Nuclear Excitation by a Zeptosecond Multi-MeV Laser Pulse

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(Received 26 November 2010; published 23 March 2011)

A zeptosecond multi-MeV laser pulse may either excite a "plasma" of strongly interacting nucleons or a collective mode. We derive the conditions on laser energy and photon number such that either of these scenarios is realized. We use the nuclear giant dipole resonance as a representative example, and a random-matrix description of the fine-structure states and perturbation theory as tools.

DOI: 10.1103/PhysRevLett.106.122502 PACS numbers: 42.50.Ct, 24.30.Cz, 24.60.Dr

Purpose. Qualitative Considerations.—With the start of the construction of ELI (the "extreme light infrastructure") [1] or with existing ultraintense laser facilities like the National Ignition Facility (if reconfigured as femtosecond pulse systems) [2,3], nuclear spectroscopy using intense high-energy laser beams with short pulses has become a realistic possibility. Indeed, it is envisaged to generate in the decade ahead pulsed laser light with photon energies of several MeV and pulse lengths of 10^{-19} seconds by coherent Thomson backscattering [4–6]. This will be possible provided that present intense experimental and theoretical efforts will validate the concept of an electron mirror [6]. These very exciting developments call for a theoretical exploration of the expected nuclear excitation processes. In the framework of the nuclear shell model (a mean-field approach with a residual nucleon-nucleon interaction), two scenarios come to mind, distinguished by time scales. (i) The time scale for the residual interaction is large compared to the time scale for laser excitation of individual nucleons. Then a single laser pulse containing $N \gg 1$ photons excites many nucleons more or less simultaneously. The resulting "plasma" of excited and interacting nucleons (distantly similar to the initial stage of a precompound reaction) is instable. Nucleons excited above particle threshold with low angular momenta are emitted instantaneously. The remainder of the system is equilibrated by the residual interaction. Exciting questions are: What are the mean mass number, the mean excitation energy, and the mean angular momentum of the resulting compound nucleus? How big are the spreads of these quantities? Presumably it will be possible to study compound nuclei at excitation energies and spin values not accessible so far. (ii) The time scale for the residual interaction is sufficiently short compared to the time between two successive photon absorption processes. Then the nucleus relaxes after each photon absorption process. Single-photon absorption leads to a collective mode (typically the giant dipole mode), and multiple photon absorption within the same laser pulse may lead to the formation of higher harmonics of that mode. Thus, scenarios (i) and (ii) lead to extremely different forms of nuclear excitation.

In this Letter we establish the time scales and the resulting conditions on the mean photon energy E_L and the number N of photons relevant for scenarios (i) and (ii). We do so by studying scenario (ii) in detail. We show that scenarios (i) and (ii) both occur for realistic choices of E_L and N. We also show that scenario (ii) is dominated by single-photon absorption.

We focus attention on dipole absorption, the dominant photon absorption process in nuclei. The dipole mode $|10\rangle$ is the normalized product of the dipole operator and the wave function $|0\rangle$ of the nuclear ground state. The dipole mode is not an eigenstate of the nuclear Hamiltonian $H_{\rm nuc}$ and is spread over the eigenstates $|\mu\rangle$ of $H_{\rm nuc}$ with eigenvalues E_μ, $\mu = 1, \ldots$. Gross features versus excitation energy E of that spreading are measured by the strength function $S(E) = \sum_\mu \overline{|\langle 10|\mu\rangle|^2 \delta(E - E_\mu)}$. The average (overbar) is taken over an energy interval large compared to the average nuclear level spacing d. In the simplest model adopted here, $S(E)$ has a Lorentzian shape and is characterized by two parameters [7]: The peak energy $E_{\rm dip} \approx 80\ A^{-1/3}$ MeV (where A is the nuclear mass), and the width $\Gamma^\downarrow \approx 5$ MeV (the "spreading width"). The resulting broad peak of $S(E)$ is referred to as the giant dipole resonance (GDR). By the uncertainty relation, the time for the dipole mode to spread over the eigenstates of $H_{\rm nuc}$ (the "equilibration time") is $\tau_{\rm eq} = \hbar/\Gamma^\downarrow$.

