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An estimate of the dynamical electric field gradient acting on the nuclear-recoiling atom in cubic crystal

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Abstract

It was shown that a detectable amount of the dynamical electric field gradient could be produced by the recoil motion of atom following the various types of the nuclear decay.

The numerical evaluation was displayed for the case of the electron capture decay of ^{111}In embedded substitutionally in Ag single crystal.

§ 1. Introduction

The values of the dynamical electric field gradient in fcc single crystal due to thermal vibration of lattice atom were evaluated in ref 1) on the basis of the classical model of the crystal. We recognized that a small amount of the dynamical electric field gradient appeared in some special type of phonon and at the high temperature of the crystal. We also noticed that value of the dynamical electric field gradient averaged over all allowed types of phonons disappeared completely independent of crystal temperature because of basically the symmetry property of the phonon mode.

This implies that there is no way to detect the dynamical electric field gradient only by the temperature control of the crystal.

Roughly speaking, the thermal energy of the lattice atom is the order of 0.1 eV and the amplitude of the vibration motion of lattice atom corresponding thermal energy is the order of 10^{-9} cm even at the high temperature of the crystal, as seen in numerical tables of the dynamical electric field gradient in ref 1).

On the other hand, if the lattice atom is radioactive, the recoil energy of the lattice atom following the radioactive decay can be much more than the thermal energy of lattice atom.

Table 1 Recoil energy for the various types of decay processes

Process	E_r(eV)	Typical Case		
		E(MeV)	A	E_r(eV)
γ emission neutrino emission	$533E^2/A$	0.25	100	0.33
		0.50	100	1.33
		1.00	100	5.33
electron emission β ray emission	$(139/A)[\{(E/0.511)+1\}^2-1]$	0.25	100	1.69
		0.50	100	4.05
		1.00	100	10.8

E(MeV) is γ ray energy for γ ray emission process and the energy release in the electron capture decay for neutrino emission process. For electron emission or β ray emission process, E(MeV) is electron or β ray energy.

A numerical value[2] of the recoil energy for a typical decay mode could be the order of 1 eV for gamma ray emission and neutrino emission decay and could be even the order of 10 eV for β ray emission and conversion electron emission decay as seen in Table 1.

These larger values of recoil energy could produce the larger amplitude of the oscillation due to the recoil motion of the lattice atom. Furthermore, the direction of the recoil motion can be identified by the detection of γ ray, β ray or electron for γ decay, β decay or conversion electron emission decay.

Even in the case of electron capture decay, the direction of recoil motion due to neutrino emission can be determined principally by the detection of the circular polarization of a component of K X ray[3].

Our idea to detect the dynamical electric field gradient stems from the large recoil energy and the possible identification of the direction of the recoil motion.

§ 2. The recoil motion of radioactive atom in cubic crystal

We treat the cubic crystal in which each lattice point is occupied by a host atom with a mass M′ and a charge Ze and a impurity atom with a mass M is assumed to be placed substitutionally in crystal.

The face-centered cubic (fcc) crystal was taken as a basic crystal throughout this paper in connection with our preceding paper[1], although the extention to the body-centered cubic (bcc) crystal is quite easy.

Fig. 1 shows the 12 nearest neighbors (n,n) and 6 next nearest neighbors in fcc crystal around the origin at which the radioactive atom is embedded substitutionally. In the figure, symboll "a" denotes the lattice constant and the number at each lattice point specifies the individual lattice atom.

All lattice atoms were simply assumed to be at rest except the radioactive atom at origin, because the amplitude of thermal motion of the lattice atom is much smaller than that of recoil motion and produce no dynamical electric field gradient on a average of time.

The equations of motion of 0 th atom (radioactive atom) are described as follows, using the displacement (u_0, v_0, w_0) of this atom,

$$M(d^2u_0/dt^2) = -ku_0$$
$$M(d^2v_0/dt^2) = -kv_0$$
$$M(d^2w_0/dt^2) = -kw_0 \qquad (1)$$

