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ITERATIVE MAPS FOR BISTABLE EXCITATION OF TWO LEVEL SYSTEMS

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ABSTRACT

Iterative maps on SO(3) with two stable fixed points are described. These generate bistable spectroscopic excitation sequences for isolated two level systems. From such sequences, tailored population inversion over specific ranges of parameters such as the resonance frequency or radiation amplitude can be obtained. The ideas developed here suggest ways of designing tailored excitation sequences useful in spatially selective NMR, spin-decoupling, nk-quantum selective multiple quantum NMR, and isotope selective zero field NMR.

Excitation sequences have recently been developed for nuclear magnetic resonance (NMR) and optical spectroscopy which are effective over very broad[1-8] or very narrow[6,9] ranges of transition frequencies and radiation amplitudes. Of the methods conceived for deriving these sequences, an iterative approach based on the use of sequence-refining algorithms has proven particularly useful. Treating these algorithms as iterative maps[10-12], it has been shown that stable fixed points lead to sequences with broadband properties, while unstable fixed points produce sequences which exhibit narrowband properties[8]. This Letter reports the first example of iterative maps for pulse sequences with more than one stable fixed point. From such maps, sequences for excitation over sharply defined, pre-selected ranges of parameters, e.g., frequencies or amplitudes, can be obtained. This provides the first experimental approach to tailoring the bandwidth of some nonlinear spectroscopic response, a long-desired goal in the excitation of spin and optical systems. The implementation of this passband response has potential use in a variety of applications in spectroscopy, including spatially selective NMR[13], selective n-quantum pumping of multiple quantum transitions[14], heteronuclear zero field NMR[15], and optical information storage[16].

The sequences demonstrated here selectively invert the equilibrium populations of uncoupled two-level systems (e.g., isolated spins-1/2) depending on the amplitude (commonly denoted ω_1) of the resonant radiofrequency (rf) radiation at the nuclear spin position. This general technique can, in fact, be used to select one or several discrete ranges of rf amplitudes for specific excitation.

For the analysis of these sequences, we employ a formalism drawn from the theory of iterative maps and their fixed points[10]. The effect of a pulse sequence on some system is represented by its time development operator, or propagator, U. Usually a specific propagator $\bar{U}$ is desired, for example, one which corresponds to an inversion of the equilibrium populations. Iterative schemes have been proposed as a way to achieve this end. These are essentially algorithms which prescribe the transformation F that must be performed on a pulse sequence for its propagator U to converge to the specifically desired form $\bar{U}$. This iterative procedure can be summarized by the equation:

$$U_{n+1} = FU_n \qquad (1)$$

Functions which can be applied iteratively in this manner are referred to as maps. The dynamics of iterative maps are influenced by their fixed points[10-12], which are defined by the relation:

$$\bar{U} = F(\bar{U}) \qquad (2)$$

The consequences of fixed points and their stability on the behaviour of iterative pulse sequence maps have already been discussed in detail[8]. Briefly, it was shown that pulse sequence algorithms could be considered as maps on a quantum-mechanical propagator space, with fixed points corresponding to the desired propagators $\bar{U}$.

In the absence of couplings, any propagator U, including the effects of pulse sequences, is describable as a simple rotation of the spin density operator[17,18]. It follows that only a subspace of the entire propagator space need be considered in the analysis, namely the space of pure rotations, commonly designated SO(3)[19], which can be visualized as a solid sphere of radius π. All rotations are uniquely defined in this space by a unit vector drawn from the origin (corresponding to the axis of the rotation) and a radius (corresponding to the angle of the rotation). All π rotations are doubly defined by antipodal points on the surface of the sphere.

In general, the convergence of a map to its fixed points depends on the initial condition U_o and the stability of the fixed points in various directions. The initial condition U_o can itself be a function of several parameters, designated here as λ_i, such as the resonance frequency or the rf amplitude ω_1. In devising a broadband sequence, e.g., a broadband inverting sequence, the objective is to make a single fixed point stable for as wide a range of the parameter λ_i as possible. For narrowband sequences, the aim is to specify a single fixed point which is unstable over the parameter λ_i. For the present case of bistable passband sequences, however, two stable fixed points are required, so that for some values of λ_i, U converges to one fixed point $\bar{U}_1$, while for other values of λ_i, U converges to the other fixed point $\bar{U}_2$.

