SOVIET PHYSICS JETP VOLUME 21, NUMBER 1 JULY, 1965

STUDY OF THE SLOWING DOWN OF SLOW ATOMS IN GASES BY THE γ-RAY RESONANCE SCATTERING METHOD

A. F. AKKERMAN, V. L. KOCHETKOV and V. N. CHEKANOV

Nuclear Physics Institute, Academy of Sciences, Kazakh S.S.R.

Submitted to JETP editor May 12, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) **48**, 13-18 (January, 1965)

The cross section for resonance scattering of 325 keV γ rays by V^{51} nuclei has been studied as a function of the density of a gaseous CrO_2Cl_2 source. The experimental density dependence agrees satisfactorily with the theoretical prediction based on the elastic collision model. The collision diameter for a V^{51} atom and a CrO_2Cl_2 molecule is 6.8 Å. Comparison of the results of the present investigation with those of other resonance experiments indicates that at low recoil nucleus energies ($E_{\text{kin}} < 100$ eV) elastic collisions are the main factor in the slowing down.

THE scattering of slow atoms or molecules ($E_{\text{kin}} < 100$ eV) in gases involves little studied questions of molecular physics. The few data in this energy region have been obtained mainly by the molecular beam method.[1] The collision diameters of several atoms and simple molecules have been determined in this way.

In the present paper we examine the results of applying the γ-ray resonance scattering method to study of the slowing down of slow atoms in gases. This method is widely used in nuclear spectroscopy to determine lifetimes of excited nuclear states. The possibility of using this method for the problem under discussion is due to the dependence of the resonance scattering cross section on the energy loss by the recoil nucleus on interaction with the surroundings.[2]

The transition $Cr^{51} \rightarrow V^{51}$ was chosen as a suitable subject for study. Resonance scattering of 325-keV γ rays by V^{51} nuclei is possible since the absorption line is overlapped by the emission line, which has been broadened by the Doppler effect on emission of a 425 keV neutrino. Furthermore, since the lifetime of the $5/2^-$ (325 keV) level in V^{51} is rather large, $\tau = 3.0 \times 10^{-10}$ sec,[3] we can expect a noticeable influence of collisions on the size of the resonance effect in a low-density gaseous source. Performance of the experiment is also assisted by the high relative abundance of V^{51} in the natural isotopic mixture (99.8%) and the large half-life of the source ($T = 27.8$ days).

EXPERIMENTAL DETAILS

1. Chromyl chloride (CrO_2Cl_2), b.p. = 116° C, was used as a volatile compound of Cr. To obtain this compound we used a slight simplification of the method described by Delyagin and Preĭs.[3] After double distillation the CrO_2Cl_2 was collected in a 39.4 ml Pyrex ampoule cooled by dry ice. The ampoule was evacuated, sealed off, and rapidly placed in darkness to avoid decomposition of the chromyl chloride. The initial activity of the source was ~45 mCi. It sublimed completely at a temperature of 109° C. Control measurements showed that no appreciable decomposition of the source was observed in the entire course of the experiment (40 days).

2. The resonance effect measurements were carried out in the automatic equipment sketched in Fig. 1. The scatterers were powdered samples

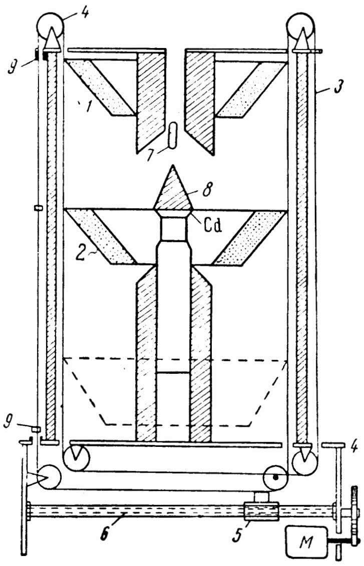

FIG. 1. Drawing of the experimental apparatus: 1,2 – scatterers, 3 – cable, 4 – pulleys, 5,6 – lead screw with nut, 7 – source, 8 – lead cone, 9 – limit switches, 10 – lead shield, M – motor with reduction gear.

of V_2O_5 or V (resonance) and Fe_2O_3 (nonresonance), pressed in thin-walled containers of conical shape so that the filling density was the same. The conical shape of the scatterer provided the optimum source-scatterer solid angle with the minimum quantity of scattering material. The mean scattering angle $\psi = 135°$. A 40 × 40 mm NaI(Tl) crystal and FÉU-11 photomultiplier were used as a detector.

