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QUASIELASTIC KNOCK-OUT OF AN ELECTRON BY A FAST ELECTRON FROM ATOMS, MOLECULES, AND VERY THIN CRYSTALLINE FILMS

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The possibilities of obtaining new information on electron states of atoms, molecules and thin (~60–100 Å) crystalline films in experiments on quasielastic knock-out of valence electrons from a target by 10–keV electrons are discussed. It is assumed that both emerging electrons are recorded by a coincidence technique (the (e, 2e) process, which is an analog of the (p, 2p) nuclear reaction). The feasibility of the method is demonstrated in several concrete cases, such as the hydrogen atom, H_2 and N_2 molecules, and Al, KCl, and Cu single crystals. The calculations are performed in the impulse approximation. The angular correlation function of the emitted electrons is found to be very sensitive to such details of the electron wave functions as hybridization of s- and p-orbits in the molecules, type of bond between the electrons and crystal lattice, etc. Although high energy resolution and high beam intensities are required, the experiment appears to be quite feasible.

1. INTRODUCTION

THE use of processes similar to direct nuclear reactions uncover great possibilities for the investigation of the electronic structure of atoms, molecules, and solids. As noted in a preliminary communication,[1] great interest attaches to the reaction of quasielastic knock-out of an electron by an electron (e, 2e), which is an analog of the nuclear quasielastic knock-out reactions (p, 2p), (α, 2α), etc.[2] It is important[1] that the angular-correlation curves of the two final electrons in the (e, 2e) reaction give the Fourier spectra of the wave functions of the knock-out electron, and measurement of the energies of these electrons makes it possible to determine the binding energy of the knock-out electron (see also [3]).

From among the other methods, the closest to the method using the (e, 2e) reaction is the positron-annihilation method.[4] It gives the integral momentum distributions of the electrons, but not their energies, and in addition, it has a unique selectivity: for example, in ionic crystals, the positron annihilates only with an electron captured by a halide.

Absorption of ultraviolet makes it possible (when dealing with a solid) to determine essentially the energy intervals between the bands at points with high symmetry.[5] X-ray scattering,[6] the Mössbauer effect,[7] etc. also yield valuable information, but only concerning the density of the electrons and the form of its distribution, but not concerning directly the wave functions of the individual states.

Thus, it can be hoped that the method discussed in detail here, based on the quasielastic knock-out process (e, 2e) will come into its own among the other methods of investigation of electronic states.

2. GENERAL DESCRIPTION OF THE METHOD

A beam of fast monochromatic electrons is incident either on an exceedingly thin (60–100 Å) free-standing film, or on a beam of atoms or molecules, and two counters connected for coincidence register the two final electrons—the knock-out electron and the scattered one (Fig. 1). It is simplest to use a symmetrical version of the experiment. In this case the pulses of the initial and final particles p_0, p_1, and p_2 are complanar, the angles θ_1 and θ_2 are equal, i.e., close to 45° each (the kinematics is close to free scattering), and the energies of the final electrons are the same, $E_1 = E_2 \approx E_0/2$.

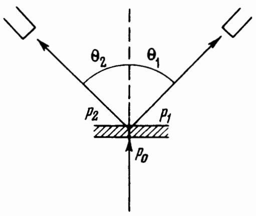

FIG. 1

The dependence of the cross section of the process (e, 2e), say on an atom on the total energy $E_1 + E_2$ will reveal several maxima, each of which has a certain finite width ("attenuation") and corresponds to the knock-out of electrons from a definite shell of the atom. The binding energy of the knock-out electron is $\varepsilon = E_0 - E_1 - E_2$. If we now measure, at a value $E_1 + E_2$ corresponding to a definite maximum (hole level), the angular correlation curve $d\sigma(\theta_1, \theta_2)$, varying the angles θ_1 and $\theta_2 = \theta_1$, then the curve will have the form of the Fourier spectrum of the wave function of the knock-out electron.

The most interesting are valence electrons with binding energies ~10 eV. A theoretical interpretation of the data is particularly simple and valuable, when there is practically no multiple scattering, and the process of quasielastic knock-out itself is well described in the impulse approximation (pole diagram). Therefore it is necessary to have very thin targets and high initial electron energies (in the investigation of light elements one can use $E_0 \sim 10$ keV; with increasing Z, the energy E_0 should increase to prevent excessive distortion of the waves of the initial and final particles).

