# LEVEL MIXING RESONANCES ON ORIENTED NUCLEI*

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## Summary

In this paper a new effect, the resonant destruction of nuclear orientation through level mixing is described. The obtained signals are similar to those known in the radiofrequency nuclear magnetic resonance. The effect can be used for the precise determination of electric quadrupole interactions of nuclei in solids, i.e. to measure nuclear electric quadrupole moments or electric field gradients in solids.

## Introduction

The measurement of a physical quantity can be performed in two ways: either the amplitude of a signal depends more or less proportionally on the quantity to be determined, or a resonant signal informs on the correct set of a reference value. A well-known example is the determination of an inductance through direct current measurement through an LC resonance-bridge. As in the resonant methods the precise knowledge of the amplitude of the resonance-signal is not necessary, they are usually one to two orders of magnitudes more precise, than the analog ones.

In this article we present a new resonant method for the determination of electric quadrupole interactions between nuclei and solids. The method was elaborated by the collaboration of the universities of Leuven and Lyon; more detailed information on it can be found in [1–4] and references therein.

## Angular distribution of the electromagnetic radiation emitted by oriented nuclei

In the most common cases an excited nucleus looses its excitation energy by emitting an electromagnetic radiation.

Let us consider a simple nucleus, where one proton is outside a closed-shell core, e.g. $^{57}$Co (see Fig. 1a). The electric field of the nucleus can be

* Dedicated to Professor Károly Simonyi on the occasion of his Seventieth Birthday

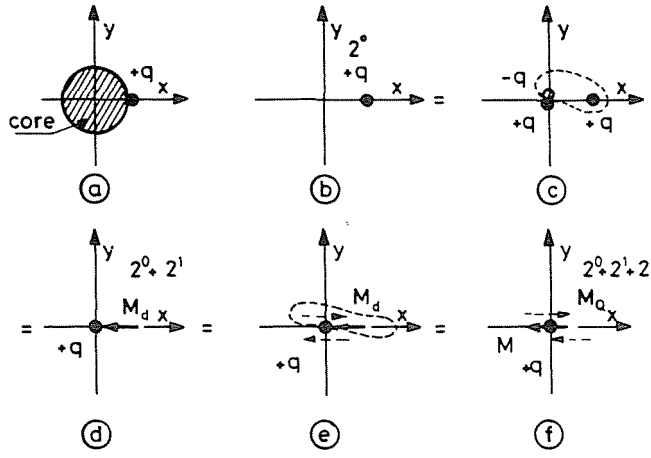


*Fig. 1.* Graphical representation of multipole order development of a charge distribution. The appearing multipoles are marked in figures b, d and g. When the origin of the axes is in the center of mass of the charges, the static dipole moment is zero

described as a pure, non-central Coulomb-field. Taking away the core, the $+q$ charge can be centred by adding a $(+q\ -q)$ pair of charges (c), which becomes equivalent to a centred charge plus a non-centred dipole (d), which can be centred also leaving a non-centred quadrupole (f), etc. When now the proton moves with respect to the core, the intensities of all the dipole-, quadrupole-, etc, moments vary, and dipole, quadrupole, and even higher order radiations are emitted. All these radiations have characteristic, non-isotropic angular distributions with respect to the nuclear spin-axis [5]. The multipole radiation field can be calculated from the variable electric current density and magnetic vector potential of the electric charges and magnetic moments of the nucleons composing the nucleus. The procedure and the results are very similar to the treatment of multiple antenna systems [6].

The electromagnetic power radiated through a surface of 1 $m^2$ by a moving electric charge $q$ subject to an acceleration $a$ (+) is:

$$S(t+R/c)=q^2\frac{\mu_0^2}{16\pi^2 c}\frac{a^2}{R^2}\bar{R}_0[1-\cos^2(\bar{R}_0\bar{a})] \tag{1}$$

Let us suppose that the nucleus decays from a state of spin $I=0$ to the fundamental state $I=0$ (Fig. 2a). The $m\pm1$ substates decay through the $(1, \mp1)$ mode (rotating dipole):

$$\bar{S}_{\text{eff}}=S_0\bar{R}_0\frac{1+\cos^2\Theta}{2},$$

where $\Theta$ is the angle between the rotation axis and the observation direction $R_0$, while the $m=0$ substate decays through the $(1, 0)$ mode (oscillating dipole,

the classical dipole-antenna):

$$\bar{S}_{\text{eff}} = S_0 \bar{R}_0 \sin^2 \Theta .$$

In an ensemble of unoriented nuclei every substate is equally populated, thus the power emission is isotropique. This fact is well known: usual radiactive sources emit in every direction with the same intensity.

