in the ordinary sense of the phrase. We assume that in its initial state ($t\to-\infty$) the system is described by the density operator

$$\hat{\rho}_i\equiv\hat{\rho}(-\infty)=\sum_i|i\rangle w_i\langle i|, \tag{11}$$

where the sum is over all eigenstates of the unperturbed Hamiltonian $\hat{\mathscr{H}}_0$. Then the probability for finding the system in stationary state $|f\rangle$ of the unperturbed Hamiltonian in the limit $t\to+\infty$ is given by

$$w_i(i)=\langle f|\hat{\rho}(+\infty)|f\rangle=\langle f|\hat{S}(+\infty,-\infty)\hat{\rho}_i\hat{S}^{-1}(+\infty,-\infty)|f\rangle. \tag{12}$$

If $\hbar\omega$ is a typical eigenvalue of $\hat{\mathscr{H}}_0$, and the parameter $\omega\tau$ is small, the perturbation operator in the interaction picture can be expanded in a power series in $\omega\tau$ [here $W_n\sim(\omega\tau)^n$]:

$$\hat{W}(t)=\hat{W}_0(t)+\hat{W}_1(t)+\hat{W}_2(t)+\ldots, \tag{13}$$
$$\hat{W}_0(t)=\exp\left(\frac{i}{\hbar}\hat{\mathscr{H}}_0t_0\right)\hat{V}(t)\exp\left(-\frac{i}{\hbar}\hat{\mathscr{H}}_0t_0\right), \tag{14}$$
$$\hat{W}_1(t)=\frac{t-t_0}{i\hbar}\exp\left(\frac{i}{\hbar}\hat{\mathscr{H}}_0t_0\right)[\hat{V}(t),\hat{\mathscr{H}}_0]\exp\left(-\frac{i}{\hbar}\hat{\mathscr{H}}_0t_0\right), \tag{15}$$
$$\hat{W}_2(t)=\frac{1}{2}\left(\frac{t-t_0}{i\hbar}\right)^2\exp\left(\frac{i}{\hbar}\hat{\mathscr{H}}_0t_0\right)[[\hat{V}(t),\hat{\mathscr{H}}_0],\hat{\mathscr{H}}_0]\exp\left(-\frac{i}{\hbar}\hat{\mathscr{H}}_0t_0\right),\ldots \tag{16}$$

To solve Eq. (6), we also write the evolution operator $\hat{S}(t,t')$ as a power series in $\omega\tau$ [$\Delta_n\sim(\omega\tau)^n$]:

$$\hat{S}(t,t')=\hat{S}_0(t,t')[\hat{I}+\hat{\Delta}_1(t,t')+\hat{\Delta}_2(t,t')+\ldots]. \tag{17}$$

Since we are assuming $\hat{\xi}\equiv 0$ in this subsection, we can immediately integrate Eq. (6), using the substitution (13)–(16):

$$\hat{S}_0(t,t')=\exp\left[-\frac{i}{\hbar}\int_{t'}^{t}dt\,\hat{W}_0(t)\right] \tag{18}$$
$$=\exp\left(\frac{i}{\hbar}\hat{\mathscr{H}}_0t_0\right)\exp\left[-\frac{i}{\hbar}\int_{t'}^{t}dt\,\hat{V}(t)\right]\exp\left(-\frac{i}{\hbar}\hat{\mathscr{H}}_0t_0\right),$$

$$\hat{\Delta}_1(t,t')=-\frac{i}{\hbar}\int_{t'}^{t}dt\,\hat{S}_0^{-1}(t,t')\,\hat{W}_1(t)\,\hat{S}_0(t,t'), \tag{19}$$

$$\hat{\Delta}_2(t,t')=\frac{[\hat{\Delta}_1(t,t')]^2}{2}-\frac{i}{\hbar}\int_{t'}^{t}dt\,\hat{S}_0^{-1}(t,t')\,\hat{W}_2(t)\,\hat{S}_0(t,t'),\ldots \tag{20}$$

In the zeroth approximation in $\omega\tau$, in which we have $\hat{S}\approx\hat{S}_0$, the transition amplitude is written as follows,

aside from an inconsequential phase factor:

$$\mathfrak{M}_{fi}^{0}=\langle f|\exp\Big[-\frac{i}{\hbar}\int_{-\infty}^{\infty}dt\,\hat{V}(t)\Big]|i\rangle. \tag{21}$$

In terms of the quantum density operator, the probability is thus

$$w_f^0(i)=\langle f|\exp\Big[-\frac{i}{\hbar}\int_{-\infty}^{\infty}dt\,\hat{V}(t)\Big]\hat{\rho}_i\exp\Big[\frac{i}{\hbar}\int_{-\infty}^{\infty}dt\,\hat{V}(t)\Big]|f\rangle. \tag{22}$$

Let us examine in particular that jarring case which is most common in practice, in which we can assume that the system acquires a definite momentum δp as the result of a collision. Under this assumption, in zeroth order in $\omega\tau$, the action of the operator $\hat{S}_0(+\infty,-\infty)$ on the function $|f\rangle$ reduces to simply a displacement δp in momentum space:

$$\langle f|\hat{S}_0(+\infty,-\infty)=\langle f|\exp\Big(-\frac{i}{\hbar}\delta\mathbf{p}\cdot\mathbf{r}\Big). \tag{23}$$

Accordingly [see also Refs. 2 and 12 regarding Eq. (24)],

$$\mathfrak{M}_{fi}^{0}=\langle f|\exp\Big(-\frac{i}{\hbar}\delta\mathbf{p}\cdot\mathbf{r}\Big)|i\rangle, \tag{24}$$

$$w_f^0(i)=\langle f|\exp\Big(-\frac{i}{\hbar}\delta\mathbf{p}\cdot\mathbf{r}\Big)\hat{\rho}_i\exp\Big(\frac{i}{\hbar}\delta\mathbf{p}\cdot\mathbf{r}\Big)|f\rangle. \tag{25}$$

Equations (24) and (25) follow directly from Eqs. (21) and (22) if we set

$$\hat{V}(t)=\mathbf{f}(t)\,\mathbf{r},\qquad \delta\mathbf{p}=\int_{-\infty}^{\infty}dt\,\mathbf{f}(t),$$

where $\mathbf{f}(t)$ is the perturbing force.

If the quantity $\mathbf{f}(t)$ falls off rapidly [say, exponentially, when the perturbation theory of (17)–(20) in powers of $\omega\tau$ is applicable in any order] outside the time interval τ, the question of the corrections to the zeroth approximation in $\omega\tau$ can be solved in a quite general case. Let us assume that the Hamiltonian of the unperturbed system incorporates a momentum-independent potential term:

$$\hat{\mathcal{H}}_0=\frac{\hat{p}^2}{2m}+U(\mathbf{r}).$$

Then the evolution operator of this system takes the following form when an external perturbation $\hat{V}(t)=\mathbf{f}(t)\mathbf{r}$ is imposed (the instant of jarring is adopted as the origin of the time scale; $t_0=0$):

$$\hat{S}(+\infty,-\infty)=\exp(i\mathbf{qr})\,[\hat{I}+\hat{\Delta}_1+\hat{\Delta}_2+O((\omega\tau)^3)], \tag{26}$$

$$\hat{\Delta}_1=\frac{i}{\hbar m}\int_{-\infty}^{\infty}dt\,t\mathbf{f}(t)\,[\hbar\mathbf{q}(t)+\hat{\mathbf{p}}], \tag{27}$$

$$\hat{\Delta}_2=\frac{(\hat{\Delta}_1)^2}{2}-\frac{1}{2m\hbar^2}\int_{-\infty}^{\infty}dt\,t^2\mathbf{f}(t)\,[U(\mathbf{r}),\hat{\mathbf{p}}], \tag{28}$$

where

$$\mathbf{q}(t)=-\frac{1}{\hbar}\int_{-\infty}^{t}dt\,\mathbf{f}(t),\qquad \mathbf{q}\equiv\mathbf{q}(+\infty). \tag{29}$$

If $\mathbf{f}(t)=\mathbf{f}(-t)$, the quantity $\hat{\Delta}_1$ becomes a purely imaginary c-number, which is independent of the coordinates, and there are no corrections of first order in $\omega\tau$ in the probabilities for transitions stimulated by the perturbation $\mathbf{f}(t)\mathbf{r}$.

There can be cases in which the time integral of the perturbation $\hat{V}(t)$ is small, although the perturbation itself is not. If we knew only the equations of the zeroth jarring approximation, we could not calculate the probabilities for quantum transitions in such a case. We would have to use equations for the next orders in $\omega\tau$ in the evolution operator in (17). Let us assume, for example, that the time integral of $\hat{V}(t)$ is smaller than or of the order of $(\omega\tau)^2$. Then the equation for the transition amplitude in the limit $\omega\tau\to 0$ takes the following form ($|i\rangle\neq|f\rangle$):

$$\mathfrak{M}_{fi}\sim\frac{1}{\hbar^2}\langle f|\int_{-\infty}^{\infty}dt\,(t-t_0)\,[\hat{V}(t),\hat{\mathcal{H}}_0]\,|i\rangle. \tag{30}$$

b. Scattering in the general case ($\hat{\xi}\neq 0$)

If the operators $\hat{V}(t)$ for different times do not commute, Eq. (6) cannot be integrated as simply as in the preceding subsection, even in the limit $\omega\tau\to 0$. Since we are interested in values $V\tau/\hbar\gtrsim 1$, it is pointless to seek solutions of (6) or (7) by successive approximations. We can take the following approach, however.

Using the group property of the evolution operators,

$$\hat{S}(t,t')=\hat{S}(t,t'')\,\hat{S}(t'',t'), \tag{31}$$

we can write $\hat{S}(t,t')$ as the product

$$\hat{S}(t,t')=\hat{S}(t,t_n)\,\hat{S}(t_n,t_{n-1})\ldots\hat{S}(t_2,t_1)\,\hat{S}(t_1,t'). \tag{32}$$

Letting t_k approach t_{k+1} (more precisely, requiring $\delta t_k=t_{k+1}-t_k\ll\tau$), we can easily integrate Eq. (6) for each of the factors in (32):

$$\hat{S}(t_{k+1},t_k)=\exp\Big[-\frac{i}{\hbar}\hat{W}(t_k)\,\delta t_k\Big]. \tag{33}$$

We now collect all the terms in (32) in pairs, applying the Baker–Campbell–Hausdorff (BCH) equation[13] each time:

$$\exp(\hat{A})\exp(\hat{B})=\exp\Big\{\hat{A}+\hat{B}+\frac{1}{2}[\hat{A},\hat{B}]+\frac{1}{12}[\hat{A},[\hat{A},\hat{B}]]+\frac{1}{12}[[\hat{A},\hat{B}],\hat{B}]+\ldots\Big\}. \tag{34}$$

As a result, the finite-evolution operator in (32) leads to the following exponential equation [in the standard method[6,13-15] for deriving (35)–(38), a solution of Eq. (6) is immediately sought in the form of the expansion in (35)]:

$$\hat{S}(t,t')=\exp(\hat{A}_1+\hat{A}_2+\hat{A}_3+\ldots), \tag{35}$$

$$\hat{A}_1=-\frac{i}{\hbar}\int_{t'}^{t}dt\,\hat{W}(t), \tag{36}$$

$$\hat{A}_2=\frac{1}{2}\Big(-\frac{i}{\hbar}\Big)^2\int_{t'}^{t}dt_1\int_{t'}^{t_1}dt_2\,[\hat{W}(t_1),\hat{W}(t_2)], \tag{37}$$

$$\hat{A}_3=\frac{1}{6}\Big(-\frac{i}{\hbar}\Big)^3\int_{t'}^{t}dt_1\int_{t'}^{t_1}dt_2\int_{t'}^{t_2}dt_3\{[\hat{W}(t_1),[\hat{W}(t_2),\hat{W}(t_3)]]+[[\hat{W}(t_1),\hat{W}(t_2)],\hat{W}(t_3)]\},\ldots. \tag{38}$$

Equations (35)–(38) are called the "Magnus expansion" and are seen to be the continuous analog of the BCH equation (34). For completeness we note that the solution of Eq. (6) for the most general case is conveniently sought by using the substitution[16]

$$\hat{S}(t,t')=\exp(\hat{B}_1)\exp(\hat{B}_2)\exp(\hat{B}_3)\ldots. \tag{39}$$

The recurrence relations for $\hat{A}_n$ and $\hat{B}_n$, which are n-fold integrals of $(n-1)$-fold commutators of the operators $\hat{W}(t)$ taken at different times, and other mathemati-

cal details can be found in Refs. 6, 13–18.