We present a number of general formulas necessary for the subsequent discussion. Assume that we knock out an electron from an atom with a wave function $\Psi_i(\mathbf{r}_1\mathbf{r}_2\ldots\mathbf{r}_Z)$, so that as a result we are left with an ion with wave function $\Psi_f(\mathbf{r}_1\mathbf{r}_2\ldots\mathbf{r}_{Z-1})$. The expression for the cross section for quasielastic knock-out is[2]

$$\frac{d\sigma}{d\Omega_1 d\Omega_2 dE_1} = \frac{mp_1}{\hbar^3}|F_{if}(\mathbf{q})|^2\left(\frac{d\sigma}{d\Omega_1}\right)_{\text{lab}}, \tag{1}$$

where

$$F_{if}(\mathbf{q}) = \sqrt{\frac{Z}{(2\pi)^3}}\int d\mathbf{r}_Z \exp(-i\mathbf{q}\mathbf{r}_Z)\int \Psi_i(\mathbf{r}_1\ldots\mathbf{r}_Z)$$
$$\times\Psi_f^*(\mathbf{r}_1\ldots\mathbf{r}_{Z-1})\,d\mathbf{r}_1\ldots d\mathbf{r}_{Z-1},$$
$$\hbar\mathbf{q} + \mathbf{p}_0 = \mathbf{p}_1 + \mathbf{p}_2, \tag{2}$$

and the cross section for free electron–electron scattering through an angle $\theta_1 = 45°$ in the laboratory system is

$$\left(\frac{d\sigma}{d\Omega_1}\right)_{\text{lab}} = \frac{4e^2m^2p_1}{p_0}\left(\frac{1}{t'^4} + \frac{1}{t^4} - \frac{1}{t'^2t^2}\right) \approx 7.4E_0^{-2}\cdot 10^{-14}\ [\text{cm}^2/\text{sr}],$$

where E_0 is in electron volts, $2\mathbf{t} = \mathbf{p}_0 + \mathbf{p}_1 - \mathbf{p}_2 - \hbar\mathbf{q}$, and $2\mathbf{t}' = \mathbf{p}_0 - \mathbf{p}_1 + \mathbf{p}_2 - \hbar\mathbf{q}$.

In the self-consistent field approximation, the multielectron wave functions Ψ_i and Ψ_f are written in the form of a Slater determinant made up of the single-electron functions of the occupied states $\varphi_m(\mathbf{r})$. If the electron is knocked out from the orbit $\varphi_l(\mathbf{r})$, then the form factor (2) is given by

$$F_{if}(\mathbf{q}) = \frac{1}{(2\pi)^{3/2}}\int \varphi_l(\mathbf{r})e^{-i\mathbf{q}\mathbf{r}}\,d\mathbf{r} \equiv \varphi_l(\mathbf{q}), \tag{3}$$

i.e., it yields directly the Fourier transform $\varphi_l(\mathbf{q})$ of the wave function of the electron on a certain orbit (inasmuch as $\theta_1 = \theta_2$ and $|\mathbf{p}_1| = |\mathbf{p}_2|$, we get $\mathbf{q} \parallel \mathbf{p}_0$). In order to investigate the momentum distribution in other directions, it is necessary either to rotate the target relative to the beam, or to forego symmetry and complanarity, which, incidentally, does not raise any special problems.[8]

We have altogether ten quantities in the final state: $\mathbf{q}$, $\mathbf{p}_1$, $\mathbf{p}_2$, and the hole excitation energy ε^*. Six of them are independent, if we take into account the conservation laws. We choose these quantities to be Ω_1, Ω_2, E_1, and E_2, which should be measured experimentally. But one of the energies serves only for the determination of ε^*, so that we are left in (1) with a five-dimensional potential. The process (e, 2e) was discussed many times[9, 10] (in particular within the framework of the impulse approximation[9]) in papers devoted to the calculation of the ionization cross section of ions and molecules, when only one emitted electron is registered, and when the integration is over the angles of emission and over the energies of the second electron. However, such an averaging leads in practice to a loss of information concerning the structure of the wave function of the knocked-out particle.

In the general case, the wave functions Ψ_i and Ψ_f are not a simple Slater determinant, and it is necessary to use the fractional-parentage expansion of the function Ψ_i:

$$\Psi_i(\mathbf{r}_1\ldots\mathbf{r}_Z) = \sum_{f,l}\langle i|f,l\rangle\Psi_f(\mathbf{r}_1\mathbf{r}_2\ldots\mathbf{r}_{Z-1})\varphi_l(\mathbf{r}_Z). \tag{4}$$

Here $\langle i|f, l\rangle$—fractional parentage coefficient, which characterizes the parentage relation between the electron on the orbit φ_l with different states Ψ_f of the remainder.

