

INTERACTION BETWEEN ATOMIC SYSTEMS IN CONDENSED MEDIA

D. K. KAIPOV, Yu. G. KOSYAK, and Yu. K. SHUBNYI

Nuclear Physics Institute, Kazakh Academy of Sciences

Submitted June 7, 1967

Zh. Eksp. Teor. Fiz. 54, 19-24 (January, 1968)

Nuclear  $\gamma$ -quantum resonance scattering is used to determine the effective interaction range between recoil atoms and molecules of a condensed medium as a function of the charge of the recoil nucleus. The measurements are carried out at energies from 10 to 200 eV. The dependence obtained is compared with calculations based on Bohr, Firsov, and Thomas-Fermi-Dirac potentials. The Firsov potential yields the best agreement.

INTRODUCTION

INFORMATION on interatomic and intermolecular interactions in gaseous media can be obtained by different means, for example, by studying the kinetic properties of gases, by investigating the thermodynamic properties of noble gases in the crystalline states, etc.<sup>[1]</sup> A direct source of information on the forces acting between the atoms and molecules of a gas is also the study of elastic scattering of atoms by the molecular-beam method<sup>[2]</sup>. At the same time, there are only few experimental data on the interaction of atomic systems in condensed media. Yet knowledge of the character of the interaction of atoms and molecules in condensed media is essential for an explanation, say, of the dynamics of radiation damage in solids, the behavior of hot atoms in different media, and certain problems of molecular physics. In this connection, the application of the physical methods (particularly methods of nuclear physics), which yield additional information on the parameters of interaction of atomic systems in condensed media, is of timely interest.

In this paper we consider the possibility of using for this purpose the method of resonance scattering of  $\gamma$  quanta by nuclei by means of radioactive isotopes in condensed substances.

1. MICROSPECTRUM OF  $\gamma$  RAYS WITH ALLOWANCE FOR THE DECELERATION OF THE RECOIL ATOMS

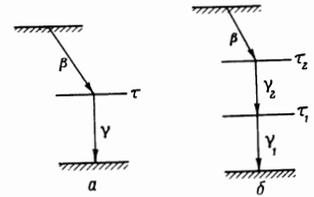
Recent investigations<sup>[3,4]</sup> have shown that the effect of resonance scattering of  $\gamma$  quanta by nuclei takes place also when  $\gamma$ -radiation sources are used in the liquid and solid states. The magnitude of the effect greatly decreases in this case compared with a gaseous source. This is the result of the considerable change in the energy distribution of the emitted  $\gamma$  quanta, owing to the deceleration of the recoil atoms.

Indeed, the expression for the average resonance-scattering cross section is<sup>1)</sup>:

$$\sigma_{av} = \frac{g_2 \lambda^2}{g_1 4} \Gamma_\gamma P(E_\gamma), \tag{1}$$

where  $g_1, g_2$  - statistical factors of the excited and ground states of the nucleus,  $\lambda$  - wavelength of the  $\gamma$  quantum in the resonance region,  $\Gamma_\gamma$  - natural level

FIG. 1.  $\beta - \gamma$  and  $\beta - \gamma_2 - \gamma_1$  cascades



width,  $P(E_R)$  - fraction of  $\gamma$  quanta in the energy distribution (microspectrum) of the radiation in a 1 eV interval with resonant energy  $E_R$ .

The quantity  $P(E_R)$  depends in the general case on the decay scheme of the isotope, on the lifetime of the excited states of the nucleus, on the aggregate state of the source, and on the character of the interaction between the recoil atoms and the surrounding atoms and molecules. If  $\Gamma_\gamma$  is known, say from experiments with self absorption, then  $P_{exp}(E_R)$  can be determined by measuring the cross section for the resonant scattering of the  $\gamma$  rays by the nuclei. On the other hand,  $P(E_R)$  can be calculated theoretically<sup>[5-7]</sup>. Besides the known parameters, it depends on the mean free path  $l$  of the recoil atoms in the substance. Comparison of  $P_{exp}(E_R)$  with  $P_{theor}(E_R)$  makes it possible to determine  $l$ .

The energy spectrum  $P(E)$  with allowance for the deceleration of the recoil nuclei was calculated as follows. If the  $x$  axis is chosen in the direction of the emission of the observed  $\gamma$  quantum, then

$$P(E) = a^{-1} \zeta(p_x), \tag{2}$$

where  $\zeta(p_x)$  is the distribution of the recoil nuclei with respect to the momentum projections,  $a = E_0/Mc$ ,  $E_0$  is the energy of the excited state of the nucleus,  $M$  the mass of the nucleus, and  $c$  the speed of light.