The width $\Gamma_{\rm dip}$ for the gamma decay of the GDR to the nuclear ground state is estimated below and has a typical value of 5–10 keV. With N coherent photons in the laser pulse, the characteristic time scale for photon absorption is $\tau_{\rm dip} = \hbar/(N\Gamma_{\rm dip})$. One would expect that excitation of the GDR [as opposed to scenario (i) considered above] dominates whenever $\tau_{\rm dip} > \tau_{\rm eq}$, i.e., whenever $N\Gamma_{\rm dip} < \Gamma^\downarrow$. That simple estimate is modified by two factors, however. (i) For a short laser pulse with energy spread σ (where we take $\sigma \approx 10$ keV corresponding to a pulse length of $\approx 10^{-19}$ s), the Lorentzian shape of the GDR produces for $E_L < E_{\rm dip}$ an additional factor $[\Gamma^\downarrow/(E_L - E_{\rm dip})]^2$. (ii) The characteristic cubic dependence of $\Gamma_{\rm dip}$ on energy yields an additional factor $(E_L/E_{\rm dip})^3$. In total, the criterion for collective excitation of the GDR at energy E_L reads

$N < (E_L/E_{\rm dip})^3[(E_L - E_{\rm dip})^2/(\Gamma_{\rm dip}\Gamma^\downarrow)]$. With $\Gamma_{\rm dip} = 10$ keV, $\Gamma^\downarrow = 5$ MeV, $E_{\rm dip} = 14$ MeV, $E_L = 7$ MeV that yields $N < 5 \times 10^3$. That bound on N is significantly larger than the bound $N < \Gamma^\downarrow/\Gamma_{\rm dip} \approx 700$ obtained from the naive estimate and shows that even for an intense laser pulse, excitation of the collective GDR is a realistic alternative in nuclei to multiple excitation of individual nucleons provided only that E_L is sufficiently far below $E_{\rm dip}$. Thus, varying both N and E_L provides the exciting opportunity to investigate scenarios (i) and (ii) separately as well as the dynamical interplay between.

We support these qualitative arguments by calculating the probabilities P_1 for single–quantum dipole excitation and P_2 for double-quantum dipole excitation as functions of N, E_L, and σ. To calculate P_2 we use the Brink-Axel hypothesis [8,9]. The hypothesis implies that single excitation of the dipole mode may be followed either by double excitation of that mode (i.e., formation of the second harmonic) or by dipole excitation of the configurations mixed with the single-dipole mode. We account for both possibilities and show that for $\sigma \ll \Gamma^\downarrow$ the contribution from the Brink-Axel mechanism dominates and yields $P_2 = (1/2)P_1^2$. For values of N and of E_L such that $P_1 \ll 1$ that relation implies that single-photon absorption is the dominant process even if $N \gg 1$. Our result suggests that the probability for nuclear excitation by n-fold dipole absorption may be approximately given by $P_n \approx 2^{-n}P_1^n$. That would imply that in the regime where our approximations apply ($P_1 < 1/2$ or so) multiple collective nuclear excitation is unlikely.

For the complex configurations that mix with the single or double-dipole modes, we use a random-matrix model. Every such model is based upon the implicit assumption that the equilibration time (here $\tau_{\rm eq}$) is short compared to the time scale of the physical process of interest (here $\tau_{\rm dip}$). Our use of random-matrix theory is justified if the above-mentioned conditions for collective excitation of the GDR are met. We also use perturbation theory to calculate P_1 and P_2. That is justified if P_1 and P_2 are sufficiently small compared to unity. The resulting constraint is the same as for the use of the random-matrix approach itself.