In the zeroth order in $\omega\tau$ one must make in expression (35)–(39) the replacement $\hat{W} \to \hat{W}_0$ [cf. Eq. (14)]. Thus, in this approximation $\hat{A}_n$ and $\hat{B}_n$ contain $(n-1)$-fold commutators of the operators $\hat{V}(t)$ taken at different times. In the case when $\hat{\xi} \equiv 0$ only $\hat{A}_1$ and $\hat{B}_1$ differ from zero in Eqs. (35) and (39) and this corresponds to the result of the preceding subsection. If it should turn out that the commutator $\xi \ll 1$, then within the framework of the expansions (35)–(39) it will be easy to write out the perturbation theory in powers of ξ.

The presentation of the solution of Eq. (6) in the form of the expansion (35) or (39) is particularly convenient in those situations when the $(n-1)$-fold commutators contained in $\hat{A}_n$ and $\hat{B}_n$ finally become c-numbers as n increases. If this occurs, for example, at $n = n^*$ then all $\hat{A}_{n>n^*} \equiv \hat{B}_{n>n^*} \equiv 0$ and, consequently, the scattering amplitude can be written in finite terms. An interesting and useful example of just such a situation is the harmonic oscillator acted upon by an arbitrary external force (in this case $n^* = 2$; cf. Sec. 5).

In conclusion we emphasize that since the operators $\hat{A}_n$ are Hermitian no manner of breaking off the infinite series in the exponent of (35) will violate the unitarity of the evolution operator $\hat{S}(t, t')$.

c. "Jarring" with a change in the Hamiltonian of the system

Let us assume that at $t \to -\infty$ the quantum system is in one of the stationary states (we are now considering a pure ensemble) which are the eigenfunctions of the Hamiltonian $\hat{\mathscr{H}}_i$:

$$\hat{\mathscr{H}}_i |i\rangle = E_i |i\rangle.$$

Near the time t_0, the system is subjected to a sudden external agent $\hat{V}(t)$, and in the limit $t \to +\infty$ the system is accordingly in some stationary state of the (generally different) Hamiltonian $\hat{\mathscr{H}}_f$:

$$\hat{\mathscr{H}}_f |f\rangle = E_f |f\rangle$$

(Fig. 3). The perturbation is "sudden" because the characteristic time interval over which is acts is much shorter than the reciprocals of the typical frequencies, $\omega^{-1} \sim \hbar/E_i \sim \hbar/E_f$.

Let us redefine the perturbation operator in such a manner that it vanishes at $t \to \pm\infty$:

$$\hat{\mathscr{H}} = \hat{\mathscr{H}}_0 + \hat{V}(t) = \begin{cases} \hat{\mathscr{H}}_i + \hat{V}(t) & \text{for } t \leqslant t_0, \\ \hat{\mathscr{H}}_f + \hat{V}(t) & \text{for } t \geqslant t_0. \end{cases} \tag{40}$$

The probability amplitude for a transition between states $|i\rangle$ and $|f\rangle$ is found through the obvious transformation

$$\langle \Phi(t_+) | \hat{U}(t_+, t_0) \hat{U}(t_0, t_-) | \Phi(t_-) \rangle = \langle \Psi_f(t_+) | \hat{S}_f(t_+, t_0) \exp\left(\frac{i}{\hbar} \hat{\mathscr{H}}_f t_0\right) \exp\left(-\frac{i}{\hbar} \hat{\mathscr{H}}_i t_0\right) \hat{S}_i(t_0, t_-) | \Psi_i(t_-) \rangle, \tag{41}$$

$$t_- \leqslant t_0 \leqslant t_+ . \quad \mathfrak{M}_{fi} = \langle f | \hat{G} | i \rangle. \tag{42}$$

The exact equation for this amplitude is

$$\hat{G} = \hat{S}_f(+\infty, t) \exp\left(\frac{i}{\hbar} \hat{\mathscr{H}}_f t_0\right) \exp\left(-\frac{i}{\hbar} \hat{\mathscr{H}}_i t_0\right) \hat{S}_i(t_0, -\infty). \tag{43}$$

The subscripts "i" and "f" on the wave functions and operators in (41) and (43) mean that these quantities are written in the interaction picture—once with the unperturbed Hamiltonian $\hat{\mathscr{H}}_i$ and once with $\hat{\mathscr{H}}_f$.

In the formulation of the collision process in which the quantum density operator is used (for a mixed ensemble), the analogous equation for the probability for finding the system in one of the eigenstates $|f\rangle$ of the final Hamiltonian $\hat{\mathscr{H}}_f$ is

$$w_f(i) = \langle f | \hat{G} \hat{\rho}_i \hat{G}^{-1} | f \rangle. \tag{44}$$

We emphasize that the results in (42)–(44) are exact, and they are valid for any value of $\omega\tau$. If, on the other hand, $\omega\tau \ll 1$, all the equations of the theory for jarring of the scattering type can be used immediately to evaluate the evolution operators in (42)–(44).

As mentioned above, in contrast to jarring of the scattering type, the transition amplitude, even in zeroth order in $\omega\tau$ and $V\tau/\hbar$,

$$\mathfrak{M}_{fi}^{00} = \langle f | i \rangle, \tag{45}$$

and the corresponding probability

$$w_f^{00}(i) = \langle f | \hat{\rho}_i | f \rangle \tag{46}$$

are generally not small in this case.

d. Example: spin flipping in a magnetic field

In this subsection we will illustrate certain aspects of the jarring approximation by calculating the probability for the flipping of the spin of a particle in a varying magnetic field. In particular, we will demonstrate that the simple result in (21) is not applicable when the perturbation operators taken at different times do not commute. We can obtain this result because this problem can be solved exactly in the nonrelativistic limit.

We assume that a particle with a spin $\hbar/2$ and a magnetic moment μ is in a constant and homogeneous magnetic field H_0, which is directed along the z axis. At time $t = 0$, a homogeneous magnetic field $\mathbf{H}_1(t)$ is applied; this field is in the x, y plane and is rotating in it at an angular velocity Ω. This field is removed at the time $t = \tau$. In the time interval $0 < t < \tau$ the particle is in the resultant field

$$\mathbf{H} = \{H_1 \cos \Omega t,\ H_1 \sin \Omega t,\ H_0\}. \tag{47}$$

Let us assume for definiteness that the spin of the particle at $t < 0$ is directed along the positive z axis, i.e., that the wave function is $\binom{1}{0}$. The wave function which describes the state of the particle at any time in the interval $(0, \tau)$ is

$$\Psi(t) = \begin{pmatrix} a(t) \\ b(t) \end{pmatrix}, \tag{48}$$

$$a(t) = \frac{(\mu H_0 - \hbar\omega_2) \exp(i\omega_1 t) + (\hbar\omega_1 - \mu H_0) \exp(i\omega_2 t)}{\hbar(\omega_1 - \omega_2)}, \tag{49}$$

$$b(t) = \frac{(\hbar\omega_1 - \mu H_0)(\mu H_0 - \hbar\omega_2) \exp(i\Omega t) [\exp(i\omega_1 t) - \exp(i\omega_2 t)]}{\mu H_1 \hbar(\omega_1 - \omega_2)}, \tag{50}$$

where

$$\hbar\omega_{1,2} = -\frac{\hbar\Omega}{2} \pm \sqrt{\mu^2 H_1^2 + \left(\mu H_0 + \frac{\hbar\Omega}{2}\right)^2}. \tag{51}$$

The probability for spin flipping by the time $t = \tau$ is

$$|b(\tau)|^2 = \left[2 \frac{(\hbar\omega_1 - \mu H_0)(\mu H_0 - \hbar\omega_2)}{\mu H_1 \hbar(\omega_1 - \omega_2)} \sin\left(\frac{\omega_1 - \omega_2}{2}\tau\right)\right]^2. \tag{52}$$

Now let us assume that the time interval τ is so short that the jarring condition holds:

$$\frac{\mu H_0 \tau}{\hbar} \ll 1. \quad (53)$$

We also assume that inequality (53) is so strong that it would be sufficient to use the zeroth approximation in $\mu H_0 \tau/\hbar$ under normal conditions. Since, however, the interaction operator

$$\hat{V}(t) = \mu H_1 \begin{pmatrix} 0 & \exp(-i\Omega t) \\ \exp(i\Omega t) & 0 \end{pmatrix} \quad (54)$$

does not commute with itself at different times,

$$\hat{\xi} \equiv [\hat{V}(t), \hat{V}(t')] = 2i\mu^2 H_1^2 \hat{\sigma}_z \sin[\Omega(t-t')] \quad (55)$$

($\hat{\sigma}_z$ is the Pauli matrix), the theory of Subsection a) is not applicable, and it is generally necessary to take into account all the terms in the Magnus expansion in (35) or the Fer expansion in (39) for estimating the time-evolution operator.

A calculation of the probability amplitude for spin flipping from Eq. (21) yields

$$\mathfrak{M}^0_{\downarrow\uparrow} = \frac{1-\exp(i\Omega\tau)}{2\sin(\Omega\tau/2)} \sin\left(\frac{2\mu H_1}{\hbar\Omega} \sin\frac{\Omega\tau}{2}\right). \quad (56)$$

The commutator in (55) depends on the parameter $\Omega\tau$. Physically, this parameter determines the angle through which the variable field $\mathbf{H}_1(t)$ manages to rotate during "jarring". For arbitrary values of $\Omega\tau$, the probability $|\mathfrak{M}^0_{\downarrow\uparrow}|^2$ has nothing in common with the exact result in (52), even in the limit $\omega\tau \to 0$. If, on the other hand, the field $\mathbf{H}_1(t)$ does not have time to change direction substantially, i.e., if $\Omega\tau \ll 1$, then it is easy to see that under condition (53) the probability $|\mathfrak{M}^0_{\downarrow\uparrow}|^2$ is equal to its exact equivalent in both the zeroth and first orders in $\Omega\tau$.