Assume that the  $\gamma$ -quantum emission is preceded by  $\beta$  decay (Fig. 1a). The momentum distribution density of the recoil nuclei following the  $\beta$  decay will be denoted  $g(p)$ . If we assume that the deceleration in the condensed medium is the result of elastic collisions, then the time variation of the momentum can be described by the differential equation<sup>[4]</sup>

$$\frac{dp}{p^2 dt} = -\frac{f}{Ml} = -\frac{1}{LM}, \tag{3}$$

where  $L = l/f$  is the characteristic range and  $f$  the relative loss of velocity per collision:

$$f = \left| 1 - \frac{2(1-k^3)}{3(1-k^2)} \right|, \quad k = \frac{M-M_2}{M+M_2}.$$

Solution of (3) yields

$$p' = \frac{MLp}{ML + pt},$$

where  $p'$  is the momentum of the recoil nucleus with allowance for deceleration. It is obvious that

$$g(p', t) = g\left(\frac{MLp'}{ML - p't}\right) \frac{M^2L^2}{(ML - p't)^2}.$$

Consequently the density of the distribution of nuclei with momentum  $p'$ , decaying by the instant of emission of the  $\gamma$  quantum, will be

$$g(p') = \frac{M^2L^2}{\tau} \int_0^{\infty} g\left(\frac{MLp'}{ML - p't}\right) \frac{e^{-t/\tau}}{(ML - p't)^2} dt,$$

where  $\tau$  is the lifetime of the excited level. We go over from this momentum distribution of the recoil nuclei to the distribution with respect to the momentum projections, which is of interest to us. We have

$$\zeta(p_x') = \frac{M^2L^2}{2\tau} \int_{|p_x'|}^{p_x \max} \int_0^{\infty} g\left(\frac{MLp'}{ML - p't}\right) \frac{e^{-t/\tau}}{(ML - p't)^2} dt,$$

where  $p_\beta^{\max}$  is the maximum momentum of the recoil nucleus after the  $\beta$  decay.

In the case of the  $\beta - \gamma_2 - \gamma_1$  cascade (Fig. 1b), with allowance for the deceleration of the recoil nuclei during the lifetime of the excited states, we obtain in similar fashion

$$\zeta(p_{x_2}'') = \frac{M^5L^5}{4p_{\gamma_1}\tau_1\tau_2} \int_{|p_{x_2}''|}^{p_2 \max} dp_2'' \int_0^{\infty} \frac{e^{-t/\tau_1} dt}{(ML - p_2''t)^3} \times \int_{|p_{x_2}' - p_{\gamma_1}|}^{p_{\gamma_1} \max} \frac{dp_{\gamma_1}'}{p_{\gamma_1}'} \int_0^{\infty} \frac{e^{-t'\tau_2} g\left(\frac{MLp'}{ML - p't}\right) dt'}{(ML - p't)^2};$$

here

$$p_2^{\max} = p_{\gamma_2} + p_\beta^{\max}, \quad p_2'' = \frac{MLp_2'}{ML - p_2't},$$

$\tau_1$  and  $\tau_2$  are the lifetimes of the first and second excited states. Going over to finite integration limits, we obtain convenient computation formulas<sup>[5-7]</sup> for the cascades  $\beta - \gamma$ ,  $\beta - \gamma_2 - \gamma_1$ , and  $k - \gamma_2 - \gamma_1$ .

## 2. REDUCTION OF EXPERIMENTAL DATA AND DISCUSSION OF RESULTS

The table lists the isotopes whose salts, in the form of aqueous solutions, were used as  $\gamma$  sources for the investigation of the resonant scattering of  $\gamma$  quanta by nuclei<sup>[3,4,8,9]</sup>. The lifetimes of the first excited states were determined by the "self-absorption" method. For each of these isotopes we calculated with the "Minsk-1" computer the quantity  $K = P(E_R)_{\text{gas}} / P \times (E_R)_{\text{liquid}}$  - the attenuation of the resonance effect for the liquid source compared with the gaseous source at the different values of  $L$ . In each concrete case  $L$  can be determined by comparing the experimentally obtained value of  $K_{\text{exp}}$  with the theoretical  $K(L)$  curves. Figure 2 shows by way of an example curves of this type for the  $V^{52} \rightarrow Cr^{52}$  decay.

