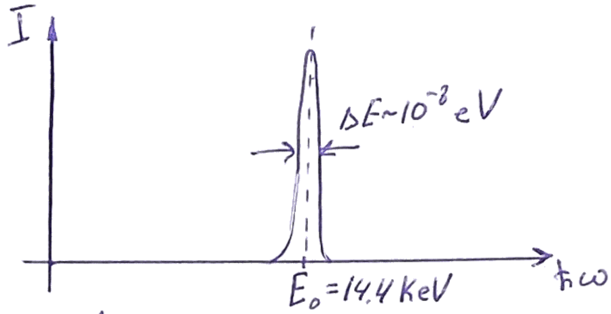
Mößbauer effect: recoilles resonance fluorescence

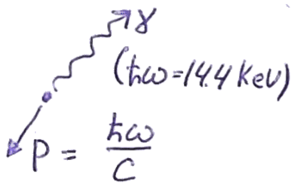
Natural width of the ^{57}Fe line: $\Delta E \cdot \Delta t \gtrsim \frac{\hbar}{2} \Rightarrow \Delta E \sim \frac{\hbar}{\Delta t} \sim \frac{6.6 \cdot 10^{-16}\, eV \cdot s}{10^{-7}\, s} \sim 10^{-8}\, eV$

Emission / absorption in nuclear frame:

Quality factor: $Q = \frac{E_0}{\Delta E} \sim 10^{12}$ (!)



If the nucleus is free, we have to take recoil into accout:

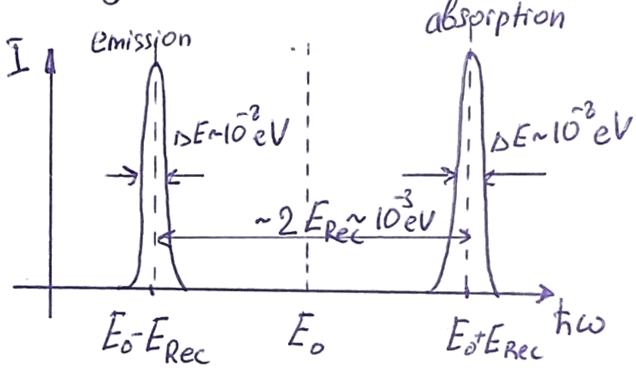


Recoil energy: $E_{Rec} = \frac{p^2}{2M} = \frac{(\hbar\omega)^2}{2Mc^2} \sim \frac{(14.4\, KeV)^2}{2 \cdot 5.6 \cdot 10^{10}\, eV} \sim 2 \cdot 10^{-3}\, eV$

Emission line will be shifted by E_{Rec} to lower energies.
Absorption line will be shifted by E_{Rec} to higher energies.

Emission / absorption in laboratory frame:

No resonance is possible, because the emission and absorption lines do not overlap.

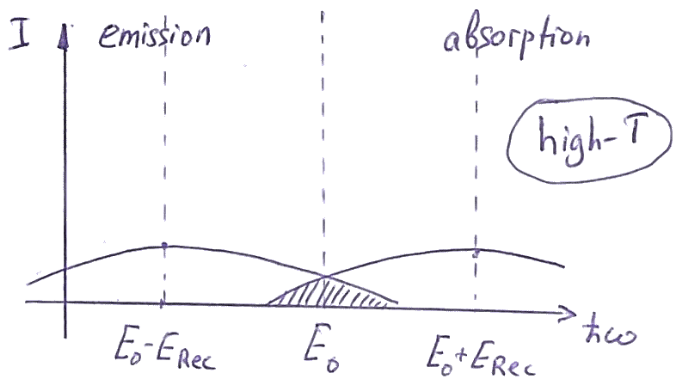


Expectation: the emission and absorption lines can be broaden by $\sim KT$ because of the thermal motion (Doppler effect).