Pulse sequences which excite a passband population inversion in ω_1 can be obtained from maps which have the origin and the equator of SO(3) in the xy plane as stable fixed sets of points. These points correspond to the identity operator and the set of π rotations which take +z to -z, respectively. In addition, the fixed points should also be stable with respect to displacements in the xy plane. The significance of this bistability can be appreciated by referring to figure 1. This figure shows schematically that maps with the origin and the equator of SO(3) as fixed sets stable in the xy plane necessarily possess an unstable circle of points also in the xy plane. Points in SO(3) inside this circle move towards the origin upon iteration of the algorithm, while points outside the circle move towards the equator.

Two phase shift algorithms derived to satisfy these stability conditions are:

(a) [0, 270, 120, 165, 120, 270, 0]

(b) [0, 15, 180, 165, 270, 165, 180, 15, 0]

Following the notation of reference 8, these algorithms are comprised of two basic operations, a series of phase shifts, shown in the brackets, followed by concatenation of the phase shifted parts. Figure 2 depicts the basin images[8] of the two algorithms. The basin image shows how many iterations are required for points in SO(3) to be mapped into the two known stable fixed sets. The regions of SO(3) that are convergent to the fixed sets are known collectively as the basin of the map, and appear as the light areas of the image. The images in this instance are cross-sections of SO(3) containing the z axis. Since algorithms comprised of simple phase shifts exhibit axial symmetry around the z axis, no information is lost in displaying a single cross-section.

The most conspicuous difference between the two images displayed is the size of their respective basins. The reason for this is that the nine shift algorithm (b) was designed so that the equator would be stable in all directions. The seven shift algorithm (a), however, is not stable at the equator for points off the xy plane. The basin for the nine shift algorithm is therefore larger and exhibits a much more intricate structure than does the seven shift sequence. A consequence of this additional stability is that the nine shift algorithm is broadband over resonance frequencies.

The implications of applying an algorithm described by a bistable map to a single nominal π pulse can be understood by referring to figure 3. This figure shows that such algorithms, in this case the 7 shift algorithm, produce sequences which display distinctive passband characteristics in the ω_1 domain. This implies that only spins that lie within specified bandwidths in ω_1 will be inverted by these sequences. Spins outside these bands remain in their initial equilibrium state. The result is a pulse sequence which is highly amplitude selective in inverting nuclear spins. Moreover, as the algorithm is iterated, the passband becomes sharper and more pronounced, indicating the refinement of the sequence by iteration. The experimental points on this curve demonstrate the practical feasibility of these algorithms for obtaining ω_1 selective inversion of the magnetization. Generalized tailoring of the population inversion can be achieved across a range of ω_1 values with the appropriate choice of an initial sequence. Sequences which favor certain basins in SO(3), or cross from one basin to another, create this kind of tailored response.

The origin of the passband behaviour is the bistability of the map. The effect of iterating this algorithm on some rotation R_{xy} around an axis in the xy plane is to transform R_{xy} into either the identity operator or a z inverting rotation. The only two responses to the radiation for a nuclear spin, then, are no response, or inversion. This binary response function manifests itself in the sharp passband behaviour of the I_z vs. ω_1 plots of figure 3.

The fixed point analysis presented here has provided guidance in deriving sequences with unusual and useful spin excitation properties. The perspective and insight this method offers furnish mathematical tools for the handling of complex nonlinear problems. Extensions of this approach are currently being undertaken for iterative mappings with multiple fixed points for general nonlinear excitation.

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FIGURE CAPTIONS

Figure 1: Cross-section of SO(3) through the xy plane indicating the movement of points in this plane as an algorithm designed to produce a bistable bandpass sequence is iterated. The origin and equator are stable fixed sets of this mapping. This entails the existence of an unstable fixed circle between the two stable fixed sets in the xy plane. The position of the unstable set defines the effective bandwidth of the excitation. Points initially in the xy plane remain in this plane even after iteration of the algorithm as a result of the symmetry of the sequence.