Substitution of (4) in (2) yields

$$F_{if}(\mathbf{q}) = \sqrt{Z}\sum_l\langle i|f,l\rangle\varphi_l(\mathbf{q}). \tag{5}$$

Thus, the measurement of the cross sections (1) for transitions to different final states Ψ_f makes it possible to obtain the quantities $\langle i|f, l\rangle$, i.e., to investigate the structure of the many-particle wave function Ψ_i (the character of the fractional-parentage relation in it, the mixing of the states). We note that for molecular orbitals $Z\Sigma_l\langle i|f, l\rangle^2 = 2$ for σ orbits and $Z\Sigma_l\langle i|f, l\rangle^2 = 4$ for π orbits.

We now consider the process (e, 2e) on a thin single crystalline film. If we use for the description of the electronic states in the crystal the single-electron functions in the form of Bloch sums (the strong-coupling approximation)

$$\Psi_{l,\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{N}}\sum_{\mathbf{R}} e^{i\mathbf{k}\mathbf{R}}\varphi_l(\mathbf{r}-\mathbf{R}), \tag{6}$$

(where $\mathbf{k}$ is the quasimomentum of the electron in a certain electron band l, $\varphi_l(\mathbf{r})$ is the atomic function of the electron, N is the number of atoms in the crystal, and $\mathbf{R}$ are the coordinates of the crystal-lattice sites), then the form factor (2) becomes

$$F_{l,k}(\mathbf{q}) = \frac{1}{\sqrt{N(2\pi)^3}}\varphi_l(\mathbf{q})\sum_{\mathbf{R}} e^{i(\mathbf{k}-\mathbf{q})\mathbf{R}} = \frac{\sqrt{N}}{(2\pi)^{3/2}}\varphi_l(\mathbf{q})\,\delta_{\mathbf{k}+\mathbf{B},\,\mathbf{q}}. \tag{7}$$

Here $\mathbf{B}$ is the reciprocal-lattice vector. Substitution of this result in (1) yields the following result for the cross section of the (e, 2e) process:

$$\frac{d\sigma}{d\Omega_1\,d\Omega_2\,dE_1} = \frac{2mp_1}{\hbar^3}\,|\varphi_l(\mathbf{q})|^2 n\left(\frac{d\sigma}{d\Omega_1}\right)_{\text{lab}}\left[1 - \frac{\mathbf{k}_2\nabla_{\mathbf{k}}\varepsilon_l(\mathbf{k})}{2E_2}\right]^{-1}\delta_{\mathbf{q},\,\mathbf{k}+\mathbf{B}}, \tag{8}$$

where 2n is the number of valence electrons per unit volume of the crystal; the dependence of the electron binding energy $\varepsilon_l(\mathbf{k})$ on $\mathbf{k}$ determines the dispersion law for the band l; $\hbar\mathbf{k}_2 = \mathbf{p}_2$. In order for the cross section (8) to be different from zero, it is necessary to satisfy simultaneously the following conditions:

$$\mathbf{p}_1 + \mathbf{p}_2 = \mathbf{p}_0 + \hbar\mathbf{q}, \tag{9a}$$
$$E_1 + E_2 + \varepsilon_l(\mathbf{k}) = E_0, \tag{9b}$$
$$\mathbf{q} = \mathbf{k} + \mathbf{B}. \tag{9c}$$

Thus, if we fix the energies E_1 and E_2, then we register only electrons that are located at that point $\mathbf{k}$ of the first Brillouin zone, for which the condition (9b) is satisfied. If we now measure, at the same value of $E_1 + E_2$, the function of the angular correlation of the electrons, then the coincidences, unlike the case of the process (e, 2e) on atoms, will be registered not in a certain continuous region of angles θ_1 near 45°, but at discrete values, determined by relation (9c). Usually the form factor $\varphi_l(\mathbf{q})$ is sufficiently large only when $\mathbf{q} = \mathbf{k}$. Therefore, by establishing the angle θ_1 at which the coincidences are observed, we determine the magnitude of the quasimomentum **k** of the knocked-out electron and its binding energy $\varepsilon(\mathbf{k})$:

$$\cos\theta_1 = (p_0 - \hbar q) \,/\, 2[m(E_0 - \varepsilon(\mathbf{k}))]^{1/2}. \tag{10}$$