Hamiltonian.—We write the total Hamiltonian as

$$\mathcal{H}(t) = H_{\rm nuc} + H(t) \tag{1}$$

where $H(t)$ stands for the time-dependent interaction with the laser pulse. In constructing $H_{\rm nuc}$ we are guided by the following qualitative picture [10]. In a closed-shell nucleus, the dipole mode $|10\rangle$ is a superposition of one-particle one-hole ($1p\ 1h$) excitations. That mode is embedded in a sea of $2p\ 2h$ excitations $|0k\rangle$ where $k = 1, \ldots, K$ and $K \gg 1$. (Here and in what follows the first label of the state vector counts the number of absorbed dipole quanta and the second enumerates the states). The mixing of both kinds of excitations causes the dipole mode to be distributed over the eigenstates of $H_{\rm nuc}$. The absorption of a second dipole quantum may either lead from the dipole mode $|10\rangle$ to the double dipole mode $|20\rangle$ (a $2p\ 2h$ state), or it may lead from one of the $2p\ 2h$ states $|0k\rangle$ to the dipole mode $|1k'\rangle$ of that same state (a $3p\ 3h$ state). The double dipole mode $|20\rangle$ is similarly embedded in a sea of $3p\ 3h$ states $|0\alpha\rangle$ with $\alpha = 1, \ldots, L$. All of the states $|1k'\rangle$ are embedded in a sea of $4p\ 4h$ states $|0\rho\rangle$ where $\rho = 1, \ldots, M$ and $M \gg K$. The residual interaction of the nuclear shell model mixes these configurations, and both the double dipole mode and the states $|1k'\rangle$ are spread out over the eigenstates of $H_{\rm nuc}$. In modeling this qualitative picture we disregard the fact that single or double dipole excitation may populate states with different spin and isospin values. $H_{\rm nuc}$ is accordingly schematically written in matrix form as follows.

$$H_{\rm nuc} = \begin{pmatrix} E_0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & E_1 & V_{1l} & 0 & 0 & 0 & 0 \\ 0 & V_{k1} & \tilde{H}^{(1)}_{kl} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & E_2 & V_{2\beta} & 0 & 0 \\ 0 & 0 & 0 & V_{\alpha 2} & \tilde{H}^{(2)}_{\alpha\beta} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & \tilde{\mathcal{H}}_{k'l'} & W_{k'\sigma} \\ 0 & 0 & 0 & 0 & 0 & W_{\rho l'} & \tilde{h}_{\rho\sigma} \end{pmatrix}. \tag{2}$$

Here E_0 is the energy of the nuclear ground state, while E_1 and E_2 are the mean excitation energies of the single and of the double dipole modes. For simplicity we use a harmonic-oscillator picture so that $E_2 - E_1 = E_1 - E_0 = E_{\rm dip}$. Moreover we put $E_0 = 0$. The real matrix elements V_{1l} mix the dipole mode with the $2p\ 2h$ states $|0l\rangle$. These are governed by the K-dimensional Hamiltonian matrix $\tilde{H}^{(1)}_{kl}$. Similarly, the matrix elements $V_{2\beta}$ mix the double dipole mode with the $3p\ 3h$ states $|0\beta\rangle$. These are governed by the L-dimensional Hamiltonian matrix $\tilde{H}^{(2)}_{\alpha\beta}$. We write $\tilde{H}^{(1)}_{kl} = E_1\delta_{kl} + H^{(1)}_{kl}$ and $\tilde{H}^{(2)}_{\alpha\beta} = E_2\delta_{\alpha\beta} + H^{(2)}_{\alpha\beta}$ and assume that both $H^{(1)}_{kl}$ and $H^{(2)}_{\alpha\beta}$ are random matrices, members of the Gaussian orthogonal ensemble, with no correlations between the elements of $H^{(1)}_{kl}$ and of $H^{(2)}_{\alpha\beta}$. The spectra of $E_1\delta_{kl} + H^{(1)}_{kl}$ and of $E_2\delta_{\alpha\beta} + H^{(2)}_{\alpha\beta}$ both have the shape of a semicircle centered at E_1 and E_2, respectively. The last diagonal block in Eq. (2) describes similarly the mixing of the states $|1k'\rangle$ with the $4p\ 4h$ states $|0\rho\rangle$. We write $\tilde{\mathcal{H}}_{k'l'} = E_2\delta_{k'l'} + \mathcal{H}_{k'l'}$ and $\tilde{h}_{\alpha\beta} = E_2\delta_{\alpha\beta} + h_{\alpha\beta}$. We implement the Brink-Axel hypothesis by putting $\mathcal{H} = H^{(1)}$. Again, the M-dimensional matrix $h_{\rho\sigma}$ is assumed to be a member of the Gaussian orthogonal ensemble. We calculate the excitation probabilities P_1 and P_2 as ensemble averages for K, L, $M \to \infty$. In that limit, the spreading widths of the single and double dipole mode and of each of the states $|1k'\rangle$ are given by the generic expression [11] $\Gamma^\downarrow = 2\pi v^2\rho$ where v^2 stands for the mean square of the relevant mixing matrix elements and ρ for the mean level density in the center of the semicircle. To avoid unnecessary complexity, we assume that all spreading widths have the same value $\Gamma^{\downarrow}$. That schematic picture can be refined if the need arises. We disregard the fact that the states excited by gamma absorption may decay by particle or by gamma emission. That is justified because the time scales associated with such decay are orders of magnitude larger than both $\tau_{\rm eq}$ and $\tau_{\rm dip}$.