Automatic changing of the scatterer was accomplished by means of a motor with a switched starting winding. The motor was turned on by a special control circuit working in conjunction with a type AZ-1 single-channel analyzer, and turned off by limit switches. The ampoule containing the source was placed in a container of duraluminum covered with a thin layer of foam plastic for thermal isolation. The container was fixed at a definite position in the shield and was connected to the external surface of an ultrathermostat. By means of the latter, a current of water-glycerine mixture was passed through the entire system, which assured maintenance of the source temperature with high accuracy. Through a 3 mm aperture in the shield (not shown in Fig. 1) it was possible, using an auxiliary detector consisting of a 20 × 30 mm NaI(Tl) crystal with an FÉU-13 photomultiplier connected to an AADO-1 single-channel analyzer, to follow the evaporation of the source, to take into account its decay, etc.

3. The resonance scattering cross section $\bar{\sigma}$ is related to the relative resonance effect N_{res}/N by the formula

$$\frac{N_{\text{res}}}{N} = n_0\bar{\sigma}\frac{\rho}{\rho_{max}}\frac{R^2}{r^2}\left[\exp\left(-\frac{0{,}693\Delta t}{T_{1/2}}\right)\right]^{-1}\frac{[\overline{\varepsilon s}]_r}{[\overline{\varepsilon s}]_R}\int\frac{N_1}{N_2}(\psi)W(\psi)$$
$$\times\frac{\exp\{-(\mu_1 s_1+\mu_2 s_2)\}}{4\pi r_1{}^2}\,dv. \qquad (1)$$

Here n_0 is the number of V^{51} nuclei per cm^3 in the scatterer; the exponential function in front of the integral takes into account the decay of the source during the measurement; $[\overline{\epsilon s}]_r/[\overline{\epsilon s}]_R$ is the ratio of γ-ray counting efficiencies of the crystal for the two source-to-crystal distances. The integral in (1) is evaluated by integration over the scatterer volume and takes into account the scatterer geometry, the finite size of the source, and the angular distribution $W(\psi)$ of the resonance-scattered γ rays; μ_1 and μ_2 are the linear attenuation coefficients in the scatterer for narrow and broad γ-ray beams; s_1 and s_2 are the distances traveled by the γ-ray in the scatterer before and after scattering; r_1 is the distance from the source to the point of scattering. The ratio ρ/ρ_{max} (ρ_{max} = 3.89 mg/ml) takes into account the fraction of the CrO_2Cl_2 in gaseous form and is found by means of an auxiliary detector. However, for $t < 80°$ we used values of ρ/ρ_{max} calculated from the vapor pressure of CrO_2Cl_2,[4] since at low temperatures, as the measurements showed, adsorption of the vapor by the walls of the ampoule occurs.

4. Table I lists values of resonance scattering cross sections for different source densities and for two scatterers, V_2O_5 and V.

Table I. Resonance scattering cross section (mb) for different source densities

ρ, mg/ml	Scatterer		ρ, mg/ml	Scatterer	
	V_2O_5	V		V_2O_5	V
0,057	11.4±2,1		0.50	5.6±0.7	6.8±1.0
0.129	9,1±1.4		0,83	3,4±0.4	3.3±0.5
0.23	8.2±1.2	9.2±1.4	1.21	2,7±0.3	
0,38	6,3±0,8	6.8±1.0	1,76	1,9±0.3	

The very existence of resonance scattering in metallic V (see also[5]) can be explained only by assuming that the chemical bonds in the CrO_2Cl_2 molecule are broken and the emission of the 325-keV γ ray occurs from a free V^{51} nucleus. In fact, on the one hand, when the entire CrO_2Cl_2 molecule undergoes recoil,[6] the emission line does not overlap the absorption line even if we take into account the thermal motion:

$$E_\nu E_0\,/\,M_1c^2 < E_0^2\,/\,2M_1c^2 + E_0^2\,/\,2M_2c^2$$

(M_1 and M_2 are the masses of the CrO_2Cl_2 molecule and V atom; E_0 = 325 keV, E_ν = 425 keV). On the other hand, among the complexes which could be formed from the excited atom V^* and the atoms of oxygen and chlorine in the gaseous phase, there are no stable products. Formation of complexes is also hindered by the large excess positive charge of the V atom, arising after K-capture and the subsequent changes in the electron shell.[7] Therefore it is natural to assume that complete rupture of the chemical bonds occurs in the CrO_2Cl_2 molecule.