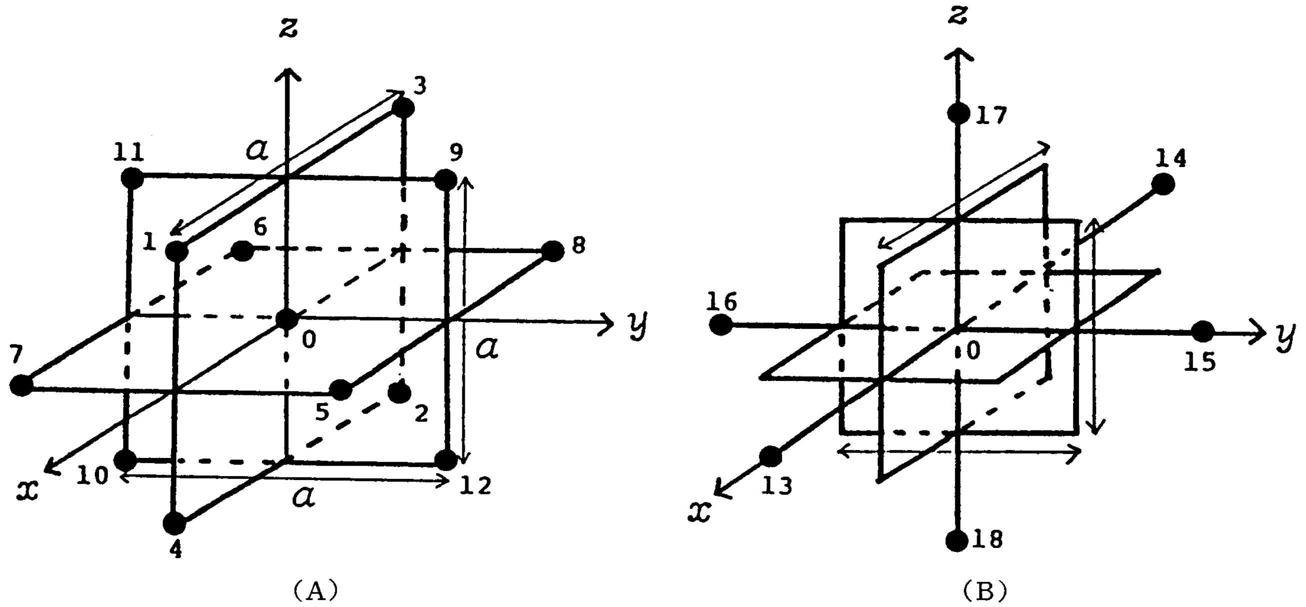

Fig. 1. 12 nearest neighbors (A) and 6 next nearest nearest neighbors (B) in fcc crystal lattice. Symbol "a" denotes the lattice constant and the number at each lattice point specifies the individual lattice atom.

,where

$$k=4\alpha_1+2\alpha_2 \tag{2}$$

Equation (1) is obtained directly from the equation (1) of ref 1) by replacement of $u_j=v_j=w_j=0$ $(j\neq 0)$.

The α_1 and α_2 are the force constant between 0 th atom and (n,n) atom and that between Oth and next (n,n) atom, respectively.

At time t=0, the radioactive atom at origin starts to move with the recoil velocity (v) to the direction specified (α, φ) as seen in Fig. 2. The solutions of the equation (1) satisfying this initial condition, take the following forms:

$$\begin{aligned} u_0 &= (v/\omega)\sin\alpha\cos\varphi\sin\omega t \\ v_0 &= (v/\omega)\sin\alpha\sin\varphi\sin\omega t \\ w_0 &= (v/\omega)\cos\alpha\sin\omega t \end{aligned} \tag{3}$$

,where the angular frequency (ω) is determined by

$$\omega=2\pi\nu=(k/M)^{1/2} \tag{4}$$

§ 3. Dynamical electric field gradient due to recoil motion of radioactive atom

For the case in which only 0 th atom and nth atom exist, the electric field gradient $(V_{zz}(n))$ acting on Oth atom displaced by $(\Delta x, \Delta y, \Delta z)$ from its equilibrium position is expressed as

$$\begin{aligned} V_{zz}(n) =& V''_0(n) + V''_x(n)\Delta x + V''_y(n)\Delta y + V''_z(n)\Delta z \\ &+ V''_{xx}(n)(\Delta x)^2 + V''_{yy}(n)(\Delta y)^2 + V''_{zz}(n)(\Delta z)^2 \\ &+ V''_{xy}(n)(\Delta x)(\Delta y) + V''_{yz}(n)(\Delta y)(\Delta z) \\ &+ V''_{zx}(n)(\Delta z)(\Delta x) + \cdots\cdots\cdots \end{aligned} \tag{5}$$

assuming nth atom being at rest at (x_n, y_n, z_n).

