EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

QUADRUPOLE INFLUENCE ON THE DIPOLAR FIELD WIDTH FOR A SINGLE INTERSTITIAL IN A METAL CRYSTAL

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ABSTRACT

The dipolar broadening of the magnetic field sensed by an interstitial impurity in a rigid lattice is calculated with the electric field gradient set up by the impurity taken into account. This is shown to give a strong dependence of the dipolar width on the applied magnetic field. The theory is especially applicable to the line width of precessing muons in metals.

Geneva - 25 April 1977

(Submitted to Phys. Rev. Letters)

The broadening, due to dipolar coupling, of magnetic resonance lines of nuclear spins I in solids was derived in 1948 by Van Vleck[1]:

$$\overline{\Delta\omega_I^2} = \frac{3}{4}\gamma_I^4 \hbar^2 I(I+1) \sum \frac{(3\cos^2\theta - 1)^2}{r^6} \qquad (1)$$

for the broadening due to like spins, and

$$\overline{\Delta\omega_I^2} = \frac{1}{3}\gamma_I^2 \gamma_S^2 \hbar^2 S(S+1) \sum \frac{(3\cos^2\theta - 1)^2}{r^6} \qquad (2)$$

for the broadening due to unlike spins, i.e. gyromagnetic ratios $\gamma_I \neq \gamma_S$. The equations are valid if the spins are subject to a static magnetic field $B_0 = B_z$ substantially larger than the dipolar fields, which are typically of the order of 1 G for nuclear spins in solids.

In Eq. (2), spin-flip terms of the type I_+S_- are absent, and it can be considered as the random sum of the dipolar fields from all the spins S at the site of spin I:

$$\overline{\Delta B_z^2} = \frac{1}{3}\gamma_S^2 \hbar^2 S(S+1) \sum \frac{(3\cos^2\theta - 1)^2}{r^6}\,, \qquad (3)$$

where ΔB_z is the contribution from each spin S to the field along the external field B_0, r is the vector from spin S to spin I, and θ the angle between r and B_0. In a single crystal the sum in Eq. (3) will depend on the orientation of the crystal with respect to the external magnetic field B_0.

If Eq. (3) is used to calculate the field distribution sensed by an interstitial in a metal, e.g. a positive muon, certain care must be taken. The muon acts as a charged impurity in the lattice and creates an electric field gradient at the nearby host nuclei. If these nuclei possess a quadrupole moment, then the magnetic field B_0 is no longer a unique quantization axis. In these circumstances Eq. (3) is no longer valid, and the orientational dependence of the dipolar width is drastically changed. In this case the dipolar fields from the lattice nuclear spins S at the position of the interstitial spin I have to be calculated with the spins S being subject to a combined electric and magnetic interaction.

The general behaviour of the nuclear spins S under the influence of a static magnetic field B_0 and an electric field gradient $V_{z'z'}$, in general not parallel with B_0, can be calculated by transforming the electric interaction to the coordinate system defined by B_0 [2]. The diagonalization of the resultant Hamiltonian matrix of order $2S+1$ gives the energies E_k and eigenfunctions $\phi_k = \sum c_m u_m$ for the stationary states, where the basic functions u_m represent the $2S+1$

m-states for spin S. Alternatively, the magnetic interaction can be transformed[3]. The components of the magnetic moment of spin S are obtained from the expectation values of the spin operators, i.e. $M_z = \gamma_S \hbar \langle\psi|S_z|\psi\rangle$ with $\psi = \sum_k \phi_k$.

a) $V_{z'z'} = 0$

The basic functions u_m are also eigenfunctions, and the spin S will have a static z-component in each of the 2S + 1 eigenstates:

$$\langle S_z \rangle_m = m \ , \tag{4}$$

while the components in the xy-plane oscillate with the Larmor frequency:

$$\langle S_x \rangle = \sum c_i^* c_{i'} \langle m|S_x|m'\rangle e^{-i/\hbar(E_{m'} - E_m)t} \ . \tag{5}$$