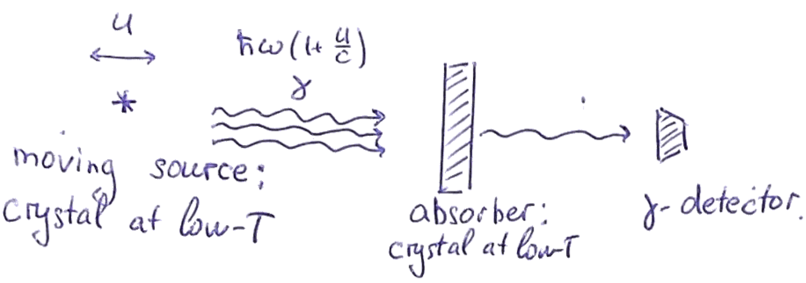
$KT \sim 2.6 \cdot 10^{-2}\, eV > E_{Rec}$

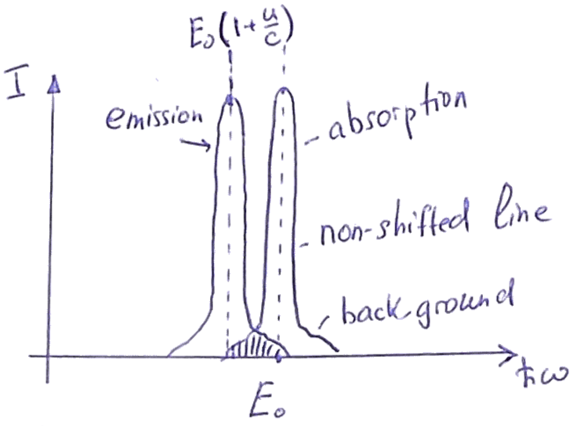
$K = 8.6 \cdot 10^{-5}\, eV/K$

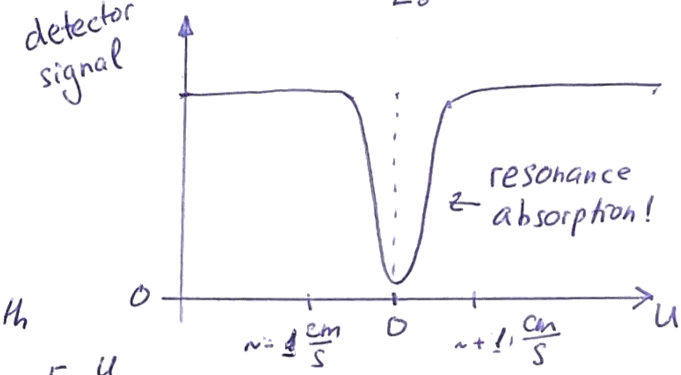
$T \sim 300\, K$



Mössbauer experiment







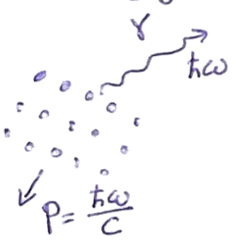
Experimental observations:

1) Resonance absorption is stronger at low temperatures
2) There is unshifted line with natural width
3) The whole spectrum can be shifted by $E_0 \cdot \frac{u}{c}$ moving the source (or absorber) with the speed u.

to shift by ΔE (natural width): $u \approx \frac{\Delta E}{E_0} \cdot c \sim 10^{-12} \cdot 3 \cdot 10^{10} \frac{cm}{s} \approx 3 \cdot 10^{-2} \frac{cm}{s}$

Explanation:

In solids, at low temperatures, the recoil momentum is transferred to the whole crystal ($N \sim 10^{23}$ atoms). Therefore, the recoil energy is vanishingly small:



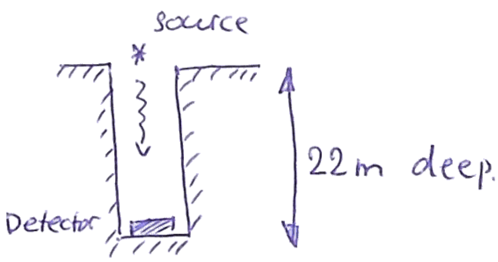
$$E_{Rec} = \frac{p^2}{2MN} = \frac{(\hbar\omega)^2}{2Mc^2} \cdot \frac{1}{N} \xrightarrow[N \to \infty]{} 0$$

It means that atoms in a cryst<u>all</u> do not start to move in such a recoilless process.
No phonons are created or absorbed.

