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Subnatural-width spectroscopy by time-delayed coincidence technique

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We show that the absorption/transmission spectrum obtained by gamma-photons, detected with a delay, is narrowed due to the interference of the spectral components of the radiation field. Experimental spectra of absorption of Mössbauer radiation, obtained by coincidence technique, confirm this conclusion.

A serious obstacle in Mössbauer spectroscopy is the comparable size of line widths and line splittings of interest. The minimum linewidth achievable experimentally is determined by the lifetime of the nuclear level emitting the recoilless γ -ray. The existence of this limitation in Mössbauer spectroscopy often hinders proper interpretation of experimental spectra, as in some cases the line widths exceed the line separations.

For some Mössbauer source nucleus (like ^{57}Co), that undergoes electron capture to form a second excited state, which decays via two-photon cascade emitting sequentially the precursor and the recoilless resonance photons, there is possibility to overcome this limitation. Detection of the precursor photon and recoilless resonance photons allows one to use the coincidence-Mössbauer spectroscopy selecting only those resonance photons that are emitted during some preset time interval after detecting precursor photon. Such a "time filtering" results in substantial modification of the experimentally observed absorption/transmission-line shapes. For the first time the coincidence-Mössbauer spectroscopy was applied by Hamill and Hoy [1] to improve the spectral resolution by taking spectra counting only those gamma photons coming from the source nucleus that have lived longer than one lifetime. Appreciable line narrowing was explained by the argument that photons, emitted by nuclei that have lived longer time, have a better defined energy, i.e. it was explained by a decrease of the energy uncertainty of photons emitted at later time.

In this work we show that the narrowing of the delayed-photon spectrum is due to the quantum interference of the spectral components of the radiation field. The resolution enhancement of the time-delayed coincidence technique will be demonstrated by the example of the poorly resolved Mössbauer spectrum of Fe(III) cations chemically bounded to the surface of the graphene oxide which was measured by standard and time-delayed coincidence technique.

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