For the time-dependent interaction Hamiltonian $H(t)$, we use a semiclassical description (justified for $N \gg 1$) and write

$$H(t) = \sqrt{N} g(t) H_{\rm dip}. \tag{3}$$

Here $H_{\rm dip}$ is the time-independent electromagnetic interaction operator for a single-photon dipole transition. The factor $\sqrt{N}$ accounts for the presence of $N \gg 1$ photons and the ensuing factor N in the transition rate. The dimensionless function $g(t)$ describes the time dependence of the short laser pulse. We use the ansatz

$$g(t) = \exp[-\sigma^2 t^2/(2\hbar^2) - i\omega_L t]. \tag{4}$$

The Fourier transformation of $g(t)$ shows that the mean energy of the laser pulse is $E_L = \hbar\omega_L$, and the spread in energy has a width σ. Actually, the interaction $H_{\rm dip}$ depends on energy, too, via the wave number k. For $\sigma \approx 10$ keV we may put $k \approx k_L$ where $k_L = E_L/(\hbar c)$.

In the scheme of Eq. (2) the nonzero matrix elements of the dipole operator are $\langle 10|H_{\rm dip}|0\rangle$, $\langle 20|H_{\rm dip}|10\rangle$, and $\langle 1k'|H_{\rm dip}|0k\rangle$. We use the Brink-Axel hypothesis to write $\langle 1k'|H_{\rm dip}|0k\rangle = \delta_{kk'}\langle 1k|H_{\rm dip}|0k\rangle$. We assume that all nonzero matrix elements of the dipole operator have the same value written as $\langle H_{\rm dip}\rangle$. That corresponds to a harmonic-oscillator approximation. To estimate $\langle H_{\rm dip}\rangle$, we write the Hamiltonian $H_{\rm int}$ describing the interaction with the electromagnetic field in the Coulomb gauge as $H_{\rm int} = -(1/c)\vec{j}\vec{A}$. Here $\vec{j}$ is the current and $\vec{A}$ the vector potential. In our time-dependent approach the latter has the form of a wave packet,

$$\vec{A}(\vec{x}, \Omega, t) = \alpha \int d\omega \exp[-i\omega t]\tilde{g}(\omega)\exp[i\vec{k}\vec{r}]\vec{\chi}. \tag{5}$$

The unit vector $\vec{\chi}$ describes the polarization, Ω indicates the direction of the vector $\vec{k}$, and $k = \sqrt{\vec{k}^2}$ and ω are related by $k = \omega/c$. The function $\tilde{g}$ is the Fourier transform of $g(t)$ in Eq. (4). We determine the normalization constant α from the requirement that the energy carried by $\vec{A}$ be equal to E_L. We use the dipole approximation. That yields $\alpha^2 = (\sigma E_L)/(\pi^{1/2}\hbar c)$. Quantization of the electromagnetic field for individual quanta that have the form of the wave packet (5) yields for the energy density the expression $n(E) = 1/(4\pi^{3/2}\sigma)$. From Fermi's golden rule, the total width for dipole decay is $\Gamma_{\rm dip} = 2\pi n(E_L)|\langle H_{\rm dip}\rangle|^2$. Thus,

$$|\langle H_{\rm dip}\rangle| = \sqrt{2\pi^{1/2}\Gamma_{\rm dip}\sigma}. \tag{6}$$

For the dipole width we use the Weisskopf estimate, $\Gamma_{\rm dip} = \frac{3}{4}\frac{e^2}{\hbar c}(kR)^2 E_L$. With $R = 3 \times 10^{-13}$ cm and $E_L = 15$ MeV that gives $\Gamma_{\rm dip} \approx 10$ keV, so that $|\langle H_{\rm dip}\rangle| \approx 10$ keV, too. A somewhat larger value for $\Gamma_{\rm dip}$ results when the Thomas-Reiche-Kuhn sum rule is taken into account. Here we are interested in order-of-magnitude estimates only, however.