Oscillating components from Eq. (5) cannot influence spin I with its different Larmor frequency, but the static z-components of the magnetic moment give rise to a dipolar field at the site of spin I:

$$\Delta B_z = \gamma_S \hbar \langle S_z \rangle \frac{3\cos^2\theta - 1}{r^3} \ . \tag{6}$$

By noting that

$$\overline{\langle S_z \rangle^2} = \frac{1}{3} S(S+1) \ ,$$

we obtain the random contribution from spin S:

$$\overline{\Delta B_z^2} = \frac{1}{3} \gamma_S^2 \hbar^2 S(S+1) \frac{(3\cos^2\theta - 1)^2}{r^6} \ . \tag{7}$$

b) $V_{z'z'}$ making an angle θ with B_0

We consider now the case where the electric field gradient $V_{z'z'}$ created by the interstitial is radially directed and axially symmetric. The stationary states of the lattice nuclei S described by the eigenvectors $\phi_k^\circ = \sum c_m u_m$ now have static z-components given by

$$\langle S_z \rangle_k^{stat} = \sum c_i^* c_i m_i \ , \tag{8}$$

but also static x-components

$$\langle S_x \rangle_k^{stat} = \sum c_i^* c_j \langle u_i|S_x|u_j\rangle \tag{9}$$

due to the mixture of m states in each eigenstate. There will also be a number of oscillating x- and z-components containing $\exp[-i/\hbar\,(E_{k'} - E_k)t]$. The fluctuating fields can be neglected as long as they do not coincide with the Larmor frequency of spin I, and we will have a resultant static contribution to the field:

$$\Delta B_z = \gamma_S \hbar \langle S_z \rangle_k^{stat} \frac{3\cos^2\theta - 1}{r^3} + \gamma_S \hbar \langle S_x \rangle_k^{stat} \frac{3\sin\theta\cos\theta}{r^3}, \qquad (10)$$

with $\langle S_z \rangle_k^{stat}$ and $\langle S_x \rangle_k^{stat}$ for each eigenstate given by Eqs. (8) and (9), and the x-axis chosen in the plane containing r and B_0.

RESULTS

The dipolar broadening at interstitial points in fcc and bcc lattices have been calculated as a function of the relative interaction strength $y = \omega_B/\omega_E$. ω_B is the Larmor frequency of the lattice nuclei determined by $w_B = \gamma_S B_0$, and ω_E the electric interaction frequency given by $\omega_E = (eQ/\hbar)\{V_{z'z'}/[4S(2S-1)]\}$. The parameter b shown in Figs. 1 to 5 is the r.m.s. sum of the contributions in Eq. (10) with $\overline{\Delta B^2} = b^2\gamma_S^2\hbar^2/a^6$, where a is the lattice constant. In general about 70 lattice points around the interstitial were included.

The field gradient has been assumed to be radially directed from the interstitial and to decrease with r^3. This choice is not critical as the major part of the sums in Eq. (10) arises from the nearest neighbours. The scale has been chosen such that the parameter $y = \omega_B/\omega_E$ is the interaction strength at the distance $r = a/2$ from the interstitial site.

The derivation assumes that the frequencies of the oscillating components of the dipolar fields are separated from the interstitial's Larmor frequency; in the opposite case, certain resonance phenomena (spin flips) could occur. For the case of muons in copper $\gamma_\mu \approx 12\,\gamma_{Cu}$, and the distance of the nearest neighbour is $r = a/2$ (octahedral sites). The curves in Fig. 1 should then be valid for $\omega_B/\omega_E \gtrsim 0.5$.

Figures 3 and 4 show the situation for bcc lattices. With the field B_0 along the [100] or [110] direction, two inequivalent positions exist both in the tetrahedral and octahedral sites. Of these positions, one kind is twice as common as the other, and is indicated by II in the figures.

Figure 5 shows the dependence of the line broadening on the spin value S for fcc octahedral positions, [111]-direction. The general shape of the dependence on ω_B/ω_E is seen to be similar for the different spin values.