Applications:
- very accurate ($Q \sim 10^{12}$) measurement of the nuclear emission and absorption lines (chemical isomer shift, quadrupole splitting, magnetic splitting)
- Proof of the general relativity (Pound-Rebka, 1959)

gravitational red-shift was measured

$\Delta(\hbar\omega) \sim 2 \cdot 10^{-2} \Delta E$ - sophisticated but possible



Let us calculate the probability of recoilless absorption (emission)

Fermi's golden rule: $\Gamma_{i\to f} = \frac{2\pi}{\hbar} |\langle\{f\}|\hat{H}_{int}|\{i\}\rangle|^2 \cdot \rho(E_f)$

rate (probability)

interaction between the γ-photon and the nucleus.

density of final states

$\{f\}$ - final phonon state of the crystal

$\{i\}$ - initial phonon state of the crystal

„recoilless" means that $\{f\} \equiv \{i\}$, i.e phonon state does not change

Let us find $\hat{H}_{int}$. The full Hamiltonian (interaction of a charged particle with an electromagnetic field):

$$\hat{H}_{full} = \sum_{\alpha} \frac{1}{2m_\alpha} \left(\vec{p}_\alpha - \frac{e_\alpha}{c}\vec{A}\right)^2 + \sum_{\alpha} e_\alpha \varphi$$

over nuclons — Vector-potential — scalar potential.

Using the Weyl gauge ($\varphi = 0$),

$$\hat{H}_{full} = \underbrace{\sum_{\alpha} \frac{1}{2m_\alpha} (\vec{p}_\alpha)^2}_{\text{Kinetic energy of nuclons}} + \underbrace{\sum_{\alpha} \frac{1}{2m_\alpha} \left(\frac{e_\alpha}{c} \cdot \vec{A}\right)^2}_{\text{Small correction}} - \underbrace{\frac{1}{c} \sum_{\alpha} \frac{e_\alpha}{m_\alpha} (\vec{p}_\alpha \cdot \vec{A})}_{\hat{H}_{int} \text{ in a dipole approximation}}$$

in an electromagnetic wave, $\vec{A}(\vec{r}) = \vec{A}_0 \cdot e^{i\vec{k}\vec{r} - i\omega t}$

Position of a nuclon is: $\vec{r}_\alpha = \vec{r}_n + \vec{\rho}_\alpha$

position of nucleus (center of mass) — position of a nuclon inside the nucleus

$$\hat{H}_{int} \propto \underbrace{\frac{1}{c} \sum_{\alpha} \frac{e_\alpha}{m_\alpha} \cdot \vec{p}_\alpha \cdot \vec{A}_0 \cdot e^{i\vec{k}\vec{\rho}_\alpha - i\omega t}}_{H_0 \text{ - interaction of the } \gamma\text{-photon with the nucleus}} \cdot \underbrace{e^{i\vec{k}\vec{r}_n}}_{\text{position of the nucleus}} = H_0 \cdot e^{i\vec{k}\vec{r}_n}$$

Finally, the probability of recoilless absorption (emission) is determined by:

$$\Gamma_{i\to i} = \frac{2\pi}{\hbar} \cdot |H_0|^2 \cdot \rho(E_f) \cdot |\langle\{i\}| e^{i\vec{k}\vec{r}_n} |\{i\}\rangle|^2 \propto \underbrace{|\langle\{i\}| e^{i\vec{k}\vec{r}_n} |\{i\}\rangle|^2}_{\text{Lamb - Mössbauer factor}}$$

$\langle\{i\}| e^{i\vec{k}\vec{r}_n} |\{i\}\rangle \equiv \langle e^{i\vec{k}\vec{r}_n} \rangle$ - average value (over phonons at any T)

Position of a nucleus in a crystal: $\vec{r}_n = \underbrace{\vec{R}_n}_{\text{equilibrium position}} + \underbrace{\vec{u}_n}_{\text{displacement due to phonons}}$

$$\Gamma_{i\to i} \propto |\langle e^{i\vec{K}\vec{r}_n}\rangle|^2 = |e^{i\vec{K}\vec{R}_n}\cdot\langle e^{i\vec{K}\vec{u}_n}\rangle|^2 = \underbrace{|e^{i\vec{K}\vec{R}_n}|^2}_{=1}\cdot|\langle e^{i\vec{K}\vec{u}_n}\rangle|^2 = |\langle e^{i\vec{K}\vec{u}_n}\rangle|^2$$

At low temperatures, $\vec{u}_n \approx 0 \Rightarrow e^{i\vec{K}\vec{u}_n} \approx 1$, which means that the probability of the Mössbauer effect is non-zero.