In equation (5), $V''_0(n)$, $(V''_x(n), V''_y(n), V''_z(n))$ and $(V''_{xx}(n), V''_{yy}(n), V''_{zz}(n), V''_{xy}(n), V''_{yz}(n), V''_{zx}(n))$ are the 0 th order, first order and second order term of the displacement, respectively.

The time average of the dynamical electric field gradient in the crystal $(\bar{V}_{zz})$ summed up n from 1 to 18 is reduced to the following form in the approximation of the neglecting the higher than the third order term,

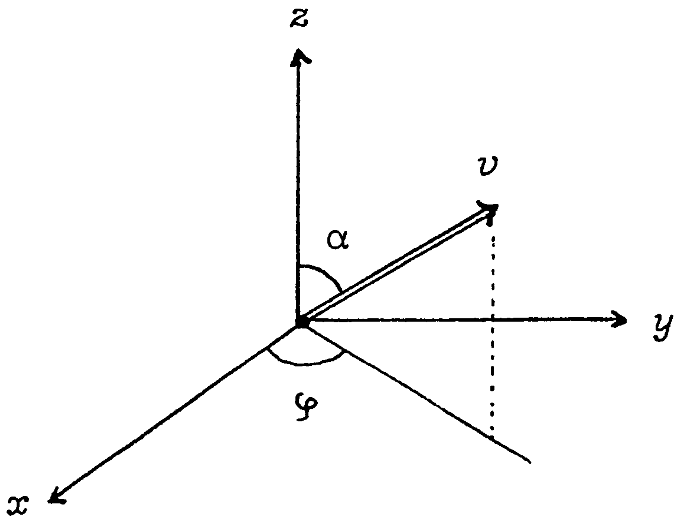

Fig. 2. Nuclear-recoiling atom is emitted to the direction (α, φ) with the velocity v.

$$\bar{V}_{zz} = (\sum_{n=1}^{18} V''_{xx}(n))\overline{(\Delta x)^2} + (\sum_{n=1}^{18} V''_{yy}(n))\overline{(\Delta y)^2} + (\sum_{n=1}^{18} V''_{zz}(n))\overline{(\Delta z)^2}$$
$$+ (\sum_{n=1}^{18} V''_{xy}(n))\overline{(\Delta x)(\Delta y)} + (\sum_{n=1}^{18} V''_{yz}(n))\overline{(\Delta y)(\Delta z)} + (\sum_{n=1}^{18} V''_{zn}(n))\overline{(\Delta z)(\Delta x)} \quad (6)$$

The upper bar of individual symbol in equation (6) means the time average over the period of oscillation due to recoil motion.

In equation (6), it should be noticed that each term appear as the simple product of summing-up term and time-averaged term.

In this equation 0 th order term disappears due to the symmetry property of the cubic crystal and first order terms also vanish completely because the displacement averaged over the period of the oscillation always lead to zero.

The elemental terms appearing in summing-up terms in equation (6) are explicitly expressed as follows:

$$V''_{xx}(n) = (48Ze/a^5)(1/\bar{r}_n^5)[1-5\bar{z}_n^2/\bar{r}_n^2-5\bar{x}_n^2/\bar{r}_n^2+35\bar{z}_n^2\bar{x}_n^2/\bar{r}_n^4]$$
$$V''_{yy}(n) = (48Ze/a^5)(1/\bar{r}_n^5)[1-5\bar{z}_n^2/\bar{r}_n^2-5\bar{y}_n^2/\bar{r}_n^2+35\bar{z}_n^2\bar{y}_n^2/\bar{r}_n^4]$$
$$V''_{zz}(n) = (48Ze/a^5)(1/\bar{r}_n^5)[3-30\bar{z}_n^2/\bar{r}_n^2+35\bar{z}_n^4/\bar{r}_n^4]$$
$$V''_{xy}(n) = (48Ze/a^5)(10\bar{x}_n\bar{y}_n/\bar{r}_n^7)[7\bar{z}_n^2/\bar{r}_n^2-1]$$
$$V''_{yz}(n) = (48Ze/a^5)(10\bar{y}_n\bar{z}_n/\bar{r}_n^7)[7\bar{z}_n^2/\bar{r}_n^2-3]$$
$$V''_{zx}(n) = (48Ze/a^5)(10\bar{z}_n\bar{x}_n/\bar{r}_n^7)[7\bar{z}_n^2/\bar{r}_n^2-3] \quad (7)$$