The line widths shown in Figs. 1 to 5 are what would be observed in a muon procession experiment at different fields B_0, assuming that the muon creates a radially directed electric field gradient. Strong effects from field gradients created by impurities (substitutional) have been seen by NMR methods in copper and aluminium (4,5). In fact the presence of electrical field gradients explains the experimental results on positive muon precession in copper, where different crystal orientations give almost equal line widths $\sigma = 2^{-1/2}\gamma_\mu \Delta B$ at low fields B_0 [6,7], and where the line width dependence at higher fields[7] shows a behaviour very similar to the curves in Fig. 1.

REFERENCES

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Figure captions

Fig. 1 : Calculated line widths b in fcc lattice, octahedral sites, as a function of the relative interaction strength ω_B/ω_E for different orientations of the external field B_0. Broken lines to the right indicate b values in the limit of large ω_B.

Fig. 2 : Calculated line widths b as in Fig. 1 for fcc lattice, tetrahedral sites.

Fig. 3 : Calculated line widths b as in Fig. 1 for bcc lattice, octahedral sites.

Fig. 4 : Calculated line widths b as in Fig. 1 for bcc lattice, tetrahedral sites.

Fig. 5 : Calculated line widths b as in Fig. 1 for fcc lattice, octahedral sites, for different values of spin S.

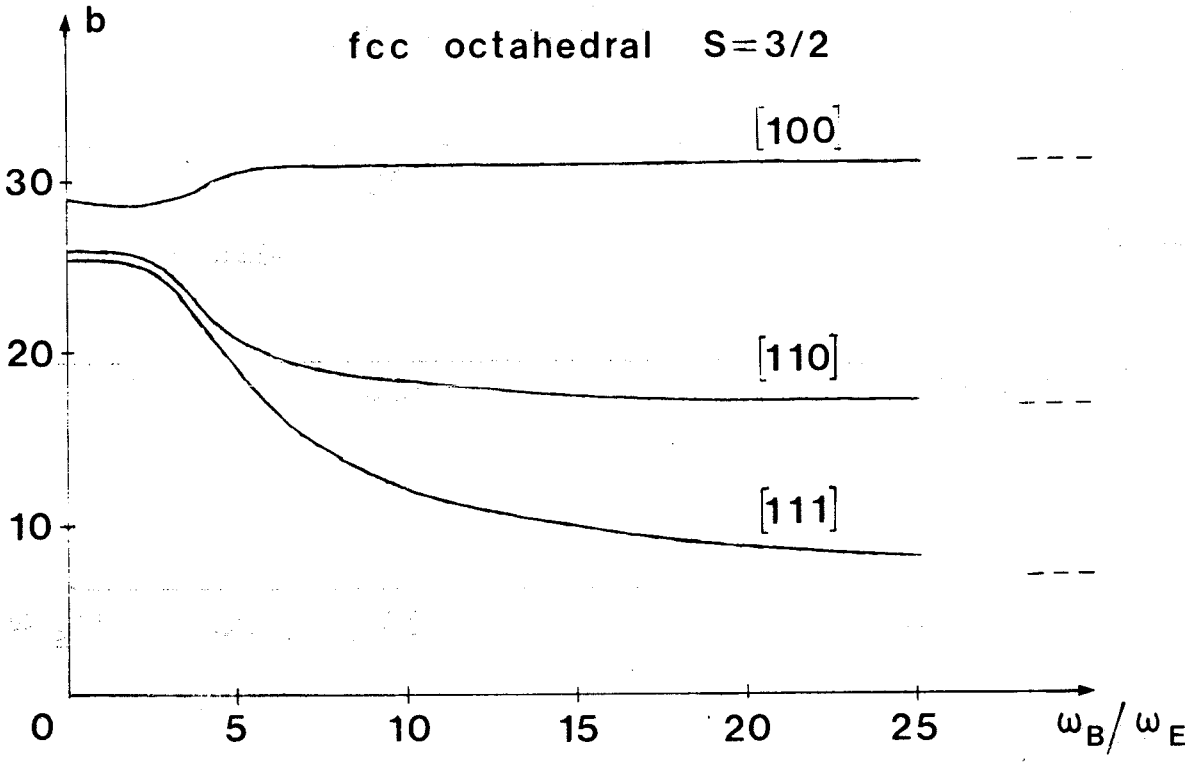
b
fcc octahedral S=3/2
[100]
[110]
[111]
30
20
10
0
5
10
15
20
25
ω_B/ω_E