An inequal substate-population of the state $I$ can be created by a preceeding nuclear transformation, by optical (laser) pumping or by static low temperature nuclear orientation. In this latter case the decoupling of the substates $m$ of the state $I$ can be due either to the magnetic interaction between the nuclear magnetic moment $\bar{\mu}$ and an extranuclear magnetic field $\bar{B}$, or by the electrostatic interaction between the nuclear electric quadrupole moment $Q$ and an extranuclear electric field gradient $V_{zz}$. The substate-energies are:

for magnetic interaction:

$$W_{\text{mag}}(m) = -m\mu B/I \tag{2}$$

for axially-symmetrical electric interaction

$$W_{\text{el}}(m) = \frac{eQV_{zz}}{4I(2I-1)} [3\,\text{m}^2 - I(I+1)] . \tag{3}$$

The population of the substates is described by a Boltzmann-distribution $\rho(m) \alpha \exp[-W(m)/kT]$. Unfortunately, the available interaction energies $W(m)$ are very small, of the order of $10^{-26} - 10^{-25}$ J, so, in order to obtain reasonable ($> 0.1$). Boltzmann-factors the sample to be studied has to be cooled down to $3 - 30$ mK. In Figure 2 we show for a simplified case the angular intensity distribution of the emitted radiation for each substate and the weighted average of this radiation at high and low temperature.

Usually the directly oriented $I_0$ state is not identical to the initial state $I$ of the observed radiation. This latter state is a daughter state of the oriented $I_0$ state, connected to through several other transitions, as $\beta$-rays, other $\gamma$-rays, etc... The emission of these unobserved rays depolarises partially the ensemble of nuclei; nevertheless, the nuclei conserve a certain degree of orientation. The substate-populations of the state $I$ can be calculated directly from those of the initial state $I_0$.

## Intermediate-state perturbation

Up till now we considered that the orientation of the state $I$, inherited from that of the state $I_0$ remains unchanged, i.e. the same at the moment of the creation of the state $I$ as at the moment of emission of the observed $\gamma$-ray. The

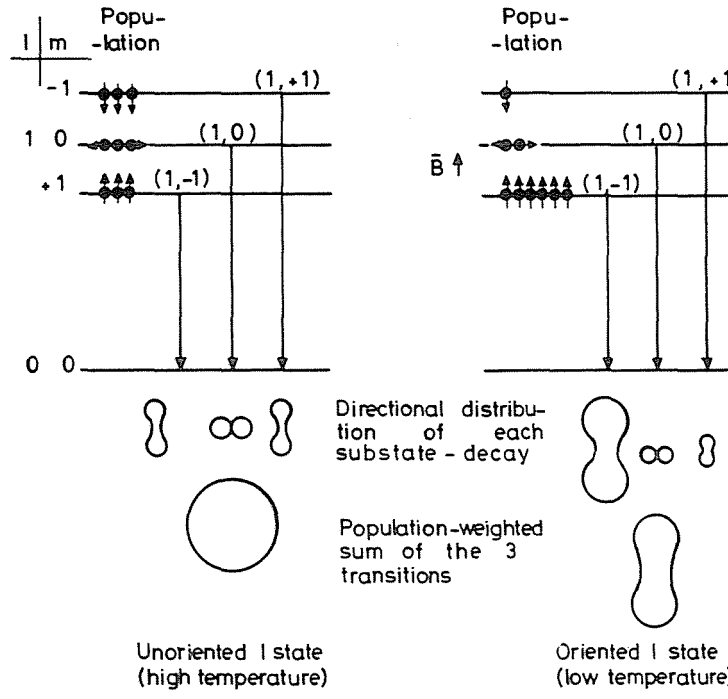


*Fig. 2.* The angular distribution of the electromagnetic radiation emitted by oriented nuclei. The number of full dots on the sublevels is proportional to the population of the sublevel. The arrows on them indicate the orientation of the nuclear magnetic dipoles with respect to the magnetic field, in the classical limit. Typical energies are: between substates $10^{-26}-10^{-25}$ J, between different $I$ states $10^{-15}-10^{-13}$ J. The radiation modes (L, 1) are indicated above the transition arrows

intermediate state can, however, have a long lifetime, and during this time extranuclear forces, as electric and magnetic fields can change this orientation, especially, when the main force acting on the intermediate nucleus is not colinear to that responsable for the initial orientation.