Figure 2: Basin images for the maps generated by the algorithms [0, 270, 120, 165, 120, 270, 0] and [0, 15, 180, 165, 270, 165, 180, 15, 0] showing a cross-section of SO(3) containing the z axis. The number of iterations of the algorithm required to map a point in SO(3) to one of the two known stable fixed points is given by the color key to the left of the image. A substantial portion of the space for the nine shift algorithm is convergent because of its stability in the z direction, in contrast with the basin image for the seven shift algorithm.

Figure 3: Extent of nuclear spin population inversion as a function of normalized on-resonance rf field amplitude for a single π pulse (curve 1), one iteration of the indicated algorithm (curve 2), and two iterations of the algorithm (curve 3). -1 on the y axis denotes the normalized equilibrium spin population (bulk magnetization aligned with the magnetic field), +1 denotes the normalized population inverted state (magnetization antiparallel to the magnetic field). The effect of iteration is seen to sharpen the passband of

the response function. Such a bistable response can be tailored to different regions of ω_1. Experimental data were obtained from the proton resonance of a distilled water sample at a Larmor frequency of 180 Mhz.

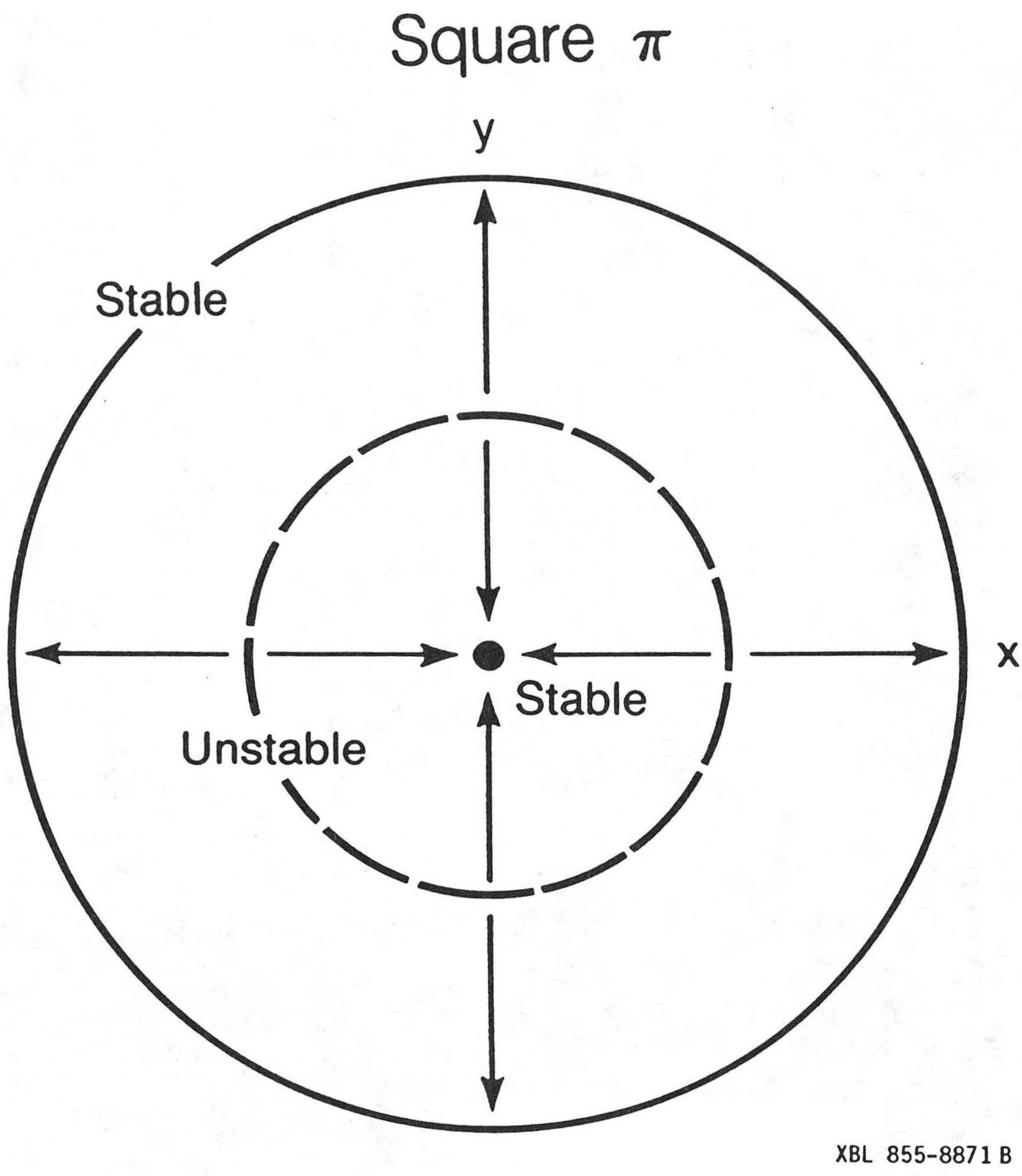

Fig. 1, Cho, et al.

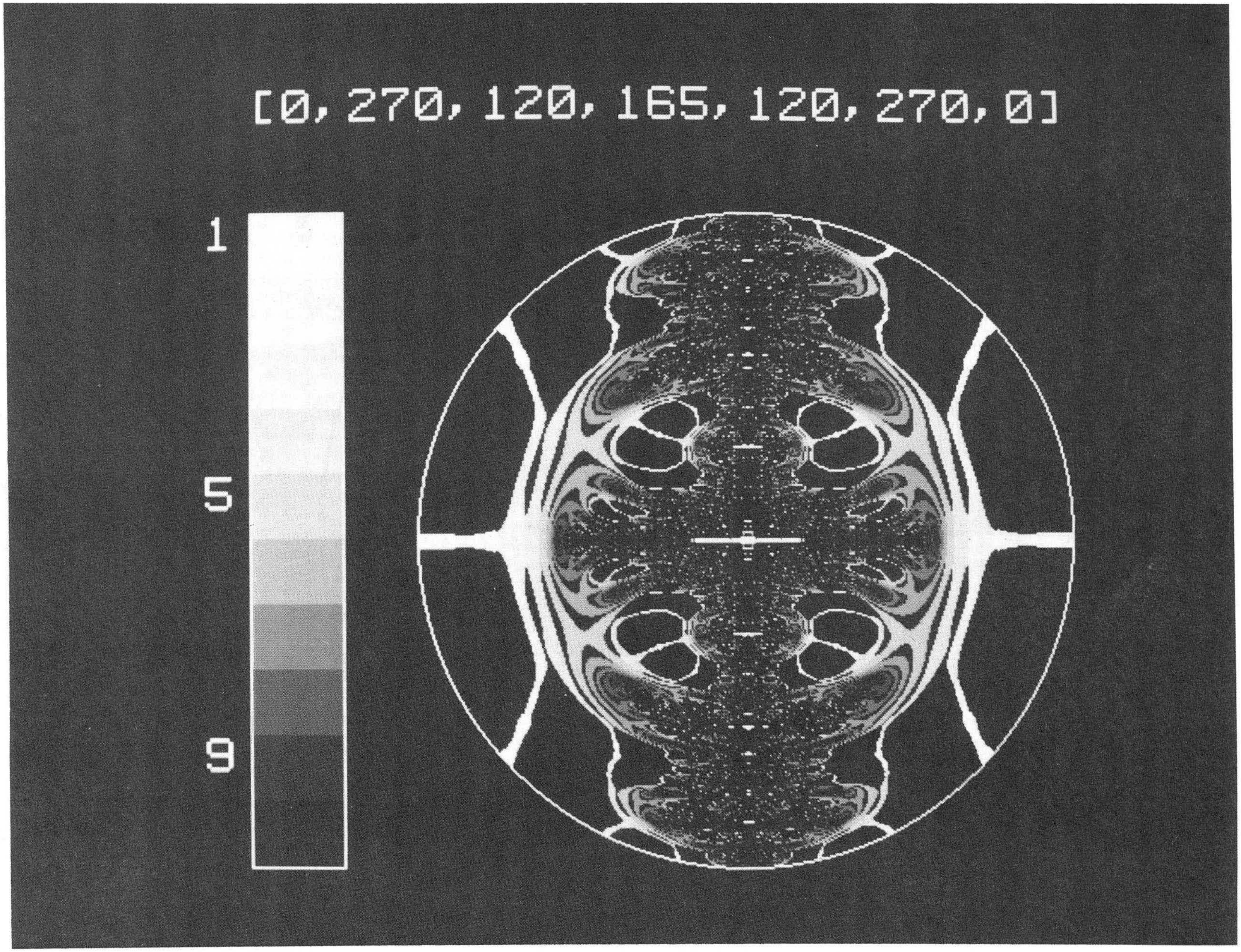

Fig. 2a, Cho, et al.