In the case of weak solutions, knowing the mean distance between the molecules and the mean free path, we can determine the total effective radius of the in-

Radioactive isotope and decay product	Resonance level (MeV)	Limits of effective recoil energy in eV		Preceding ( $\beta$ and $\gamma$ ) emissions
		$E_r$	$E_{\text{max}}$	
$Na^{24} \rightarrow Mg^{24}$	1.38	84.4	281	$\beta$ (1.39) $\gamma$ (2.75)
$V^{52} \rightarrow Cr^{52}$	1.43	42	93.5	$\beta$ (2.73)
	0.663	7.5	23.7	$\beta$ (1.67)
$Zn^{68} \rightarrow Cu^{68}$	0.963	15.6	29.6	$\beta$ (1.38)
$Ni^{65} \rightarrow Cu^{65}$	1.114	20.5	26.3	$\beta$ (1.00)
	1.29	15.5	28	$\beta$ (1.00) $\gamma$ (1.10)
$In^{116} \rightarrow Sn^{116}$	1.29	15.5	21	$\beta$ (0.34) $\gamma$ (1.77)
	1.29	15.5	18	$\beta$ (0.60) $\gamma$ (1.51)
$La^{140} \rightarrow Ce^{140}$	1.60	19.5	32.4	$\beta$ (2.20)

teraction  $R_{\text{eff}}$ . The obtained values of  $R_{\text{eff}}$  as functions of the charge  $Z$  of the investigated elements are shown in Fig. 3, where results by other authors<sup>[4,8,9]</sup> were processed together with our experimental data.

This was followed by comparison of the experimental dependence of  $R_{\text{eff}}$  on  $Z$  with the theoretical calculations based on the Bohr, Firsov, and Thomas-Fermi-Dirac (TFD) interaction potentials. The potential curves were calculated for different values of  $R_{\text{eff}}$  and  $Z_2$  ( $Z_1 = \text{const}$ )<sup>2)</sup>. At internuclear distances smaller than 0.1 Å, a good approximation is a Coulomb repulsion potential acting between the nuclei with charges  $Z_1$  and  $Z_2$ , proposed by Bohr:

$$U(R) = (Z_1 Z_2 e^2 / R) e^{-R/\beta},$$

$$\beta = a_0 / (Z_1^{1/2} + Z_2^{1/2}),$$

$a_0$ —Bohr radius,  $e$ —electron charge. Since we have the region of large internuclear distances, obviously, this potential is not applicable in this case (Fig. 3, Bohr).

Another expression for the potential, based on the Thomas-Fermi (TF) model of the atom, was proposed by Firsov<sup>[10]</sup>:

$$U(R) = (Z_1 Z_2 e^2 / R) \chi(x),$$

$$x = (Z_1^{1/2} + Z_2^{1/2})^{2/3} R / a, \quad a = 0.8553 a_0, \quad R \leq 1.89 a_0,$$

$\chi(x)$  is a screening function (its values were taken from the tables of Gombas<sup>[11]</sup>). In this model we took into account only the electrostatic interaction between the nucleons. The exchange effects were excluded from consideration. The electron density then decreases very slowly with increasing radial distance from the

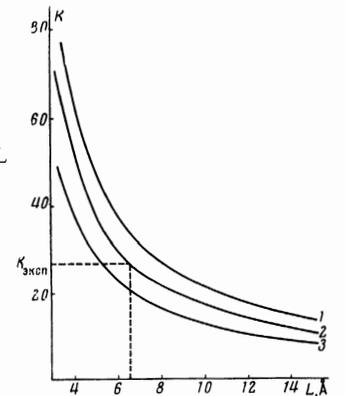


FIG. 2. Dependence of  $K$  on  $L$  for the  $V^{52} \rightarrow Cr^{52}$  decay at  $\tau = 10 \times 10^{-13}$  sec (1),  $8 \times 10^{-13}$  sec (2), and  $6 \times 10^{-13}$  sec (3).

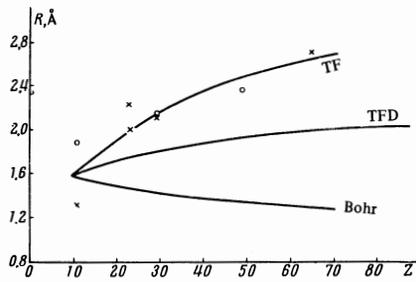


FIG. 3. Dependence of the effective interaction radius on  $Z$ : curves - theoretical, points - experimental:  $\circ$  - present work,  $\times$  - data by others.

atomic nucleus<sup>[11]</sup> (see Fig. 4, curve TF). Therefore the atom is unrealistically large, and the interaction energy between two such atoms decreases slowly with increasing internuclear distance.