Displacement is small ($\vec{K}\vec{u}_n \ll 1$): $e^{i\vec{K}\vec{u}_n} \approx 1 + i(\vec{K}\vec{u}_n) - \frac{1}{2}(\vec{K}\vec{u}_n)^2 + \dots$

$$\langle e^{i\vec{K}\vec{u}_n}\rangle \approx 1 + i\langle\vec{K}\vec{u}_n\rangle - \frac{1}{2}\langle(\vec{K}\vec{u}_n)^2\rangle + \dots$$

Let us introduce a unit vector $\tilde{\vec{K}} = \frac{\vec{K}}{K}$ pointing in the direction of the wave vector of the γ-photon.

$$\vec{K}\vec{u}_n = K\cdot(\tilde{K}^x u_n^x + \tilde{K}^y u_n^y + \tilde{K}^z u_n^z)$$

$$\langle\vec{K}\vec{u}_n\rangle = K\cdot(\tilde{K}^x \underbrace{\langle u_n^x\rangle}_{=0} + \tilde{K}^y \underbrace{\langle u_n^y\rangle}_{=0} + \tilde{K}^z \underbrace{\langle u_n^z\rangle}_{=0}) = 0.$$

$$\langle(\vec{K}\vec{u}_n)^2\rangle = K^2\cdot\langle(\tilde{\vec{K}}\vec{u}_n)^2\rangle$$

$$\langle e^{i\vec{K}\vec{u}_n}\rangle \approx 1 - \frac{1}{2}K^2\langle(\tilde{\vec{K}}\vec{u}_n)^2\rangle \approx e^{-\frac{K^2}{2}\langle(\tilde{\vec{K}}\vec{u}_n)^2\rangle}$$

It can be shown, that $\langle u_n^x u_n^y\rangle = \langle u_n^x u_n^z\rangle = \dots = 0$.

Therefore, $\langle(\tilde{\vec{K}}\vec{u}_n)^2\rangle = \tilde{K}^{x2}\langle u_n^{x2}\rangle + \tilde{K}^{y2}\langle u_n^{y2}\rangle + \tilde{K}^{z2}\langle u_n^{z2}\rangle$

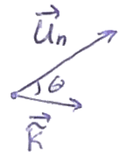
for a cubic crystal, $\langle u_n^{x2}\rangle + \langle u_n^{y2}\rangle + \langle u_n^{z2}\rangle = \langle u_n^2\rangle$

and all directions are equivalent, so $\langle u_n^{x2}\rangle = \langle u_n^{y2}\rangle = \langle u_n^{z2}\rangle = \frac{\langle u_n^2\rangle}{3}$

$$\langle(\tilde{\vec{K}}\vec{u}_n)^2\rangle = \underbrace{(\tilde{K}^{x2} + \tilde{K}^{y2} + \tilde{K}^{z2})}_{|\tilde{K}|^2=1}\cdot\frac{\langle u_n^2\rangle}{3} = \frac{\langle u_n^2\rangle}{3}.$$

Alternatively, averaging over all possible directions of a unit vector $\tilde{\vec{K}}$:

$$\overline{(\tilde{\vec{K}}\vec{u}_n)^2} = u_n^2\cdot\frac{1}{4\pi}\int_0^{\pi}\cos^2\theta\cdot\sin\theta\, d\theta\cdot\int_0^{\pi}d\varphi = \frac{1}{3}u_n^2$$



Now let us calculate $\langle u_n^2 \rangle$.