If we measure the cross section (8) at different values of $E_1 + E_2$, i.e., we move over a populated band, then we can obtain a representation of the form factor $\varphi_l(\mathbf{q})$, since the last factor in (8) changes little when **k** is varied. In the opposite case of a weak coupling of the electrons with the crystal lattice (model of almost free electrons), we have

$$\Psi_{\mathbf{k}} = \frac{1}{\sqrt{NV}} e^{i\mathbf{kr}}, \quad F_{\mathbf{k}}(\mathbf{q}) = \sqrt{\frac{(2\pi)^3}{NV}}\, \delta(\mathbf{k} - \mathbf{q}),$$

$$\varepsilon(\mathbf{k}) = \varepsilon(0) - (\hbar \mathbf{k})^2/2m_{\text{eff}}, \tag{11}$$

where V is the volume of the unit cell of the crystal and m_{eff} is the effective mass of the electron. The cross section (1) then becomes

$$\frac{d\sigma}{d\Omega_1 d\Omega_2 dE_1} = \frac{2mp_2}{(2\pi\hbar)^3} \left(\frac{d\sigma}{d\Omega_1}\right)_{\text{lab}} nV \delta_{\mathbf{k},\mathbf{q}}, \tag{12}$$

inasmuch as the quantity $\nabla_{\mathbf{k}}\varepsilon(\mathbf{k}) \cdot \mathbf{k}_2/2E_2 = \hbar^2(\mathbf{k} \cdot \mathbf{k}_2)/2E_2 m_{\text{eff}}$ is usually much smaller than unity. Again, the conditions (9a) and (9b) should be satisfied. From (8) and (11) we see that the form factor of the process (e, 2e) (i.e., the form of the electron angular-correlation curve) depends strongly on the type of coupling between the electron and the crystal lattice.

3. CERTAIN CONCRETE EXAMPLES

For the simplest targets, hydrogen atoms and molecules, we obtain from (1)–(3) respectively

$$\frac{d\sigma}{d\Omega_1\, d\Omega_2\, dE_1} \approx \frac{4.2 \cdot 10^{-16}}{E_0^{3/2}(1 + q^2 a_0^2)^4} \left[\frac{\text{cm}^2}{\text{eV-sr}}\right], \tag{13}$$

$$\frac{d\sigma}{d\Omega_1\, d\Omega_2\, dE_1} \approx \frac{5 \cdot 10^{-16}}{E_0^{3/2}(Z^2_{\text{eff}} + q^2 a_0^2)^4} \left(1 + \frac{\sin qR}{qR}\right) \left[\frac{\text{cm}^2}{\text{eV-sr}}\right], \tag{14}$$

where the energy E_0 is in electron volts, $Z_{\text{eff}} = 1$ or 2 (Heitler–London wave function), and R is the distance between the atoms.

Thus, for the atomic s-state and for the orbit σ_g at the base of this state, the maximum of the cross section will occur at zero momentum transfer q = 0. For atomic states with $l > 0$, the cross section vanishes where q = 0. Because of this difference, the form factors are very sensitive to hybridization of the orbits. By way of illustration, Fig. 2 shows the result of the calculation for the N_2 molecule at $E_0 = 10$ keV for two states: $3\sigma_g(\varepsilon = 15.6 \text{ eV})$—curve 1—and $2\sigma_u(\varepsilon = 18.7 \text{ eV})$—curve 3—with wave functions taken from [11]. Comparison of curves 1 and 2 shows clearly the strong effect of the transition from the orbital $3\sigma_g(2z)$, which corresponds to curve 2, to the hybridized "real" wave function (curve 1)

$$|3\sigma_g\rangle = 0.85|3\sigma_g(2z)\rangle - 0.56|3\sigma_g(2s)\rangle.$$

Such a sharp difference between curves 1 and 2 is connected with the fact that when q = 0, for the orbital $3\sigma_g(2z)$, both the atomic form factor $F_{2p}(\mathbf{q})$ and the interference factor

$$(qR - \sin qR) \,/\, qR \tag{15}$$

vanish. Therefore even a small admixture of the s-component of the orbitals σ_g causes an appreciable change of the form factor in the region of small q. For the orbit $2\sigma_u$ (curve 3), the hybridization has little effect, owing to the factor (15) of the function $|2\sigma_u(2s)\rangle$.

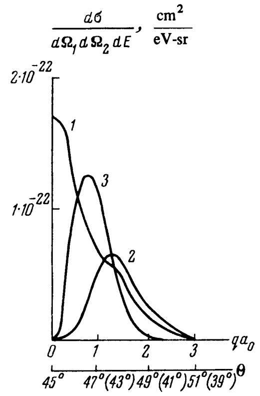

FIG. 2. Curves of angular correlation for the molecule N_2 at $E_0 = 10$ keV. For explanations see the text.

Proceeding to the consideration of solids, let us examine several different types of electronic wave functions.