Perturbation Expansion.—We solve the time-dependent Schrödinger equation in the interaction representation where the perturbation has the form

$$\tilde{H}(t) = \exp[iH_{\rm nuc}t/\hbar]H(t)\exp[-iH_{\rm nuc}t/\hbar]. \tag{7}$$

We assume that at time $t = -\infty$ the nucleus is in the ground state $|0\rangle$. We determine perturbatively the probabilities P_1 and P_2 that at time $t = +\infty$ one or two dipole quanta have been absorbed.

At $t = +\infty$, the probability amplitude for occupation of the state $|10\rangle$ reached after single-dipole absorption is

$$\begin{aligned} b_1 &= \frac{1}{i\hbar}\langle 10|\int_{-\infty}^{+\infty} dt\tilde{H}(t)|0\rangle \\ &= \frac{\sqrt{N}}{i\hbar}\langle H_{\rm dip}\rangle \int_{-\infty}^{+\infty} dt g(t)\langle 10|\exp[iH_{\rm nuc}t/\hbar]|10\rangle, \end{aligned} \tag{8}$$

and analogously (with $\langle 10|$ in the last line replaced by $\langle 0k|$) for b_{0k}. The corresponding amplitudes for the occupation of the states $|20\rangle$, $|0\alpha\rangle$ and $|1k'\rangle$, $|0\rho\rangle$ reached after double-dipole absorption are denoted by b_2, b_α, $b_{1k'}$, and $b_{0\rho}$. For example, we have

$$\begin{aligned} b_{0\rho} &= \left(\frac{1}{i\hbar}\right)^2 \langle 0\rho|\int_{-\infty}^{+\infty} dt_1 \tilde{H}(t_1)\int_{-\infty}^{t_1} dt_2 \tilde{H}(t_2)|0\rangle \\ &= \left(\frac{\sqrt{N}}{i\hbar}\right)^2 \langle H_{\rm dip}\rangle^2 \int_{-\infty}^{+\infty} dt_1 g(t_1) \\ &\quad \times \int_{-\infty}^{t_1} dt_2 g(t_2)\sum_{ll'}\langle 0\rho|\exp[-iH_{\rm nuc})t_1/\hbar]|1l'\rangle \\ &\quad \times \delta_{ll'}\langle 0l|\exp[i\{H_{\rm nuc}(t_1 - t_2)\}/\hbar]|10\rangle. \end{aligned} \tag{9}$$

The average probabilities for single and double dipole absorption are, thus, given by

$$\begin{aligned} P_1 &= \left\langle |b_1|^2 + \sum_k |b_{0k}|^2 \right\rangle, \\ P_2 &= \left\langle |b_2|^2 + \sum_\alpha |b_{0\alpha}|^2 + \sum_{k'} |b_{1k'}|^2 + \sum_\rho |b_{0\rho}|^2 \right\rangle. \end{aligned} \tag{10}$$

The big angular brackets indicate the ensemble average. The first (last) two terms that contribute to P_2 are due to double excitation of the dipole mode and to the Brink-Axel hypothesis, respectively.

Averages.—By way of example we perform the ensemble average for P_1 and focus attention on the sum of the squares of the time-dependent matrix elements in Eq. (8). Using completeness and a simple identity we obtain for these

$$\left\langle\langle 10|\exp[iH_{\rm nuc}(t_1-t_2)/\hbar]|10\rangle\right\rangle$$
$$= \int_{-\infty}^{+\infty} d\varepsilon \exp[i\varepsilon(t_1-t_2)/\hbar]\Bigg(\frac{1}{2i\pi}\Bigg\langle\langle 10|\frac{1}{\varepsilon^- - H_{\rm nuc}}|10\rangle$$
$$\times -\langle 10|\frac{1}{\varepsilon^+ - H_{\rm nuc}}|10\rangle\Bigg\rangle\Bigg). \tag{11}$$

We use Eq. (2) to write

$$\left\langle\langle 10|\frac{1}{\varepsilon^{\pm} - H_{\rm nuc}}|10\rangle\right\rangle$$
$$= \left\langle\langle 10|\frac{1}{\varepsilon^{\pm} - E_{\rm dip} - V_1(\varepsilon^{\pm} - H^{(1)})^{-1}V_1^{\dagger}}|10\rangle\right\rangle$$
$$= \frac{1}{\varepsilon - E_{\rm dip} \pm (i/2)\Gamma^{\downarrow}}. \tag{12}$$