,whre $(\bar{x}_n, \bar{y}_n, \bar{z}_n, \bar{r}_n)$ is a reduced quantity of (x_n, y_n, z_n, r_n) expressed as

$$\bar{x}_n = ax_n/2$$
$$\bar{y}_n = ay_n/2$$
$$\bar{z}_n = az_n/2$$
$$\bar{r}_n = (\bar{x}_n^2+\bar{y}_n^2+\bar{z}_n^2)^{1/2} \quad (8)$$

Applying the values of the reduced quantities $(\bar{x}_n, \bar{y}_n, \bar{z}_n, \bar{r}_n)$ into equation (7) and (8), the equation (6) finally reduces to the following simple form:

$$\bar{V}_{zz} = 21(2\sqrt{2}-1)(Ze/a^5)[\overline{(\Delta x)^2}+\overline{(\Delta y)^2}-2\overline{(\Delta z)^2}] \quad (9)$$

As the quantities $(\Delta x, \Delta y, \Delta z)$ in equation (9) equal to (u_0, v_0, w_0) in equation (3), the time averaged dynamical electric field gradient $(\bar{V}_{zz})$ in crystal for the recoil atom specified by (v, α, φ) is expressed as

$$\bar{V}_{zz}(v, \alpha, \varphi) = (21/2)(2\sqrt{2}-1)(ze/a^5)(v/\omega)^2(3\sin^2\alpha-2) \quad (10)$$

Fig 3 shows the α-dependence of $\bar{V}_{zz}(v, \alpha, \varphi)$.

§ 4. Recoil direction of radioactive atom following various types of decay mode

In γ decay, the recoil direction is just the opposite direction of emission of γ ray, in electron capture decay neutrino and in conversion electron emission decay conversion electron.

In β decay, however, the direction of recoil has a direction related to the direction of β ray emission, because the β particle share the decay energy with neutrino particle.

In all decay processes except electron capture decay, accompanied radiation can be easily detected. So the direction of the recoil motion can be identified uniquely (except β decay) or statistically (β decay). In the electron capture decay, however, it is quite difficult to identify the direction of the recoil motion of nucleus as the neutrino rarely interacts with the matter. In spite of this restricted situation for the electron capture decay, it has been pointed out[3] that the direction of the recoil motion of nucleus in electron capture decay proccess could be identified by the measurement of the circular polarization of

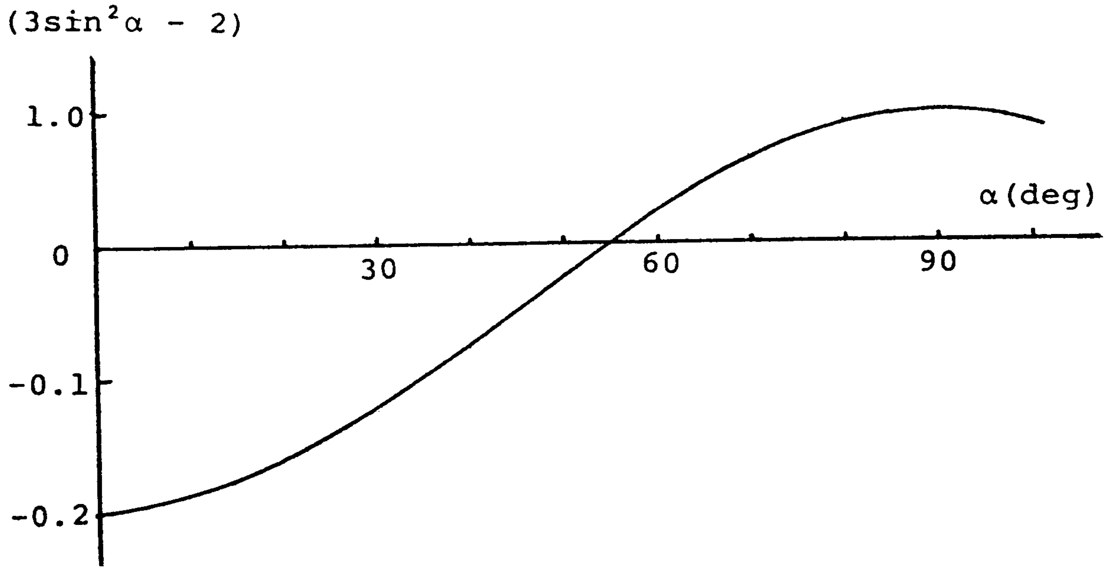

Fig. 3. The α-dependence of $\bar{V}_{zz}$ Note the value of $\bar{V}_{zz}$ is proportional to $(3\sin^2\alpha-2)$.