CBB 861-523

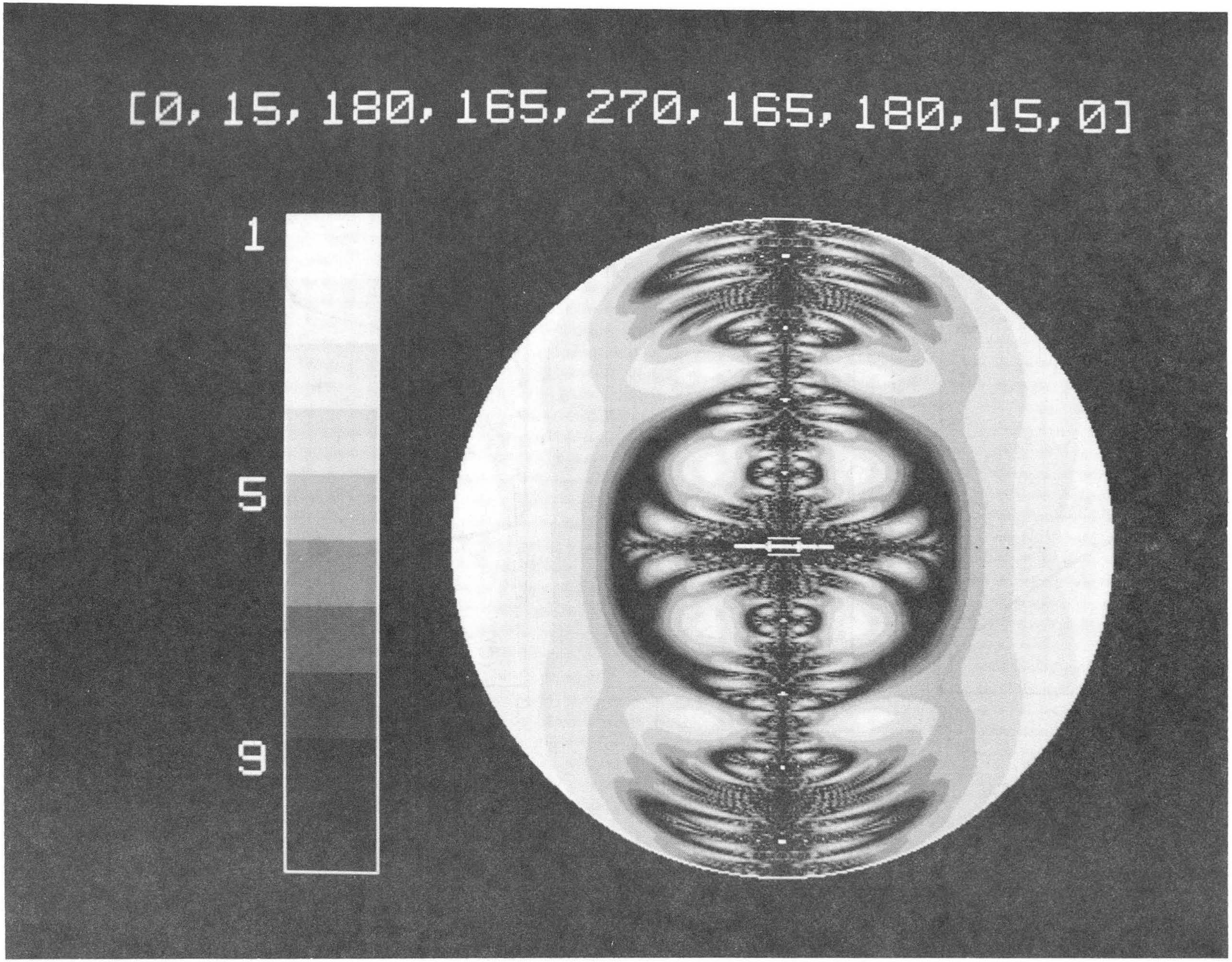

Fig. 2b, Cho, et al.

