Spin waves in boron-implanted polycrystalline diamond films

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Spin waves have been detected for the first time in nonmetallic systems lacking a magnetic order: in quasi-1D carbon structures. These waves are observed at room temperature. They are caused by paramagnetic topological solitons. Oriented quasi-1D structures were fabricated through ion implantation in diamond films.

Reduced-dimensionality systems are attracting increasing scientific and practical interest. Theoretical work on the electronic structures of the reduced-dimensionality systems which have been studied most thoroughly, such as polyacetylene and polyyne (a 1D allotropic form of carbon),[1,2] has shown that these entities exhibit several properties which are quite different from the properties of 3D systems. For example, in the 1D structures based on carbon, in contrast with diamond, a 3D modification of this material, it is a simple matter to arrange conditions for the formation of solitons.[1,2] The existence of solitons in these structures agrees well with a series of magnetoresonance experiments on polyacetylene, e.g., those of Ref. 3, corresponding experiments on oriented samples of polyyne,[4] and ESR studies of quasi-1D structures formed in diamond and diamond films[5,6] by ion implantation (a modification effect, i.e., the formation of quasi-1D ordered carbon structures under certain implantation conditions, was observed in diamond in Ref. 5). It follows from Ref. 7 that the magnitude and nature of the electron-electron correlations play a leading role in the realization of the specific type of electronic structure of 1D carbon systems, i.e., in the realizations of states which represent either coupling-order waves, spin density waves, or charge density waves.

In the present letter we are reporting the first observation of electronic excitations of a new type in quasi-1D carbon systems. These new excitations are a direct consequence of electron-electron correlations, namely, spin waves coupled with paramagnetic topological solitons. As the quasi-1D systems we used some modified structures formed in implanted layers of polycrystalline diamond films. The diamond films, 0.5 μm thick, were grown by CVD. The boron implantation (150 keV, 10^{16} cm^{-2}) was carried out in the direction perpendicular to the film surface. The ESR measurements were taken on a Bruker BE-420 spectrometer at room temperature.

In the ESR spectrum of the original polycrystalline diamond film we see a low-intensity singlet isotropic line with a g-factor of 2.0027 and a half-width of 6.2 G. Figure 1 shows ESR spectra observed in this film after the boron implantation. The

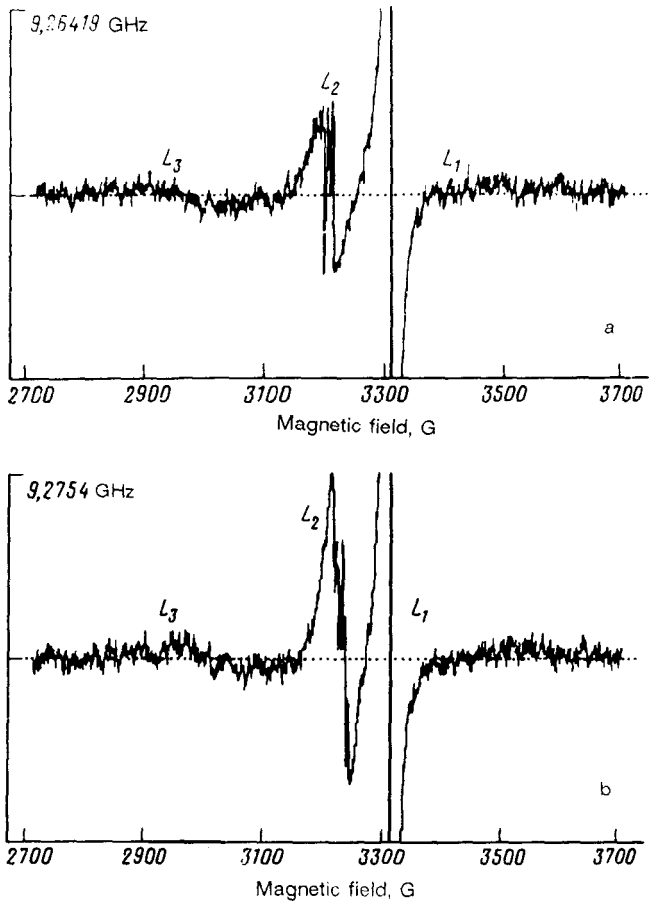

FIG. 1. ESR spectra of a polycrystalline diamond film after implantation of boron (150 keV) in a magnetic field **H**. The field is (a) parallel to and (b) perpendicular to the film surface.