The experimental values were compared also with the theoretical potential based on the statistical TFD model of the atom, used by A. Abrahamson for pairs of colliding noble-gas atoms<sup>[12]</sup>. In this case, in addition to the TF model, account is taken of exchange effects. We have

$$U(R) = \frac{1}{2} \frac{Z_1 Z_2 e^2}{R} \left[ \psi \left( \frac{Z_1^{1/3} R}{a} \right) + \psi \left( \frac{Z_2^{1/3} R}{a} \right) \right] + \Lambda,$$

where  $\psi$  is the TFD screening function,

$$\Lambda = \frac{1}{6} \int_{D_{12}} \left\{ 2.87 e^2 a_0 [(\rho_{01} + \rho_{02})^{1/3} - (\rho_{01}^{1/3} + \rho_{02}^{1/3})] - 1.476 e^2 [(\rho_{01} + \rho_{02})^{1/3} - (\rho_{01}^{1/3} + \rho_{02}^{1/3})] \right\} dv,$$

$\rho_{0i}(r_i)$  is the exact undistorted TFD electron distribution density, due to the  $i$ -th atom, as a function of the radial distance measured from the center of this atom ( $i = 1, 2$ ), and  $D_{12}$  is the region of overlap of the two electron shells. The electron distribution density  $\rho(r)$  (Fig. 4, curve TFD) agrees in this case better with the quantum-mechanical (QM) description of the atom. An unfavorable aspect of this model is the excessively abrupt cut-off of the electron density at the limiting value of the radius  $r_{lim}$ .

Using the values of  $U(R)$ , calculated by Abrahamson for pairs of elements with different charges  $Z_2$  and  $Z_1 = 10$  (neon) at different distances  $R$ , we calculated the dependence of  $R_{eff}$  on the charge of the recoil atoms (Fig. 3, curve TFD).

Our investigations allow us to assume that the Firsov potential is the best approximation in the case of interaction of recoil nuclei with surrounding molecules in a condensed medium.

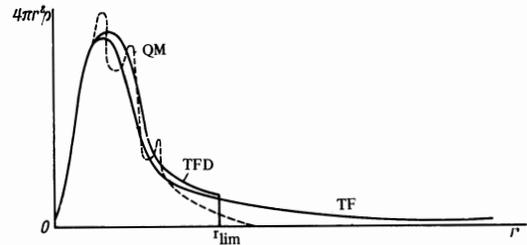


FIG. 4. Electron distribution density for the Thomas-Fermi, Thomas-Fermi-Dirac, and the quantum mechanical models of the atom.

In conclusion, the authors are grateful to O. B. Firsov and V. B. Leonas for a discussion of the work.

<sup>1</sup> J. Hirschfelder, C. Curtiss, and R. Bird, *Molecular Theory of Gases and Liquids*, Wiley, 1964.

<sup>2</sup> V. B. Leonas, *Usp. Fiz. Nauk* **82**, 287 (1964) [*Sov. Phys.-Usp.* **7**, 121 (1964)].

<sup>3</sup> D. K. Kaipov, Yu. K. Shubnyĭ, Yu. G. Kosyakov, and R. B. Begzhanov, *Zh. Eksp. Teor. Fiz.* **45**, 443 (1963) [*Sov. Phys.-JETP* **18**, 305 (1964)].

<sup>4</sup> J. B. Cumming, A. Schwarzschild, and A. W. Sunyar, *Phys. Rev.* **120**, 2128 (1960).

<sup>5</sup> D. K. Kaipov, Yu. K. Shubnyĭ, V. M. Amerbaev, A. Kzangalov, and Yu. G. Kosyakov, *Zh. Eksp. Teor. Fiz.* **48**, 1221 (1965) [*Sov. Phys.-JETP* **21**, 815 (1965)].

<sup>6</sup> D. K. Kaipov, Yu. G. Kosyakov, and Yu. K. Shubnyĭ, *Yad. Fiz.* **4**, 493 (1966) [*Sov. J. Nucl. Phys.* **4**, 351 (1967)].

<sup>7</sup> D. K. Kaipov, Yu. G. Kosyakov, and Yu. K. Shubnyĭ, *Tr. Instituta yadernoi fiziki AN KazSSR (Proc., Institute of Nuclear Physics AN KazSSR)*, **7**, 12 (1967).

<sup>8</sup> S. Ofer and A. Schwarzschild, *Phys. Rev. Lett.* **3**, 384 (1959).

<sup>9</sup> J. Kaus, *Z. Naturforsch.* **20a**, 391 (1965).

<sup>10</sup> O. B. Firsov, *Zh. Eksp. Teor. Fiz.* **33**, 696 (1957) [*Sov. Phys.-JETP* **6**, 534 (1958)].

<sup>11</sup> P. Gombas, *Statistical Theory of the Atom and Its Applications* (in German), Springer, 1949.

<sup>12</sup> A. A. Abrahamson, *Phys. Rev.* **133A**, 990 (1964).