Fig. 1

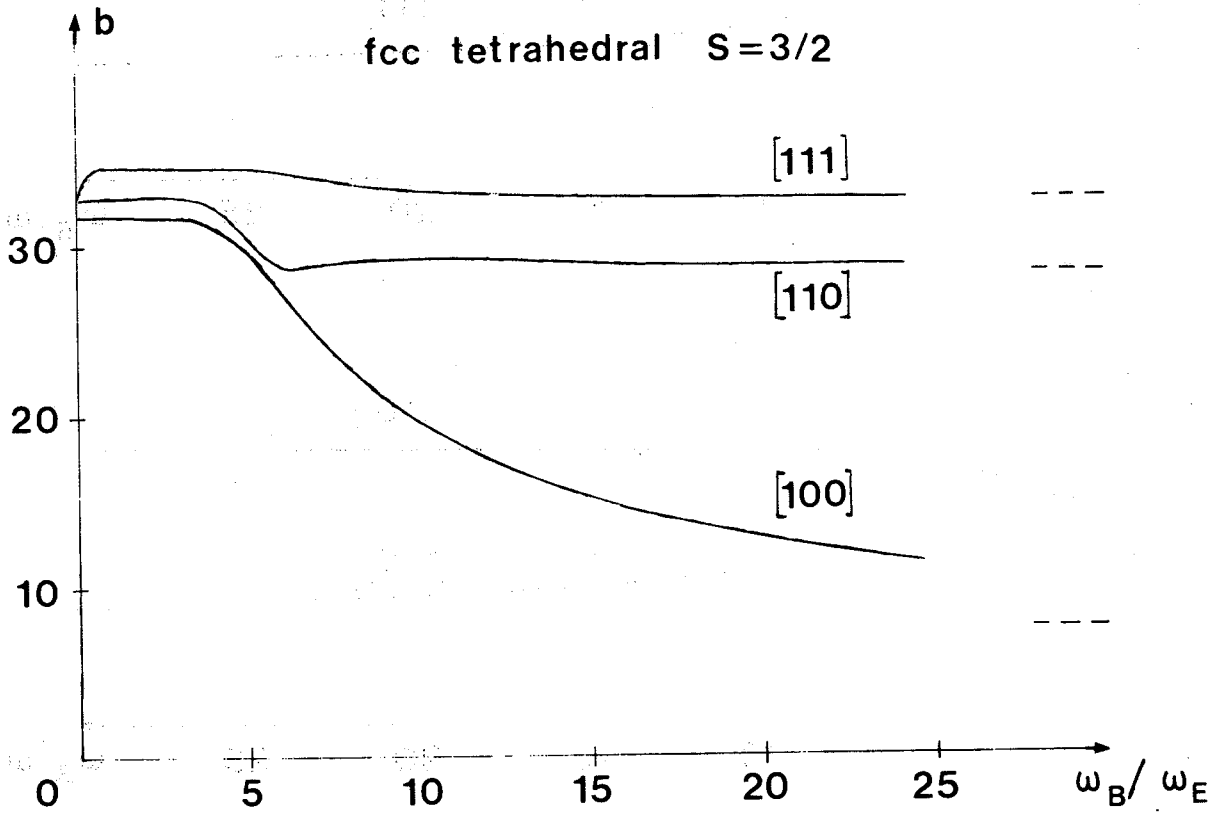
b
fcc tetrahedral S=3/2
[111]
[110]
[100]
30
20
10
0
5
10
15
20
25
ω_B/ω_E