3. "JARRING" IN THE CASE OF A NEARLY RESONANT EXTERNAL EFFECT ON A QUANTUM SYSTEM

If the atoms or molecules are excited by a varying external field instead of in a fast collision, the jarring condition $\omega\tau \ll 1$ is not easily satisfied since the necessary interaction times τ become too short. Conversely in most cases in which a quantum system is excited by an external field the adiabatic condition $\omega\tau \gg 1$ holds, and the transition probabilities are small. An exceptional case (although again in this case the condition $\omega\tau \gg 1$ holds) is that in which a resonant laser field is used to pump the system; this is the most effective pumping method presently available. The jarring-theory explanation for this situation is that the problem of a nearly resonant excitation of a quantum system in an electromagnetic pulse of finite length reduces to a jarring problem although the frequency of the external agent is comparable to the internal frequencies of the unperturbed system. We will now show that whether the jarring-theory approach is applicable depends on whether the product $\Delta \cdot \tau$ is small, where Δ is the frequency difference in the case of a nearly resonant excitation, and τ is the pulse length of the external agent. In this sense the zeroth jarring approximation ($\Delta \cdot \tau \to 0$) is equivalent to the resonant approximation.

Experimentally, molecular systems are usually quite complicated, so that the parameter $\Delta \cdot \tau$ can take on arbitrary values. As soon as it reaches values of the order of unity or higher, however, the probabilities for the allowed transitions to the corresponding levels fall off rapidly, and the entire spectrum break up into separate groups of levels, for each of which the resonant-excitation problem can be solved separately.

We therefore consider an arbitrary n-level system in which the level spacing is nearly uniform and approximately equal to the photon energy of the external field ($\hat{\mathcal{H}}_0$ is the Hamiltonian of the system, and E_k and $|k\rangle$ are its eigenvalues and eigenvectors):

$$\hbar\omega \sim E_{k+1} - E_k, \quad k = 1, 2, \ldots, n.$$

Let us assume that the external perturbation $\hat{V}\varphi(t)\cos\omega t$ causes transitions only between adjacent levels, that the operator $\hat{V}$ is independent of the time, and that the pulse envelope $\varphi(t)$ is confined primarily to the time interval τ near the instant $t = 0$. The maximum value of the function $\varphi(t)$ is unity, and, of course, the condition $\omega\tau \gg 1$ holds.

Solving the Schrödinger equation by means of the substitution

$$\Psi(t) = \sum_{k=1}^{n} C_k(t) \exp(-ik\omega t) |k\rangle \quad (57)$$

after the high-frequency terms are eliminated in the standard manner (we assume $|E_k - \hbar k\omega| \ll \hbar\omega$), we find the system of equations

$$i\hbar \frac{\partial C_k}{\partial t} = (E_k - \hbar k\omega) C_k + \varphi(t)(V_{k,k-1}C_{k-1} + V_{k,k+1}C_{k+1}). \quad (58)$$

These equations describe the time evolution of a quantum system which is different from the original system and which has the quasi-Hamiltonian

$$\hat{\mathcal{H}}' = \hat{\mathcal{H}}'_0 + \hat{V}\varphi(t). \quad (59)$$

The energy spectrum of $\hat{\mathcal{H}}'_0$ is a set of quasilevels $E_k - \hbar k\omega$. If the adiabatic condition $\omega\tau \gg 1$ holds for the original system, with the Hamiltonian

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{V}\varphi(t)\cos\omega t, \quad (60)$$

then the jarring condition can hold for a quantum system with quasi-Hamiltonian (59) provided that

$$|E_k - \hbar k\omega| \frac{\tau}{\hbar} \ll 1. \quad (61)$$

Inequality (61) means that, because of the finite time interval τ, all the levels are actually excited in a resonant manner. In jarring-theory terms, the resonant approximation is equivalent to the zeroth order approximation in $\Delta \cdot \tau$. The methods of Section 2 can thus be applied to the quantum-transition problem.

The transformation to the interaction picture used in Section 2 corresponds in this case to a transformation from the amplitudes $C_k(t)$ [see (57)] to the amplitudes $a_k(t)$, which are determined from the expansion

$$\Psi(t) = \sum_{k=1}^{n} a_k(t) \exp\left(-\frac{iE_k t}{\hbar}\right) |k\rangle. \quad (62)$$

In place of (58) in the interaction picture we have

$$i\hbar \frac{\partial \hat{A}}{\partial t} = \varphi(t)\hat{W}(t)\hat{A}, \quad \hat{A} = \begin{pmatrix} a_1 \\ \vdots \\ a_n \end{pmatrix}. \quad (63)$$

The only nonvanishing elements in $\hat{W}(t)$ are those which are near the main diagonal:

$$W_{k,k+1} = V_{k,k+1}\exp(-i\Delta_k t), \quad W_{k,k-1} = V_{k,k-1}\exp(i\Delta_{k-1}t), \quad \hbar\Delta_k \equiv E_{k+1} - E_k - \hbar\omega. \quad (64)$$

Equation (63) has precisely the same form as the equation for the time-evolution operator in (6), and it is solved in precisely the same manner. In the zeroth order approximation in $\Delta \cdot \tau$, we find

$$\hat{A}(+\infty)=\exp\left[-\frac{i}{\hbar}\hat{V}\int_{-\infty}^{\infty}dt\,\varphi(t)\right]\hat{A}(-\infty). \tag{65}$$

The only nonvanishing elements in the time-independent operator $\hat{V}$ are $V_{k,k+1}$ and $V_{k,k-1}$.

The result in (65) is valid for arbitrary values of $V\tau/\hbar$, i.e., for an arbitrary intensity of the electromagnetic pulse stimulating the transitions. We will not write out the solutions of Eq. (63) for high-order approximations in $\Delta \cdot \tau$. Let us examine some illustrative calculations of the probabilities for stimulated transitions on the basis of Eq. (65). In the case of a two-level system which is in one of its stationary states at $t \to -\infty$, the probability for a transition to the other state at $t \to +\infty$ is given by the familiar equation (cf., for example, Ref. 12)

$$w_{21}=\sin^2\left[\frac{|V_{21}|}{\hbar}\int_{-\infty}^{\infty}dt\,\varphi(t)\right]. \tag{66}$$

Analogously, for a three-level system which is initially in, say, its lowest-lying state, $|1\rangle$, the transition probabilities are

$$w_{11}=\left(\frac{|V_{12}|^2\cos\Phi+|V_{23}|^2}{|V_{12}|^2+|V_{23}|^2}\right)^2,\quad w_{31}=\left(\frac{|V_{12}V_{23}|(1-\cos\Phi)}{|V_{12}|^2+|V_{23}|^2}\right)^2,$$
$$w_{21}=\frac{|V_{12}|^2\sin^2\Phi}{|V_{12}|^2+|V_{23}|^2},\qquad \Phi=\frac{\sqrt{|V_{12}|^2+|V_{23}|^2}}{\hbar}\int_{-\infty}^{\infty}dt\,\varphi(t). \tag{67}$$

Calculations for the probabilities of nearly resonant transitions are frequently carried out in the resonant approximation, which can be used to transform from the original Hamiltonian in (60) to the quasi-Hamiltonian in (59) and then to take the limit $\Delta \cdot \tau \to 0$. To evaluate the limits of applicability of this approach it is necessary, for example, to compare the exact equation for the transition amplitude in the simple harmonic oscillator [see Eq. (79) and (80) below] with the corresponding amplitude in the resonant case, in which the equation for $f(\omega)$ contains the time integral of $\varphi(t)\exp(-i\Delta t)$ or a time integral of $\varphi(t)$ instead of $\varphi(t)\exp(-i\omega t)$. We immediately see that the high-frequency terms in the Fourier component $f(\omega)$ which have been discarded make an exponentially small contribution.

4. THE "JARRING" PARAMETER N

An examination of the various models for many kinds of fast collisions of interest in physics shows that most of them are formulated in one of two ways: either the corresponding perturbation operators $\hat{V}(t)$, taken at various times, are assumed to commute, or (most frequently) the results of the collision can be interpreted as a fast transfer of momentum to the quantum system.

In the second case, in the zeroth order in $\omega\tau$, it is easy to see that no restrictions are imposed on the commutation relations for the operators $\hat{V}(t)$. This fact can be understood most simply if we interpret the momentum transfer as a "turn-on" jarring, i.e., if we consider the displacement in momentum space in (23) to be a consequence of the transformation to the wave functions of a different Hamiltonian.[2,12] Since no assumptions are made about the commutation relations in the derivation of (45) and (46), the results which follow from these equations, (24) and (25), are also valid for any jarring processes in which there is a fast momentum transfer.

This circumstance considerably expands the range of interactions for which Eqs. (24) and (25) can be used to determine the probabilities of the corresponding quantum transitions. Much of the present paper is devoted to an analysis of processes involving a fast momentum transfer and to an identification of the common features of these processes, which actually stem from the general nature of the results in (34) and (25).

We first recall some results from Ref. 1. It was shown there that various elementary processes which involve the transfer of a large momentum δp to an electron and which occur in a laser field stimulate the absorption and emission of photons of this field (we will refer to reaction channels with definite sets of such photons as "satellites"). The first stage of this process (for example, the scattering of an electron by an atom, the motion of an electron in an inhomogeneous medium, the emission or absorption of a photon by an electron, the Compton effect, β decay, or the photoelectric effect) occurs in several cases in a time τ which is much shorter than the period of the low-frequency motion of the electron caused by the external field, $2\pi/\omega$. The second stage (stimulated emission or absorption) does not depend on the physical nature of the first stage and is a universal stage, regardless of the process.[1] The problem of stimulated effects of this type can be solved once and for all as the problem of the jarring of an electron in an external field.

In nonrelativistic external fields in which the additional variable velocity of the electron, $\tilde{\mathbf{v}}$ is small in comparison with the speed of light, the probabilities for the stimulated reaction channels are governed by the parameter[1]

$$N\sim\frac{|\tilde{\mathbf{v}}\delta\mathbf{p}|}{\hbar\omega}, \tag{68}$$

which is a measure of the rms number of emitted or absorbed photons.

The intensity of the transitions stimulated by the jarring should of course be governed not only by the time interval over which the external perturbation acts on the system but also by the intensity of this perturbation, i.e., its magnitude. It can be seen from (68) that N increases with increasing intensity of the laser beam, since there is an increase in the value of $\tilde{\mathbf{v}}$.

In the limiting cases $N \gg 1$ and $N \lesssim 1$, the jarring problem can be solved by classical and quantum-mechanical methods, respectively. In the Compton effect, with $\delta\mathbf{p}=\hbar(\mathbf{k}_1-\mathbf{k}_2)$, and also in jarring due to emission accompanying absorption of a hard photon, with $\delta\mathbf{p}=\hbar\mathbf{k}$, the parameter N does not contain Planck's constant. The appearance of satellites in the emission spectrum can thus be explained from the purely classical standpoint for any value of N.