We can see from Table I that the resonance scattering cross sections for the same source density but for different scatterers, V_2O_5 and V, agree within experimental error. This indicates that the positions of the absorption lines also coincide, i.e., the whole V_2O_5 molecule does not undergo recoil,[7] but only a quasifree V atom. The effect of chemical binding of the latter in the V_2O_5 and V lattices is manifested through the

Debye temperature in calculation of the resonance absorption in the scatterer. In our case the corrections due to chemical binding are negligible. The result that at high γ-transition energies (in comparison with Mössbauer energies) an atom entering into the composition of a molecule of the scatterer undergoes recoil as if it were free follows also from the elementary theory of the Mössbauer effect.[8]

THEORETICAL RESULTS

To obtain the theoretical resonance scattering cross section σ_{theor} as a function of source density ρ, it is necessary to assume a definite law for the slowing down of the recoil nuclei. We shall assume that the slowing down occurs by means of elastic collisions [1] with CrO_2Cl_2 molecules. Then, using the formulas given previously[10] and also assuming that the free V^{51*} atom undergoes recoil on emission of a neutrino, we obtain for the resonance scattering cross section the expression

$$\sigma(\rho, d), \text{mb} = \frac{33.7 M_2 c^2}{E_0 E_\nu} \sum_{n=0}^{\infty} \left(\frac{2}{k} \ln \frac{\lambda + p_{max}\tau/M_2}{\lambda + p_{min}\tau/M_2} \right)^n \frac{1}{n!} \frac{\lambda}{\lambda + p_{max}\tau/M_2}, \quad (2)$$

where c is the velocity of light, E_0 is the γ-ray energy, $k = 4M_1M_2/(M_1 + M_2)^2$ (M_1 and M_2 are the masses of the CrO_2Cl_2 molecule and the V^{51} atom), τ is the lifetime of the level, $\lambda = \mu/\sqrt{2}\pi d^2 \rho N_A$ is the path length (N_A is Avogadro's number, d is the collision diameter, μ is the molecular weight of CrO_2Cl_2), $p_{min} = E_0/c$, $p_{max} = E_\nu/c$.

For the densities used in the experiment it is sufficient to take into account five terms in the summation in Eq. (2).

Figure 2 shows the theoretical dependence $\sigma_{\text{theor}}(\rho)$ together with the experimental points. It can be seen that there is good agreement of experiment and theory. The collision diameter in this case turns out to be d = 6.6 Å.

The dependence of the resonance scattering cross section on source density has also been found from experiments on determination of the lifetime of the second level of certain nuclei.[11-13] The elastic collision model calculation also expresses the experimental dependence $\sigma_{\text{exp}}(\rho)$ satisfactorily in these cases. The effective collision diameters determined from resonance experiments are listed in Table II. For $Ge^{74*} + As_4$ we computed the collision diameter from the experimental data of Metzger.[14] As can be seen from the table, the collision diameters d_{exp} turn out to be close to the diameters calculated using the constant b of the Van der Waals equation.[15] Of course, it is impossible to neglect a priori the contribution of inelastic processes in collisions of molecules or atoms with molecules. These processes involve excitation of internal motions (vibrational and rotational) of the molecule.

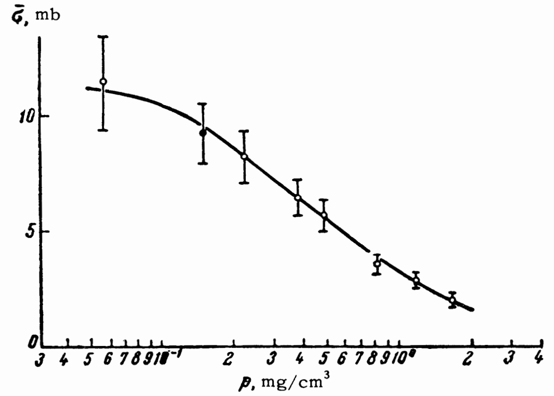

FIG. 2. Cross section for resonance effect as a function of source density. Solid curve – calculated from elastic collision model; points – experiment.