Fig. 2

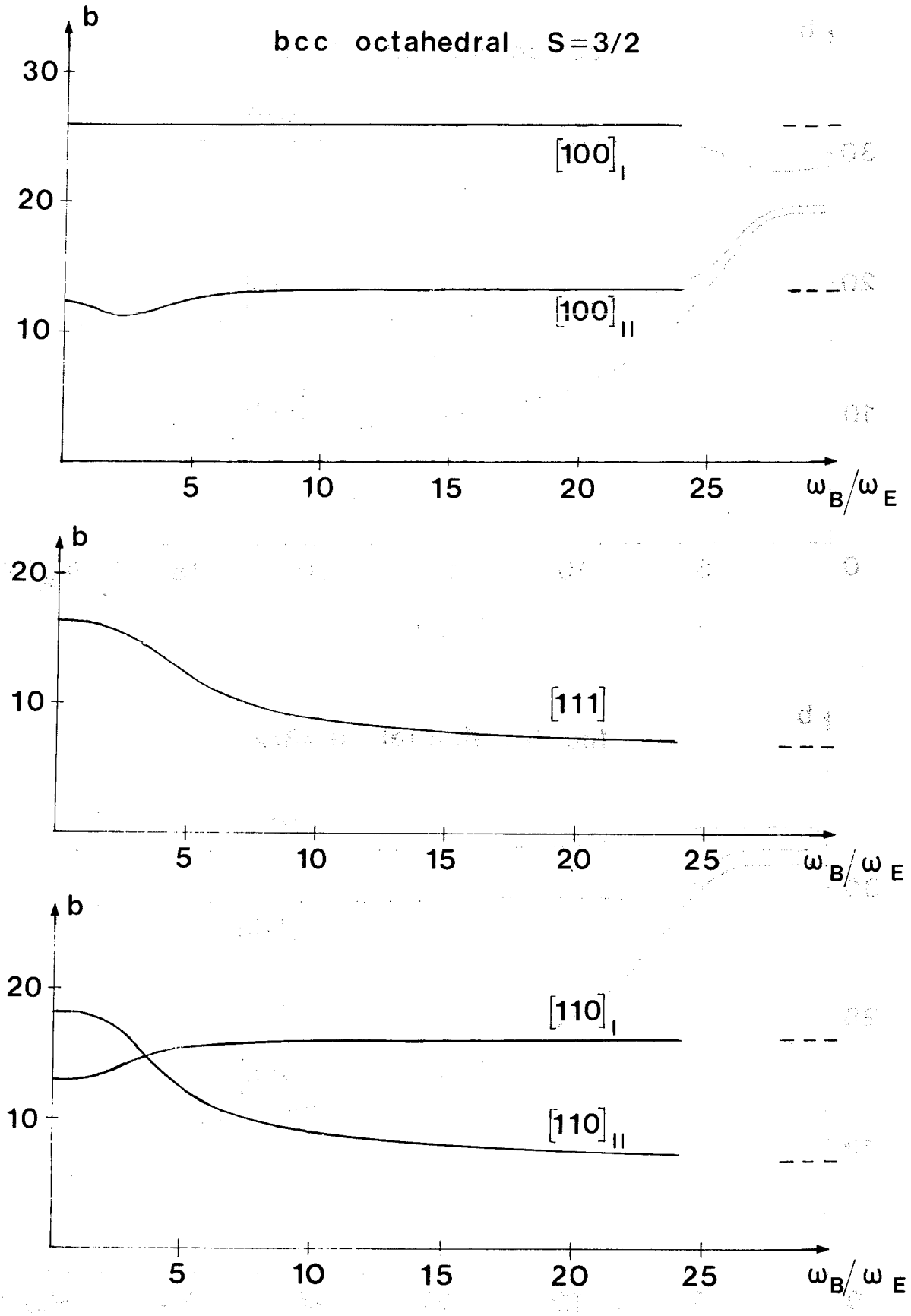
bcc octahedral S=3/2
b
30
20
10
[100]I
[100]II
5
10
15
20
25
ωB/ωE
b
20
10
[111]
5
10
15
20
25
ωB/ωE
b
20
10
[110]I
[110]II
5
10
15
20
25
ωB/ωE