When a probe-nucleus is imbedded into a non-cubic or non-octahedric single crystal, the electric field at the nuclear site is inhomogeneous and the energy levels of the nucleus are split according to eq. 3. The nucleus can be oriented at low temperature by this electric field gradient, and the emission of the electromagnetic radiation becomes anisotropic. Let us apply now an external magnetic field tilted with an angle $\beta$ with respect to the previous electric field gradient $V_{zz}$ (usually crystal $c$-axis). The sublevel-energies of the intermediate state are given by eqs 2 and 3 as $W=W_{\text{mag}}\cos\beta+W_{\text{el}}$. The evolution of these energy-levels versus the external magnetic field is represented on Fig. 2a. This is the well-known Breit-Rabi diagram; the case $\beta=0$ is called in optical spectroscopy anticrossing, or level mixing, while for

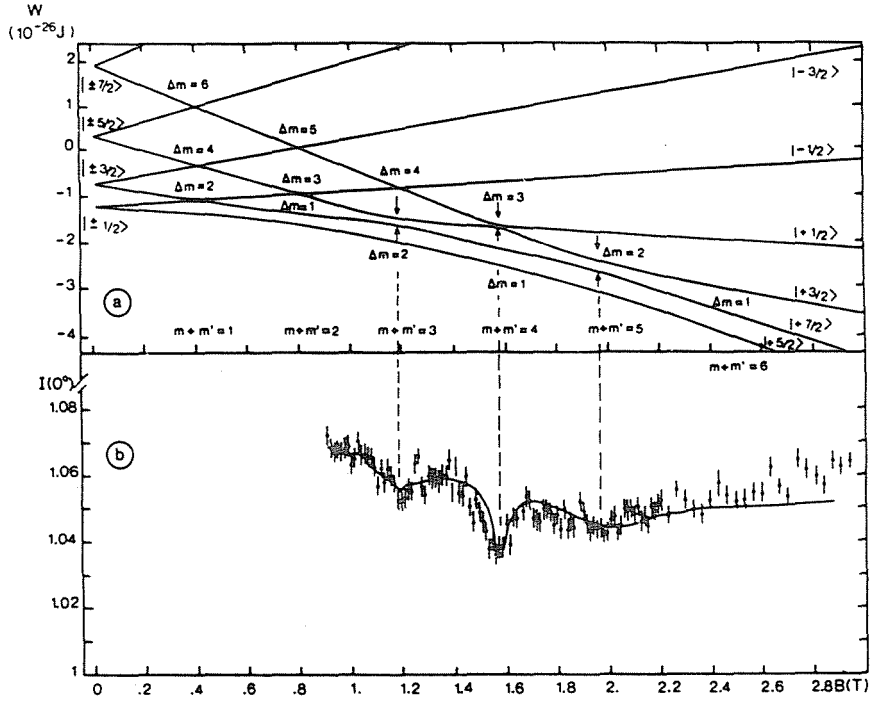


*Fig. 3.* Breit-Rabi diagram in the $^{109}$CdZn experiment with $\beta = 7°$ misalignment angle (a) and the anisotropy of the 88 keV gamma rays versus magnetic field at T = 7 mK (b). The ordinate scale is normalized to the high-temperature (isotropic) countings. The continuous line is not a fit, but a theoretically calculated one with $eQV_{zz}(^{109}Ag^{m}Zn) = 6.6 \times 10^{-26}$ J and spin-lattice relaxation time = 410 s. The line broadening of the $\Delta m = 3$ resonance is probably due to sample imperfections

$\beta = 0$ the levels cross. At magnetic field values for which at $\beta = 0$ the level cross, the levels repel for $\beta = 0$. This is clear on the figure for e.g. the $m + m' = 3\Delta m = 2$ or $m + m' = 5\Delta m = 2$ anticrossings, where $\Delta m = |m - m'|$. The nearest approach between two repelling levels is proportional to $(\sin \beta)^{\Delta m}$. At a magnetic field, set to the resonant value, the populations of the repelling sublevels mix, and if the lifetime of the state is sufficiently long, the inequality of the populations disappear and the emitted electromagnetic radiation becomes isotropic. The counting rate of a nuclear radiation detector exhibits thus resonance like variations versus the magnetic field, and the widths of these resonances are proportional to the nearest approach of the repelling levels.