The parameter N is equal in order of magnitude to the ratio of the amplitude of the electron oscillations in the

external field, δR ($\delta\mathbf{R} = e\mathbf{E}_0/m\omega^2$ in a laser field), to the de Broglie wavelength of the electron calculated from the momentum transfer, $\lambda = \hbar/\delta p$. In this case, in which we can speak in terms of the directions of the quantities $\delta\mathbf{p}$ and $\delta\mathbf{R}$, the jarring parameter is determined by the scalar product

$$N \sim \frac{1}{\hbar} |\delta\mathbf{p}\cdot\delta\mathbf{R}|. \tag{69}$$

Up to this point we had in mind the low-frequency motion of an electron which is caused by an external laser field. We have also stated that the physical reason for the low-frequency motion of the electron which is jarred is inconsequential for our purposes. The fact that a laser field is present is not the crucial consideration; similar motions can occur without laser fields, e.g., in the vibrations of nuclei in molecules or cyrstals. The electronic shells naturally follow the motion of the nuclei in an adiabatic manner, much as the electrons move in a laser wave. The basic distinction between the oscillatory motion of the electronic shell together with the nuclei and the corresponding motion in a laser field is that in the former case the motion is quantized. The problem of stimulated transitions in this case thus reduces to the problem of the excitation of a quantum oscillator which receives a momentum δp in a collision. The uncertainty in the coordinates, δR, is due to the motion of the nuclei, which are involved in vibrations and rotations. In the most complicated situation, in which δR is not a well-defined vector, the value of N can still be estimated from

$$N \sim \frac{1}{\hbar}\delta p\cdot\delta R \sim \frac{\delta p}{\delta p_0}, \tag{70}$$

where δp_0 is the momentum spread corresponding to the low-frequency motion of the electron.

The essential reason that the parameter N is universal is the fact that the equation for the transition amplitude, (21) or (24), is universal. In by no means all cases, however, does this equation appear explicitly in the problem; it is usually masked by other, faster, processes. This is the case, for example, in problems involving the effect of nuclear vibrations on the cross sections for collisions with the electronic shell of a molecule (see Sec tion 7 below). Separating the electronic and nuclear motions by means of the adiabatic principle, and calculating the electronic part of the amplitude, we can reduce the calculation of the probability for the excitation of vibrations or rotations to a calculation of an amplitude like that in (21) or (24).

In the simplest cases, a study of these effects reduces to a solution of the problem of the jarring of a linear oscillator or rotator. In the case of a simple harmonic oscillator which is subjected to an arbitrary external force, the Schrödinger equation can be solved exactly. The transition amplitudes can also be found exactly (see, for example, Refs. 19–21). If exact equations for the transition probabilities under the influence of an external force are found with the help of the Magnus and Fer expansions, it is possible to draw a close analogy between the exact result and the jarring-theory result and to find the range of applicability of the approximation of zeroth order in $\omega\tau$ (see Section 5 for more details). The model of a simple harmonic oscillator subjected to an arbitrary external force can be used to trace the transformation from the problem of the excitation of a quantum oscillator to the problem of the excitation of a classical oscillator.

For molecules, the model of a simple harmonic oscillator is satisfactory for only the first few vibrational levels, for which the anharmonic component of the nuclear motion is small. For large values of the jarring parameter N, however, higher-lying vibrational levels (with $v \sim N$) are also excited appreciably, and to describe these levels it is necessary to take into account the difference between the harmonic-oscillator potential and the actual interatomic potential. Accordingly, in Section 5 we derive equations for the probabilities for quantum transitions in the jarring of certain nonlinear oscillators.

In collisions involving molecules, an important feature of the total cross section (the total cross section for all the stimulated reaction channels) is the radical change in its interference attributes in that range of scattering parameters in which N reaches values of the order of unity and larger, when the nuclear vibrations are taken into account (Section 7). This interference behavior stems from the fact that the electron density in molecules is distributed around more than one center, but this behavior is found only in the region $N \ll 1$; for other parameters, the collisions are completely smoothed out. In speaking of a change in the interference of the emitted or scattered particles, we can assign the jarring parameter N a simple and graphic meaning: the uncertainty in the phases of the interfering particles which is due to the uncertainty in the nuclear positions.

We have seen that the scattering actually occurs at stationary atoms, whose positions are "smeared out" in space in some manner. The nature of the cross section is governed by the nature of the interference pattern, which depends on the spatial distribution of the electron density, which is turn depends on the positions of the nuclei. The phase relations for the interfering particles play the dominant role in this interference pattern. For simplicity we will speak in terms of scattering by an electron shell which is bound to a single "smeared out" nucleus. The parameter governing the effect of the vibrations should reflect the appearance of uncertainties in the phases of each interfering particle,

$$\delta\varphi_j = \frac{1}{\hbar}\mathbf{p}_j\delta\mathbf{R}, \tag{71}$$

due to the uncertainty in the nuclear position, $\delta\mathbf{R}$ (we are now considering the simplest case, in which $\delta\mathbf{R}$ is a vector).

Let us examine the changes in the phase relations when a particle is scattered from state $|i\rangle$ to state $|f\rangle$, as shown in Fig. 4. The displacement of the nucleus from position A to A' leads to a phase difference between points a_f and a_f' (the phases are measured from the points a_i and a_i'); this difference is

$$\delta\varphi = \delta\varphi_i - \delta\varphi_f = \frac{1}{\hbar}(\mathbf{p}_i - \mathbf{p}_f)\,\delta\mathbf{R}. \tag{72}$$

This parameter which we are seeking, as a measure of the "smearing out" of the nucleus, should evidently be independent of the sign of $\delta\varphi$. From (70) we have

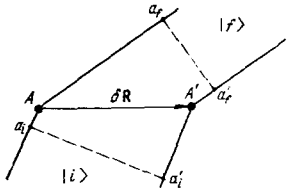

FIG. 4.

$$N \equiv |\delta\varphi| = \frac{1}{\hbar}|\delta\mathbf{p}\cdot\delta\mathbf{R}|. \tag{73}$$

Although we have been speaking so far in terms of the effect of nuclear vibrations on the scattering cross sections, similar arguments concerning phase relations can be made to characterize other effects when the spatial uncertainty δR is due to a completely different factor, say the motion of an electron in an atomic shell. In this case δR is no longer a vector, but we can still use an estimate like that in (70):

$$N \equiv |\delta\varphi| \sim \frac{1}{\hbar}\delta p\cdot\delta R. \tag{74}$$

A good example, which shows that the simple interference picture is not directly applicable but that the uncertainty in the position of the nucleus (δR) nevertheless affects the total momentum transferred to the nucleus (δp), is the photoelectric effect at a molecular electron. In the absorption of a hard photon $(\mathbf{k},\Omega)$ by an atom which is vibrating in a molecule or in a crystal, the equation for the jarring parameter follows immediately from (70) if we set $\delta\mathbf{p} = \hbar\mathbf{k} - \mathbf{p}$, where $\mathbf{p}$ is the momentum of the photoelectron. This result is found in the plane-wave approximation for the wave function of the emitted electron, with the help of (24).

In discussing the effect of nuclear vibrations on the cross section for the molecular photoelectric effect, we should emphasize that the corrections may not be at all small in this case; instead, they may substantially change the overall behavior of the cross section. A study[22] of the relativistic photoelectric effect at the H_2 molecule shows that, as the photon energy $\hbar\Omega$ increases, the number of oscillations in the plot of the photoelectric cross section as a function of the electron emission angle increases without bound if the nuclear vibrations are ignored (this behavior is the primary molecular aspect of the cross section, due to the multicenter nature of the electron cloud). When the vibrations are taken into account correctly, the oscillations in the angular distribution of the photoelectrons are smoothed over an angular range which increases with increasing energy of the incident photon. Thus, it is these vibrations which shape the characteristic angular distribution of the photoelectrons at high energies of the absorbed photons.

The cross section for the photoelectric effect at a molecule oscillates as a function of $\delta\mathbf{p}$ only under the condition $N \ll 1$. The angular dimensions of the oscillation region (the angles between $\delta\mathbf{p}$ and $\delta\mathbf{R}$) decrease with increasing photon energy $\hbar\Omega$, since the boundaries of this oscillation are governed by the condition $N \approx 1$. The nuclei of the molecule can be assumed fixed only if $N \ll 1$. This inequality determines, for example, the range of applicability of a familiar approach in calculations of the photoelectric effect at large molecules: that in which the wave function of the photoelectron is constructed from combinations of the wave functions of the electrons which would be emitted by each of the atoms in the molecule.

5. EXCITATION OF AN OSCILLATOR

a. Simple harmonic oscillator

For all the processes studied in Ref. 1, the expressions for the relative contribution of a satellite as well as the jarring parameter N are universal functions. The reason for this universality is that the entire discussion in Ref. 1 dealt with the jarring of a classical harmonic oscillator: an electron in a light wave. For a fast momentum transfer ($\omega\tau \ll 1$) to such an oscillator, the structure of this fast stage and the nature of the interaction Hamiltonian corresponding to it are unimportant, as mentioned earlier. The result depends only on the vector representing the resultant momentum transfer.

In problems involving nuclear vibrations there is a universality of an analogous type, because we are actually always dealing with the jarring of a quantum oscillator. Let us examine this problem in detail for the case of a harmonic oscillator.

We adopt a one-dimensional model in which the oscillator acquires a momentum δp in a short time. The amplitude for a transition from vibrational state $|v\rangle$ to the state $|v,+n\rangle$ is, according to (24),

$$\mathfrak{M}^0_{v\to v+n} = \langle v+n|\exp\left(-\frac{i}{\hbar}\delta p\cdot x\right)|v\rangle. \tag{75}$$

What is the range of applicability of the jarring approximation in (75) for calculating the transition amplitude $\mathfrak{M}_{v\to v+n}$? To answer this question we must either calculate the corrections of higher orders in $\omega\tau$ by using Eqs. (18)–(20), or we must completely discard the jarring concept and attempt to find the transition amplitude by means of the Magnus expansion in (35) or the Fer expansion in (39).

Fortunately, the latter approach rapidly leads to the exact solution of the problem in which a harmonic oscillator is subjected to an arbitrary external force $f(t)$. The reason is that the operator $xf(t)$, when written in the interaction picture, in which the unperturbed Hamiltonian is the harmonic-oscillator Hamiltonian, has a structure such that the commutator $[\hat{W}(t),\hat{W}(t')]$ is independent of both the coordinates and the momenta. Accordingly, all terms in the Magnus and Fer expansions beginning with the third vanish identically, and the second terms, A_2 and B_2, lead only to an inconsequential phase factor in the exact amplitude. We shall in future discard these factors, writing

$$\mathfrak{M}_{v\to v+n} = \langle v+n|\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{\infty} dt\,\hat{W}(t)\right]|v\rangle. \tag{76}$$

Equation (76) simplifies considerably if we also single out and discard one more unimportant phase factor.

