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V. S. Letokhov. Use of Lasers in Nuclear Spectroscopy

The paper considers the possible use of coherent laser light for: 1) production of narrow frequency-tunable emission and absorption resonances in nuclear γ emission, 2) highly accurate measurement of nuclear recoil energies on emission or absorption of γ quanta, 3) highly accurate measurement of the energy of metastable (isomeric) states of nuclei, 4) rapid separation of excited and unexcited nuclei, i.e., sorting of nuclei by states.

Two obvious physical facts underlie all of these effects:

a) the state of the nucleus (spin, excitation energy, mass) is seen in its atomic and molecular spectra;

b) motion of the atom or molecule simultaneously causes a Doppler frequency shift for both optical and nuclear transitions, together with one less obvious fact:

c) buildup of molecular oscillations causes frequency modulation of the nuclear transition.

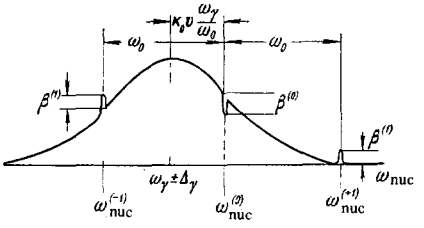
The production of narrow nuclear γ-emission resonances, unlike that of Mössbauer resonances[1], is based on the action of a coherent light wave on an infrared vibrational transition of a molecule in a gas at comparatively low pressure ($10^{-2}-10^{-1}$ Torr). A coherent wave of frequency ω builds up the vibrations of molecules having a certain projection of their velocity v onto the direction of the wave vector k_0 ($k_0v = \omega - \omega_0 = \Omega$, ω_0 is the center frequency of the Doppler absorption line)[2]. The mean oscillator displacement $\tilde{a}$ of the nucleus on excitation of molecular vibration is of the same order as or even larger than the wavelength $\lambdabar_\gamma$ of γ quanta with energies from 10 keV to 1 MeV. The vibration of a nuclear radiator with amplitude $\tilde{a} \gtrsim \lambdabar_\gamma$ should cause considerable frequency modulation of the nuclear emission. The result is distortion of the Doppler-broadened line of the nuclear transition as observed collinearly with the light wave[3,9] (see the figure). A resonance dip appears at the frequency

$$\omega_{nuc}^{(0)} = (\omega_\gamma \pm \Delta_\gamma) + \frac{\omega_\gamma}{\omega_0}\mathbf{k}_0\mathbf{v}$$

and resonance peaks at the frequencies

$$\omega_{nuc}^{(m)} = \omega_{nuc}^{(0)} \pm m\omega_0$$

($\omega_\gamma \pm \omega_\gamma$ is the center frequency of the Doppler line with consideration of the recoil energy $\Delta\gamma$). Vibrations



of molecules with a desired velocity projection $\mathbf{k}_0\mathbf{v}/k_0$ can be built up by changing the frequency deviation of the light wave with respect to ω_0, thus effecting frequency retuning of the narrow resonances within the limits of the Doppler contour of the nuclear transition. Schemes for observing narrow γ resonances in emission and absorption have been examined.

A narrow γ-radiation resonance can be obtained in a nuclear Doppler line by the action of a light wave on an atomic electron transition (double γ and optical resonance[4]). Excitation of the atom's electron shell has no influence whatever on the nuclear transitions. The situation changes materially if "runoff" of atoms that have been selectively excited by the light wave is provided (atoms with $\mathbf{k}_0\mathbf{v} = \omega - \omega_0$) are excited. This can be done by photoionization, of excited atoms only, by additional laser radiation, with subsequent extraction of the ions by a small electric field. Because of the resulting shortage of nuclei with the resonance velocity, a narrow dip appears in the Doppler line of the nuclear transition and can be observed as an absorption minimum of the γ radiation from the Mössbauer source at $(\omega_\gamma/\omega_0)\mathbf{k}_0\mathbf{v} = -\Delta\gamma$, This permits measurement of the recoil energy with a relative error of 10^{-5}.

Excitation of an atomic nucleus in a molecule should result in a change in the vibrational frequencies of the molecule, since the excitation energy ΔE is equivalent to an increase in nuclear mass by an amount[5] $\Delta m = c^{-2}\Delta E$. The resulting isomeric shift in the vibrational-rotational spectrum is smaller than or of the same order as the Doppler broadening. It can therefore be observed only by modern methods of laser spectroscopy within the Doppler width. This method can be used to resolve nuclear levels separated by ~1 keV and to determine the energy of the nucleus accurate to ~10 eV.

The isomeric shift due to excitation of the nucleus also appears in the atomic spectra as a result of the weak electron-neutron interaction. The magnitude of isomeric shifts in the atomic spectrum may greatly exceed the Doppler width, and this opens the way to selective laser excitation only of atoms with excited nuclei. Again, physical separation of the isomers can be accomplished by photoionization of the excited atoms with additional laser radiation and collection of the ions[6]. We note that selective two-stage photoionization of atoms of a given species by laser radiation was first demonstrated in[7]. This method can be used for fast separation of isomers with lifetimes down to $10^{-4}-10^{-6}$ sec, i.e., faster by a factor of 10^7-10^9 than is possible by existing methods of nuclear chemistry. It can be used to create inversions on nuclear transitions with short excited-state lifetimes[8].

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