In the nuclear magnetic resonance (NMR) technique the population equalisation between the sublevels is induced by the radiofrequency electromagnetic field. In the level mixing resonance (LMR) the population-exchange is spontaneous due to the nearby approach of the sublevels. As in

NMR, in LMR the signal-versus-field relation exhibits both an absorption type (Lorentzian) form (A) or a dispersion-type form (D); the relative importance of each signal depends on the direction of observation with respect to the magnetic field and electric field gradient directions.

## Experiments in Zn and Cd single crystals

The radioactive isotopes $^{111}$In, $^{107,109}$Cd were diffused into small blocks of Zn or Cd single crystals. This diffusion gives a good chance to introduce the impurities into substitutional sites of the host matrix. The samples are cooled down in a $^3$He—$^4$He dilution refrigerator to about 7 mK. The electric quadrupole interaction between the electric field gradient and the electric quadrupole moment of the impurity orients the radiactive isotopes. At this temperature the emitted electromagnetic radiations will exhibit $4-8\%$ anisotropies. A superconducting magnet supplied an homogeneous, variable intensity magnetic field, which is superposed at $\beta = 4-12°$ tilting angles to the electric field gradient. Nuclear radiation detectors were disposed in the direction of the magnetic field and in the plane perpendicular to it. Fig. 4 shows the dispersion and the absorption type signals in the $^{111}$In Zn experiment.

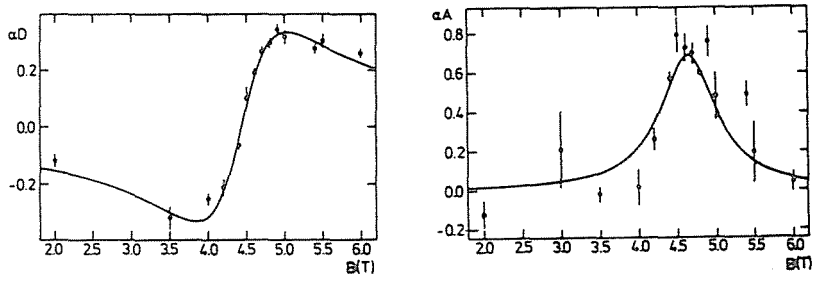


*Fig. 4.* Absorption (A) and dispersion (D) type resonances versus magnetic field on the 247 keV level in the $^{111}$InZn experience

In this experiment the lifetime of the intermediate state subject to the level mixing peturbation has a 84 ns lifetime. We have seen in paragraph 3 that this lifetime must be sufficiently long in order to allow the equalization of the populations of the mixing substates. What is the upper limit of the intermediate state lifetime compatible with this technique?

The nucleus is in interaction with the electronic shells of his own atom, which are coupled also to the neighbouring atoms of the crystal. In metallic lattices the most important contribution to these interactions comes from the overlapping of the conduction-electrons and the nucleus. This overlapping

assures the thermal equilibrium between the nucleus and the lattice. The time necessary to this thermalisation, i.e. the reorientation of the nuclear substate populations is characteristic to the impurity-host combination. It is described by the spin-lattice relaxation time, which is inversely proportional to the temperature. It is evident that, when the lifetime of the intermediate state $I$ is not short as compared to the spin lattice relaxation time, the initial orientation is wiped out and LMR cannot work.

The resonances displayed on Fig. 3b are observed in the magnetic field direction on the 88 keV state of $^{109}$Ag, the daughter-state of $^{109}$Cd. The half-life of this state is 40 s, more than eight orders of magnitude greater than that of the preceeding experiment. The correspondance between the two parts of Fig. 3 shows the equidistance of the resonances: the position of each resonance is $|m+m'|$ times the distance between to successive resonances. It can also be observed that higher order (greater $\Delta m$) resonances are narrower. This fact can be easily understood: the observed linewidth is due to the minimal approach of the two repelling levels, which is proportional to $(\sin \beta)^{\Delta m}$.