To demonstrate how to do this, we rewrite Eq. (76) by expanding the exponential function in a Taylor series and introducing a sum over the intermediate states $|s_j\rangle$, for computational purposes:

$$\mathfrak{M}_{v\to v+n} = \sum_{k=0}^{\infty}\frac{1}{k!}\left(-\frac{i}{\hbar}\right)^k M_{k,} \tag{77a}$$

$$M_k = \langle v+n \mid \Big\{ \int_{-\infty}^{\infty} dt\, \hat{W}(t) \Big\}^k \mid v \rangle$$
$$= \sum_{1 \leqslant j \leqslant k-1}^{s_j} \Big\{ \langle v+n \mid \int_{-\infty}^{\infty} dt\, \hat{W}(t) \mid s_1 \rangle \ldots \langle s_{k-1} \mid \int_{-\infty}^{\infty} dt\, \hat{W}(t) \mid v \rangle \Big\}. \quad (77b)$$

The nonvanishing factors in (77) are

$$\langle l \pm 1 \mid \int_{-\infty}^{\infty} dt\, \hat{W}(t) \mid l \rangle = \langle l \pm 1 \mid x \mid l \rangle \int_{-\infty}^{\infty} dt\, f(t) \exp(\pm i\omega t)$$
$$= \langle l \pm 1 \mid x \mid l \rangle \mid f(\omega) \mid \exp(\pm i\Phi), \quad (78)$$

where ω is the oscillator frequency. In any term in the sum in (77) we can single out the same phase factor $\exp(in\Phi)$, as is easily shown. Omitting this factor, we finally find

$$\mathfrak{M}_{v \to v+n} = \langle v+n \mid \exp\left(-\tfrac{i}{\hbar} \mid f(\omega) \mid x\right) \mid v \rangle \quad (79)$$

$$f(\omega) = \int_{-\infty}^{\infty} dt\, f(t) \exp(-i\omega t). \quad (80)$$

The exact equation for the amplitude $\mathfrak{M}_{v \to v+n}$ thus differs from the amplitude in the zeroth order in $\omega\tau$, $\mathfrak{M}^0_{v \to v+n}$, only in that the modulus of the Fourier component $|f(\omega)|$ is replaced by the momentum transfer δp, so that calculations from Eqs. (75) and (79) are identical. Substituting into (79) the wave functions of the unperturbed harmonic oscillator, we find [cf. Refs. (19)–(21)]

$$\mathfrak{M}_{v \to v+n} = \sqrt{\frac{v!}{(v+n)!}} \left(\frac{\mathcal{N}_\delta^2}{2}\right)^{n/2} \exp\left(-\frac{\mathcal{N}_\delta^2}{4}\right) L_v^n\left(\frac{\mathcal{N}_\delta^2}{2}\right), \quad (81)$$

$$\mathcal{N}_\delta = \frac{|f(\omega)|}{\sqrt{m_0 \hbar\omega}}, \quad (82)$$

where $L_v^n(z)$ are the Laguerre polynomials, and m_0 is the oscillator mass.

In the zeroth order approximation in $\omega\tau$, $\mathfrak{M}^0_{v \to v+n}$ differs from $\mathfrak{M}_{v \to v+n}$ only in that $\mathcal{N}_0$ is replaced by the jarring parameter

$$N_0 = \frac{\delta p}{\sqrt{m_0 \hbar\omega}}. \quad (83)$$

The subscript on N_0 (as well as that on $\mathcal{N}_0$) means that if we write the jarring parameter as in (70) the corresponding oscillation amplitude $\delta R_0 \equiv \sqrt{\langle \delta R^2 \rangle_0}$ refers to the ground state ($v = 0$) of the oscillator. For an arbitrary state we would have to set

$$N_v \equiv \frac{1}{\hbar} \sqrt{2}\, \delta p \sqrt{\langle \delta R^2 \rangle_v} = \delta p \sqrt{\frac{2v+1}{m_0 \hbar\omega}}. \quad (84)$$

$$\mathcal{N}_v \equiv |f(\omega)| \sqrt{\frac{2v+1}{m_0 \hbar\omega}}. \quad (85)$$

All these conclusions remain valid for the stimulated transitions in a classical oscillator. The classical limit corresponds to infinitely large values of v in Eqs. (81), (84), and (85). The amplitude in (81) is transformed to the limit $v \gg 1$ by using the expansions[23]

$$z^{n/2} e^{-z/2} L_v^n(z) = \frac{\Gamma(v+n+1)}{v!} \left(v + \frac{n+1}{2}\right)^{-n/2} \times \sum_{l=0}^{\infty} a_l \left(\frac{z}{4v+2n+2}\right)^{l/2} J_{n+l}\left(\sqrt{z(4v+2n+2)}\right), \quad (86)$$

where $a_0 = 1$, $a_1 = 0$, $a_2 = (n+1)/2$, and the other coefficients are found from the recurrence relation

$$(l+2)\, a_{l+2} = (l+n+1)\, a_l - (2v+n+1)\, a_{l-1}, \quad l = 1, 2, \ldots.$$

For values of N/v which are not too small, and for large values of v, the amplitude $\mathfrak{M}_{v \to v+n}$ can thus be written as a combination of Bessel functions with different indices, and in the limit $v \to \infty$ we have

$$\mathfrak{M}_{v \to v+n} \to J_n(\mathcal{N}). \quad (87)$$

Appearing in (87) is the classical value of the parameter $\mathcal{N}$,

$$\mathcal{N} \equiv \sqrt{2v}\, \mathcal{N}_0, \quad (88)$$

which, together with the corresponding classical value of the jarring parameter N (for which the equation is precisely the same as that derived in Ref. 1), is found from (84) and (85) by taking the limit $v \to \infty$.

The problem of determining the range of applicability of the zeroth order approximation in $\omega\tau$ reduces to comparing the parameters $\mathcal{N}$ and N in this case. For small values $\omega\tau$, the corrections in $\mathcal{N}$ to the value of N are strongly governed by the time variation $f(t)$. As a rule, these corrections are of the order of $(\omega\tau)^2$ or smaller [see the discussion in the text regarding Eqs. (26)–(29)]. However, if the tails in the $f(t)$ distribution are important, as they are in the case of a Lorentzian pulse, the corrections become comparable to $\omega\tau$ and sometimes even larger.

b. Parametric "Jarring" of a harmonic oscillator at an arbitrary temperature

Let us assume that there are changes in the mass, $m_i \to m_f$, and in the frequency, $\omega_i \to \omega_f$, in a very short time τ ($\omega\tau \ll 1$) (corresponding, for example, to the absorption or emission of a neutron by a vibrating light nucleus in a molecule or to the β decay of such a nucleus). The probability of finding the oscillator in a particular quantum state $|f\rangle$ after this parametric jarring is given for an arbitrary temperature by Eq. (46).

This problem can be solved completely for the jarring of a harmonic oscillator. The quantum-density operator for a one-dimensional harmonic oscillator, written in the x representation, is

$$\hat{\rho}(x, x') = \sqrt{\frac{m\omega}{2\pi\hbar \operatorname{sh}(\hbar\omega/T)}} \exp\left[-(x^2 + x'^2)\frac{m\omega}{2\hbar} \coth\frac{\hbar\omega}{T} + xx' \frac{m\omega}{\hbar \operatorname{sh}(\hbar\omega/T)}\right], \quad (89)$$

where m and ω are the oscillator mass and frequency, and T is the temperature (in energy units).

Using this equation, we obtain the probabilities for finding the oscillator in any of the even states,

$$w_{2n} = \frac{(2n)!}{2^{2n}(n!)^2} \left(\frac{b}{a}\right)^{1/2} \left(1 - \frac{c}{a^2}\right)^n s^{-2n-1/2} \times (s-1)^n (s-\lambda)^n F\left(-n, -n; \tfrac{1}{2}; \frac{\lambda}{(s-1)(s-\lambda)}\right), \quad (90)$$

and in any of the odd states,

$$w_{2n+1} = \frac{(2n+1)!}{2^{2n}(n!)^2} \left(\frac{b}{a}\right)^{3/2} \left(1 - \frac{c}{a^2}\right)^n s^{-2n-3/2} \times (s-1)^n (s-\lambda)^n F\left(-n, -n; \tfrac{3}{2}; \frac{\lambda}{(s-\lambda)(s-1)}\right). \quad (91)$$

The subscripts on w correspond to the usual indexing of the oscillator levels; F is the hypergeometric function; $n = 0, 1, \ldots$;

$$a^2 = \frac{c}{2} + \frac{m_i\omega_i}{2\hbar} \coth\frac{\hbar\omega_i}{T}, \quad ab = \frac{m_i\omega_i}{2\hbar \operatorname{sh}(\hbar\omega_i/T)},$$
$$c = \frac{m_f\omega_f}{\hbar}, \quad s = \frac{a^2 - b^2}{c}, \quad \lambda = \frac{b^2}{a^2 - c}.$$

In particular, the probabilities for the filling the ground and first excited states after jarring are

$$w_0 = \sqrt{\frac{bc}{a(a^2-b^2)}}, \tag{92}$$

$$w_1 = w_0^3. \tag{93}$$

c. Anharmonic oscillators

With the ultimate goal of applying the problem of the jarring of an oscillator to real molecular systems, we will briefly discuss some particular features of stimulated transitions when the nuclear vibrations are anharmonic. We mentioned above that the anharmonicity must definitely be taken into account at large values of the jarring parameter N, at which the high-lying vibrational levels are excited efficiently.

The most important distinguishing feature of the real interatomic potential $U(R)$ (for definiteness, we assume a diatomic molecule) is that it is asymmetric with respect to the equilibrium nuclear separation R_0. To avoid "losing" this asymmetry, we must study the expansion of $U(r)$ in powers of $1/R$ or $(R-R_0)/R$ instead of in the traditional power series in $(R-R_0)$ (see, for example, Refs. 24–27).

Using even the simple potential

$$U(R) = U_0 - \frac{\alpha}{R} + \frac{\beta}{R^2}, \quad \alpha, \beta > 0, \tag{94}$$

we can take into account the anharmonicity of the vibrations in molecules relatively easily. It is also important to note that the Schrödinger equation with a central potential like that in (94) can be solved exactly for arbitrary vibrational—rotational states [by changing the constants in (94), we can convert this problem to the hydrogen-atom problem]. By using potential (94) we can thus rigorously analyze the vibrational—rotational interaction in a molecule, while this could not be done on the basis of, for example, the harmonic potential.

As in subsection a), we consider the excitation resulting from the jarring of a one-dimensional oscillator with a potential energy of the type in (94):

$$U(x) = U_0 - \frac{\alpha}{x} + \frac{\beta}{x^2}, \quad \alpha, \beta > 0. \tag{95}$$

We denote the quantum states of the discrete spectrum of this oscillator by the integers $v = 0, 1, 2, \ldots$. The corresponding energies are

$$E_v = U_0 - \frac{\hbar^2 q^2}{2m_0}, \quad q = \frac{\alpha m_0}{\hbar^2 (a+v)}, \quad a = \frac{1}{2} + \sqrt{\frac{1}{4} + \frac{2\beta m_0}{\hbar^2}}, \tag{96}$$

and the corresponding wave functions are

$$\Psi_v(x) = \frac{1}{\Gamma(2a)} \sqrt{\frac{q\Gamma(v+2a)}{v!(v+a)}} (2qx)^a e^{-qx} F(-v, 2a; 2qx), \tag{97}$$

where F is the degenerate hypergeometric function.