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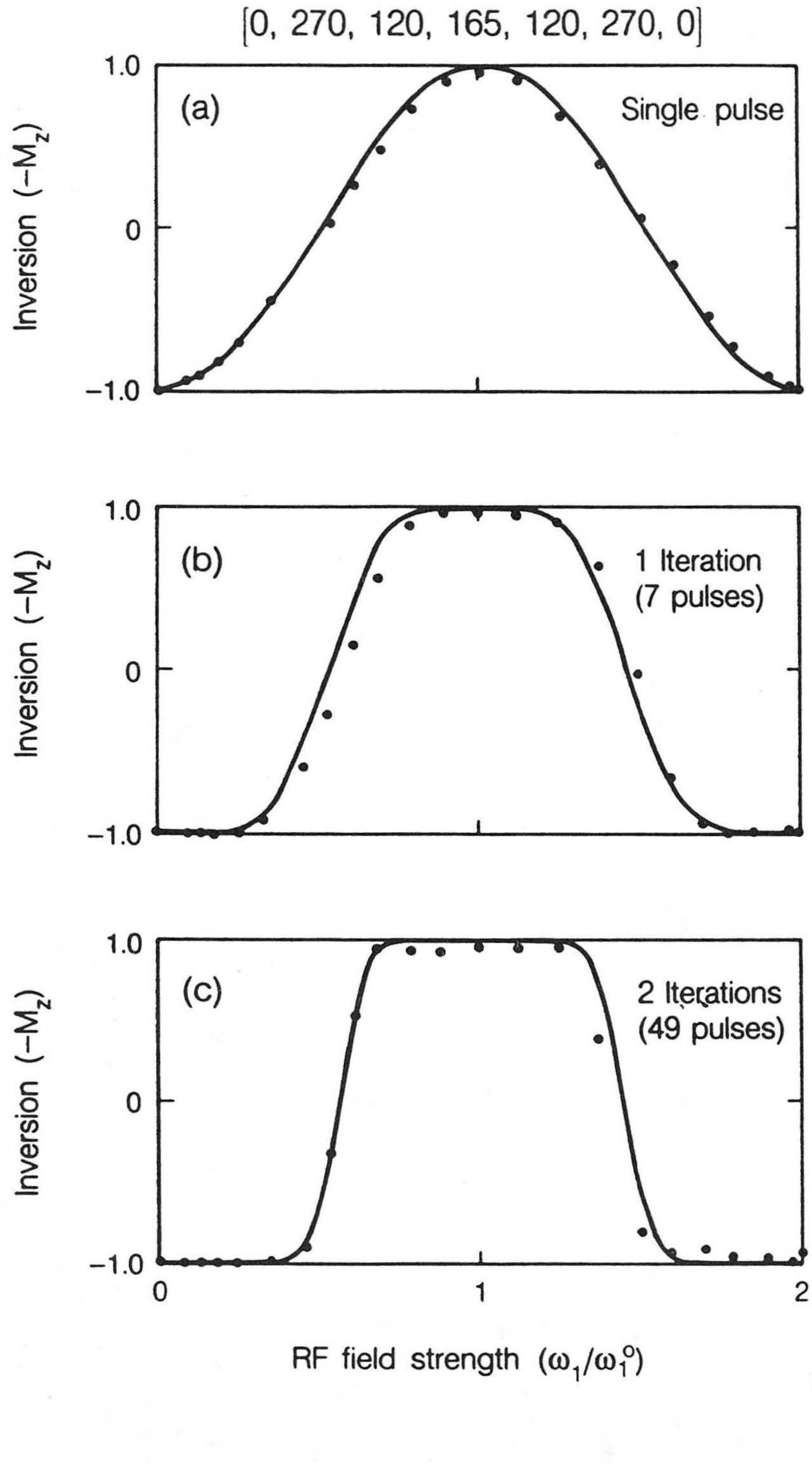

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Fig. 3, Cho, et al.

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