For this we will consider phonon modes:

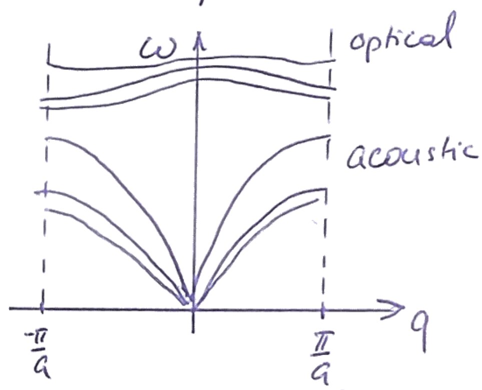
$$\vec{u}_n = \sum_{\xi} \vec{u}_{n\xi}$$ ← displacement of n^{th} atom due to phonons.

sum over all phonon modes

Phonon modes are indicated by index $\xi = \{\vec{q}, s\}$

$\vec{q}$ - wave vector

s - branch (acoustic or optical)



Let us consider a simple cubic crystal (however, the obtained result will be general)

For each phonon mode: $\vec{u}_{n\xi} = |u_\xi| \cdot \vec{e}_\xi \cdot \cos(\vec{q}\vec{R}_n - \omega_\xi t)$

amplitude ↑ ↑ polarization

Let us estimate $|u_\xi|$: Energy of the phonon mode, when n_ξ phonons are excited:

$$E = \hbar\omega_\xi \left(n_\xi + \frac{1}{2}\right)$$

Energy of N atoms oscillating with magnitude $|u_\xi|$ and frequency ω_ξ:

$$E = N \cdot \frac{M\omega_\xi^2 \cdot |u_\xi|^2}{2}$$

These energies are equal $\Rightarrow |u_\xi| = \sqrt{\frac{2\hbar\left(n_\xi + \frac{1}{2}\right)}{MN\omega_\xi}}$

$$\langle u_n^2 \rangle = \left\langle \sum_\xi \vec{u}_{n\xi} \cdot \sum_{\xi'} \vec{u}_{n\xi'} \right\rangle = \sum_{\xi\xi'} |u_{n\xi}| \cdot |u_{n\xi'}| \cdot \underbrace{\vec{e}_\xi \cdot \vec{e}_{\xi'} \cdot \left\langle \cos(\vec{q}\vec{R}_n - \omega_\xi t) \cdot \cos(\vec{q}'\vec{R}_n - \omega_{\xi'} t) \right\rangle}_{\delta_{\xi\xi'} \cdot \frac{1}{2}}$$

$$= \frac{1}{2} \sum_\xi |u_{n\xi}|^2 = \sum_\xi \frac{\hbar\left(n_\xi + \frac{1}{2}\right)}{MN\omega_\xi}$$

Finally, $\langle e^{i\vec{K}\vec{u}_n}\rangle \approx e^{-\frac{1}{2}K^2\cdot\frac{1}{3}\langle u_n^2\rangle} = e^{-\frac{1}{2}K^2\cdot\frac{1}{3}\cdot\sum\limits_{\xi}\frac{\hbar(n_\xi+\frac{1}{2})}{MN\omega_\xi}} = e^{-\frac{1}{2}W}$

Debye-Waller factor: $W = \frac{K^2}{3}\cdot\sum\limits_{\xi}\frac{\hbar(n_\xi+\frac{1}{2})}{MN\omega_\xi}$

Debye-Waller factor can be also expressed using phonon density of states:

$$W = \underbrace{\frac{\hbar^2K^2}{2M}}_{E_{Rec}}\cdot\frac{1}{3N}\cdot\sum_{\xi}\frac{2n_\xi+1}{\hbar\omega_\xi} = E_{Rec}\cdot\int_0^{\omega_{max}} d\omega\cdot\underbrace{\frac{1}{3N}\sum_{\xi}\delta(\omega-\omega_\xi)}_{g(\omega)}\cdot\frac{2n_\omega+1}{\hbar\omega}$$

$$= E_{Rec}\int_0^{\omega_{max}} d\omega\cdot g(\omega)\cdot\frac{2n_\omega+1}{\hbar\omega}\,,\quad \text{where } n_\omega = \frac{1}{e^{\frac{\hbar\omega}{kT}}-1}$$