The "smearing out" of the ground-state momenta of the oscillator in (95)–(97) is of the order of $\hbar q$. In the problem of the excitation of this oscillator as momentum is rapidly transferred to it a jarring parameter like that in (70) or (83) should accordingly arise:

$$N_0 = \frac{\delta p}{\hbar q}. \tag{98}$$

In fact, the probability for excitation involving the $0 \to v$ transition is calculated from Eq. (24) to be

$$|\mathfrak{M}_{0\to v}|^2 = \frac{a}{a+v} \frac{\Gamma(v+2a)}{v!\Gamma(2a)} [4a(a+v)]^{2a+1} \frac{a^2 N_0^2 (v^2 + a^2 N_0^2)^{v-1}}{[(2a+v)^2 + a^2 N_0^2]^{v+2a+1}}. \tag{99}$$

We can also find a simple equation for the probability that the oscillator of (95)–(97) does not become excited when it is jarred:

$$|\mathfrak{M}_{0\to 0}|^2 = \left(1 + \frac{N_0^2}{4}\right)^{-2a-1}. \tag{100}$$

By way of comparison we note that the probability for this event in the case of a harmonic oscillator decreases exponentially with increasing N [see Eq. (81)], while in the case of the one-dimensional Morse potential

$$U(x) = U_0 \left[\exp\left(-\frac{2x}{x_0}\right) - 2\exp\left(-\frac{x}{x_0}\right)\right] \tag{101}$$

it is given by

$$|\mathfrak{M}_{0\to 0}|^2 = \prod_{n=0}^{\infty} \left[1 + \left(\frac{N_0}{n+\varepsilon-1}\right)^2\right]^{-1}, \tag{102}$$

$$N_0 = \frac{1}{\hbar} \delta p \cdot x_0, \quad \varepsilon = \frac{1}{\hbar} x_0 \sqrt{2m_0 U_0}. \tag{103}$$

It can be seen from these examples that the probabilities for the stimulated transitions in very different situations are governed by the same jarring parameter, given in (70). This jarring parameter is thus a principal characteristic of these problems.

6. "JARRING" OF ELECTRONS FROM ATOMIC SHELLS

a. Collisions of neutrons with light atoms

Apparently the first direct application of the jarring concept in calculating the amplitudes of quantum transitions was in the problem of the ionization of atoms in collisions with neutrons.[28] If the atomic nucleus acquires a momentum $\hbar\mathbf{q}$ in a short time τ, the transition amplitude in zeroth order in $\omega\tau$ can be calculated from Eq. (24) (here $\hbar\omega$ is a typical electron energy).

Of practical interest are the ionization probability, integrated over the momentum directions of the emitted electron,

$$\frac{dw}{dp} = (2\pi\hbar)^{-3} \int d\left(\frac{\mathbf{p}}{p}\right) p^2 |\langle f | e^{i\mathbf{q}\mathbf{r}} | i \rangle|^2, \tag{104}$$

and the total ionization probability for a given q,

$$w(q) = \int dp \frac{dw}{dp}. \tag{105}$$

If the question is posed in such a manner that we can also introduce some distribution function $g(q)$ [the quantity $g(q)dq$ determines the probability for the transfer to the nucleus of a momentum between q and $q+dq$], then the ionization probability is

$$w = \int dq\, g(q)\, w(q). \tag{106}$$

For a hydrogen-like atom, in which case the electrons are jarred from the K or L shell, the probability dw/dp can be found analytically. The jarring parameter, which is a measure of the transition probability in this problem, can be defined as follows:

$$N \equiv \frac{nq}{\eta} = \frac{nq\hbar^2}{Ze^2 m}, \tag{107}$$

where n is the principle quantum number of the shell from which the electron is ejected, Z is the nuclear charge, and m is the electron mass. We also introduce the dimensionless ratio $\gamma \equiv np/\hbar\eta$. The probability for the jarring of an electron from the K shell is[28]

$$\frac{dw_K}{d\gamma} = \frac{2^8 \cdot N^2\gamma}{3[1-\exp(-2\pi/\gamma)]} \frac{3N^2+\gamma^2+1}{(\alpha^2+4\gamma^2)^3} \exp\left(-\frac{2}{\gamma}\tan^{-1}\frac{2\gamma}{\alpha}\right), \tag{108}$$

where $\alpha = N^2 - \gamma^2 + 1$, and the function $\tan^{-1}$ is defined to

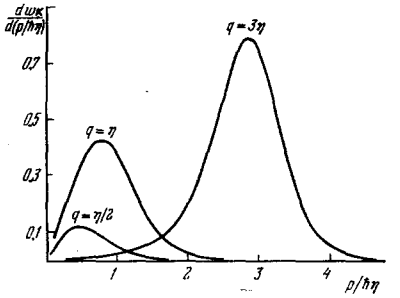

FIG. 5. Spectrum of electrons "jarred" from the K shell.

be $\pi+\tan^{-1}(x)$ for $x<0$ or $\tan^{-1}(x)$ for $x\geq 0$.

Figure 5 shows the spectra of electrons jarred from the K shell for fixed values of N. Figure 6 shows the total probability as a function of N.

The probabilities for the jarring of electrons from any of the L subshells are found from the general equation (it is shown below that the mathematical problem of calculating $dw/d\rho$ is equivalent to the problem of calculating the cross section for the nonrelativistic Compton effect[29-31])

$$\frac{dw_L}{d\gamma}=\frac{2^{11}N^2\gamma}{15\varkappa^5}S\frac{\exp[-(4/\gamma)\tan^{-1}(2\gamma/\alpha)]}{1-\exp(-4\pi/\gamma)},\qquad \varkappa=(1+\gamma^2+N^2)^2-4N^2\gamma^2. \tag{109}$$

The equations for the function S in (109) are different for the different subshells. For the $2S$ state, for example,

$$S_{2S}=5\varkappa^3(3N^2+\gamma^2+4)-2^4\,15\varkappa N^2(N^2+1)+2^6N^2(15N^4+10N^2+2\gamma^4+5\gamma^2-5N^2\gamma^2+3). \tag{110}$$

For the jarring of an electron initially in the $2P^{(\pm1)}$ level, we have

$$S_{2P}^{(\pm1)}=4\varkappa(\gamma^2+4)(5N^2+\gamma^2+1). \tag{111}$$

Finally, for a $2P^{(0)}$ electron, we have ($\beta=\gamma^2-N^2+1$)

$$S_{2P}^{(0)}=\beta^2(7\gamma^4+40\gamma^2+48)+\frac{5}{2}\beta(\gamma^2+4)(4\beta(\gamma^2+4N^2)-\varkappa)+\frac{15}{16}[4\beta(\gamma^2-4N^2)-\varkappa]^2. \tag{112}$$

The results calculated from Eqs. (109)–(112) are shown in Figs. 7 and 8. The cases in which electrons are jarred from the K and L shells can be distinguished experimentally by detecting the radiation emitted by the excited ions which are formed when the electrons are jarred from the different inner shells.

b. Compton effect at a weakly bound electron

In the scattering of a photon by a bound electron, a jarring situation arises if the electron energies in the initial and final states are small in comparison with the photon energy $\hbar\Omega$. In this case the jarring time is estimated as follows: The absorption of the incident photon leads to a deviation $\Delta E\sim\hbar\Omega$ from energy conservation, so the scattered photon must be emitted in a time $\tau\sim\hbar/\Delta E\sim 1/\Omega$.

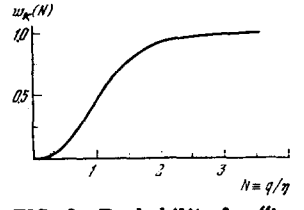

FIG. 6. Probability for "jarring" of electrons from the K shell.

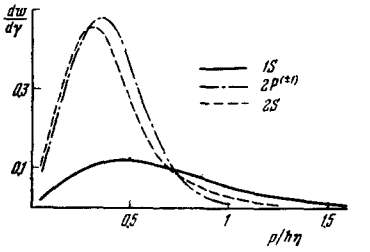

FIG. 7. Spectra of electrons "jarred" from the K and L shells ($q=\eta/2$).

That the elastic scattering of x rays by light atoms is a jarring process was emphasized by Trammell[32] in connection with the difference in the Debye–Waller factors for the resonant and nonresonant scattering of radiation in crystals. For the case in which the energies of the incident photons are not relativistic, and the corrections of second order in $\omega\tau$ and $\hbar\Omega/mc^2$ are negligible, the cross section for the Compton effect can always be written in the nonrelativistic jarring form for any scattering channels, accompanied by both bound–bound and bound–free transitions of the electron.[30] As an example, for the ionization channel involving the ejection of an electron from an arbitrary nS shell, in first order in $\omega\tau$ and $\hbar\Omega/mc^2$, we have the usual result [in Eqs. (113)–(116), we use the relativistic units, with $\hbar=c=1$]:

$$\frac{d^2\sigma_S}{dt\,d\Omega_2\,do_2}=\frac{r_e^2(1+\cos^2\theta)}{8\pi^2}\frac{\Omega_2}{\Omega_1}\,pm\,|\langle f|e^{i\mathbf{kr}}|i\rangle|^2. \tag{113}$$

Here $\mathbf{k}=\mathbf{k}_1-\mathbf{k}_2$, $\mathbf{k}_{1,2}$ are the wave vectors of the incident and scattered photons, $\Omega_{1,2}$ are their frequencies, θ is the scattering angle, $\mathbf{p}$ is the momentum of the emitted electron, $\mathbf{pk}=pkt$, $r_e=e^2/mc^2$ is the classical radius of the electron, $d\theta_2$ is the element of solid angle containing the vector $\mathbf{k}_2$, and the wave function of the final state of the electron is assumed normalized to a unit volume. For the Compton effect at nP electrons, it is convenient to write[30]

$$\frac{d^2\sigma_P}{dt\,d\Omega_2\,do_2}=\frac{r_e^2(1+\cos^2\theta)}{8\pi^2}\,pm\frac{\Omega_2}{\Omega_1}\sum_{j=1}^{3}\rho_j^2|M_j|^2, \tag{114}$$

$$\mathbf{M}=\langle f|e^{i\mathbf{kr}}\frac{\mathbf{r}}{r}|R_P\rangle. \tag{115}$$

For subshells with different angular momenta and different angular-momentum projections ($nP_{1/2}$, $nP_{3/2,1/2}$, $nP_{3/2,3/2}$), the vector $\boldsymbol{\rho}$ has the components (1, 1, 1),

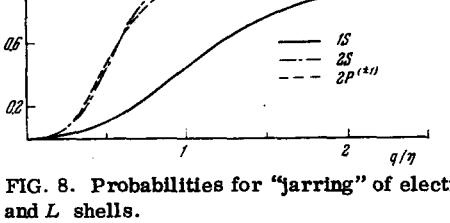

FIG. 8. Probabilities for "jarring" of electrons from the K and L shells.

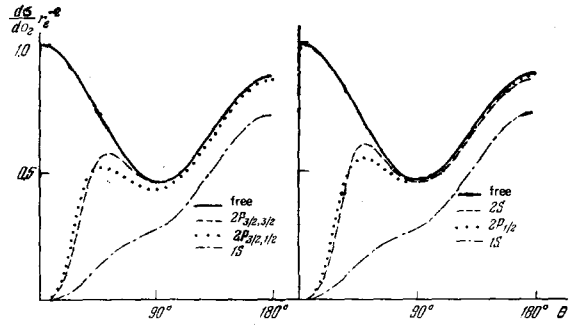

FIG. 9. Angular distributions of scattered photons in the Compton effect at a free electron at rest (solid curves); other curves-the same, for the electrons of the K and L shells of the carbon atom ($Z_N=6$). The energy of the incident photon is 20 keV.

$(1/\sqrt{2})(-1,-1,2)$, and $\sqrt{3/2}\,(1,-1,0)$, respectively. Here R_p is the radial part of the wave function.