Fig. 3

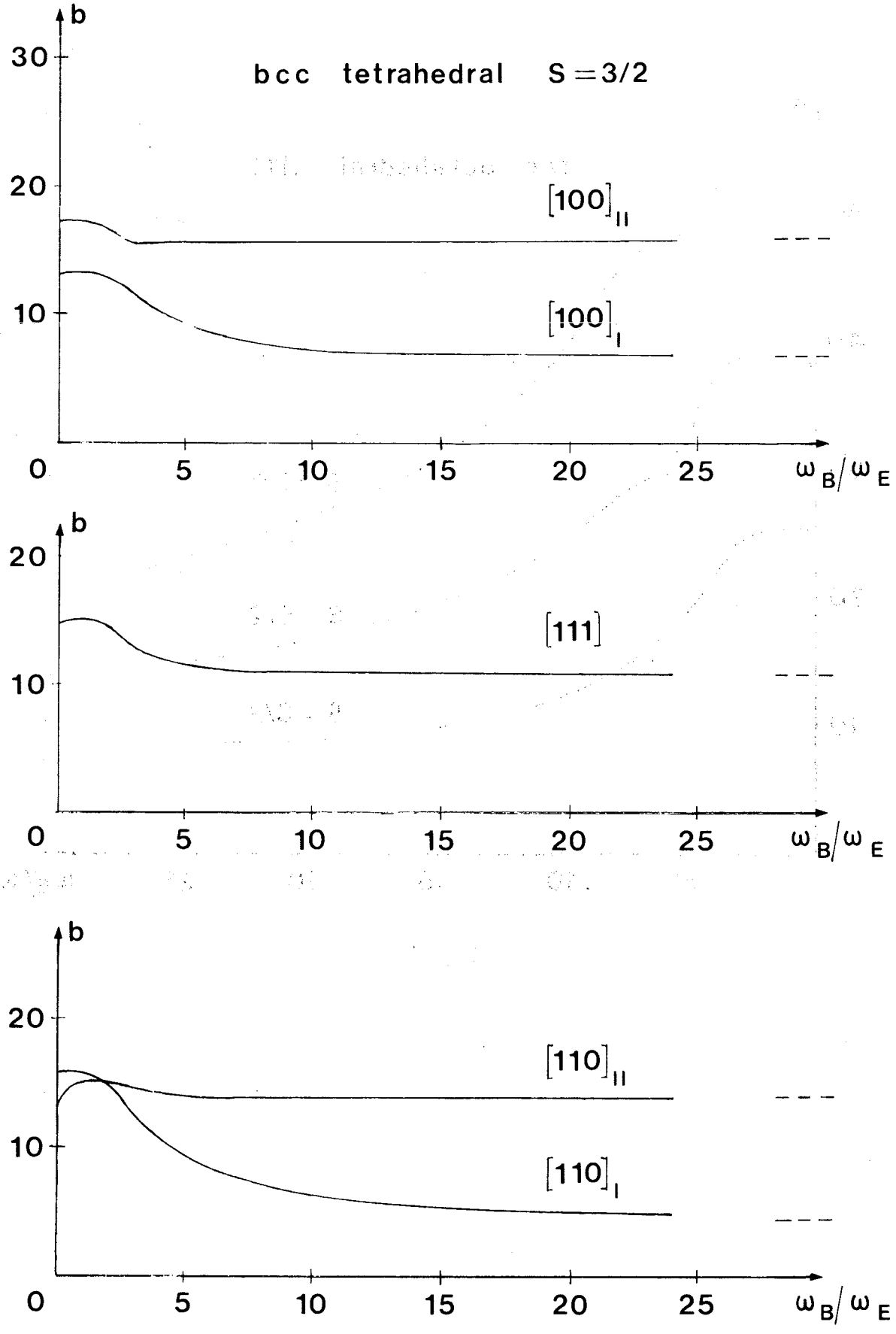
b
30
20
10
0
bcc tetrahedral S=3/2
[100]II
[100]I
5
10
15
20
25
ωB/ωE
b
20
10
0
[111]
5
10
15
20
25
ωB/ωE
b
20
10
0
[110]II
[110]I
5
10
15
20
25
ωB/ωE