(Bose-Einstein statistics of phonons)

at low temperatures: $n_\omega \approx 0$

$$W(T\to 0) = E_{Rec}\cdot\int_0^{\omega_{max}}\frac{1}{\hbar\omega}\cdot g(\omega)d\omega = E_{Rec}\cdot\underbrace{\left\langle\frac{1}{\hbar\omega}\right\rangle}_{\text{finite value in 3D.}}$$

at high temperatures: $n_\omega \approx \frac{kT}{\hbar\omega} \Rightarrow W \approx E_{Rec}\cdot\int_0^{\omega_{max}}\frac{2kT}{(\hbar\omega)^2}\cdot g(\omega)d\omega \propto T$

Probability of the Mössbauer effect:

$\Gamma_{i\to f} \propto |\langle e^{i\vec{K}\vec{u}_n}\rangle|^2 = e^{-W} \propto e^{-\alpha T}$ ← decreases with temperature

Addendum

It can be shown, that in any crystal with N unit cells and $j = 1, 2, \dots, g$ atoms in a unit cell, the displacement of the atom $\{n, j\}$ due to the phonon mode $\{\vec{q}, s\}$ can be written as

$$\vec{u}_{jn\xi} = \frac{1}{\sqrt{M_j N}} \cdot \left\{ \vec{e}_{j\xi} \cdot Q_\xi e^{i\vec{q}\vec{R}_n - i\omega_\xi t} + \vec{e}^{\,*}_{j\xi} \cdot Q^*_\xi e^{-i\vec{q}\vec{R}_n + i\omega_\xi t} \right\}$$

here $\vec{e}_{j\xi}$ is polarization vector

Q_ξ is the magnitude of oscillations.

Different phonon modes are orthonormal: $\sum_q e^{\alpha}_{j\{q,s'\}} e^{*\beta}_{j\{q,s\}} = \delta^{\alpha\beta} \delta_{ss'}$ $\quad \alpha = \{x, y, z\}$

$$\vec{u}_{jn} = \frac{1}{\sqrt{M_j N}} \cdot \sum_{\vec{q}, s} \left\{ \vec{e}_{j\xi} \cdot Q_\xi \cdot e^{i\vec{q}\vec{R}_n - i\omega_\xi t} + \vec{e}^{\,*}_{j\xi} \cdot Q^*_\xi \cdot e^{-i\vec{q}\vec{R}_n + i\omega_\xi t} \right\}$$

$$\langle u^\alpha_{jn} u^\beta_{jn} \rangle = \frac{1}{M_j N} \cdot \left\langle \sum_{\vec{q}, s} \left\{ e^\alpha_{j\xi} \cdot Q_\xi e^{i\vec{q}\vec{R}_n - i\omega_\xi t} + e^{*\alpha}_{j\xi} Q^*_\xi \cdot e^{-i\vec{q}\vec{R}_n + i\omega_\xi t} \right\} \cdot \sum_{\vec{q}', s'} \left\{ e^\beta_{j\xi'} Q_{\xi'} e^{i\vec{q}'\vec{R}_n - i\omega_{\xi'} t} + e^{*\beta}_{j\xi'} \cdot Q^*_{\xi'} e^{-i\vec{q}'\vec{R}_n + i\omega_{\xi'} t} \right\} \right\rangle$$

$$\langle u^\alpha_{jn} u^\beta_{jn} \rangle = \frac{1}{M_j N} \cdot \sum_{\vec{q}, s} \left\{ e^\alpha_{j\xi} \cdot Q_\xi e^{*\beta}_{j\xi} \cdot Q^*_\xi + e^{*\alpha}_{j\xi} Q^*_\xi \cdot e^\beta_{j\xi'} Q_{\xi'} \right\}$$

$$= \frac{1}{M_j N} \sum_{\vec{q}, s} |Q_\xi|^2 \cdot \left\{ \underbrace{e^\alpha_{j\{q,s\}} \cdot e^{*\beta}_{j\{\vec{q},s\}}}_{\delta^{\alpha\beta}} + \underbrace{e^{*\alpha}_{j\{q,s\}} \cdot e^\beta_{j\{q,s\}}}_{\delta^{\alpha\beta}} \right\} \propto \delta^{\alpha\beta}.$$