To calculate the cross sections for the Compton effect at K and L electrons from Eqs. (113)–(115), we can clearly use the results of (108)–(112). For example, in the case of a $2P_{3/2,3/2}$ electron we must use Eq. (109), with Eq. (111) for S. For electrons from the $2P_{1/2}$ and $2P_{3/2,1/2}$ subshells, we must use (109) with the combinations $(2S_{2P}^{(1)}+S_{2P}^{(0)})/3$ and $(S_{2P}^{(1)}+2S_{2P}^{(0)})/3$. As an example, Fig. 9 shows the angular distributions of the Compton photons in scattering by free electrons and by K and L electrons.

The states of the electrons in the $2P$ shell could of course also be classified according to the usual nonrelativistic scheme, as in the preceding section. In this classification, the Compton cross sections are directly related to the probabilities for the jarring of electrons from the K and L shells, in (108)–(112):

$$\frac{d^2\sigma}{d\Omega_2 d\omega_2}=r_e^2(1+\cos^2\theta)\,\frac{\Omega_2}{\Omega_1}\frac{m}{p}\frac{dw}{dp}. \tag{116}$$

Figure 10 shows the angular distributions of the Compton photons found by integrating (116). The interpretation of the scattering as a jarring process has a special heuristic value for the range of collision parameters such that

$$\alpha Zmc^2 \gtrsim \hbar\Omega \gg |E_b|$$

(α is the fine-structure constant, Z is the effective nuclear charge, and E_b is the electron binding energy in the atom). For this energy range, the Compton cross section is strongly affected by the interaction of the electron with the ion core in the final state, since the emitted electron has a relatively small momentum, on the average. As a rule, there is no hope of taking this effect into account exactly, but this is not the case in nonrelativistic calculations of the cross sections for ionization involving the ejection of electrons from inner shells. We will now show how these exact calculations can be used to test the jarring approach: a principal characteristic of the angular distribution of the scattered photons in the Compton effect at a bound electron should again by a jarring parameter N, chosen in an appropriate manner. The behavior found here remains the same in other, more comlicated situations, which are not amenable to a rigorous analysis.

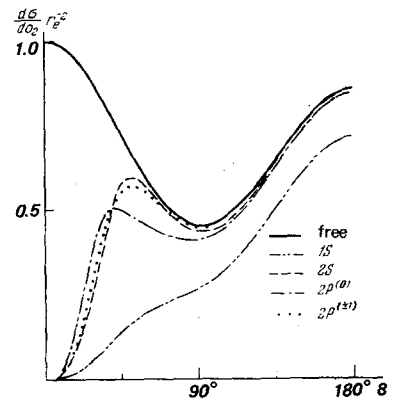

FIG. 10. The same as in Fig. 9, according to an integration of Eq. (116).

In the limit of an infinitely heavy nucleus, momentum is not conserved in the Compton effect at a bound electron, and we are dealing with precisely that case in which the quantity δp, which is incorporated in the corresponding jarring parameter, becomes indefinite in the calculation of the amplitude in (24). Nevertheless, in the Compton effect it is possible[30,41] to introduce a jarring parameter of the type in (70),

$$N\sim\delta k\cdot\delta R\sim\frac{\hbar\delta k}{\delta p_0}, \tag{117}$$

if we take into account the fact that the Compton line for a weakly bound electron is relatively narrow [$\hbar\delta k$ in Eq. (117) is understood as the rms value of the momentum transferred to the electron in the scattering].

It is simplest to seek δk in first order in αZ, using the Compton equation for this purpose:

$$\Omega_1=b\Omega_2,\quad b=1+\frac{\hbar\Omega_1}{mc^2}(1-\cos\theta). \tag{118}$$

For example, if the electron initially occupies an atomic shell with the principal quantum number n, we can conveniently write the result of the calculations for any energy of the incident photon, $\hbar\Omega_1$, in the following form [cf. Refs. 30 and 31 and Eq. (107)]:

$$N=\frac{n\hbar\Omega_1\sqrt{1+b^2-2b\cos\theta}}{\alpha Zmc^2 b}. \tag{119}$$

The magnitude of the parameter N is governed by the difference between the angular distribution of the scattered photons in the case of a bound electron and the Klein–Nishina distribution:

$$\frac{d\sigma_{K\text{-}N}}{d\omega_2}=\frac{r_e^2}{2b^2}\left(b+\frac{1}{b}-\sin^2\theta\right). \tag{120}$$

As shown in Refs. 30 and 31, the ratio of the cross section for the Compton effect at a free electron, $d\sigma_b/d\omega_2$, to the Klein–Nishina cross section in (120) is in fact a convenient measure of the parameter in (119). This conclusion is in complete agreement with the jarring interpretation of the scattering process. All the diagrams like Figs. 9 and 10 can be described by a few curves for the ratio $d\sigma_b/d\sigma_{K\text{-}N}$, depending on the value of the jarring parameter N (see Figs. 11–15). The reason for this universality is that the cross section is governed primarily by the peak of the Compton line, whose width becomes smaller as $\omega\tau$ becomes smaller. Here we are seeing the profound analogy between the behavior of

$d\sigma_b/d\sigma_{K-N}$ and w [see subsection a)] as functions of the corresponding jarring parameters, in (119) and (107).

The jarring approach is convenient for finding a systematic arrangement for the experimental data on the scattering of hard γ rays by atomic electrons. The theory for the angular distribution of Compton photons for relativistic γ-ray energies ($\hbar\Omega \gtrsim mc^2$) is extremely fragmentary and unreliable (the best discussion of this question is in Ref. 33). Even numerical estimates are very laborious,[33] so all the situations which arise experimentally cannot be analyzed, and general aspects of the behavior cannot be discerned.

The jarring interpretation of Compton scattering by weakly bound electrons, in contrast, yields the most important aspect of the behavior: that the parameter N is a measure of the cross section. The product $\omega\tau$ must of course be small. Figures 11 and 12 show the experimental values of the ratio $d\sigma_b/d\sigma_{K-N}$ as a function of N, found from data on the scattering of γ rays with energies between 279 keV and 1.12 MeV by the K shell of Sn and Sm atoms, through angles θ between 20° and 160°. In the calculations of N from Eq. (119), here and below, the screening of the nuclear field by other electrons is taken into account by the Slater rules. The theoretical curves in Figs. 11 and 12 are taken from Ref. 31; they correspond to nonrelativistic γ-ray energies, $\hbar\Omega \ll mc^2$.

The theoretical curve required for analyzing the experimental data on scattering by the filled L shell can be found from the results for other subshells. Yudin[30] has studied the behavior of $d\sigma_b/d\sigma_{K-N}$ as a function of the jarring parameter N for scattering by subshells, classified in accordance with relativistic theory. The results are shown in Fig. 13. The corresponding results for the nonrelativistic level scheme can be found in a straightforward manner (Fig. 14). The average Compton cross section, divided by the Klein–Nishina cross section, is shown in Fig. 15, along with experimental data on the Compton effect at Pb and Th atoms.

On the basis of Figs. 11, 12, and 15, we can state that the nonrelativistic theoretical curves generally agree well with the experimental results. There are some discrepancies, caused by several factors. We are concerned here primarily with those factors which affect the theoretical calculations.

First, the inequality $\omega\tau \ll 1$ is barely satisfied, so it would improve the situation to take into account the higher-order corrections to the zeroth-order jarring approximation. Furthermore, we recall that Eq. (119) for the jarring parameter is derived in the first approximation in $\alpha Z/n$ (the results for the approximations of zeroth and first order in $\alpha Z/n$ are the same). The higher-order corrections can be taken into account in the following manner.

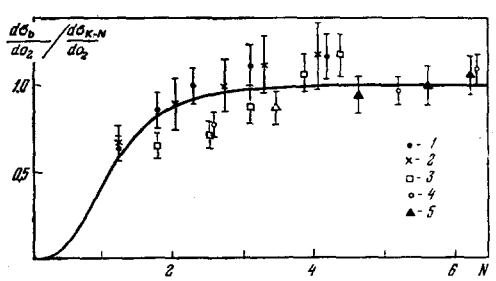

FIG. 11. Compton effect at the K shell of Sn ($Z_N = 50$). Experimental data: 1) Ref. 34; 2) Ref. 35; 3) Ref. 36; 4) Ref. 37; 5) Ref. 38.

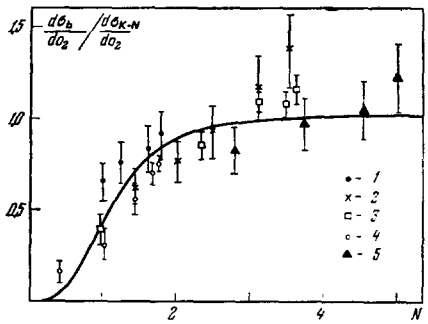

FIG. 12. Compton effect at the K shell of Sm ($Z_N = 62$). Experimental data: 1) Ref. 39; 2) Ref. 36; 3) Ref. 40; 4) Ref. 41; 5) Ref. 38.

The scattering of a photon by a weakly bound electron at a large value of N can be interpreted as scattering by a free electron which has precisely the same momentum distribution as the bound electron. The cross section for the Compton effect at a free electron which is moving with the 4-momentum $(E, \mathbf{p})$ is

$$\frac{d\sigma_{\text{free}}}{d\omega_2} = \frac{1}{2}\left[\frac{r_e mc^2}{a+(b-1)mc^2}\right]^2 \times\left\{\frac{b-1}{a}mc^2+\left[1-\frac{m^2c^4(1-\cos\theta)}{a(E-pc\cos\theta_1)}\right]^2+\frac{a}{a+(b-1)mc^2}\right\}, \tag{121}$$

where $a = E - pc\cos\theta_2$ [the quantity b is defined in (118)], and $\theta_{1,2}$ are the angles between $\mathbf{p}$ and the wave vectors of the incident and scattered photons, $\mathbf{k}_{1,2}$. The angular distribution of the γ rays in the Compton effect at the bound electron should be compared with the cross section in (121), averaged in the appropriate manner, rather than with the Klein–Nishina cross section. It would also be a simple matter to refine the parameter N to take into account the corrections of the various orders in $\alpha Z/n$ in the equation for δk.

However, it is not necessary to pursue this point theoretically at present, because of the large errors and the discrepancies found even in identical measurements by different investigators. The difficulties confronting the

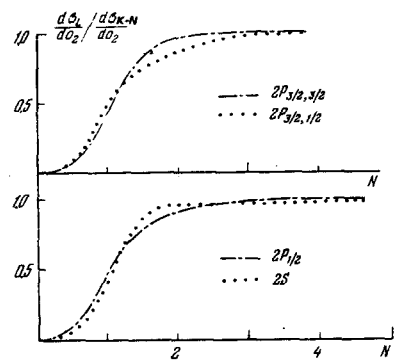

FIG. 13. Ratio of the cross section for the Compton effect at L-shell electrons to the Klein–Nishina cross section as a function of the "jarring" parameter N.