X ray emitted in the decay process as an effect of the parity non-conservation in β decay.

For example, if the circular polarization of X ray following the electron capture decay is measured, the direction of the recoil motion of a nucleus could be determined with respect to the propagation direction of X ray following the distribution function[3)]

$$W(\theta,\tau)=(1/24\pi)[1+H\tau\cos\theta/(1+b)] \quad \text{for } K\alpha_2\ (2p_{1/2}-1s_{1/2})$$
$$W(\theta,\tau)=(2/24\pi)\ [1-H\tau\cos\theta/(1+b)] \quad \text{for } K\alpha_1\ (2p_{3/2}-1s_{1/2}) \qquad (11)$$

In equation (11), θ stands for the angle between the direction of recoil motion and that of X ray emission, and $\tau=+1$, or -1 correspond to the right or left circular polarized X ray.

The constant b is the Fierz interference term[4)] and usually considered to be negligibly small.

The constant H is a factor showing the effect of the parity non-conserving interaction[4)].

The K α_1 and K α_2 X ray come from the atomic transition $2p_{3/2}$-$1s_{1/2}$ and $2p_{1/2}$-$1s_{1/2}$, respectively.

Even in the electon capture decay of heavy atom, the energy difference between K α_1 and K α_2 is very small and the separation of two X rays is not so easy but some possibility still remains.

On the other hand, in the detection of circular polarization of X ray, we encounter with another difficulty connected with the detection principle at the energy range of K α X ray.

With this energy range, we have to use the detection principle of the photoelectric effect because the Compton scattering cross section is negligibly small. For the effective detection of circular polarization of X ray, it is necessary to utilize the polarized inner atomic shell electrons.

Generally speaking, inner atomic levels with electron spin up and spin down degenerate completely. It has been pointed out theoretically and experimentally[5-7)], however, that the magnetized iron produces the split of the energy levels of inner shell with spin up and spin down.

So, in principle, the way to detect the circularly polarized Kα_1 and Kα_2 X ray is still open. This means that the detection of the circular polarization of Kα_1 or Kα_2 X ray identifies the direction of the recoil motion of the radioactive atom.

Any way, we could get in principle some experimental method to detect the direction of the recoil motion for all types of the decay precesses.

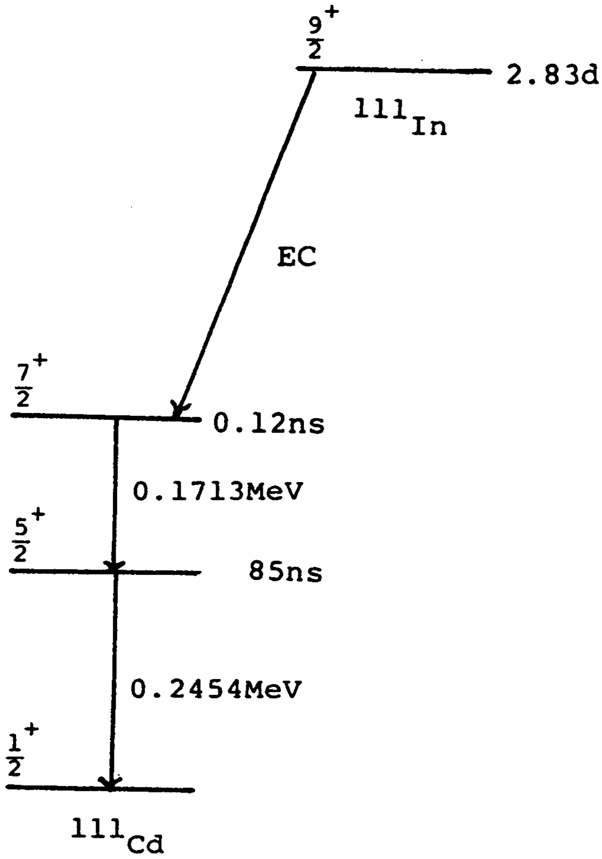

Fig. 4. The decay scheme of ^{111}In
The times written on the right hand side of levels mean the half-life of the corresponding levels.