spectrum consists of three comparatively broad, intense lines, L_1, L_2, and L_3, along with several relatively narrow lines which overlap L_2. A study of the saturation of the resonance spectrum revealed that lines L_1, L_2, and L_3 belong to the same magnetic system. At the same time, the behavior of the widths of the components in this series is characteristic of spin waves. Specifically, line L_1, which is furthest to the right in the spectrum in Fig. 1, can be thought of as an analog of the $n=0$ mode of the spin-wave signal in a ferromagnetic film.[8] Lines L_2 and L_3 correspond to the $n=3$ and 5 modes. The positions of the lines in the resonance spectrum are also described by quadratic dispersion relations like the Kittel relation for ferromagnetic metal films:

$$\omega = Ak^2 + \omega_0, \tag{1}$$

$$k = n\pi/d. \tag{2}$$

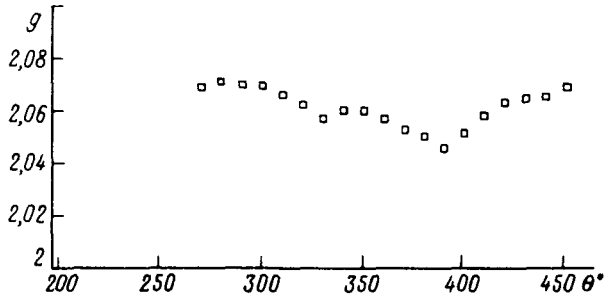

FIG. 2. Angular profile of the effective g-factor of line L_2. The angle $\theta = 10°$ corresponds to the case in which **H** is perpendicular to the film surface.

Here d is the effective thickness of the structure created by the incident ion, n is a quantum number, and the coefficient A characterizes the electron-electron interaction.

From expressions (1) and (2) we find the ratio A/d^2. It turns out to have values of 2.1×10^7 and 1.6×10^7 s^{-1} for magnetic fields directed respectively parallel and perpendicular to the surface of the film. Further evidence that L_1, L_2, and L_3 are spin-wave modes comes from the angular profile of their effective g-factor. This profile is similar to that of the linewidth observed during high-energy implantation of neon in diamond single crystals.[5] This result is understandable, since the linewidth[5] and the position of the modes of a spin-wave resonance are determined by the same mechanism, namely, electron-electron correlations.

There is an important difference between the spin waves observed in these experiments and those observed in alkali metals and ferromagnetic materials. Spin waves in alkali metals differ from those which we observed in that they can be detected only at very low temperatures, and their resonance spectrum consists of modes of alternating polarity.[9] The line with $n=0$ in the spin-wave spectrum of a ferromagnetic film is a ferromagnetic-resonance line. The value of the corresponding resonance field is highly anisotropic when the sample is not spherical.[10] This situation is quite different from the nearly isotropic value of the resonant field for the L_1 line which we observed (Fig. 1). In addition, the intensities of spin modes in both alkali metals and ferromagnetic films decrease markedly with increasing quantum number n. On the other hand, as was shown in Ref. 6, the resonance line L_1 can be attributed to paramagnetic solitons. The results found here thus suggest that the spin waves appear because of the presence of solitons which are spin carriers with an extended distribution of spin density. The motion of the solitons and the propagation of the spin waves are interrelated. Since solitons can transport spins or charges with essentially no loss, the intensity of the spin-wave modes with various quantum numbers should also be essentially the same. Indeed, an estimate of the concentrations showed that the value of $V\Delta H_{pp}^2$ (V is an amplitude) is, within the measurement error, the same for the lines studied, and the overall effective spin concentration is estimated to be $\approx 10^{22}$ cm^{-3}. The fact that a system of paramagnetic solitons can be a source of spin waves in the presence of electron-electron correlations means that we can also explain the very high effective concentration of the spins participating in the resonant absorption: In addition to the energy transfer out of the spin system to the lattice through spin-lattice relaxation, there is a rapid direct conversion of the energy absorbed by the spins of the solitons

into kinetic energy of solitons,[4–6] because of the gyromagnetic coupling of their spin and mechanical moments under conditions of a very small activation energy [0.002 eV (Ref. 1)] for the motion of solitons. The time scale of this process would be expected to be comparable to the relaxation time of the electron kinetic energy, i.e., $\sim 10^{-13}$ s.[11] Consequently, a given soliton can absorb many times in the course of spin-lattice relaxation; the natural result is a high effective concentration of spins. Note that this mechanism agrees well with the experimentally observed times $\sim 10^{-13}$ s (Ref. 3) for a 1D "diffusion" of the energy of spins in a system of solitons in polyacetylene. (The relaxation times set by a different mechanism for a magnetic interaction should be no shorter than 10^{-10} s.[11])

The possibility of simultaneously observing nonlinear properties (solitons) and linear properties (spin waves) of quasi-1D carbon chains can be explained in the following way. For 1D carbon chains, Peierls and Mott-Hubbard metal-insulator transitions are favored from the energy standpoint.[2,7,12–14] There is a region of values of the parameters of the electron-electron correlations in which the two transitions can be observed simultaneously.[13,14] As a result of a Peierls transition, there is an alternation of bond lengths, which is the physical basis for the formation of topological solitons. In the case of a Mott–Hubbard transition, on the other hand, the conditions favor the appearance and propagation of spin waves. The appearance of a gap in the energy spectrum of a carbon chain as a result of these transitions and the deep energy state of this chain corresponding to the presence of a soliton make it possible to understand why spin waves can be observed at room temperature.

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