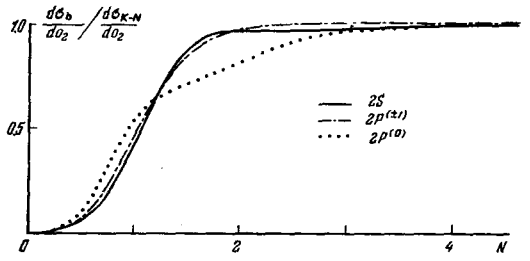

FIG. 14. Ratio of the cross section for the Compton effect at L-shell electrons to the Klein-Nishina cross section as a function of N.

experimentalists are related to the complex nature of the measurements of the cross sections for ionization events involving the ejection of an electron from a specific atomic shell. The lifetime of the vacancy left in an inner shell when an electron is ejected is $t \sim 10^{-16}$ sec. This vacancy is then filled by an electron from an outer shell, and the x ray emitted in the process is measured in coincidence with the scattered γ ray. Unfortunately, it is difficult to arange these coincidence measurements, since some of the γ rays and some of the x rays are absorbed in the target, and there is also the additional background bremsstrahlung of the Compton electrons. These and other side effects make the measured ratio $d\sigma_b/d\sigma_{K-N}$ very sensitive to the shape of the sample, the scattering angle, and other specific experimental conditions [the errors reported by many investigators, shown in Figs. 11, 12, and 15, frequently refer only to the counting of detected events; the experimental angular distributions of γ rays in scattering by Al ($Z_N = 13$) or Be ($Z_N = 4$) are used as $d\sigma_{K-N}/d\sigma_2$].

7. OTHER COLLISION PROCESSES

a. Emission from a nucleus in a molecule or crystal

Stimulated transitions in processes involving a vibrating atom occur in collisions not only with an electronic shell but also with the nucleus itself. Familiar examples are the absorption, emission, and scattering of neutrons by atoms in a crystal lattice[44] or in a molecule.[45]

Far more important in practice than the capture of neutrons by atoms in a crystal is the Mössbauer effect, which is an equivalent effect from the theoretical standpoint.[46-48] In reviewing the basic features of this effect, we distinguish among three very different time intervals: 1. the collision time τ, which is the time required for an element of the wave of length $l \sim c/\Omega$ to traverse the nucleus, whose dimensions are far smaller than l in all

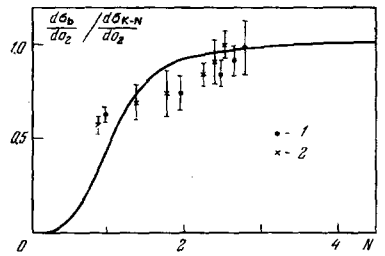

FIG. 15. Compton effect at the L-shell of Pb ($Z_N = 82$) and Th ($Z_N = 90$). Experimental data: 1) Ref. 42; 2) Ref. 43.

real cases. Thus $\tau \sim 1/\Omega$; 2. the characteristic time of the motion of the nuclei in the molecule or the solid, $1/\omega$; 3. the lifetime of the excited state, $T \sim 1/\Gamma$, where Γ is the Mössbauer line width. The average times T usually lie between 10^{-7} and 10^{-9} sec.

It is not difficult to establish the relations among these times:

$$\tau \ll \frac{1}{\omega} \ll T. \tag{122}$$

The first inequality in (122) means that the emission process can be treated as a jarring process with respect to the slow nuclear motions. In this case the parameter N is

$$N = |\mathbf{k}\delta\mathbf{R}| \sim k\delta R. \tag{123}$$

Since this parameter does not contain Planck's constant, it is of purely classical origin.[1] The probability for the emission from a vibrating nucleus, with any change whatever in the vibrational state of this nucleus, is determined in the harmonic approximation from the results in (81) and (83) for quantum vibrations or (87) for classical vibrations.

The second inequality in (122) figures in the resonant scattering of the Mössbauer radiation. The long lifetime of the nucleus in its excited state, in comparison with τ and $1/\omega$, means that two steps of the process—the resonant absorption and the recoilless emission—can be treated as absolutely independent. As a result, the probability for resonant scattering is proportional to the product of the probabilities for the transitions due to the two independent jarrings. For a crystal, this probability is proportional to the product of two Lamb–Mössbauer factors, which contain the jarring parameters $N_1 = |\mathbf{k}_1\delta\mathbf{R}|$ and $N_2 = |\mathbf{k}_2\delta\mathbf{R}|$.

Up to this point we have been talking about the "natural" vibrations of γ-emitting nuclei in molecules or crystals and the appearance of satellites in the γ spectrum against the background of this relatively slow motion. Recent advances in the field of lasers are drawing attention to the effect of strong fields on the pumping of atomic and molecular systems, usually through stimulated internal transitions under the influence of a resonant field. It is thus pertinenet to examine jarring transitions against the background of the stimulated slow motions of a quantum system as it is pumped in an external field.

For selective pumping with, say, an increase in δR for a certain type of motion, we can arrange a situation such that the corresponding jarring parameter $N \sim \delta p \delta R/\hbar$ and the transition probabilities undergo changes far greater than in the absence of the laser field. It is thus possible to distinguish some particular channel in a narrow spectral interval (the width of this interval is governed by the resonant-excitation conditions, in particular, the wavelength spread of the laser beam). Noteworthy among the studies in this direction are Refs. 49 and 50.

b. Collisions of molecules with electrons and heavy particles

Among the stimulated effects in a laser field, the one that has been studied in greatest detail is "stimulated

bremsstrahlung,"[51] which is an emission or absorption of laser photons which is stimulated by electron scattering. A direct analog of stimulated bremsstrahlung is quasielastic scattering of an electron by a vibrating atom. If the atomic vibrations are classical, this scattering is very similar in nature to stimulated bremsstrahlung. In most cases, on the other hand, there is a quantum analog of stimulated bremsstrahlung, in which the nucleus is in quantized motion, for example, in a molecule.

However, there is also an important distinction between these processes: in the quantum case, the fact that the incident electron is identical to the target electrons comes into play. The presence of $\delta\mathbf{p}$ in (24) does not prove that the amplitude depends only on the momentum transfer. Differences should evidently be expected in collisions of atoms or molecules with electrons in those cases when exchange effects must be taken into account in calculating the cross sections.

On the example of electron scattering by the K shell of a homonuclear diatomic molecule it can be shown that when the exchange interactions are taken into account the influence of the nuclear vibrations (for, say, only those scattering channels which involve the excitation of vibrations) is characterized not simply by the single parameter N, which contains the momentum transfer, but also by three other parameters, which contain instead of $\delta\mathbf{p}$, the combinations $\mathbf{p}_1$, $\mathbf{p}_2$, and $\mathbf{p}_1+\mathbf{p}_2$ ($\mathbf{p}_{1,2}$ are the momenta of the incident and scattered electrons).

To see how the jarring parameter N arises in the most general case of a collision of a molecule with a fast charged particle, we assume that the fast particle, with charge Ze, mass m, and momentum $\mathbf{p}_0$, collides with the target molecule and ionizes it. We denote the energy and momentum of the ejected electron by E_2 and $\mathbf{p}_2$, while the momentum of the scattered particle in the final state is $\mathbf{p}_1$. The triply differential cross section for this process is written as follows in the first Born approximation[52]:

$$\frac{d^3\sigma}{dE_2\,do_1\,do_2}=\frac{p_1}{p_0}\left(\frac{2mZe}{p^2}\right)^2\langle\Psi_f|\sum_j e^{i\mathbf{p}\mathbf{r}_j/\hbar}|\Psi_i\rangle|^2, \tag{124}$$

where Ψ_i and Ψ_f are the wave functions of the initial and final states of the scattering system, $\mathbf{p}=\mathbf{p}_1-\mathbf{p}_2$, $do_{1,2}$ are the elements of solid angle containing the vectors $\mathbf{p}_{1,2}$, and the summation over j is carried out over all the electrons in the molecule.

In the adiabatic approximation we have

$$\Psi=\Phi(r,R)\,\Lambda(Q)\,\Theta(\theta), \tag{125}$$

where $\Phi(r,R)$, $\Lambda(q)$, and $\Theta(\theta)$ are the electronic, vibrational, and rotational wave functions, respectively; r and R represent the sets of electronic and nuclear coordinates; Q represents the coordinates of the normal vibrational modes; and θ represents the Euler angles, which define the spatial orientation of the molecule. The electronic coordinate wave function of the final state is written as a linear combination of products of the type $\Phi_{ion}\Phi(\mathbf{p}_2)$.

We are generally interested in the cross section summed over all the final states of the resulting ion:

$$\frac{d^3\sigma}{dE_2\,do_1\,do_2}\approx\frac{p_1}{p_0}\left(\frac{2mZe}{p^2}\right)^2\langle\Lambda_f\Theta_f||\mathscr{M}|^2|\Lambda_i\Theta_i\rangle, \tag{126}$$

$$\mathscr{M}(R)=\langle\Phi(\mathbf{p}_2)|\sum_j e^{i\mathbf{p}\mathbf{r}_j/\hbar}|\Phi_i(r,R)\rangle. \tag{127}$$

The notation $\mathscr{M}(R)$ is adopted to emphasize that the matrix elements between the electronic states depends on the set of nuclear coordinates. The variation with R in (127) is governed by the vector $\delta\mathbf{p}\approx\mathbf{p}_0-\mathbf{p}_1-\mathbf{p}_2$, so the characteristic parameter

$$N\sim\frac{1}{\hbar}\delta p\cdot\delta R$$

appears in the integration over the vibrational coordinates in (126).

Let us consider the particular case of channels involving the excitation of vibrations in a diatomic molecule without a change in electronic state; we assume that the axis of the molecule is oriented along a definite direction. The cross section for a collision involving the transition of the molecule to a fixed vibrational state, $|v\rangle\to|v+n\rangle$, incorporates the characteristic two-center factor

$$\chi=1+(\delta_{n,2l}-\delta_{n,2l+1})\cos(\mathbf{q}\mathbf{R}_0), \tag{128}$$

where $\hbar\mathbf{q}$ is the momentum transferred to the nuclei, and $\mathbf{R}_0$ is the equilibrium nuclear separation. The cross section for a collision involving the transition from $|v\rangle$ to all vibrational states is given in the harmonic approximation by

$$d\sigma^v=d\sigma_0[1+e^{-N^2}L_v(2N^2)\cos(\mathbf{q}\mathbf{R}_0)]. \tag{129}$$

The oscillatory term in the cross section in (129) (the quantity added to unity in brackets) is peculiar to molecules and does not arise in the case of scattering by atoms. It is seen from (129) that this specific feature of the molecular case vanishes completely at large values of the jarring parameter. Then there is absolutely no manifestation of the multicenter nature of the electron distribution in the molecule.

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Note added in proof. 1. The only modification of the theory of sudden perturbations for the most general case, in which the commutator $[\hat{V}(t),\hat{V}(t')]$ is any arbitrary operator, is that the time-evolution operator $\hat{S}_0(t,t')$ is written in zeroth order in $\omega\tau$ by means of the Magnus or Fer expansion, which incorporates successive commutators of the operators $\hat{V}(t)$ taken at various times, as mentioned earlier. The entire calculation scheme and, in particular, the equations for calculating the higher orders in $\omega\tau$, (19)–(20), remain the same.

2. All the results of Section 2 can be generalized directly to the case in which the unperturbed Hamiltonian also depends on the time by formally replacing $\hat{\mathscr{H}}_0(t)$ by $\int^t dt\hat{\mathscr{H}}_0(t)$ [provided, of course, that $[\hat{\mathscr{H}}_0(t),\hat{\mathscr{H}}(t')]\equiv 0$]. To find the transition amplitudes in the case of sudden perturbations in this case it is more convenient to use the Schrödinger picture, transforming back from the time-evolution $\hat{S}(t,t')$ (calculated with the necessary accuracy) to the operator $\hat{U}(t,t')$.

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