For aluminum, the orthogonalization additions to the plane wave in the OPW method[12] make a contribution of only several per cent, so that in practice the form factor F(**q**) is given by formula (11). The width Γ of the hole levels, as shown by estimates given in [13], does not exceed a fraction of an electron volt for real metals even at the bottom of the band. On moving from the point Γ to the point X, the form factor will have at all times the form of a narrow peak of constant height, and the quantity $|F(\mathbf{q})|^2$ start to decrease only in the direct vicinity of the point X, owing to the mixing of the states $|\mathbf{k}\rangle$ and $|-\mathbf{k}\rangle$ under the influence of the pseudopotential, and at the point X the value of $|F(\mathbf{q} = 2\pi a^{-1} \times [1, 0, 0])|^2$ will be half as large as inside the band (in analogy with curve 2 of Fig. 3).

In lattices of the diamond type at the point X, for example, there is a merging of two electron bands, and the states $(\sqrt{2})^{-1}[|\mathbf{k}\rangle \pm |-\mathbf{k}\rangle]$ of the almost-free-electron approximation[14] are degenerate. Therefore the form factor no longer changes in magnitude on moving from the point Γ to X. In copper, the form factor is greatly influenced by the hybridization of the s-states (conduction band) and the d-states. Curve 3 of Fig. 3, calculated using the model functions of Mueller and Phillips,[15] characterizes the change of the form factor F(**q**) when moving along the lower strip at the bottom of the band from the point Γ to the point (L = [1, 1, 1]). This curve differs essentially from both the result obtained for the almost-free electrons (curve 2) and from the form factor for the Bloch function of the d-electrons (curve 1).

Finally, for the ionic crystal KCl, the wave functions have the form of Bloch sums (6), so that the form factor takes on the form (7). We have performed the calculation for the upper 3p-band of Cl^-.[16] The results are shown in Fig. 4 for two points Γ and X (at the point X—for the lower nondegenerate state). We see that if we move, shifting the values of the registered energy $E_1 + E_2$, on a strip from Γ to X, then the counting rate at the angles $\theta_1 = \theta_2$ corresponding to the discrete val-

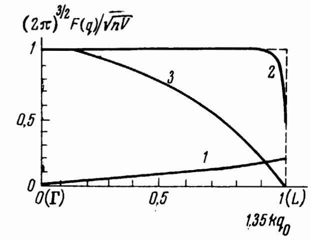

FIG. 3

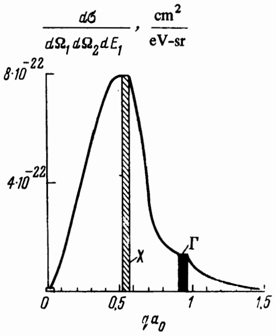

FIG. 4

FIG. 3. Form factor for copper. For explanations see the text.

FIG. 4. For factor for KCl. The point $\Gamma - qa_0 = 0$ and 1.06; the point $X - qa_0 = 0.53$.

ues of $q = k + B$ will change in accordance with the atomic form factor (7), thus duplicating this form factor as a whole.

The related problem of quasielastic knock-out of atoms or ions from molecules by neutrons, protons, etc. is also of interest.[17] Unfortunately, a further discussion of this question by Sayasov and Ivanov[18] contains serious errors—there is no plane wave corresponding to the momentum of the recoil relative to the mass center of the remainder radical, the employed Fourier amplitudes do not refer to the required degrees of freedom, etc.

The cross section of free ee-scattering through an angle $\theta_1 = \theta_2 = 45°$ amounts to 7.4×10^{-22} cm²/sr at $E_0 = 10$ keV, making it possible to use extremely thin free-standing targets 60–100 Å thick, which leave more than 50% of the passing electrons of the primary beam (at $E_0 \geq 10$ keV) unscattered.[19] Apparently the accuracy limits imposed by the finite thickness of such targets is ~0.2 eV in energy and ~0.05° in angle.

For a beam of H_2 molecules, equivalent in mass to an aluminum film 60 Å thick, at an angular resolution of each counter is $\Delta\Omega = 0.2° \times 0.2°$, and at an energy resolution $\Delta E_1 = 1$ eV for $E_0 = 10$ eV the counting rate of the coincidence at the maximum of the form factor ($q = 0$) will amount to $N = 600$ coincidences/sec per mA of current in the beam. For nitrogen molecules under the same conditions there will be $N = 17$ coincidences/sec. For aluminum at $E_0 = 10$ keV, we obtain $d\sigma/d\Omega_1 d\Omega_2 dE_1 = 3.7 \times 10^{-22}$ cm²/eV-sr² per atom, i.e., $N = 35$ coincidences/sec for the aluminum film itself. The danger of damaging the film can be avoided by shifting the target under the beam. On the whole, the experiments are perfectly realistic, although difficult.

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