Using Eq. (4) for $g(t)$ and carrying out the time integrals [see Eqs. (8)], we find that ε is confined to an interval of size σ around E_L. Since $\sigma \ll \Gamma^{\downarrow}$, the argument of the expression in Eq. (12) can be taken at $\varepsilon = E_L$. The remaining integration can be done. With the help of Eq. (6) that yields

$$P_1 = \frac{2\pi N\Gamma_{\rm dip}\Gamma^{\downarrow}}{(E_L - E_{\rm dip})^2 + (1/4)(\Gamma^{\downarrow})^2}. \tag{13}$$

The result (13) is intuitively appealing and clearly displays the suppression factors $\Gamma^{\downarrow 2}/(E_L - E_{\rm dip})^2$ and $(E_L/E_{\rm dip})^3$ mentioned above that come into play for $E_L < E_{\rm dip}$.

The calculation of P_2 proceeds similarly but is more involved. We use operator identities such as

$$\langle 10|\frac{1}{\varepsilon_2^- - H_{\rm nuc}}|0k\rangle$$
$$= \langle 10|\frac{1}{\varepsilon_2^- - E_{\rm dip} - V_1(\varepsilon_2^- - E_{\rm dip} - H^{(1)})^{-1}V_1^{\dagger}}|10\rangle$$
$$\times \langle 10|V_1\frac{1}{\varepsilon_2^- - E_{\rm dip} - H^{(1)}}|0k\rangle. \tag{14}$$

That leads to products of terms each containing $H^{(1)}$ in the denominator. We neglect the correlations between eigenvalues of $H^{(1)}$ in different factors because such correlations extend over an energy range measured in units of the mean level spacing d while the range of the terms in Eq. (14) is given by $\Gamma^{\downarrow} \gg d$. For the last two terms in the second of Eq. (10) we obtain

$$\left\langle \sum_{k'}|b_{1k'}|^2 + \sum_{\rho}|b_{0\rho}|^2\right\rangle = \frac{1}{2}P_1^2 \tag{15}$$

with P_1 given by Eq. (13). The calculation of the first two terms yields a contribution that in comparison to Eq. (15) is small of order $\sigma/\Gamma^{\downarrow}$. Thus for all values of E_L the contribution to P_2 from double excitation of the dipole mode is negligibly small in comparison with that from the Brink-Axel mechanism in Eq. (15). As a result we find

$$P_2 = \tfrac{1}{2}P_1^2. \tag{16}$$

The factor $1/2$ in Eq. (16) is due to the time ordering in Eqs. (9). Thus, we expect that for arbitrary positive integer n we have $P_n = 2^{-n}P_1^n$.

Conclusions.—We have established the time scales and the resulting values for mean photon energy E_L and mean photon number N required for the realization of either of the two scenarios mentioned in the Introduction. This was done with the help of a random-matrix model for scenario (ii) for which we have calculated the probabilities P_1 and P_2 for single and double nuclear dipole absorption. Our assumptions and approximations require both P_1 and P_2 to be small compared to unity. In that case, scenario (ii) applies. Equation (13) shows that in the tails of the GDR that condition is easily met even for an intense laser pulse. Ways of detecting such collective nuclear excitation experimentally are discussed in Ref. [12]. Double photon absorption is dominantly due to the Brink-Axel mechanism (as opposed to double excitation of the dipole mode). For P_1 small compared to unity, single-photon absorption is the dominant mechanism while $P_2 \ll P_1$.

With increasing N, the time for dipole absorption $\tau_{\rm dip}$ eventually becomes small compared to the nuclear equilibration time $\tau_{\rm eq}$, and the competition between collective excitation and the formation of a strongly interacting nucleon plasma is decided in favor of the latter. Equation (13) shows that in the center of the GDR ($E_L \approx E_{\rm dip}$), that will happen already for fairly small values of $N \approx 10$ or so. As N is increased, the process spreads to the tails of the GDR. It is a challenge to attain a theoretical understanding of scenario (i), and of the interplay between both scenarios.

I thank P. Thirolf for valuable advice, D. Habs for stimulating discussions, and A. Richter and B. Dietz for helpful suggestions.

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