THE ORIENTATION OF NUCLEI

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The possibility of additional nuclear transitions in electron nuclear double resonance (ENDOR) under the influence of the electric component of a radio frequency field is considered. A system of nuclei with a preferred spin orientation can be obtained by this method.

FEHER[1] has proposed a method of electron nuclear double resonance (ENDOR) which enables one, in particular, to obtain in a system of nuclei situated at local centers a preferred spin orientation. The corresponding quantum transitions were brought about by the action of the magnetic components of UHF and radio frequency (rf) fields. The selection rules for transitions in the nuclear subsystem are as follows: $\Delta m = \pm 1$, $\Delta M = 0$; and for the electron subsystem the selection rules are $\Delta m = 0$, $\Delta M = \pm 1$. Here m is the quantum number of the component of the nuclear spin along the direction of the magnetic field, M is the same for the electron spin.

We consider below quantum transitions in ENDOR under the action of the electric component of a radio-frequency field. To provide an example we restrict ourselves to a system of symmetry T_d with effective electron angular momentum $J = 3/2$ and arbitrary nuclear spin I. We shall take the z axis to be directed along a crystallographic axis of type [100].

An external static magnetic field ($H = H_z$) removes the degeneracy of the system and produces $4(2I + 1)$ levels. The energy levels of the system and the corresponding wave functions, taking into account the contact hyperfine interaction (which acts as a perturbation), were determined with an accuracy up to second order perturbation theory. The perturbing operator which gives rise to the quantum transitions has the form

$$\hat{V} = -\mu_n \mathbf{H} - \mu_e \mathbf{H} - \mathbf{dE}, \qquad (1)$$

where μ_n and μ_e are respectively the magnetic moments of the nucleus and of the electron, d is the operator for the dipole moment of the electron, H and E are the intensities of the magnetic and the electric fields.

Utilizing the perturbing matrix[2] and the wave functions of the stationary states we can obtain the matrix elements for the transitions in the system. Thus, for the transition $-1/2, m \rightarrow -1/2, m + 2$ the matrix element is equal to

$$M_1 = -i\alpha E_z \frac{A^2\sqrt{3}}{4g^2\mu_0^2H^2}[(I+m+2)(I-m-1) \times (I+m+1)(I-m)]^{1/2},$$

where g is the g-factor for the electron, μ_0 is the Bohr magneton. It is of interest to compare M_1 with the value of the matrix element M_2 for the transition $m \rightarrow m + 1$ under the action of the operator $-\mu_n \mathbf{H}$:

$$\frac{|M_1|}{|M_2|} = \frac{\sqrt{3}}{2}\sqrt{(I+m+2)(I-m-1)}\frac{E_z}{H_x}\frac{\alpha}{\mu_0}\frac{\mu_0}{\mu_n} \times \left(\frac{A}{g\mu_0H}\right)^2.$$

Taking $\alpha = \mu_0$, $\mu_0/\mu_n = 10^3$, $A/g\mu_0H = 10^{-2}$, we obtain $|M_1|^2/|M_2|^2 = 0.01(E_z/H_x)^2$. In favorable cases (large A, α), and also for sufficiently high power of the RF field E_z^2 this ratio will be greater, and, therefore, observation of such transitions appears feasible.[1)]

The matrix elements for the transitions $\Delta m = \pm 1$ under the influence of the components E_x and E_y will be proportional to $A/g\mu_0 H$[2] and, therefore, will be more intense. But the transitions $\Delta m = \pm 1$ can also take place under the influence of the magnetic component of the rf field.

If by means of electron spin transitions $\Delta M = \pm 1$, $\Delta m = 0$ one achieves a saturation of levels, and then inverts the population of the nuclear spin levels ($\Delta M = 0$, $\Delta m = \pm 1, \pm 2$), then the product P of the component of the nuclear spin and the corre-

1)The transitions $\Delta m = \pm 2$ can be brought about also as a result of other mechanisms: a change in the quadrupole interaction constant under the influence of an external electric field[3] or the presence of a nuclear dipole moment in a crystalline field.

sponding number of nuclei will be different from zero. At the same time

$$P_{\Delta m=\pm 1,\ \Delta M=\pm 1} < P_{\Delta m=\pm 2,\ \Delta M=\pm 1}.$$

In particular, for $g\mu_0 H/kT \ll 1$

$$P_{\Delta m=\pm 2,\ \Delta M=\pm 1} = 2P_{\Delta m=\pm 1,\ \Delta M=\pm 1}.$$

In the system under consideration transitions $\Delta M = \pm 2$ are possible under the influence of the z-component of the electric field. If these transitions are utilized to saturate the electron spin levels[2)] and then the transition $\Delta m = \pm 2$ is made to take place, it is possible to increase P.

In the approximation utilized above the following relations are valid:

$$P_{\Delta m=\pm 2,\ \Delta M=\pm 2} = 2P_{\Delta m=\pm 2,\ \Delta M=\pm 1} = 2P_{\Delta m=\pm 1,\ \Delta M=\pm 2} = 4P_{\Delta m=\pm 1,\ \Delta M=\pm 1}.$$

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[2)] The possibility of such saturation has been pointed out by Ludwig and Ham.[4]

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