§ 5. Numerical evaluation of the dynamical electric field gradient for the case of ^{111}In decay in Ag single crystal

Fig. 4 shows the decay scheme[8] of ^{111}In. The electron capture transition from ^{111}In ($9/2^+$) to ^{111}Cd ($7/2^+$) is classified to be unfavoured allowed transition[9]. The energy release (E_{er}) of this transition is calculated as 0. 404 MeV. This value leads to the recoil energy (E_r) to be 0. 78 eV, using the formula seen in Table 1.

This recoil energy is about several times larger than the thermal vibration energy of atom in crystal at room temperature.

The recoilless fraction (f) is estimated as follows.

The function (f) is expressed as[10]

$$f=\exp[-(E_r/(kT_D))\{3/2+(\pi T/T_D)^2\}] \tag{12}$$

, where k, T_D and T are Boltzman constant, Debye temperature of impurity in host material and the temperature of whole material, respectively. The Debye temperature of impurity (T_D) is related to[11] that of host (T'_D) as

$$T_D=T'_D(M'/M)^{1/2} \tag{13}$$

, here M (M′) is the mass of the impurity (host) atom. Inserting the values $M'=107.88$, $M=111$, $T'_D=229$ (K) into equation (13), we get the value $T_D=225.8$ (K). The recoilless fraction (f) takes the maximam value at $T=0$. Putting the value $E_r=0.78$ eV and $T_D=225.8$ (K) into equation (12), it becomes clear that the value of (f) is completely zero even at $T=0$.

From these consideration, it is justified that the recoil atom oscillate under the recovering force following the equation (1).

The force constant (k) is estimated using lattice constant (a) and stiffness constant (c_{11}, c_{12}, c_{44}) of Ag crystal because α_1 and α_2 are expressed as

$$\alpha_1 = a c_{44}$$
$$\alpha_2 = (a/4)(c_{11} - c_{12} - c_{44}).$$

The recoil velocity (v) of the radioactive nucleus is obtained from the recoil energy (E_r) as

$$v = 1.17 \times 10^5 \ \mathrm{cm \cdot s^{-1}}. \qquad (14)$$

The maximum amplitude (A) of the oscillation motion of the recoil atom is calculated as

$$A = 0.59 \times 10^{-8} \ \mathrm{cm} \qquad (15)$$

by the equation

$$(1/2)\ M v^2 = (1/2)\ k\ A^2$$

and using the constant $a = 4.078 \times 10^{-8}$ cm, $c_{11} = 1.240 \times 10^{12}$ dyne•cm^{-1}, $c_{12} = 0.937 \times 10^{12}$ dyne•cm^{-1}, $c_{44} = 0.461 \times 10^{12}$ dyne•cm^{-1}.

This amplitude is about 3.5 times larger than that of the thermal vibration at 1200K. Therefore nuclear-recoiling atom could experience very large dynamical electric field gradient.

The frequency (ν) of the oscillation due to the nuclear-recoil motion is calculated as

$$\nu = (\omega/(2\pi)) = (1/(2\pi))(k/M)^{1/2} = 0.314 \times 10^{13} \mathrm{s^{-1}}. \qquad (16)$$

The dynamical electric field gradient acting on the nuclear-recoiling atom is a function of the direction of the recoil motion and takes the maximum value at $\alpha = 0$. Inserting the calculated constant mentioned above and Ze=1 into equation (10), we get

$$\bar{V}_{zz}(\alpha = 0, \varphi) = 0.34 \times 10^{17} \ \mathrm{V \cdot cm^{-2}}. \qquad (17)$$

This value is large enough to be detected.

From these numerical evaluation, we conclude that if we are able to get a effective method to identify the direction of the recoil motion, we can always detect the dynamical electric field gradient acting on nuclear-recoiling atom in single crystal.

Although discussion described above is based on the classical model of the lattice and the crude approximation is accepted, we expect that the main features of the evaluation will be alive even in further refinement of model and approximation.

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