The position of the resonance in magnetic field units gives for the resonant state the ratio $eQV_{zz}/\mu V_{zz}$ is a solid-state parameter, while $\bar{\mu}$ and $Q$ are nuclear moments. The precise determination of the nuclear magnetic moments is technically easier—in our case they are already known—than that of the quadrupole interaction $eQV_{zz}$, so LMR is useful for the determination of the quadrupole interaction energy. In fact, the precision of the older methods is of the order the $5-20\%$. The precision of this determination is in any cases better than the full width at half maximum (FWHM) of the absorption-resonance. A great advantage of the method is, that the FWHM can be tuned without intensity-loss through the misalignment angle $\beta$: for a first scan, when the positions of the resonances are still unknown, a great tilting angle is applied. So, lower order resonances are wiped out and only the higher order ones appear. In the $^{109}$CdZn experiment the $\Delta m=4$ resonance of the $|m+m'|=3$ position (superposed to the large $\Delta m=2$ resonance) has been resolved and the FWHM was 7 mT, i.e. in corresponding energy units $2.10^{-28}$ J. As to appreciate this resolution, we must remark that the FWHM of $^{57}$Fe in Mössbauer-effect, a today classical high resolution tool for solid state and metallurgical investigations, is of the order of $9\times10^{-27}$ J.

As we mentioned, the inequality of the populations can be created also by a nuclear reaction. In this case the target-temperature is considerably higher than in the previous low-temperature experiments, so the spin-lattice relaxation time is shorter, which limits the range of the intermediate-state lifetimes. In such an experiment a Zn single crystal has been bombarded with $\alpha$-particles, and the resonance was observed for a nearly zero tilting angle [7].

## Conclusions

The level mixing resonance technique is a new useful tool to study nucleus-solid state interactions. As compared to existing methods the following new features appear:

i) In the lifetime range of LMR, i.e. $10^{-7}$ s $-50$ s, most of the existing techniques fail to be efficient.

ii) The high resolution of the resonance assures a high precision, but it is not an obstacle when e.g. the solid state inhomogeneities cause an inhomogeneous line broadening, because the resonance width can be tuned with the misalignment angle.

iii) As lower and higher order resonances have different line-widths, it is always possible to find an easily accessible one.

iv) The population transfer near the level mixing conditions can be favored by a radiofrequency electromagnetic field [8]. This combined LMR-NMR technique may be useful to find the resonance signal, when the nearest approach of the repelling levels cannot be attained.

## References

1. COUSSEMENT, R.–PUT, P.–HERMANS, L.–ROTS, M.–BERKES, I.–BRENIER, R.–MAREST, G.: Phys. Lett. *97A* 301 (1983)
2. PUT, P.–COUSSEMENT, R.–SCHEVENEELS, G.–HARDEMAN, F.–BERKES, I.–HLIMI, B.–MAREST, G.–SAYOUTY, E. H.: Phys. Lett. *103A* 151 (1984)
3. BERKES, I.–HLIMI, B.–MAREST, G.–SAYOUTY, E. H.–COUSSEMENT, R.–HARDEMAN, F.–PUT, P.–SCHEVENEELS, G.: Phys. Rev. *C30* 2026, (1984)
4. PUT, P.–COUSSEMENT, R.–SCHEVENEELS, G.–HARDEMAN, F.–BERKES, I.–HLIMI, B.–MAREST, G.–SAU, J.: Hyperfine Interactions *22* 131 (1985).
5. BERKES, I.: Directional Distribution of Nuclear Gamma Radiation, in Site Characterisation and Aggregation of Implanted Atoms in Materials (ed. Perez, A. and Coussement, R. Plenum Press, New York–London, 1978).
6. SIMONYI, K.: Foundations of Electrical Engineering (Pergamon Press, London, 1963).
7. SHIBUVA, S.–FUJIOKA, M.–KAWAMURA, N.–MATSUMOTO, A.–HAYASHIBE, S.–KIMURA, Y.–ISHIMATSU, T.–COUSSEMENT, R.–ROTS, M.–HERMANS, L.–PUT, P.: Hyp. Int. *14* 315, (1983).
8. BACK, L.–DEMEURE, R.–DESCHEPPER, P.–GRENÁCS, L.–LEHMANN, J.–LEBRUN, P.–LEROY, C.–MAIO, A.–PÁLFFY, L.–POSSOZ, A.: Phys. Lett. *105A* 255, (1984).

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