Fig. 4

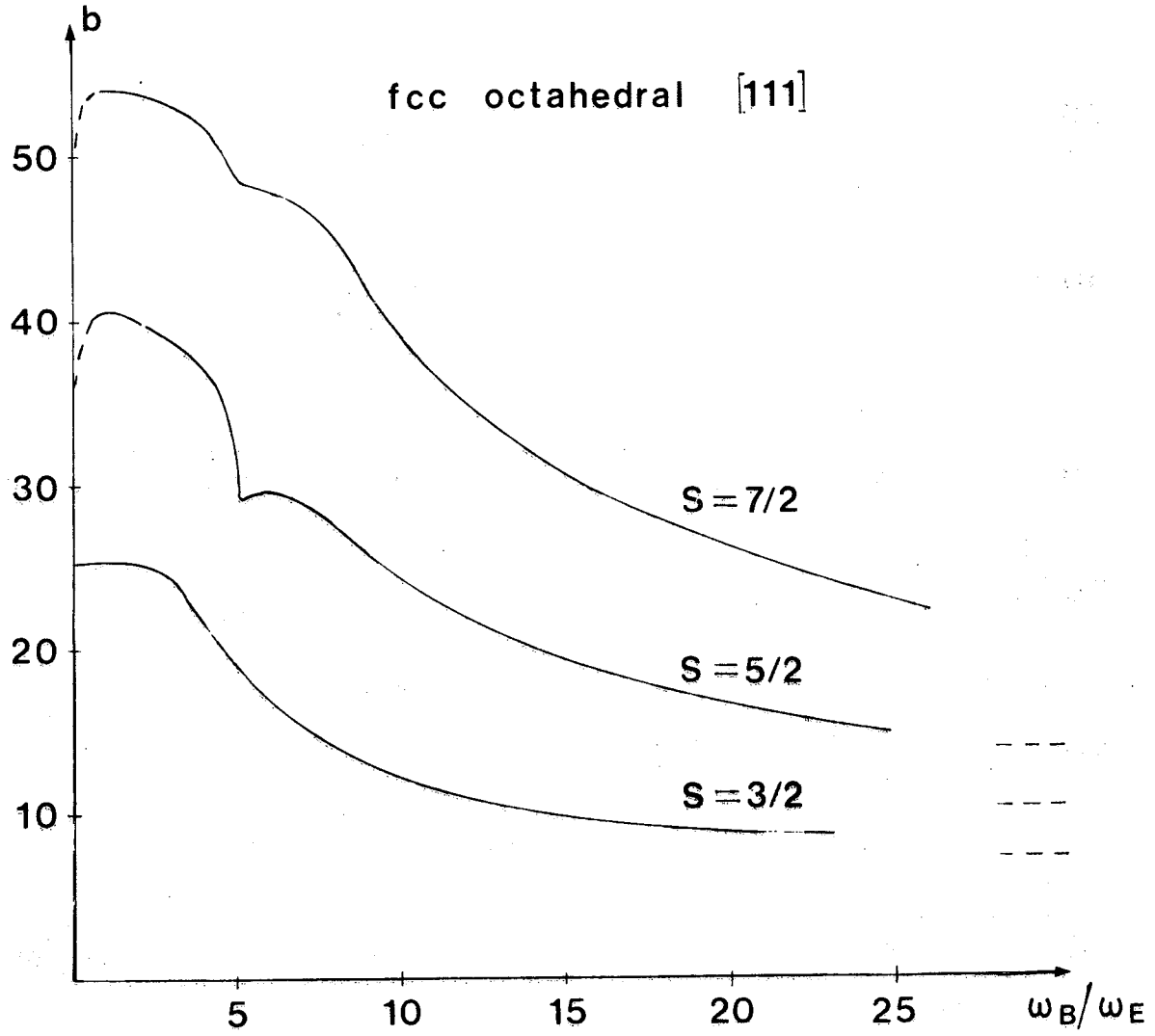
b
fcc octahedral [111]
50
40
30
20
10
S = 7/2
S = 5/2
S = 3/2
5
10
15
20
25
ω_B/ω_E

Fig. 5