The probability of excitation of molecular rotational levels is very small, since the kinetic energy of the incident particle considerably exceeds the distance between these levels (≤ 0.05 eV).

The excitation of vibrational levels of a molecule in collisions was recently discussed by Ivanov and Sayasov.[16] [2] Application of their results to one of the cases considered by us—collision of $Ti^{48*} + TiCl_4$—shows that the cross section for excitation of vibrations gives a small (< 10%) contribution to the total collision cross section; the main contribution is due to elastic scattering. A similar conclusion can be drawn also for the collisions $Ge^* + As_4$ and $As^*H_2 + SeH_2$. The application of the formulas of Ivanov and Sayasov[16] to the collision $V + CrO_2Cl_2$ is not so obvious. However, the results of the present work allow us to assume that the role of inelastic processes is small in these cases. Actually, a noticeable contribution of inelastic processes would lead to a different angular distribution of the scattered recoil nuclei, which would directly change the observed behavior of $\sigma_{\text{exp}}(\rho)$.

Thus, from resonance experiments using dense

---

[1] We note that the elastic collision model has been used by Cummings et al[9] to explain the weakening of the resonance effect in condensed sources.

[2] We are grateful to F. L. Shapiro who directed our attention to this work.

Table II. Comparison of experimental (d_{exp}) and theoretical (d_{theor}) collision diameters

	d_{exp}, A	d_{theor}, A		d_{exp}, A	d_{theor}, A
$Ti^{48*} + TiCl_4$ [13]	3.2	3.9	$As^{75*}H_2 + SeH_2$ [11]	2.6	3.5
$V^{51*} + CrO_2Cl_2$	6.6	4.2	$Cd^{114*}Cl_3 + InCl_3$ [12]	2.4	4.4
$Ge^{74*} + As_4$ [14]	4.6	5.1			

gaseous sources it follows that the slowing down of recoil nuclei with energies $E_{kin} < 100$ eV occurs principally through elastic collisions.

We express our sincere gratitude to N. N. Delyagin for his very helpful discussions of our results.

---

[1] N. F. Ramsey, Molecular Beams, Oxford, Clarendon Press, 1956. (Russian translation, IIL, 1960).

[2] S. É. Frish, UFN 68, 3 (1959), Soviet Phys. Uspekhi **2**, 343 (1959).

[3] N. N. Delyagin and M. Preĭs, JETP **36**, 1586 (1959), Soviet Phys. JETP **9**, 1127 (1959).

[4] L. Gmelin, Handbuch der anorganischen Chemie, 8 Auflag, Chrom. teil B., 1962, p. 269.

[5] I. Y. Krause, Phys. Rev. **129**, 1330 (1963).

[6] H. Schopper, Zs. f. Phys **144**, 476 (1956).

[7] S. Wexler, Phys. Rev. **93**, 182 (1954).

[8] Mössbauer Effect, Collection of Translations edited by Yu. Kagan, IIL, 1962.

[9] Cummings, Schwarzschild, Sunyar, and Porile, Phys. Rev. **120**, 2128 (1960).

[10] Akkerman, Vil'koviskiĭ, and Chekanov, Izv. AN Kazakh SSR, ser. fiz. **2**, 19 (1963).

[11] H. Langevin-Joliot and M. Langevin, J. phys. rad. **19**, 765 (1958).

[12] Akkerman, Vil'koviskiĭ, Kaipov, and Chekanov, JETP **43**, 1268 (1962), Soviet Phys. JETP **16**, 899 (1963).

[13] Akkerman, Kochetkov, Chekanov, Suvorov, and Shtol'ts, JETP **45**, 1778 (1963), Soviet Phys. JETP **18**, 1218 (1964).

[14] F. R. Metzger, Phys. Rev. **103**, 983 (1956).

[15] M. P. Vukalovich and I. I. Novikov, Uravnenie sostoyaniya real'nykh gazov (The Equation of State of Real Gases), GÉI, 1948.

[16] G. K. Ivanov and Yu. S. Sayasov, DAN SSSR **154**, 1314 (1964), Soviet Phys. Doklady **9**, 171 (1964).

Translated by C. S. Robinson
3